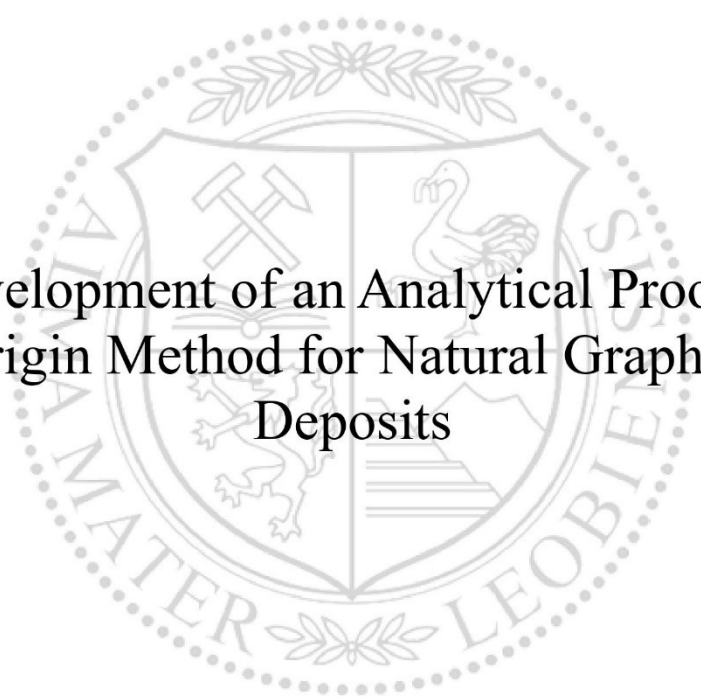




Chair of Geology and Economic Geology

Doctoral Thesis



Development of an Analytical Proof of
Origin Method for Natural Graphite
Deposits

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AFFIDAVIT

I declare on oath that I wrote this thesis independently, did not use any sources and aids other than those specified, have fully and truthfully reported the use of generative methods and models of artificial intelligence, and did not otherwise use any other unauthorized aids.

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Furthermore, I declare that the electronic and printed versions of the submitted thesis are identical in form and content.

Date 27.05.2024

Signature Author
Valentina Elisa Dietrich

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Abstract

Graphite is considered as a critical raw material due to its essential role in various industrial applications, particularly in the steel, refractories and energy storage sectors. The latter, encompassing the production of lithium-ion batteries, is especially significant, as these batteries are essential to electric vehicles and numerous electronic devices. The escalating demand for lithium-ion batteries has consequently increased the demand for graphite and the interest in the provenance of this raw material. This thesis, as part of a larger initiative, focused on the traceability of graphite as an important battery raw material, aiming to enhance the transparency, reliability and sustainability of complex supply chains.

The study introduces an innovative multi-parameter analytical approach to differentiate graphite concentrate samples from various global locations, including major graphite producing countries. Stable carbon isotopes serve as a primary parameter for distinguishing between carbon sources. Flake and amorphous graphite, originating from organic materials, contrast with hydrothermal graphite. Through this methodology, three distinct sample groups were identified: Ukraine, Sri Lanka and the rest of the world. The Raman spectrum, characterizing the microstructural state of graphite, provides information about the peak metamorphic temperatures during graphite formation and also allows a differentiation between different graphite types. However, the microstructure can be altered by mineral processing activities such as crushing and grinding, impacting the natural structure of the material. This technique enables the distinction of amorphous graphite from flake and hydrothermal graphite types.

Trace element analysis potentially differentiates the origin of graphite on the most detailed, the deposit level. The geographic provenance of individual deposits can be traced through characteristic trace elements (including V, Mo, Cr, Co, Cu, Ga, As, Rb, Sr, Ba and REE patterns). Developing methods for solution-based analysis (such as ICP-MS) was necessary due to the inherent challenges in fully dissolving graphite, given the robust carbon layers. Laser Ablation-ICP-MS offers a less time- and cost-intensive method, though it also required significant method development for sample preparation and calibration. The ablation of very fine-grained compressed materials and the lack of reference and calibration materials presented notable challenges. A significant proportion of trace elements in graphite concentrates are associated with impurities and accompanying mineralogical components. These impurities can be identified and delineated through the application of advanced statistical methods and data processing techniques.

Analytical proof of origin (APO) methods are considered incorruptible, as they directly relate to the chemical composition of the raw material, offering significant potential for verification and control purposes. Unlike methods such as documents, tracers, QR codes and barcodes, which can be manipulated or falsified, APO methods provide a robust means of authentication. The developed APO methodology serves to facilitate the implementation of supply chain laws and battery regulations within the graphite industry.

Zusammenfassung

Graphit gilt aufgrund seiner bedeutenden Rolle in verschiedenen industriellen Anwendungen, wie der Stahl- und Feuerfestindustrie, als wichtiger und auch kritischer mineralischer Rohstoff. Die aktuell signifikanteste Anwendung dieses Rohstoffs liegt aber in der Batterieerzeugung. Graphit ist eine entscheidende Komponente in Lithium-Ionen-Batterien, die weit verbreitet in Elektrofahrzeugen und anderen elektronischen Geräten eingesetzt werden. Die Nachfrage nach Lithium-Ionen-Batterien steigt rapide an, somit auch die Nachfrage nach Graphit und damit auch das Interesse an der Herkunft dieses Rohstoffs. Eingebettet in ein Projekt zur Rückverfolgbarkeit von Batterierohstoffen zielt diese Arbeit darauf ab, die Transparenz und Zuverlässigkeit komplexer Lieferketten für Graphit zu stärken.

Die Studie entwickelt einen analytischen multiparametrischen Ansatz, um Graphitproben aus verschiedenen globalen Lagerstätten, einschließlich Länder, die wesentlich zur Graphit- Weltproduktion beitragen, zu unterscheiden. Stabile Kohlenstoffisotope dienen als primärer Parameter zur Unterscheidung zwischen den unterschiedlichen Kohlenstoffquellen und Graphittypen. Flocken- und amorpher Graphit, der aus organischen Materialien stammt, kontrastiert mit hydrothermale Graphit. Drei Probenkategorien, die voneinander unterschieden werden können, wurden identifiziert: Ukraine, Sri Lanka und RoW (Rest der Welt). Das Raman-Spektrum von Graphit ist charakteristisch für seinen mikrostrukturellen Zustand und dient als Grundlage für die Berechnung der Metamorphose Temperaturen während der Graphitbildung Die Struktur von Graphit wird jedoch auch durch Verarbeitung (Brechen, Mahlen) verändert und repräsentiert nicht nur die natürliche Struktur des Materials. Mit Raman Spektroskopie kann amorpher Graphit von den anderen Graphittypen (Flockengraphit und hydrothormaler Graphit) unterschieden werden.

Die Spurenelementanalyse kann potenziell die Herkunft des Graphitkonzentrates auf der detailliertesten Ebene, der Lagerstätte, unterscheiden. Der geografische Ursprung einzelner Lagerstätten kann durch charakteristische Spurenelemente (einschließlich V, Mo, Cr, Co, Cu, Ga, As, Rb, Sr, Ba und REE-Muster) analysiert durch induktiv gekoppelte Plasma-Massenspektroskopie (ICP-MS) zurückverfolgt werden. Die Entwicklung von Methoden für lösungsbasierte Analysen war aufgrund der Herausforderungen bei der vollständigen Auflösung von Graphit, bedingt durch die robusten Kohlenstoffschichten, notwendig. Laser Ablation-ICP-MS bietet eine weniger zeit- und kostenintensive Methode, erforderte jedoch ebenfalls eine signifikante Methodenentwicklung für die Probenvorbereitung und Kalibrierung. Die Ablation von sehr feinkörnigen komprimierten Materialien und das Fehlen von Referenz- und Kalibrierungsmaterialien stellten erhebliche Herausforderungen dar. Ein erheblicher Anteil der Spurenelemente in Graphitkonzentraten ist mit Verunreinigungen und begleitenden mineralogischen Komponenten verbunden. Diese Verunreinigungen können durch die Anwendung fortschrittlicher statistischer Methoden und Datenverarbeitungstechniken identifiziert und abgegrenzt werden.

Analytische Herkunftsnachweismethoden (APO) gelten als manipulationssicher, da sie sich direkt auf die chemische Zusammensetzung des Rohmaterials beziehen und ein erhebliches Potenzial für Verifikations- und Kontrollzwecke bieten. Im Gegensatz zu Methoden wie Dokumenten, künstlich hinzugefügte Materialien oder Tracer, QR-Codes und Barcodes, die manipuliert oder gefälscht werden können, bieten APO-Methoden eine robuste Möglichkeit zur Authentifizierung von mineralischen Rohstoffen. Die entwickelte APO-Methodik dient zur Unterstützung der Implementierung von Lieferkettengesetzen und Batterieregulatorien in der Graphitindustrie.

List of Abbreviations

Abbreviations

AFP	Analytical Fingerprinting
Ag	Silver
Al	Aluminium
APO	Analytical Proof of Origin
As	Arsenic
Au	Gold
B	Boron
Ba	Barium
BAM	Bundesanstalt für Materialforschung
Be	Beryllium
Bi	Bismuth
C	Carbon
Ca	Calcium
Cd	Cadmium
Ce	Cer
CERA	Certification of Raw Materials
Co	Cobalt
Cr	Chromium
CRM(s)	Critical Raw Material(s)
Cu	Copper
DNA	Deoxyribonucleic acid
DRC	Democratic Republic Congo
Dy	Dysprosium
EIT-RM	European Institute of Innovation & Technology – Raw Materials
EMPA	Electron Micro Probe Analyzer
Er	Erbium
ETAAS	Electro Thermal Atomic Absorption
EU	European Union
Eu	Europium
FAAS	Flame Atomic Absorption Spectroscopy
Fe	Iron
FI-CVG-AAS	flow-injection chemical vapor-generation atomic absorption spectrometric
FTIR	Fourier-Transform Infrared Spectroscopy
Ga	Gallium
Gas-MS	Gas-mass Spectrometry
Gd	Gadolinium
Ge	Germanium
GTK	Finnish Geological Survey
H	Hydrogen
He	Helium
Hg	Mercury
HHI	Herfindahl-Hirschman Index
hhXRF	Handheld X-ray Fluorescence
Ho	Holmium
ICP-MC-MS	Inductively-Coupled Plasma Multi Collector Mass Spectrometry

ICP-MS	Inductively-Coupled Plasma Mass Spectrometry
ICP-OES	Inductively-Coupled Plasma Optical Emission Spectrometry
INAA	Instrumental Neutron Activation Analysis
ISMC	Iberian Sustainable Mining Cluster
ISO	International Organization for Standardization
km	Kilometer
KSD	Kolmogorov–Smirnov Test
La	Lanthanum
LA-ICP-MS	Laser Ablation Inductively-Coupled Plasma Mass Spectrometry
LA-MC-ICP-MS	Laser Ablation Multi Collector Inductively-Coupled Plasma Mass Spectrometry
LA-QQQ-ICP-MS	Laser Ablation Triple Quadrupole Inductively-Coupled Plasma Mass Spectrometry
LDA	Linear Discriminant Analysis
Li	Lithium
LIBS	Laser-Induced Breakdown Spectroscopy
LOD	Limit of Detection
LOD	Limit of Detection
LOQ	Limit of Quantification
LRM	Linear Regression Model
Lu	Lutetium
Ma	Million Years
MaDiTraCe	Material and Digital Traceability of Critical Raw Materials
MC-ICP-MS	Multicollector Inductively-Coupled Plasma Mass Spectrometry
Mg	Magnesium
MIC	Microwave Induced Combustion
MLA	Mineralogy
Mn	Manganese
Mo	Molybdenum
MW	Microwave
n	Number
na	Not Attributable
Na	Sodium
Nd	Neodymium
Nda	No Data Available
Ni	Nickel
O	Oxygen
OECD	Organisation for Economic Co-operation and Development
P	Phosphorous
Pb	Lead
PCA	Principal Component Analysis
Pr	Praseodymium
Rb	Rubidium
REE	Rare Earth Element
RFID	Radio-Frequency Identification
RJC	Responsible Jewellery Council
S	Sulfur
Sb	Antimony
Sc	Scandium

Se	Selenium
SEM	Scanning Electron Microscope
Si	Silicon
Sm	Samarium
Sn	Tin
Sr	Strontium
T	Temperature
Tb	Terbium
Te	Tellurium
TGA	Thermogravimetric Analysis
TGC	Total Graphitic Carbon
Th	Thorium
Ti	Titanium
TIMS	Thermal Ionization Mass Spectrometry
Tl	Tellurium
Tm	Thulium
U	Uranium
UN	United Nations
US	United States
USP	Unique Selling Point
V	Vanadium
VOC	Volatile Organic Compound
VPDB	Vienna Bee-Dee Peleminite
W	Tungsten
WGC	World Gold Council
WMD	World Mining Data
WP	Work Package
wt%	Weight Percent
XRF	X-ray Fluorescence
Y	Ytterbium
Yb	Ytterbium
Zn	Zinc
Zr	Zirconium

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1. Introduction

This introduction provides background information on the involvement in projects related to the traceability of mineral raw materials, the choice of graphite as the research material for the PhD thesis and the subsequent continuation of the research through the current MaDiTraCe project. It also emphasizes the importance and potential impact of traceability in the raw materials industry and sets the stage for the subsequent chapters, where I will provide details of the research and present the findings.

The thesis can be traced back to the project CERA4in1 (Certification of Raw Materials) project, which received funding from the EIT Raw Materials program. The Chair of Geology and Economic Geology assumed responsibility for work package 2, which involved the evaluation and development of proof of origin methods for mineral raw materials. Although the project commenced in early 2017, my involvement began towards the end of 2019 when I joined as a project staff member at the Chair of Geology and Economic Geology. At the beginning of 2022, thanks to a grant from the BMF (Bundesministerium für Finanzen), obtained through the RIC (Resources Innovation Center) at Montanuniversität Leoben, I was able to resume the development of methods for analytical proof of origin.

My personal motivation stems from my profound interest in the possibility of traceability for raw materials based on attributes directly related to the raw material itself, such as trace elements or isotope ratios. I strongly believe that this research topic will gain significant relevance in the coming years, as European and international legislation is rapidly emerging, necessitating compliance from companies that often lack solutions. Even industry giants are establishing departments for sustainable raw material procurement, employing geologists and sociologists together to tackle these issues. Thus, it becomes imperative for research to support industry and pave the way forward, as scientists should serve as thought leaders.

Returning to the research topic, the traceability of raw materials, particularly graphite, remains largely unexplored. In the first stage of the development of a proof of origin for graphite, the focus is on distinguishing between different economically significant graphite deposits distributed worldwide. The scope is global. Only once the deposit and the correlating traded product, namely the graphite concentrate, is identified, one can proceed to trace the processed raw materials along the whole supply chain.

This thesis primarily addresses the initial stages of the supply chain, specifically mining and the resulting processed product, known as concentrate. The raw ore corresponding to each concentrate was not available for every economically relevant graphite mine. Hence, the analysis primarily involves concentrates, which undergo basic processing steps such as grinding, milling and flotation to separate the ore from the surrounding rock. In this initial step, no chemical treatment of the ore takes place, making graphite an interesting material for analytical proof of origin, as there are minimal chemical reactions involved in the first steps.

Several parameters were utilized to differentiate between various graphite deposits, including carbon isotope ratios, trace elements, grain sizes and crystallinity. These parameters were determined through a preliminary study, conducted in 2021.

In this thesis, a comprehensive examination of the issues related to the certification of raw materials has been worked on, focusing not only on the broader context of raw material supply chains and the issues related to it, but also delving into the specific case of fingerprinting graphite. By addressing the challenges faced, developing and describing analytical methods, processing and documenting the outcomes of the research, this thesis aims to contribute to the understanding and development of proof of origin methods for mineral raw materials and provides an outlook and recommendations for advancing research on tracing graphite origins.

2. Literature Review on Existing Proof of Origin Methods and (Raw) Materials

Methods for traceability for non-mineral (raw) materials, such as food and other organic substances are presented, as well as other applications of traceability. Research on fingerprinting has gained importance, especially in the last two decades and in the last years many initiatives and research projects had been initiated. A lot of further publications, especially on critical raw materials and raw materials needed for the Green Transformation, can be expected in the coming years. The chapter highlights the evolving landscape of research, new initiatives and the increasing focus on critical raw materials and materials required for the Green Transformation. Most of the literature review on mineral commodities had been done for a deliverable for the CERA project and has now been supplemented with recent work on mineral commodities. The list and descriptions from the CERA report were published in 2021 in the article “Analytical Proof of Origin for Raw Materials” (Melcher et al. 2021). It should be noted that the list of selected non-mineral raw materials is not exhaustive and shows only prominent examples and serves as an inspiration for the current work.

2.1. Exploring the Origins of this Research Topic: Archaeometry - A Multidisciplinary Branch of Science

2.1.1. Definition

Before going into more detail about analytical proof of origin methods, the origin of analytical provenance must first be clarified. It lies in another research discipline, namely archaeometry. Archaeometry encompasses the development and also the use of scientific methods to solve cultural-historical questions. This includes the interdisciplinary participation of the biological sciences, chemistry, earth sciences and physics from the natural science side and the archaeological disciplines, art history, historic preservation, and restoration from the cultural history side (GNAA – ARCHAOMETRIE e.V 2024). In addition to important methods for age determination, such as the ¹⁴C or radiometric age determination, there are a number of other applications of the methods for the proof of origin on finds from earlier times. Common finds represent coins, e.g. gold, silver or copper, as well as ceramics or glasses, tools or swords.

2.1.2 Examples

The origin of gold found on plaques for swords from the Early Bronze Age in Saxony-Anhalt was compared to naturally occurring gold from Cornwall. Trace element analysis was carried out using LA-ICP-MS (Co, Ni, Pd, Ag, Sn and Sb) and showed similar signatures to the naturally occurring gold from the Cornon river gold deposit. This important finding would thus prove the close cultural relationship between the British Isles and Germany in the early Bronze Age and a development of trade (Ehser et al. 2011).

Trace element and Pb isotopic analyses in Celtic Gold Coins were carried out to explore whether there is a connection between gold deposits and Celtic gold coins. Important findings from this are that any

alloying addition to a gold coin makes it impossible to assign the sample to a gold source, since silver, copper, lead and antimony are the main alloying components, but on the other hand they are the main impurities/trace elements of a gold source. Only non-alloyed artifacts can thus be used to assign them to their origin. Furthermore, the overlap of geochemical signatures in placer deposits complicates the the research question (Bendall et al. 2009).

Another prominent example from the connection of geoscientific and cultural-historical research is the origin of the Venus of Willendorf, a small prehistoric figurine, discovered near Willendorf, a town in lower Austria, that remained a mystery since its discovery more than 100 years ago. Chronologically, the Venus Oolite could be assigned to the Mesozoic, based on the sampling of many other Oolite Limestone occurrences in Europe within a radius of 2500 km from the site of discovery. The grain size distribution of the natural occurring rocks is compared to an existing database (analysis of oolite from up to 2500 km distance from site of discovery) to proof the origin, which resulted to be the region around Lake Garda in Italy (Weber et al. 2022).

Pre-historic and cultural interactions during the Neolithic in Germany were analyzed by determining the provenance of amphibolitic raw material used for axes by elemental and Sr, Pb isotopic methods. This combination of geochemical and isotopic data provided a valuable tool for identifying the provenance of stone material used as various tools. Reference samples from Fichtelgebirge in Germany and Jistebsko in the Czech Republic were collected and analyzed before, to compare unknown samples to (Christensen et al. 2006).

The research that emerged from this multidisciplinary branch of science was also transferred to the modern era and one was first faced with questions about the provenance of gemstones (jewelry). The determination of identity, origin (natural or synthetic), geological or geographical origin also describes the value of the gemstone.

2.2. Literature Review on Mineral Raw Materials

2.2.1. Excerpt of Literature Examples

Gemstones are considered one of the first obviously valuable raw materials of human mankind and are therefore briefly described:

Emerald samples from 62 occurrences and 19 different countries were differentiated by using O isotope ratios, which serves as an important parameter to distinguish different deposits. The samples from Russia, Brazil, Madagascar and Zambia however overlap by just using O isotope ratios as a single parameter (Giuliani et al. 1998).

O isotope analyses using laser-fluorination techniques were carried out and combined with gemological parameters, such as color, clarity, cut and others to allow the discrimination between ruby and sapphire from different deposits worldwide (Giuliani et al. 2005).

LA-ICP-MS analyses on tourmalines from three different countries (Nigeria, Mozambique and Brasil) were performed to differentiate their origin by using bivariate and ternary diagrams of Ga, Pb, Cu, Mn, Be, Mg, Zn, Pb and interelement ratios (Abduriyim et al. 2006).

Other raw materials and the analytical methods used are described in detail in (Melcher et al. 2021). Table 1 serves as an overview for existing proof of origin methods for mineral raw materials.

Table 1: Analytical proof of origin methods used for mineral raw material provenance analysis elaborated until 2019

Raw material	Method & Parameters	Regional focus	References
3T (Ta, Sn, W) concentrates	AFP: combination of MLA (mineralogy) and ICP-MS (mineral chemistry)	Central Africa	(Melcher, 2008) ; (Martyna et al. 2018) ; (Gäbler et al. 2011) ; (Gäbler et al. 2013) ; (Gäbler et al. 2017))
3T (Ta, Sn, W) concentrates	LIBS (Mineral chemistry)		(Harmon et al. 2011) ; (Hark et al. 2012)
Gold	LA-ICP-MS (Mineral chemistry)	South Africa, South America	(Dixon 2014) ; (Dixon and Schouwstra 2017) ; (Roberts et al. 2016) (Dixon and Merkle 2019)
Gold	LA-ICP-MS (Mineral chemistry)	W Australia, South Africa	(Watling et al. 1994)
Gold	LA-ICP-MS (Mineral chemistry)	Central Europe	(Schmiderer 2008)
Diamonds, garnet, chromite,	LA-ICP-MS (Mineral chemistry)	worldwide	(Watling et al. 1995)
Diamonds	LA-ICP-MS (Mineral chemistry)		(Dalpé et al. 2010)
Diamonds	Morphology, FTIR, LA-ICP-MS	African countries	(Coney et al. 2012)
Platinum ores and intermediate products	Complex Procedure for Identification of the Nature and the Source of Origin of Precious Metals Containing Products of Mining and Metallurgical Operations	Russia, South Africa	(Perelygin et al. 2008)
Heterogenite	Raman spectroscopy; chemistry, U-Pb dating	Katanga, DR Congo	(Burlet et al. 2011) ; (Decree et al. 2014) ; (Decrée et al. 2015)
Massive sulphide deposits	Microtexture, EMPA	Urals versus Spain	(Machault 2012; Machault et al. 2014)
Uranium ore concentrates, yellow cake	ICP-MS (ore chemistry), TIMS (Pb isotopes)	Australia	(Švedkauskaitė-LeGore et al. 2008)
Uranium ore concentrates	ICP-MS, XRF, TIMS, SIMS	Australia	(Keegan et al. 2008)
Yellow cake	LIBS	worldwide	(Sirven et al. 2009)
Phosphate fertilizers	ICP-MS, MC-ICP-MS, TIMS	worldwide	(Sattouf et al. 2008)
Emerald	¹⁸ O/ ¹⁶ O isotopes	worldwide	(Giuliani et al. 1998; Giuliani et al. 2005)
Marble	C-O isotopes, trace elements, fluid composition	Mediterranean area	(Attanasio et al. 2013) ; (Attanasio et al. 2015) ; (Prochaska & Grillo 2010; Prochaska 2013)
Vermiculite	Chemical composition by INAA, XRF	USA versus foreign materials	(Wright & Palmer 2008)
Gemstone (tourmaline)	Trace elements (LA-ICP-MS)	Brazil, Nigeria, Mozambique	(Abduriyim et al. 2006)

2.2.2. Recently Published Work on Analytical Proof of Origin Methods

The effectiveness of laser-induced breakdown spectroscopy (LIBS) as a portable tool for quantifying the silver content in natural gold was examined. The study involved the analysis of commercial gold alloys, natural gold and unidentified samples, which were successfully matched to their respective origins by referencing a comprehensive database (Pochon et al. 2020).

A three-step approach was developed to identify illicit gold from French Guiana a conflict-affected area with widespread illegal mining. In the first step they revealed Hg amalgamation with either optical microscopy or handheld methods such as LIBS. The Ag content of a gold grain population referenced against KSD statistics provides information about the origin. Also, minor and trace elements assessed by LA-ICP-MS together with the use of a multivariate statistics were used as a robust tool to differentiate between different deposits in almost 70 % of the investigated cases. The remaining 30 % of the samples cannot be differentiated due to their geological similarity to other populations (Pochon et al. 2021).

Diamonds can be certified by distinguishing between two specific types of spectral features, namely elemental emission peaks and emission clusters of C-N and C-C molecules analyzed by laser-induced breakdown spectroscopy and emission peaks. The origin signal is directly contained in the carbon structure. C-N molecule emission peaks are associated with the N concentration of the diamond. The molecular C-C intensities fluctuate based on the bond strength between carbon atoms and the presence of elemental impurities (McManus et al. 2020).

The use of Li isotopy as a fingerprint for Li in raw ores and intermediates was also investigated. Although Li isotopy seems to change during beneficiation, it remains a reliable tool for determining origin and paves the way for a new method to ensure the certification of lithium in lithium-ion batteries. Although the Li isotope primarily only allows the distinction of the deposit type, namely brines and spodumene, certain deposits or geographic origins can still be distinguished from each other (e.g. South America and Australia) (Desaulty et al. 2022).

2.3. Traceability Methods for Non-Mineral Raw Materials Such as Food, Organic Substances and Beyond

2.3.1. Examples

Not only mineral raw materials, but also other products of our daily life are subjected to the analytical proof of origin.

Numerous research investigations have focused on the analysis of pumpkin seed oil. A notable study aimed to differentiate and categorize the geographical origins of pumpkin seeds sourced from Austria, China, and Russia. Their approach involved the measurement of stable isotope ratios, namely hydrogen, carbon, and nitrogen, as well as the evaluation of trace elements, particularly rare earth elements (REE). By employing a combination of two or more parameters, this methodology enables a differentiation among specific localities and, crucially, distinguishes between domestically produced and foreign-sourced materials (Bandoniene et al. 2013).

Another investigation was conducted on the incorporation of rare earth elements (REE) into tomato plants. Dissolved forms of REE were added to the substrate of tomato plants and the uptake of REE by the plants and their subsequent incorporation into the fruit (tomato) was analyzed. Remarkably, even when administered in very small quantities and for a short duration (rather than the entire growing season), the REE exhibited significant labeling effects on the tomatoes (Bandoniene et al. 2018).

REE patterns, specifically terbium and thulium, had been examined in eggs and chicken products, such as meat and bones. To achieve this, laying hens were provided with a controlled diet containing low

levels of REE. The researchers successfully achieved REE labeling in all sample types through the use of spiked chicken feed, demonstrating the effectiveness of this approach. The REE analysis was performed using both conventional techniques and laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) (Bandoniene et al. 2018).

The provenance of flower bulbs was assessed through the correlation of isotope fingerprints and chemical fingerprints. Specifically, the study focused on the analysis of $^{12}\text{C}/^{13}\text{C}$, $^{14}\text{N}/^{15}\text{N}$, and $^{16}\text{O}/^{18}\text{O}$ isotopes to differentiate between materials originating from Europe and China. The investigation successfully achieved this separation, highlighting the efficacy of isotope fingerprinting in determining the geographic origin of flower bulbs. Additionally, the study also employed volatile organic compound (VOC) patterns to further distinguish between different groups. Notably, the VOC patterns allowed for the classification of three distinct groups: flower bulb materials from The Netherlands, other European Union (EU) countries, and China. This comprehensive analysis of both isotope and chemical fingerprints proved instrumental in discerning the origin of flower bulbs with enhanced precision (van Ruth and Visser 2015).

Published proposals for proof of origin methods do not always contain analytical methods only. Often, artificially added tracers or even managerial instruments, such as documents are also used as proof of origin.

Technical, managerial and environmental key issues were defined that need to be considered in the food supply chain and propose a set of technical instruments for traceability. Their research proposes a range of technical tools for traceability. Various technologies, including alphanumeric codes, barcodes, radio frequency identification tags (RFID), and Wireless Sensor Networks, were analyzed. However, it was determined that none of these methods offers comparable efficacy to analytical fingerprinting, which enables the examination of the authentic DNA of the product. In this study, artificial tracers are utilized as "fingerprints" to facilitate accurate traceability within the food supply chain (Aung and Chang 2014).

Isotope analysis was proposed as a parameter for food traceability, while pointing out that advancing technology also brings enormous benefits for food traceability, including increased efficiency and effectiveness. Wireless monitoring devices such as RFID (radio-frequency identification) have improved enormously, but none of these methods is capable of meeting all the requirements of the heterogeneous food supply chain. New advancements such as DNA barcoding can fill some of the gaps, but are prohibitively expensive for many agents of the food chain (Badia-Melis et al. 2015).

An analysis of the impact of traceability concepts on contemporary supply chain management was conducted. The study primarily focuses on two key aspects: firstly, exploring the influence of traceability concepts on supply chain management practices, and secondly, discussing future trends and perspectives in this field. The authors emphasized that the increasing consumer demand for food products with specific attributes, such as ethical sourcing, organic production, and reduced carbon footprint, necessitates the implementation of robust and efficient traceability systems. To achieve this, they suggest integrating traceability techniques with complementary tools like Hazard Analysis and Critical Control Point (HACCP) and production planning. This integration would enhance the effectiveness and reliability of traceability systems, thereby meeting the evolving demands of the modern food supply chain (Dabbene et al. 2014).

3. The Analytical Proof of Origin

3.1. Traceability: Definitions and Perspectives

Given that the term **traceability** will be frequently referred to in this study, it becomes imperative to provide a clear and precise definition for this term (Table 2). In essence, traceability is intrinsically linked to the availability of information. However, it is crucial that this information is generated at the outset of the value chain, whether it is related to mineral resources, food products, or any other goods. The information is subsequently traced throughout the supply chain, ultimately reaching the end of the chain, ideally visible to the consumer in the final product. Thus, access to information becomes vital in achieving traceability, facilitating informed decision-making processes, which are highly valued. There is an increase in the need to know more about the where and how (raw) materials of our daily lives were mined but also as a result of intervention by regulators, such as the European Union, who are now mandating more transparency in supply chains than ever.

Another term associated with traceability is **provenance**. Provenance and traceability are not the same. Transparency and trust are not simply interchangeable. Trace and track are not the same. Provenance describes a single point of origin, while traceability is the ability to trace back. Arguably, the associated **transparency** refers to the familiar chronological proof of ownership, also known as chain of custody. All of these devices of transparency of a supply chain refer to shared information to various stakeholders in the supply chain.

3.1.1. Delimitation of This Study with Respect to the Above-Mentioned Terms

In this graphite provenance study, samples were assigned only to their origin. The verification or determination where the samples came from was made by analyzing the sample (concentrate) itself. This does not provide any information about traceability. Describing the proof of origin as traceability is fundamentally misleading and confusing for stakeholders looking for solutions to specific supply chain needs and problems. Nevertheless, knowing the origin also lays the foundation for traceability purposes.

What is the purpose of this study? Track? Trace? Verify? It is a verification of the origin of the sample (intermediate product/concentrate). No tracking and tracing were conducted in this study. But still, it is an important feature to limit the risk for consumers, but also for any retailer and also supplier. Ensuring transparency, traceability and resilience within our supply chains, necessitates the establishment of a comprehensive infrastructure.

3.1.2. Definitions and Literature

Table 2: Different definitions for traceability, listed from oldest to most recent

Author and Year	Definiton
(Moe 1998)	Traceability is the ability to track a product batch and its history through the whole, or part, of a production chain from harvest through transport, storage, processing, distribution, and sales.
(Lamming et al. 2001)	Transparency is translucent with limited exchange, transparent with full and free exchange; two-way exchange of information and knowledge between customer and supplier. Certain level of transparency is needed for achieving traceability.

(Jansen-Vullers et al. 2003) Traceability can be viewed in a passive and in an active sense based on its use. In the passive sense, traceability provides the visibility to where items are at all times and their disposition. But in an active sense, the on-line tracking information is additionally used to optimize and control processes in and between the different links of the supply chain in addition to keeping historical records by means of recorded identification.

(Skilton & Robinson 2009) Supply chain transparency, defined on a network level as a state or mode of being, is the extent to which information about sources, processes, and relationships is readily accessible to counterparties in an exchange and to outside observers.

(Olsen & Borit 2013) The ability to access, through recorded identifications, all information related to the object under consideration throughout its life cycle. They pointed out the difference between having traceability and verifying the claims in a traceability system by analytical tools and methods. This includes systematically recording properties and some of these can be verified by analytical tools and methods.

(van Ruth and Visser 2015) Authenticity of origin by analytical tools

(Souali et al. 2017) Traceability is the ability to maintain a detailed history of all activities and changes that a particular object may experience throughout its life cycle, taking into account the various relationships that may occur. This particular object can be a material, a product, a model, or even a class in a software development platform.

3.2. Does Provenance Matter? Exploring Consumers Perspectives, Regulatory Frameworks and Governmental Impact

Consumers simply lack access to information regarding the mineral raw materials contained in products they purchase, limiting their ability to make informed decisions. There is a growing demand for visibility into the movement of material throughout the entire supply chain. From the mine to the processing plant, factory, customer and beyond, there is an interest for traceability at every stage. The information generated at the very beginning of the supply chain, namely provenance must be verified, traced throughout the supply chain and shared with relevant stakeholders, which also includes customers in the end. At least since the collapse of many supply chains as a result of the Covid-crisis and the Russian war against Ukraine, the interdependence of the world economy has reached the awareness of the broad public society. Both of these miserable events shifted the focus to supply chain resilience. The systems had shown to be fragile and not resilient at all. This realization has raised concerns about consumer trust, social license and the freedom to operate. The scarcity of essential raw materials due to supply bottlenecks during these events highlighted interest in the origin of consumer goods and the provenance of raw materials in general. Furthermore, global production has undergone significant changes, with various industries being distributed across different countries within global production networks. This necessitates the establishment of global rules for companies, which will be addressed by future supply

chain laws. Governments and regulators are already acting as society's interest in responsible raw material extraction grows and legislative pressure increases (Dietrich and Melcher 2022).

The EU Conflict Minerals Regulation, based on the OECD Due Diligence Guideline (OECD 2016), applies globally and imposes supply chain obligations on importers of tin, tantalum, tungsten and gold sourced from conflict-affected and high-risk areas. The US Dodd-Frank Act, since 2010, requires disclosure and reporting from US listed companies using specific raw materials from the Democratic Republic of Congo (DRC) and neighboring countries (Dodd-Frank Wall Street Reform and Consumer Protection Act). The European Supply Chain Act, effective from 2023, mandates EU companies to address human rights violations and environmental damage in their supply chains, regardless of the raw material involved (ibid.; COM: Proposal for a Directive Of The European Parliament And Of Proposal for a Directive of the European Parliament and of the Council on Corporate Sustainability Due Diligence and amending Directive EU (2019/1937) ing Directive (EU) 2019/1937 2021). The UK Modern Slavery Act, implemented in 2015, aims to eradicate modern-day slavery in business operations and global supply chains (Modern Slavery Act 2015). In December 2020 the EU Commission proposed the EU Battery Regulation, which will replace the Battery Directive from 2006 and includes battery categories such as transport batteries, industrial batteries with a capacity above 2kWh and electric vehicle batteries (The European Parliament and the council of the European Union, battery regulation).

These legal obligations are crucial for prompting company action. More raw materials, especially critical ones, and conflict regions are expected to be included in these regulations. Certification systems and initiatives have emerged in the past 20 years to address these challenges, although they often focus on specific raw materials or segments of the value chain. Harmonization among these systems is urgently needed (Dietrich and Melcher 2022).

3.3. Geo-based Analytical Proof of Origin: Mechanisms, Functionality and Opportunities

The geo-based Analytical Proof of Origin describes analytical/geochemical methods and mathematical and statistical procedures and evaluation methods to proof the origin of (mineral) raw materials. Analytical provenance is also used to verify the origin of raw materials and is a forensic tool that can be used when the origin of a raw material needs to be verified or even when there is doubt about the origin. The result of these analytical methods is a fingerprint, which is ideally not corruptible throughout the entire supply chain and refers to the chemical/mineralogical composition of the raw material itself. Mineral raw materials in general are extracted from a wide variety of deposit types. Each deposit type is to be found in a special geological setting, with special geological properties and features, therefore they inherit unique natural characteristics to the location, deposit or mine from where the raw materials originate.

3.3.1. How? The Approach to Analytical Fingerprinting and Providing the Scientific Evidence

From reviewing literature of existing APO methods for specific raw materials (Melcher et al. 2021) a general work flow emerges that is characterized by analytical procedures and data processing and can be applied in modified forms for different raw materials. However, each raw material has to be treated separately. There is no standardized work flow and recipe for raw materials and the methods to be applied have to be re-evaluated for each raw material. If the availability of samples is given, a distinction must be made between raw ores, rocks, minerals and concentrates. According to this, the further steps of sample preparation may be as follows: crushing, grinding, sizing, preparation of thin sections or polished blocks, dissolution, pellet pressing and others if necessary. Then the analytical workflow must be discussed. This can happen in a separate preliminary study to identify the properties of the material or already emerges from the experience of the processors. The next step is the data evaluation, containing

data reduction and pre-processing, before applying various statistical methods. Different types of data can be generated. The results from analytical work must also be reproducible and a sufficient amount of data must be available, in order to be able to continue working with it and to be able to perform (multivariate) statistics. Following this statistical data evaluation and analysis a reference database containing geochemical and mineralogical properties of known deposits must be established in order to be able to compare future samples with unknown origin to this database. This database needs to be updated and enlarged regularly. APO works only with comparing unknown (daughter/reference) samples to a database (also daughter samples). The reason for the comparison between daughter samples along the same level and not in relation to mother-daughter is the fact that a mother sample would represent all parameters of a deposit. Only some aspects of a deposit can be captured through the daughter sample, never the entire deposit. Reference samples and sample to be identified contain the same level of information. The output is information about the origin and whether it could be identified (referred to a sample in the database) or not.

Figure 1 presents a concept for establishing the geo-based proof of origin through a multi-step approach. The workflow starts with the sample collection, followed by the sample preparation and the application of analytical methods. All of the obtained data is then subjected to data processing, data reduction and various statistical methods, finally leading to a robust database, which serves as the foundation for the determination of the proof of origin. Only samples that refer to the database, can be identified, otherwise they are attributed to an unknown origin.

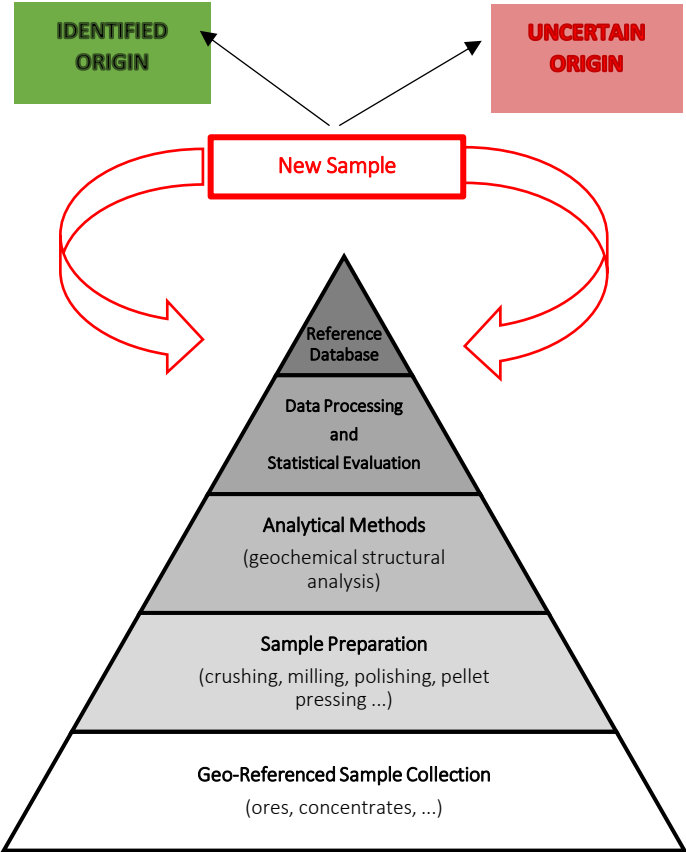


Figure 1: Concept of a geo-based proof of origin

3.3.2. Advantages and Opportunities

The application of APO methods in the mineral resources sector will bring a number of advantages. It is a powerful tool to ensure quality, safety and authenticity.

- Creates trust and enhanced raw material authentication
- Contaminant detection: Detection of undesirable by-products or elements
- Increased consumer awareness and informed-decision making for consumers: Consumers can choose products based on their budget or preferences, supporting local or sustainable companies and avoid regions with potential concerns on e.g. environmental or ethical issues, which fosters a more conscious marketplace in general
- Increased supply chain transparency and resilience: By verifying the product origin it becomes possible to establish a clear chain of custody from the mine (source) to the consumer and serves as a quality assurance
- Marketing Opportunity and USP (Unique Selling Point): The quality and distinctiveness associated with a special geographic origin can be a very powerful selling point
- Regulatory compliance: Meeting the needs from political/governmental measures
- Process optimization and quality control: manufacturing processes can be optimized by identifying critical parameters that affect the quality of the (intermediate) product
- Strong market interest and security for producers: APO helps to preserve the economic value of regionally specific raw materials/products, it allows producers to help to defend their reputation
- Sustainability aspects along the supply chain can be considered (knowing where the product comes from, can also estimate how much CO₂ it has emitted and under which conditions it has been mined and/or produced)

3.3.3. Limitations, Challenges and Pitfalls

Technical Challenges:

- Time-consuming procedure, if complex laboratory methods have to be applied: Sample preparation for analysis (e.g. solution analysis) is usually time-consuming, complex and expensive
- Equipment, expertise (trained personnel) and resources can be very expensive especially in underdeveloped regions or for small-scale producers
- Lack of Harmonization: Certified lab techniques and devices, reference databases and standardized lab protocols need to be widely available for a widespread acceptance
- Inconsistencies across different countries and labs: Establishment of certified laboratory facilities (analytical devices and staff); modern and well-equipped technical laboratory facilities must be available

Obstacles:

- Host frameworks need to be developed (harmonization and standardization)
- In early stages, with limited awareness, the benefits might be minimal (cost/use ratio); persuasion needs to be done (regulatory impact from governments might help)
- Most traceability solutions until now are based on digitalization, such as blockchain technologies
- A database needs to be built for each raw material, i.e. samples can only be identified by correlation to a reference sample.

Limitations and Pitfalls:

- The APO has only been developed for a few mineral raw materials (Table 1). There may also be no natural way of tracing for some raw materials due to various aspects, such as chemical processing in an early stage of the supply chain.
- Altered signatures: post-production treatments and mineral processing can modify or mask the original geochemical signature of a mineral raw material
- Blending of materials: Mixed signatures can hardly be traced back to one certain place of origin
- Overlapping signatures and natural variations: Multiple factors such as weathering, diagenetic processes and host environment influence geochemical signatures of raw materials. These factors are highly variable within a given region or deposit and might cause overlapping signatures.
- Garbage in-garbage out problematics: If poor or even wrong information enters the system, misleading information will come out (e.g. when sample material is not geo-referenced)
- Criminal activity might cause false information
- Access to sample material: Sample material from certain regions may not be accessible
- Equipment and expertise not available at all

To conclude it, a probability might be provided indicating the likelihood that the unknown sample matches a known sample from the database. The higher the number of producers/mine in the database, the higher the probability that an unknown origin of samples can be identified with a high degree of certainty.

3.4. Artificial Tracers: Exploring Innovative Methods for Ensuring Supply Chain Transparency

This study focuses on chemical/mineralogical compositions of mineral raw materials. It does not encompass methods such as labeling (e.g., barcodes, QR codes or RFID), documentary traceability (blockchain), or the use of artificial tracers. Artificial tracers are materials intentionally introduced into a product to enable its traceability. While these alternative methods do exist and possess their own merits, they are susceptible to potential falsification and do not directly relate to the composition of the raw material itself. Based on the chemical/physical properties of the material there are a variety of taggants available, such as chemical or physical taggants. Barcodes and QR codes, RFID tracers or even added REE are incorporated into materials. The use of a combination of blockchain and physical tracking technologies to enable traceability within raw materials supply chains was applied to bridge gaps by increasing transparency in supply chains by combining two different traceability methods (Bacchetta et al. 2021).

3.5. The Application of APO in Research Projects: CERA4in1 and MaDiTraCe

It must be stated, that APO plays a minor role in most certification systems and is not a standard application, but in some cases (with increased demand) it has already become a routine application (especially gold, gemstone and diamonds). The problem with certification schemes for mineral commodities is as follows: There are already a number of certification schemes existing, all of which either target only specific commodities, or are applicable to only part of the value chain. Examples of commodity-specific certification schemes, already on the market are the Bettercoal Code (BetterCoal), applicable to coal materials, or Fair Stone (Fair Stone 2018) and Xertifix (XertifiX 2024) for natural stones and the Aluminum Stewardship Initiative for the aluminum industry (Aluminium Stewardship Initiative 2022). Furthermore, the well-known Kimberley Process for diamonds (The Kimberley Process (KP) | KimberleyProcess 2024) and the RJC (Responsible Jewellery Council 2024) for diamonds and other gemstones. There are also a number of certification systems for gold, such as Fairmined (Alliance

for Responsible Mining) or the WGC (World Gold Council). So far, there is no single certification system that covers all mineral raw materials and all areas of the supply/value chain. Due to the above facts such as the non-harmonization among certification systems, there have been EU-wide efforts to develop and also establish certification systems for raw materials for several years. APO is expected to play a role in both of the projects mentioned below and provide important and trustworthy commodity-specific information. One of these projects was the CERA4in1 project (2017 - 2021). This project aimed to establish 4 sub-standards within one certification system. The four sub standards cover each area of the supply chain from exploration to the final product, as a label, visible to the consumer. These sub standards refer to different areas in the supply chain, which do not have the same requirements for certification, but should nevertheless be included in one system. Thus, each participant in the supply chain should receive an individual solution tailored to the area of the supply chain. CERA4in1 aimed to be the first certification system for raw materials that captures and considers its complexity, making it the first system adapted to all mineral raw materials and to each area of the supply chain.

The 4 sub standards can be described as followed:

- The CERA 4in1 Readiness Standard (CRS) governs the objectives for evaluating a deposit in terms of social and environmental issues during the exploration process.
- The CERA 4in1 Performance Standard (CPS) applies to a production facility or group of production facilities and covers mining, processing and refining.
- The CERA 4in1 Chain of Custody Standard (CCS) refers to the product and establishes criteria to ensure full traceability of responsibly sourced raw materials and traded goods.
- The CERA 4in1 Final Product Standard (CFS) certifies a final product made from raw materials that have been sustainably sourced and traded.

The overall system is supposed to lead to a certification of the entire trade chain of primary and secondary raw materials. The follow-up project is MaDiTraCe (Material and digital Traceability and Certification) (<https://lgi.earth>, LGI Sustainable Innovation - Maditrace 2024). Here, the CERA 4 in1 approach will further be pursued. The main objective of MaDiTraCe is to expand and integrate the portfolio of technological solutions to improve the reliability of critical raw material tracking (CRM) and the transparency of complex supply chains. The project aims to develop and test independent digital and geo-based approaches to CRM traceability and integrate them into a general certification system throughout the mineral supply chain from mine to manufactured and recycled products. The project aims to increase the technical readiness level of experimental or largely untested methods in both digital and materials sciences. Special attention will be given to the complexity of mineral supply chains with points of material aggregation and transformation (processing, refining etc.) including circular economy (recycling). This methodology will allow downstream industries to demonstrate the reliability of their sustainability claims while complying with current regulations (in particular the EU Battery Regulation and the German Supply Chain Act) and anticipating the implementation of future regulations (EU Corporate Due Diligence Directive). MaDiTraCe's fundament is a strong stakeholder process, with upstream and downstream industrial players from mining to manufacturing industry and large networks involved via the consortia (EIT-RM) and clusters (ISMC – Iberian Sustainable Mining Cluster) participating in the project. Continuous interaction with this industrial and policy-oriented stakeholder community on the traceability technology and the certification schemes developed in the project will ensure to stay in line with industrial needs and expectations with respect to regulatory compliance.

3.6. Conceptualization and Analytical Approaches for the Proof of Origin of Graphite

One critical aspect in the determination of the significance of graphite lies in understanding its geographical origin, which can significantly impact its properties and suitability for various applications.

The ability to trace the provenance of graphite has become crucial in today’s globalized market, where sourcing sustainable and ethically-mined materials is eminent. The concept for the proof of origin of graphite should be as robust and as consistent as possible. This approach to the Analytical Proof of Origin is the first ever for an industrial mineral. On the one hand, graphite is a raw material that undergoes only basic processing, i.e. in the first processing steps the raw ore is crushed and ground and then floated, on the other hand it is a very challenging material to work with due to its special properties (e.g. it is very brittle and prone to fracturing). This processing results in graphite concentrates with different concentrations, but mostly >94 % carbon. These concentrates are then passed on, which is the first point where the material is traded in the supply chain and leaves its place of origin. The basic processing happens mostly in or near the mine, as it is not economically viable to transport large quantities of raw ore, along with the surrounding rock. The proof of origin refers mainly to these traded concentrates.

The three-step approach described in this thesis is a powerful tool to proof the geographical origin of graphite. This approach focuses on the analysis of carbon isotope ratios, trace elements and crystallographic parameters found within the graphite sample material. Elements can be qualitatively classified into **major** (>0.4 wt %), **minor** (0.1–0.4 wt %), and **trace** elements (<0.1 wt %) (Lee 2019). Each deposit individually assembles trace elements based on geological processes and thus represents an individual tool to assign respective elements to specific geographic regions. The third factor to be considered are the crystallographic properties of the graphite assessed by Raman spectroscopy. (Rantitsch et al. 2016a) showed the differentiation of semi graphite and graphite by Raman spectroscopic investigations. It is a three-step approach whose individual parameters together result in a holistic approach for fingerprinting graphite and is therefore more robust than a one-step approach with the analysis of only one parameter. Each of these steps contributes insights to the material’s history and geological formation, enabling to make informed decisions regarding the origin and authenticity of the graphite. An outlook on how to proceed in the future will be given in the last chapter of this thesis. In summary, as with many examples in the field of proof of origin methods, multi element analytics is the most significant tool for distinguishing the geographic origin of graphite concentrates.

Figure 2 illustrates the three-step workflow for the analytical proof of origin of graphite.

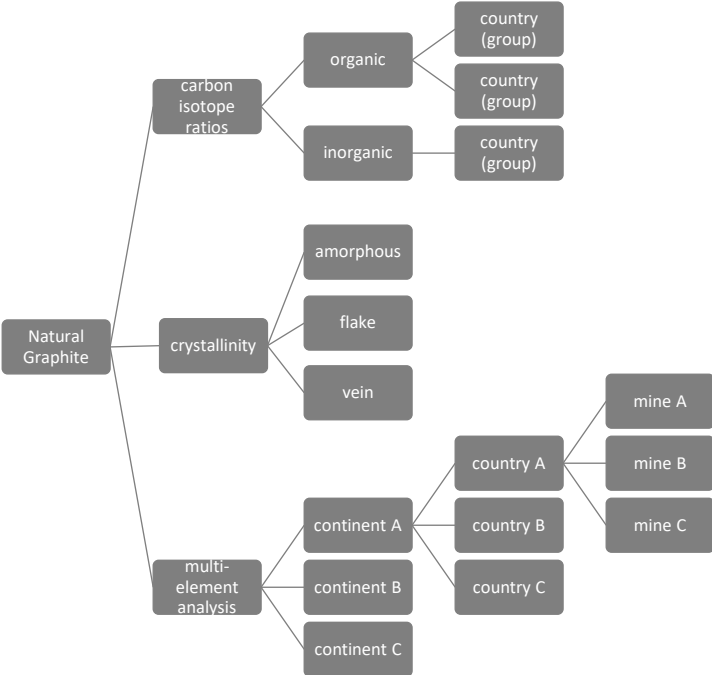


Figure 2: Workflow Analytical Proof of Origin for Graphite

4. Graphite

4.1. Introduction

Graphite, a raw material especially important for the transition to a low-CO₂ economy is used far beyond the well-known use in pencils. There is a large number of applications. The raw material is indispensable in lithium-ion batteries, which are built into laptops, smartphones and electric cars, graphite is the leading material for construction of bipolar plates which are then used in fuel cells, as well as the use in the steelmaking industry. For the production of graphene, graphite is used as electrical and thermal conductor. The rapidly growing market for Li-ion batteries has been driving the demand for graphite, thus increasing interest in the origin of this raw material. There is more graphite used in Li-ion batteries than any other raw material; the estimated demand for lithium for batteries only for e-mobility in 2030 is 32000 t, whereas the demand for graphite is 340000 t by 2030, which is more than 10 times more graphite needed than lithium in a low demand scenario. Another 99000 t of graphite (and 10000 t of Li) are needed in a low demand scenario by 2030 for batteries for renewables, such as solar and wind energy, as well as others for Europe only (Carrara et al. 2023). The use and importance of graphite in Li-ion batteries is often neglected (Tsuji 2022). More than half of the material used in Li-ion batteries is graphite. It can be replaced by synthetic graphite, which however is significantly more expensive and has a higher carbon footprint. Therefore, natural graphite will remain essential. Synthetic graphite is very costly to produce as the precursor material needs to be treated to very high temperatures >2500 °C for several days. The synthetic graphite is manufactured from calcined petroleum coke, coal tar pitch, anthracite or recycled synthetic graphite (Thrane and Kalvig 2019).

The estimated demand for graphite in the EU in 2030 for a low demand scenario is 342448 t/y and 595405 t/y by 2050 for strategic technologies and sectors only. The forecasted demand for graphite in 2030 worldwide is 1239286 t/y (world production 2022 1.7 t/y) and by 2050 9464128 t/y only for strategic technologies and sectors (Carrara et al. 2023). The growing demand for graphite, driven by the widespread application of rechargeable Lithium-ion Batteries (LIBs) into consumer goods and the electrification of mobility, has triggered a keen interest in its provenance.

4.2. World Production

In 2022 around 1,7 Mio tons of natural graphite were extracted worldwide (Reichl & Schatz 2024). Figure 3 showcases the dynamics of the global graphite production over a span of the past 38 years. Fluctuations and trends can be observed throughout the specified period. Technological advancements, economic developments and environmental factors have shaped the graphite sectors evolution.

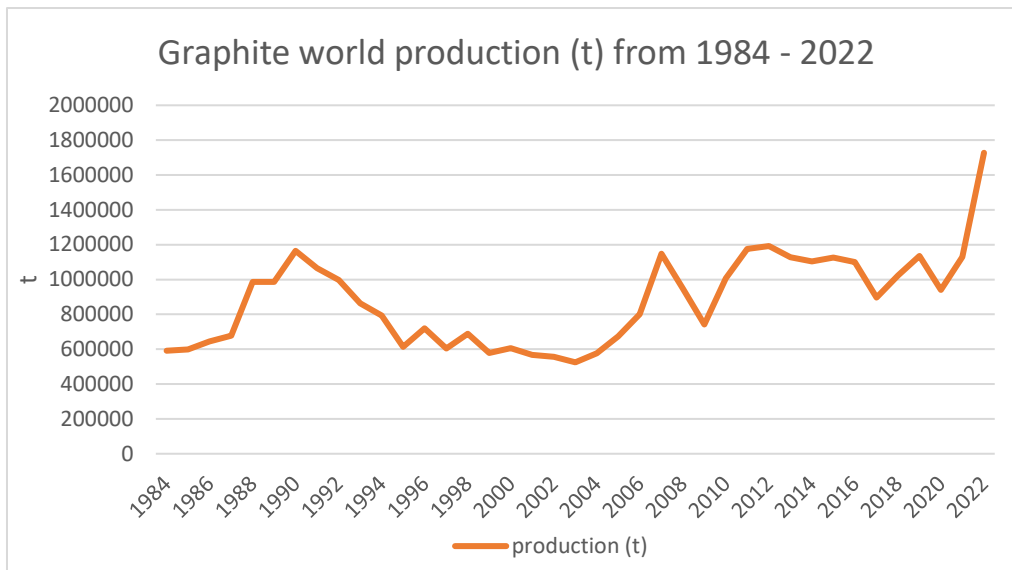


Figure 3: Evolution of graphite world production from 1984 to 2022 (WMD)

The graphite world production reaching its high in 2022 with a decrease in demand in 2009, 2017 and 2020 was due to less demand in the refractories industry. USGS reported a marked decrease from 2002 in the consumption of graphite electrodes, owing to the development of more efficient iron and steel production techniques during the late 1980s. The use of natural graphite in lubrication applications is also decreasing because of changes in requirements for lubricants and in processing technologies (Kalyoncu 2002; Olson 2009; Robinson et al. 2017; Olson 2020; Olson 2022).

4.2.1. Major Producer Countries

The supply of graphite is dominated by China with 67 % of the world graphite production in 2022, followed by Mozambique (9.61 %), Madagascar (6.76 %) and India (4.77 %). Other graphite producing countries (as seen in Table 3) have a share of 1 % or less of the world production, except Brazil ranking fourth accounting to 4.59 % of the world production. Mozambique as the second largest producing country has gained enormously in importance since 2018 thanks to the world’s largest graphite mine, Balama. The four main producing countries (China, Mozambique, Madagascar and India are responsible for 88 % of the global graphite production (Reichl & Schatz 2024).

An overview of the major graphite producing countries is presented in Table 3, along with their respective tonnages and the global production share (Figure 4). The data has been collected for the most recent period available from WMD 2024 (Reichl & Schatz 2024).

Table 3: Major graphite producing countries and corresponding tonnages (WMD 2024 and USGS 2021 and 2022)

Country	Tonnage 2021 USGS 2021	Tonnage 2022 USGS 2022	Diff 2021-2022 in t	USGS Diff 2022 in %	USGS 2021-Tonnage 2022 WMD 2024	USGS 2022 Global production share in %	WMD 2024 Global production share in %
Austria	500	500	0	0	100	0.04%	0.01%
Brazil	82000	87000	5000	6	79372	6.62%	4.59%
Canada	12000	15000	3000	20	13000	1.14%	0.75%
China	820000	850000	30000	4	1161000	64.64%	67.21%
Germany	250	250	0	0	185	0.02%	0.01%
India	7000	8300	1300	16	82439	0.63%	4.77%
Korea, North	8100	8100	0	0	20000	0.62%	1.16%
Korea, Republic	10500	17000	6500	38	23798	1.29%	1.38%
Madagascar	70000	110000	40000	36	116700	8.37%	6.76%
Mexico	2100	1900	-200	-11	3420	0.14%	0.20%
Mozambique	72000	170000	98000	58	165932	12.93%	9.61%
Norway	6290	10000	3710	37	10380	0.76%	0.60%
Russia	15000	15000	0	0	18800	1.14%	1.09%
Sri Lanka	3000	3000	0	0	3299	0.23%	0.19%
Tanzania		8000	8000	100	x	0.61%	
Turkey	2700	2900	200	7	27715	0.22%	1.60%
Ukraine	10000	3000	-7000	-233	846	0.23%	0.05%
Vietnam	5000	5000	0	0	500	0.38%	0.03%
Total	1126440 t	1314950 t	188510 t	14%	1727486 t	100%	100%

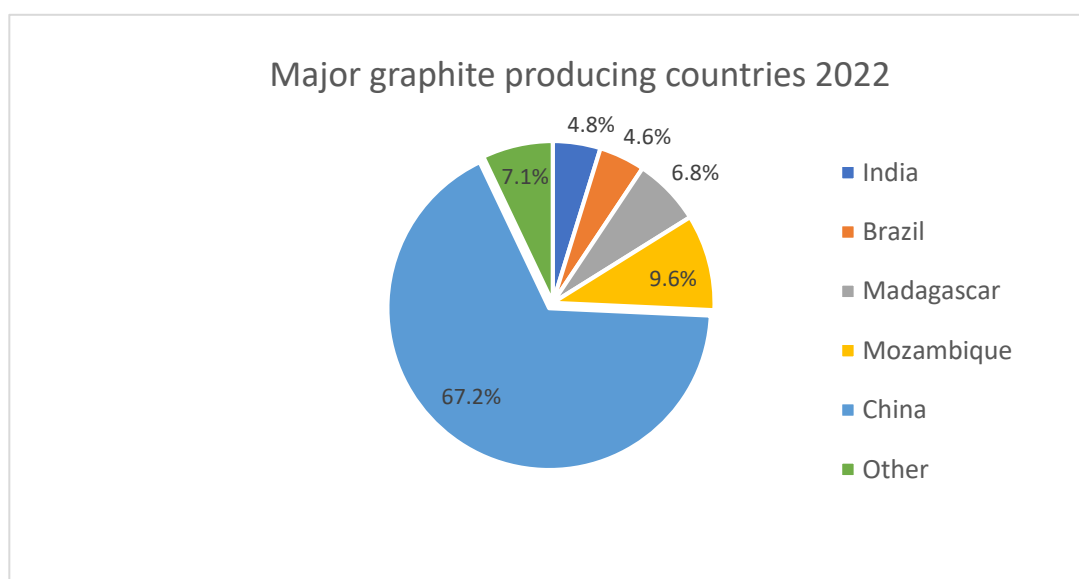


Figure 4: Major graphite production countries in 2022 (WMD, 2024)

4.2.2. Dependence on China and Diversification of Graphite Resources

China continues to maintain its position as the epicenter of the graphite industry as the largest producer, consumer and exporter of graphite on the global scale (Zhou and Damm 2020).

Wang et al. in 2018 in their simulation analysis of the spread of a supply crisis based on the global natural graphite network proposed that in order to minimize the risks of dependence on China, new technologies need to be developed to optimize the utilization of graphite resources and reduce consumption. This is not an easy task. It is advised to adopt another proposal, which advocates diversifying graphite resources instead of solely depending on China. For the market outside China, but

especially in Europe, there is a strong need to not only focus on the cost effectiveness but also on enhancing environmental sustainability and technical capability to reach supply security.

One of the noteworthy graphite mining projects outside China, is the Munglinup Graphite Project in Western Australia, which was being developed by Mineral Commodities Limited (MRC). They also operate the Skaland mine in Norway. The Munglinup project involves several open pits and flotation plants to process graphite ores. It is located in a zone of graphitic schists within a sequence of hornblende and hornblende-garnet gneisses. The latest mineral resources from 2020 prepared following the 2012 JORC Code are estimated to be at 7,99 Mio t and 12,2 % total graphitic carbon content (TGC), using a 5 % cut off and 4,24 Mio t ore reserves at 12.6 % TGC (Mineral Commodities Ltd 2022).

4.2.3. European Natural Mine Production

On a global scale, European graphite production remains constrained, comprising only 0.67 % of the World Production in 2022 (Reichl & Schatz 2024).

Ukraine is the largest producer of natural graphite in Europe. The entire production from Ukraine is coming from a single flake graphite mine operated by Zavalivskiy graphite (Zhou and Damm 2020). Meanwhile Norway is the second largest producer in Europe and the eighth largest producer worldwide, excluding Chinese companies. In 2018, approx. 12.000 t of flake graphite were produced annually with an estimated mine life of 30 years in the Skaland graphite mine. This mine shows an average carbon content of 25 %, making it the world’s highest-grade graphite mine in operation (Mineral Commodities Ltd 2022b). Due to the war in Ukraine the production amount of graphite dropped from 17000 t in 2021 to 846 t in 2022, which accounts to only 0.05 % of the world production in 2022 (Reichl & Schatz 2023, 2024).

Other graphite producing countries in Europe are Germany and Austria with one operating mine only in each country. Scandinavia tries to meet the demand for graphite using European resources and is establishing new mining operations (Table 4, Figure 5). A prominent example is the Vittangi graphite mine in Northern Sweden, which is based on the Nunasvaare graphite deposit and had been assessed by the SGU as a mineral deposit of national interest. In Sweden areas of particularly valuable mineral substances may be declared national interests (SGU, Mineral deposits of national interest 2024) and are therefore protected.

Additionally, the Woxna mine in Sweden is kept in production-ready mode, but is currently not operating (Barrera 2021).

In Finland, Beowulf Mining operates the Aitolampi flake graphite deposit, which is still under exploration and currently undergoing a pre-feasibility study. Beneficiation tests had also been performed (personal communication Rasmus Blomqvist). Graphite bearing rocks in this deposit are strongly foliated black schist and disseminated graphite flakes within banded gneiss (Al-Ani et al. 2020).

Table 4: European (Scandinavian) graphite exploration projects and their published reserves and resources, map see Fig. 5

Project name	Country	Company	Reserves (mt)	Resources (mt)	References
Jalkunen	Sweden	Talga Group Ltd.	na	4.7	(Zhou and Damm 2020; Barrera 2021)
Vittangi	Sweden	Talga Group Ltd	0.45	3.9	
Woxna	Sweden	Leading Edge Materials Corp.	na	0.6	
Raitajärvi	Sweden	Talga Group Ltd	na	0.3	
Aitolampi	Finland	Beowulf Mining	na	26.7	(Beowulf Mining plc 2024)
Skaland	Norway	Mineral Commodities Ltd	1.78	22	(Olson 2021)
Haapamäki	Finland	Beowulf Mining	na	1.3	(Zhou and Damm 2020)

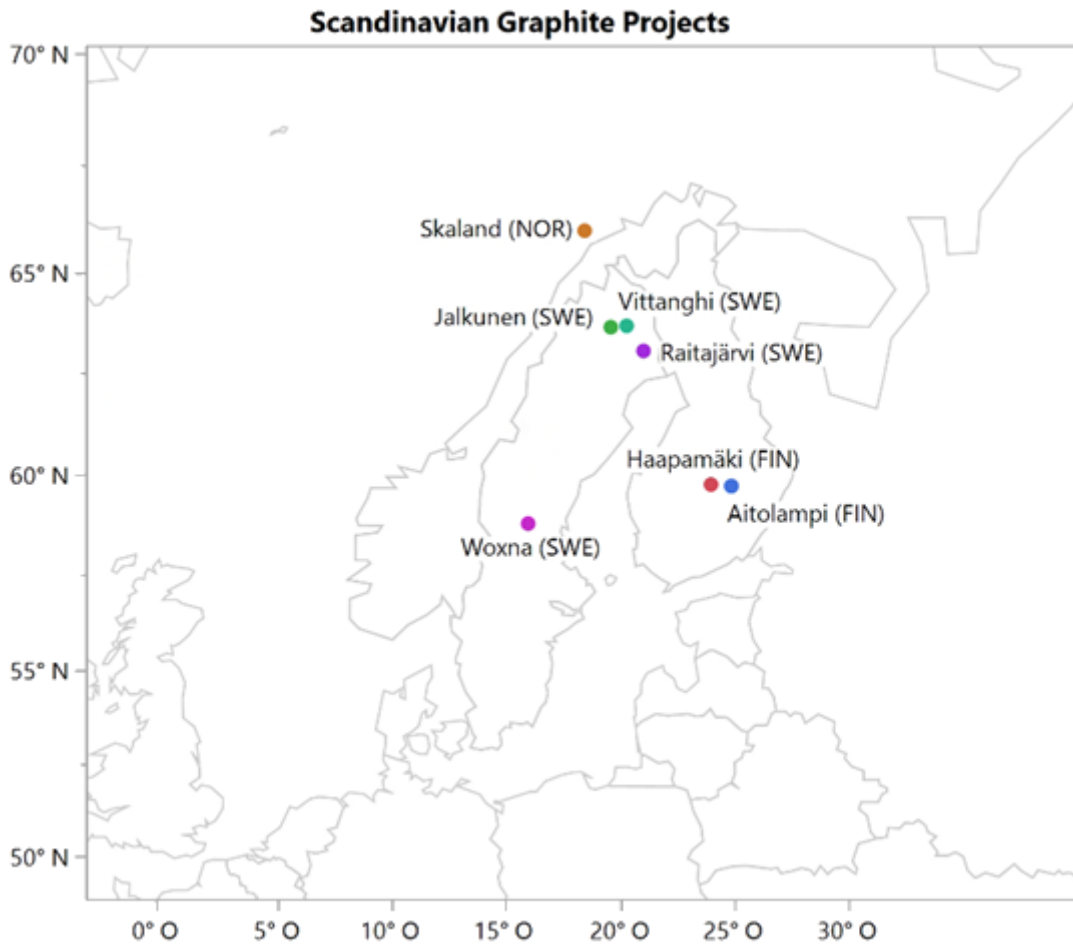


Figure 5: Map of graphite projects in Scandinavia

Graphite beneficiation, which requires a number of hazardous chemicals and acids such as HF, is currently concentrated almost exclusively in China. Other ways of processing and the construction of plants in Europe is in preparation (production and processing facilities are currently planned and tested at the Woxna mine site and at the Vittanghi graphite mine site in Northern Sweden) (Barrera 2021).

Weber et al. (2023) analyzed the likelihood of supply bottlenecks for the graphite market and compared the world mine production of graphite in terms of political stability of the producer countries. The study revealed that 98 % of the global graphite production originates from developing countries, while western industrial nations contribute only 1,8 % to graphite production. Madagascar and Mozambique are classified as developing nations. The largest graphite deposits with the highest carbon contents are located in Africa. Among them the Balama mine (Syrah Resources), which started its production in 2018, as well as projects like Nicanda Hill (Triton) in Mozambique and Anjamanga (PRGGlobal) in Madagascar (Weber 2023a). Additionally, Asia and North America are home to significant graphite deposits. However, European deposits exhibit notably lower resources.

4.2.4. Supply, Demand and Natural Graphite Reserves and Resources

In terms of quantity, graphite is the major component of modern Li-ion batteries and demand will increase dramatically in the coming years. The risks can be located primarily in the high concentration of supply and the market power of China. China controls almost the entire value chain of this important industrial mineral. The amount of graphite required by 2050 is 4 times higher than the current production from 1984 to 2020, partly due to the already mentioned and forecast demand from e.g. electromobility and other green technologies. The green transition will be significantly mineral intensive.

Global graphite reserves are spread across various regions, totaling an estimated 300 million tons in recoverable reserves by 2019. Worldwide, the recoverable graphite resources exceeded 800 million tons in 2020 (Olson 2020). China possessed substantial reserves and resources, reaching 537 million tons in 2018 (Zhou and Damm 2020). In contrast, European producers had a combined 11 million tons in reserves and resources, with reported reserves of 1.05 million tons, making up less than 1 % of the global total. Notably, Norway contributed half of Europe's reserves, reporting 560,000 tons for the Traelen graphite deposit in 2015 (Gautneb 2015).

Reserves beyond China and Europe amounted to 227 million tons, comprising a substantial 76 % of the global reserves (Olson 2020; Zhou and Damm 2020). Among these, Turkey boasts the world's largest graphite reserves, totaling 90 million t, representing 30 % of the global reserves. However, a significant portion of these reserves is composed of the amorphous type of graphite, not suitable for most applications in the EV sector. Brazilian reserves are estimated at 72 million t, earning them the third-largest position worldwide and accounting for approximately 24 % of the global reserves (Olson 2020).

In recent years, East Africa has emerged as a primary hub for graphite exploration, resulting in a notable surge in the reserve and resource levels of flake graphite, particularly in Mozambique, Tanzania, and Madagascar. As of 2019, the region's combined reserves amounted to 44.6 million tons, comprising a significant 15 % of the global reserves (Zhou and Damm 2020; Taylor 2024).

4.2.5. Herfindahl-Hirschman Index (HHI)

The HHI is a widely used and accepted measure of market concentration and thus illustrates the market competition. The closer a market is to a monopoly position, the less competition. 100 % market share of a company/country would correspond to an HHI of 10 000. If, on the other hand, there is a diverse market, the HHI would be 0, indicating perfect distribution (Brezina et al. 2016).

Some ranges are:

- HHI below 1500: a competitive marketplace
- HHI between 1500 and 2500: a moderately concentrated marketplace
- HHI of 2500 or greater: a highly concentrated marketplace

The high HHI of graphite (4707) (Reichl & Schatz 2024) therefore indicates a highly concentrated market place and the risk of bottle necks.

4.2.6. Graphite as a Critical Raw Material (CRM)

The classification of a raw material as critical refers to the importance for the EU economy, growth and jobs and the supply risks we are exposed to, due to the dependence on certain supplier countries. This classification has been launched as the first action of the European Raw Materials Initiative (RMI) in 2008. Therefore, the European Commission has established a list of critical raw materials, which is updated regularly. The very first list was published in 2011 with 14 raw materials. In 2014 the list of CRMs was updated to 20 CRMs and in 2017 the list already consisted of 27 CRMs. The most recent additions to the list include arsenic, feldspar, helium, manganese, copper and nickel. The latter are considered as strategic raw materials rather than critical raw materials. Indium and Natural rubber were removed from the list. Graphite was a CRM from the beginning in 2011 and was quoted as Natural Graphite from 2014 (Grohol et al. 2023).

4.2.7. The Graphite Supply Chain

The graphite supply chain is complex and characterized by many intermediate processing steps. However, one advantage in the processing of graphite is the basic processing at the beginning of the

value chain, where the raw ore is only crushed and ground and the ore is separated by flotation. This graphite concentrate is then traded worldwide. Graphite raw ore is not traded due to the large amount of material and the corresponding transport cost. In some graphite producing regions graphite is processed in one processing plant, however originating from different mines. This is the case for some samples present in this study. Which samples are affected by mixing materials from mines in a close proximity to each other will be explained in detail in later chapters. The following flow chart (Figure 6) shows the graphite processing value chain from exploration and mining through various processing steps up to the final use of the raw material in products.

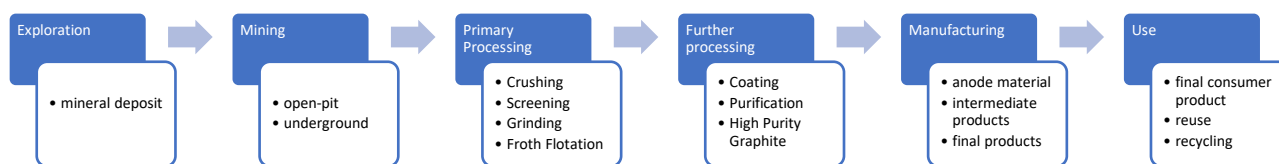


Figure 6: Simplified graphite processing flow chart (modified after Mitchell, 2022)

4.2.8. Major Producer Companies Worldwide

The following companies (Table 5) represent the most important graphite producers worldwide.

Table 5: List of the largest graphite mines worldwide including yearly capacity (DERA 2022)

Company name	Country	Capacity (t/y)
Syrah Resources	Mozambique	350.000
Luobei County Yunshan Graphite	Mozambique	100.000
Aoyu Graphite Group	Mozambique	100.000
Jixi Changyuan Mining	China	100.000
Jixi Pucheng Graphite	China	100.000
Qingdao Haida Graphite	China	100.000
Qingdao Jinhui Graphite	China	100.000

4.3. Understanding Graphite

4.3.1. Material Properties

Graphite is one of two stable crystalline forms of pure carbon. Carbon is a crucial element in the Earth's biosphere and, after oxygen, has the highest mass fraction in all living organisms. In the biosphere, carbon is always found in connection with other chemical elements (O, H, S). In nature, however, carbon also occurs in a completely pure form, namely as the two minerals diamond and graphite. By far the more common of these is graphite. On the Mohs hardness scale, graphite is assigned a value of 1-2 and is therefore among the softest minerals. Diamond, in contrast is the hardest of all. The density of graphite is 2.26 g/cm³. Due to its high melting point of 3.700 ° C, graphite is very heat-resistant and very resistant to acids and alkalis. In addition, graphite also has good thermal and electrical conductivity and reduces the kinetic energy of the neutrons released during nuclear fission (Regelous & Holzförster unknown year). The average carbon content present in the Earth's crust is approximately 0.08 wt % (Clarke and Washington 1924).

4.3.2. Carbon Origin and Graphite Formation

Graphite formation is primarily a result of graphitization e.g. the continuous transformation of organic matter, known as coalification. This process commences from peat, progressing through bituminous coals and anthracite, ultimately ending in the formation of graphite. Consequently, graphite and semi-graphite indicate a moderate to high metamorphic grade. In contrast, natural coke originates from localized elevated heat flow associated with an intrusive body, leading to its presence across a spectrum of coalification stages. Similarly, natural char occurs in various coalification stages as it is produced by the influence of heat from fire on coal or gelified organic matter in peat. These transformations demonstrate the spectrum of geological processes that shape the formation of these carbonaceous materials (Kwiecińska 2004).

The formation of graphite always requires organic or inorganic carbon, as well as high temperatures and pressures. Graphite is formed over the course of millions of years either from biological or hydrothermal sources (Luque et al. 1998; Beyssac and Rumble 2014) from substances initially deposited on the earth's surface on land or in water such as plants or algae (biological formation). The contained carbon (approx. 50 % of the plant or digested sludge) is enriched up to 90 % in the anthracite coal. Up to this point the enrichment process can run under relatively low pressure and temperature conditions.

For the final formation of graphite however, higher temperatures and pressures (up to granulite facies of metamorphism) are necessary to convert amorphous carbon to crystalline carbon.

These processes can be achieved as long as no oxygen enters the system, since at increased temperatures the carbon reacts with oxygen and escapes as gaseous carbon monoxide and carbon dioxide. Graphite therefore occurs preferentially in formerly deeply buried metamorphic rocks, e.g. within the core of orogens (Bonijoly et al. 1982).

Economically significant natural graphite deposits are typically the result of two main processes (Simandl and Kenan 1997):

1. The maturation and metamorphism of organic material
2. The precipitation of graphite from C-O-H fluids, which can occur due to changes in temperature and pressure conditions, fluid buffering, or the mixing of C-O-H fluids with varying compositions and likely different origins

4.3.3. Deposit Types

Within natural graphite, further distinction is made between microcrystalline (amorphous) and macrocrystalline graphite (Table 6, Figure 7). Within the macrocrystalline graphite category, there is a further differentiation between flake graphite and hydrothermal (vein) graphite (Luque et al. 2014). Flake graphite consists of platelets/flakes and occurs in metamorphic rocks such as marble, gneiss or schist. Amorphous graphite is a very fine-grained (semi)-graphite found in coal seams, shales and schist. Vein graphite can occur needle like or massive in igneous rocks or granulite facies rocks.

Table 6: Natural occurring types of graphite (modified after Keeling 2017; Scherba et al. 2018)

Graphite type	deposit	main host rocks	(genesis Kwiecinska, 2014)	Origin	Product grade
Amorphous (microcrystalline)		quartzites, phyllites, schists	Syngenetic, metamorphic	Contact and/or regional thermal metamorphism of coal seams	75 -85 % C
Flake (macrocrystalline)		gneiss, marble, quartzite, amphibolite	Syngenetic, metamorphic	Graphitization of organic precursors through regional metamorphosis	85 – 98 % C
Vein (macrocrystalline)		granulite, granite	Epigenetic, hydrothermal	Precipitation from carbon bearing fluids	90 – 99 % C

4.3.3.1. Amorphous Graphite

The term "amorphous" graphite can be misleading, as even amorphous graphite possesses a crystalline structure. It is often referred to as "semi-graphite" to describe the transitional phase during coalification to graphitization in organic-rich sediments (B. Kwiecińska 2004). Amorphous graphite is the most common form of graphite. Microcrystalline graphite deposits typically form through sub-greenschist to greenschist contact metamorphism or regional metamorphism of coal seams (Taylor 2006). These deposits primarily comprise small graphite particles intermixed with impurities. They often manifest as stratiform or lens-shaped formations, with beds that may exhibit deformation, folding, and faulting. Variations in bed thickness, characterized by pinching and swelling, are common. Such deposits may encompass multiple beds, each a few meters thick and extending for hundreds of meters along their strike. The graphite content in these ores ranges from 30% to 95%, frequently exceeding 80 % (Simandl et al. 2015). Typical host rocks are quartzites, phyllites, schists and metagreywackes with a schistose or massive texture of the graphite ore (Simandl and Kenan 1997).

4.3.3.2. Flake Graphite

Flake graphite is the most economically significant type of graphite due to its application in LIB and other battery types. Flake size and crystallinity depend on the temperature during metamorphism. Disseminated graphite flakes are found in various rock types, including marble, paragneiss, iron formation, quartzite, pegmatite, syenite (Simandl et al. 2015), and exceptionally rare cases of serpentinized ultramafic rocks (Crespo et al. 2006). The most common hosts for economically significant crystalline flake deposits are paragneiss and marble grading at 2-3 % graphite or lower.

The highest graphite grades in paragneiss-hosted deposits are typically found near paragneiss-marble contacts. Marbles in granulite-facies terrains typically contain less than 0.5 % crystalline flake graphite, although concentrations of 1 to 3 % crystalline graphite are common. Graphite is uniformly distributed

throughout the host rock, and the size of graphite flakes correlates with calcite or dolomite (Simandl et al. 2015).

4.3.3.3. Vein Graphite

Vein graphite deposits, also described as hydrothermal graphite deposits are found in both granulite-facies metamorphic and igneous rocks are not confined to specific rock types. These deposits are structurally controlled, typically intersecting host rocks, often forming due to hydraulic fracturing. In granulite rocks, graphite veins are typically composed of graphite alone, while in igneous rocks, they can be associated with various hydrous minerals or magmatic sulfides, depending on whether they formed from aqueous fluids or carbon-rich melts. The specific mineral associations are influenced by the deposition mechanisms. Regardless of their geological context, vein graphite exhibits large crystal sizes, commonly referred to as lump graphite in commercial terms and possesses a high degree of crystallinity. Furthermore, vein graphite typically contains a higher carbon content compared to graphite in syngenetic deposits within metamorphic rocks. The carbon source differs between granulite-hosted and igneous-hosted deposits. In granulites, carbon originates from sub-lithospheric sources or, in certain areas, results from decarbonation reactions in carbonate-bearing lithologies. In igneous rocks, graphite primarily derives from the assimilation of rocks containing organic matter. Due to the limited solubility of carbon in silicate magmas, immiscibility leads to the formation of carbon-rich magmas or fluids. Carbon is transported as CO₂ and/or CH₄ in supercritical aqueous fluids, depending on the oxygen fugacity (fO₂), both in granulite and igneous rocks through fracture systems. The principal mechanism driving graphite deposition in granulite-facies rocks involves cooling and the consumption of water through reactions with surrounding rocks. In igneous-hosted deposits, the primary factors leading to graphite deposition are the cooling of the melt or fluid and the consumption of water from these fluids. Water consumption in igneous rocks is typically linked to hydration reactions of host rocks, resulting in the formation of hydrous minerals within graphite-bearing veins (Luque et al. 1998; Luque et al. 2014)

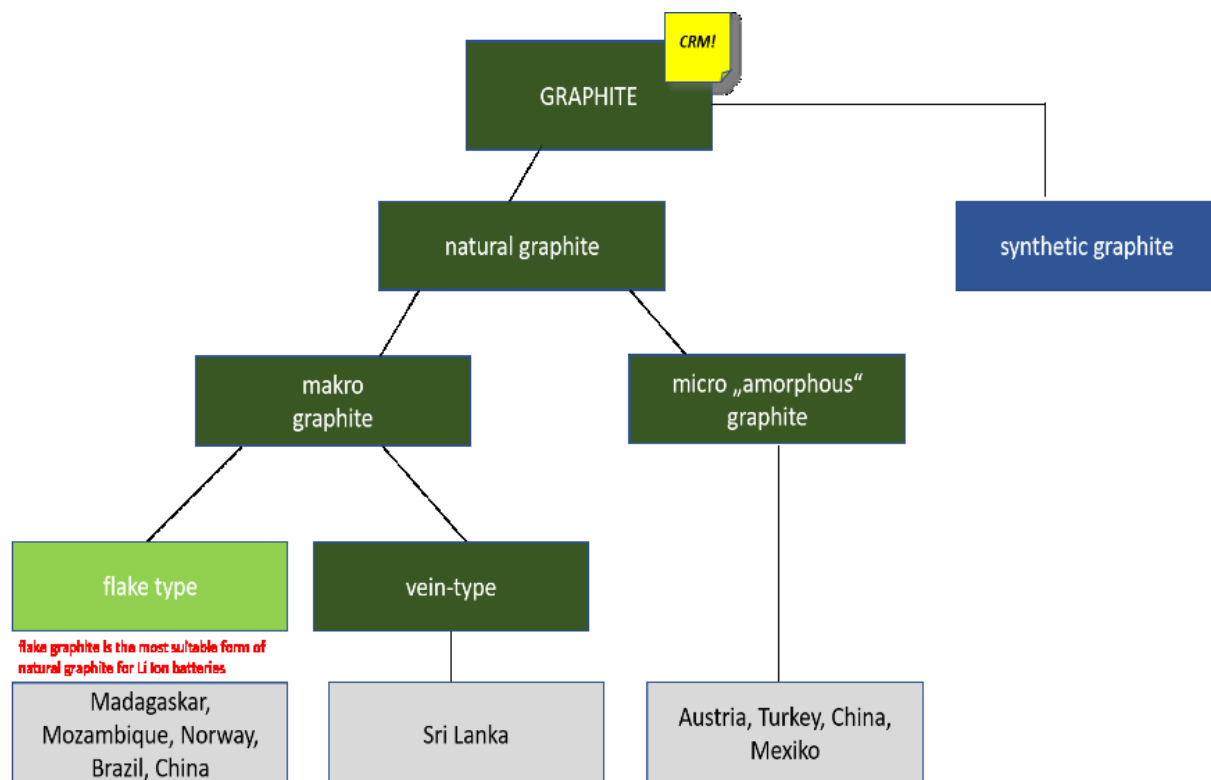


Figure 7: Classification of graphite deposit types

4.4. Sample Material

4.4.1. Explanation and Clarification

The sample collection consists of commercially available graphite concentrates covering the major graphite-producing countries worldwide (Fig. 8).

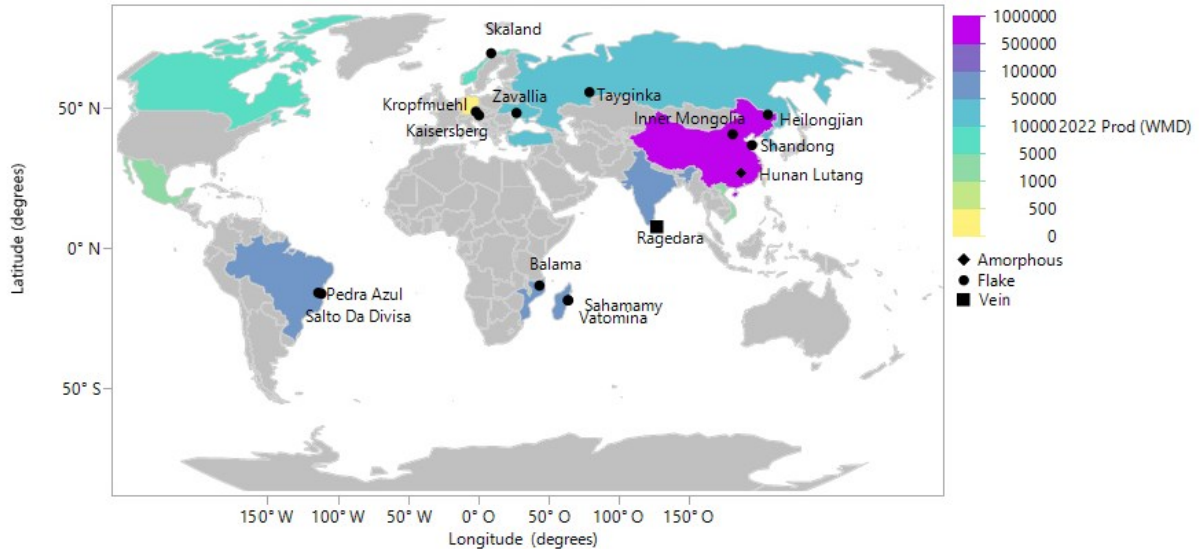


Figure 8: World map of graphite sample material and production (2022) in t

These concentrates hold global significance due to their widespread trade. Raw, unprocessed ore from some deposits could be obtained: Kaisersberg (Austria), Passau (Germany), Zavallia (Ukraine), Skaland (Norway) and Balama (Mozambique) and was included in this study. Raw ore is not transported globally due to the large quantities involved and it is processed locally before being traded worldwide in the form of graphite concentrate.

Samples from China are more related to provinces than to specific mines, as the situation in China is highly opaque. A province may have multiple processing plants and various mines, including artisanal mining, or even one processing plant for one province and multiple mines. A more detailed subdivision is not possible for the occurrences in China used in this study. The level of geographic origin for samples from China is therefore one level above the others, referring to the region/province rather than the specific mine site (except Hunan Lutang, which refers to the province Hunan and the mine Lutang). Graphite deposits in China are mainly distributed to Heilongjiang, Inner Mongolia, Sichuan, Shanxi, and Shandong (Sun, 2018). In this study, three out of the five major regions are covered (Inner Mongolia, Heilongjiang, Shandong). Precise production quantities for each province could not be obtained. Samples from Madagascar, specifically Vatomina and Sahamamy Sahaso, represent mixed concentrates from these two mines. Both mines are in close proximity and processed in a single processing plant, making further differentiation impossible. The province is called Brickaville and includes the two mines named above. The same applies to Brazil. The material from the mines Pedra Azul, Itaperica, and Salto da Divisa is processed and mixed in a single processing plant. The available concentrate samples cannot be traced back to each individual mine, therefore the province Minas Gerais is stated in the further course of the work.

Samples from all other origins directly refer to the mine site along with the associated processing plant in the same location. Commercially available samples were acquired yearly from 2020 to 2023, if available. Some samples represent earlier years (2019 and earlier), due to a collection available at the

university collection. The following list presents localities with the number of available individual samples from the same mine/province (Table 7).

It is still necessary to extend the analysis to encompass additional graphite intermediates, particularly those subjected to more advanced processing. This extension aims to define the degree to which specific identifying features, the unique signatures or fingerprints, can be traced, particularly across multiple stages within the graphite supply chain. The sample collection and analytical efforts were primarily focused on graphite concentrates sourced from various mining operations. This allows to distinguish between various deposit origins and the corresponding graphite concentrates extracted from them. This distinction serves as a starting point for the establishment of a traceability framework aimed at tracking graphite throughout supply chain for graphite.

Table 7: Summary of the available sample material (nda – no data available)

Country	Mine/province	Company	Deposit type	Resources in t (Weber, 2023)	Number of samples (conc)	Number of samples (raw ore)
Austria	Kaisersberg	Kaisersberg Grafit	Amorphous	nda	2	3
Brazil	Pedra Azul, Itaperica, Salta da Divisa (Minas Gerais)	Nacional de Grafit (NDG)	Flake	8.045.700 (Itaperica only)	5	-
China	Hunan Lutang	na	Amorphous	27.590.000	1	-
China	Heilongjian	na	Flake	na	1	-
China	Shandong	na	Flake	na	2	-
China	Inner Mongolia	na	Flake	na	1	-
Germany	Kropfmühl	AMG Graphite	Flake	na	1	1
Madagascar	Sahamamy-Sahaso, Vatomina (Brickaville)	Tirupati Graphite	Flake	7.100.000, 18.400.000	3	-
Mozambique	Balama	Syrah Resources	Flake	1.423.000.000	6	1
Norway	Skaland	MRC, Skaland Graphite	Flake	2.484.000	3	1
Russia	Taiginka	JSC Uralgraphite	Flake	11.000.000	3	-
Sri Lanka	Ragedara	Elcora	Vein/lump	na	2	-
Ukraine	Zavallia	Volt, Zavaljevsky Grafit	Flake	22.913.000	2	3

4.4.2. Description of Deposits and Samples

4.4.2.1. Kaisersberg, Austria, Europe

Kaisersberg is the only active graphite mine in Austria, operated by Grafitbergbau Kaisersberg Ges.m.b.H. in St. Stefan ob Leoben. The amount of extracted graphite is not reported (Heike 2023). The extracted material is referred to as semi-graphite (Rantitsch et al. 2016a) and is mined underground. The annual extraction quantities are approximately 100 tons (personal communication, Twrdy, G.). This graphite deposit represents one among several stratabound graphite deposits situated within the Eastern

Greywacke Zone, however currently being the only one in operation. These deposits are found within the Upper Carboniferous strata of the Veitsch nappe (Scharfe 1981).

The Veitsch nappe is the lowermost tectonic unit within the Austro-Alpine Greywacke Zone, and it overlies a suite of medium-grade metamorphic rocks and Permo-Mesozoic cover sequences (Ratschbacher 1984, Nievoll 1984, Neubauer & Vozárovà 1989, Ebner 1991, Neubauer 1994). The graphite deposits are situated within a carbonate-clastic sequence, referred to as the Sunk Formation. This formation is characterized by a coarsening-upward sequence with cyclic sedimentation patterns (Ratschbacher 1984 and 1985). These sequences, with thicknesses ranging from 50 to 150 meters, consist of graphitic schists, meta-argillites, and metaconglomerates. They are interpreted as sediments that originated in deltaic to coastal plain environments. Plant fossils found within this formation provide evidence of its Westphalian A age (Raith & Vali 1998), Rantitsch et al. 2016).

4.4.2.2. Pedra Azul, Itapecerica, Salto da Divisa, Minas Gerais, Brasil, South America

The company Nacional de Grafite operates three mines with processing facilities on-site in Brazil. These mines, namely Pedra Azul, Itapecerica, and Salto da Divisa, collectively produce 70.000 tons of graphite per year (Nacional de Grafite 2024). They are situated within the state of Minas Gerais. Samples from Brazil are concentrate blends. The company operates three mines in close geographical proximity to each other, all associated with the same graphite deposit. In this context, it is not feasible to precisely attribute each sample to its respective mine. However, due to processing practices, it can be reasonably assumed that the samples consist of homogeneous mixtures of the concentrates.

The mines are situated within the southern São Francisco Craton, specifically within the Itapecerica Supracrustal Sequence. The southern portion of the São Francisco Craton encompasses a diverse geological landscape, featuring Meso- to Neoproterozoic granite-greenstone terrains, Paleoproterozoic clastic-chemical metasedimentary rocks from the Minas Supergroup, and Neoproterozoic pelitic-carbonate sedimentary rocks of the Bambuí Group (Teixeira et al. 2017). The underlying basement comprises the Campo Belo, Divinópolis, Bonfim and Belo Horizonte metamorphic complexes. This sequence also includes the banded iron formations found in the Quadrilátero Ferrífero (Machado et al. 1996; Moreira et al. 2016).

The Itaperica schist, formed in the Paleoproterozoic is primarily composed of fine to medium-grained graphite, along with the presence of quartz and sillimanite. Graphite is the dominant constituent, establishing the foliation. Quartz, on the other hand, plays a secondary but significant role and exhibits elongated crystal shapes reminiscent of ribbons, which align with the foliation pattern. Sillimanite is also present in the schist, its proportions varying, and it may display either a fibrous or prismatic habit (Miranda et al. 2019).

4.4.2.3. Inner Mongolia, Heilongjian, Hunan Lutang, Hubei, Shandong, China, Asia

Graphite is sourced from 170 mining areas across 25 provinces in China. The predicted graphite resources are estimated at about 1.63 billion tons. The major graphite resources in the Heilongjiang province amount a billion tons representing 60 % of China's graphite resources. For the Chinese samples only the provinces are known, neither the precise location of the extraction, nor the exact mine is known.

There are two main types of graphite mineralization in China – the regional metamorphic-type, associated with Neoproterozoic and Paleoproterozoic rocks and the Mesozoic contact metamorphic-type. Among these, flake graphite formed during regional metamorphism of carbonaceous rocks are the most prominent in China. Typically, these deposits are situated in high-grade orogenic zones positioned at the peripheries of tectonic blocks or terranes. Examples are located in the Jiamusi and Alashan blocks, as well as along the boundaries of the Northern China and Yangtze cratons, where the highest levels of

metamorphic grade are observed. Regions with potential for this variety of graphite encompass Heilongjiang, Inner Mongolia, Shandong, Jilin, and Sichuan provinces, with Heilongjiang Province standing out as a location of very high potential for regional metamorphic-type graphite found within metasedimentary rocks (Cui et al. 2017).

Hunan Lutang is a typical contact metamorphic graphite deposit, which was deposited in part of marine and continental environments in the Hunan Province in the east of the country. It formed in the Late Permian Longtan Formation. The rock was altered during the Mesozoic Qitianling Granite intrusion, creating different types of rock near and far from the intrusion – calc-silicate rock near it and marble further away. Graphite is found in areas with structural openings and within altered coal-bearing layers, with varying widths from 0.1 to 11 meters. The graphite consists of small and well-shaped flakes, about 0.3 micrometers wide, and contains between 70 % and 86 % carbon content (An et al. 2016; Cui et al. 2017). Hydrothermal graphite is very rarely found in China and is primarily located in hornfels formations in the western part of the country

4.4.2.4. Kropfmühl, Germany, Europe

Graphit Kropfmühl GmbH (GK), located in Kropfmühl, near Passau in Germany processes graphite raw materials to manufacture customized graphite products and concentrates for specific uses from various sources. They also engaged in labor-intensive underground mining, which restarted in 2012 after a seven-year pause. Furthermore, the company secures its raw material supply through deposits located in various regions of Africa and Asia. Access to the primary graphite deposit is achieved through the Erhard shaft, which facilitates excavation activities extending down to the 10th level of the mine. The Kropfmühl graphite deposit is situated in the Moldanubic geological domain, within the Bohemian Massif. This deposit is characterized by a metamorphosed rock series. These predominantly encompass dark garnet (plagioclase) biotite gneisses, interspersed with light quartzitic gneisses, quartzitic intercalations, amphibolites, and calc-silicate marbles (Krüger et al. 2017).

Of particular importance are the Moldanubic graphites, which exhibit a macrocrystalline morphology, with crystal dimensions surpassing 100 micrometers. The genesis of these graphites is attributed to the regional metamorphic overprinting of sedimentary rocks. An organogenic origin, emphasizing the significance of biophilic trace elements such as molybdenum, vanadium, nickel and boron within the graphite-bearing rock formations had been postulated (Weber 1987).

4.4.2.5. Vatomina und Sahamamy Sahaso, Brickaville, Madagascar, Africa

Madagascar has consistently demonstrated its capacity to contribute significantly to the global graphite market due to its ability to supply high quality and large flake graphite from several graphite deposits in Madagascar, with currently five active graphite mining operations (Weber 2023b). The sample material from Madagascar is an aggregate from the Vatomina and Sahamamy Sahaso mines, both operated by the company Tirupati Graphite. The two mines are located very close to each other in the north east of the country. Graphite deposits in Madagascar are found within the Precambrian metasedimentary basement, comprising schist, gneiss, granitic and basic igneous rocks, forming a wide north-south-oriented belt in the eastern part of the country. These regions are rich in graphite and closely linked to the Manampotsy group. The graphite ore occurs as both, disseminated and layered forms, mainly within gneissic and migmatitic rocks, that have undergone significant decomposition, resulting in clay-rich substrates. The distribution of graphite content within the layers varies, and the primary orientation of these major veins runs in a north-south direction, with widths reaching up to approximately 100 meters (Heritiana et al. 2019).

4.4.2.6. Balama, Cabo Delgado Province Mozambique, Africa

Mozambique, a significant supplier of graphite, currently has two active mining operations extracting graphite, along with several additional deposits that are not currently exploited. The material for this study is sourced from the Balama mine, which is located within the Southern Cabo Delgado province in the district of Namuno, northern Mozambique and is regarded as being the world's largest high-grade graphite deposit. The mine is operated by Syrah Resources and the production was commenced in January 2019 after starting various pre-construction activities late 2014 and completing the feasibility study of this project in 2015 (Mining Technology 2021). The rapid execution of the approval process and the commencement of mining activities just four years after the feasibility study demonstrates the enormous demand for this raw material.

Proven and probable reserves are estimated to be 114.5 Mt at an average TGC grade of 16.6 %, containing 18.9 Mt of flake graphite. Resources are estimated to be 1,036 Mt within a life of mine of 50 years. The mining is performed in a simple, low strip open-pit mine. The product is 94 to 98 % fixed carbon graphite concentrate and 80 % fine flake (100 mesh) graphite (Syrah Resources 2024).

The Balama Graphite Mine is situated in the Cabo Delgado province of northern Mozambique. It is located within the Mozambique Belt, a geological region known for its various mineral resources. The Metamorphic Mozambique Belt (NMMB) formed during the Neoproterozoic and Cambrian periods due to the closure of the Mozambique Ocean. It consists of four major rock types: ophiolites, island arc magmatic rocks, metamorphic rocks with raw materials including gemstones and graphite deposits, one of them being the Balama mine and late intrusive formations like granitoids and anorthositic gabbros. This region is renowned for its gemstone deposits and is often referred to as the 'Gemstone belt of East Africa' or the 'Pan-African Gems and Graphite Belt (Feneyrol 2013).

4.4.2.7. Skaland - Traelen, Norway, Europe

For nearly a century, Norway has been a leading European producer of flake graphite with three operating graphite mines. Four graphite provinces can be found in Norway, starting in the north, these provinces are: the island of Senja (Skaland/Traelen mine), the Vesteralen islands, the Holandsfjorden area and the Bamble area in southern Norway. Only the Skaland/Traelen mine will be described here. Paleoproterozoic graphite deposits with a notable concentration of high-grade ore, characterized by a high total carbon content, are located on the Fennoscandian Shield, specifically, within Norway, Sweden, Finland and Russian Karelia (Parnell et al. 2021).

Being a stable and reliable supplier of high-quality flake graphite since the 1900's and being the biggest crystalline graphite producer in Europe and the fourth largest producer globally outside of China, Norway accounts for around 2 % of the global annual natural flake graphite production with a production volume of 10380 t/year in 2022 (Reichl & Schatz 2024). The Traelen mine together with the Skaland processing plant has been owned by Skaland Graphite and was acquired by the Australia based company Mineral Commodities (MRC) in October 2019, where the mine permit has been extended for another 10 years until 2029 (NS Energy 2024). Senja Island is situated in the southern region of the West Troms Basement Complex (WTBC). The WTBC consists of tonalitic and granitoid gneisses intruded by a mafic dyke swarm. The basement gneisses are overlain by a variety of supracrustal units, including metapsammites, conglomerates, metavolcanites, dolomitic carbonates, banded-iron formations, and graphite schists. During the Svecofennian orogeny around 1.8 to 1.75 billion years ago, both the basement gneisses and supracrustal rocks underwent deformation. Within this complex, the peak metamorphism levels vary, ranging from lower grades in the northern part to amphibolite to granulite facies in the southern region, particularly on Senja Island (Gautneb et al. 2020).

The deposit consists of two main lithological units: The stratigraphically lower unit is a banded gneiss with alternating biotite-rich and granitic bands (“Traelen Gneiss”) and the upper unit an amphibolitic gneiss. These hornblende gneisses contain graphite horizons, metamorphic greywacke and calcareous rocks (Mineral Commodities Ltd 2022c).

4.4.2.8. Tayginka, Chelyabinsk region, Russia, Asia

With three existing and two operating graphite mines, Russia contributes with 18800 t/year (1.09 %) to the global graphite production. Samples for this study originate from the the Tayginka graphite deposit that is situated in the Chelyabinsk region (Siberia). Graphite mining dates back to 1942 (Uralgraphite - natural graphite producer and supplier 2024)

The Middle Urals represent a significant bend in the Urals mountain range and are the most compressed part of the late Palaeozoic collision zone. This region is mainly composed of metamorphic rocks and cut by large fault lines. The Main Uralian Fault zone (MUF) is a major fault running north-south. It separates the Sysert Complex in the east from the Ufaley Complex in the west. The Sysert Complex is part of the East Uralian Zone where the graphite mineralization can be found (Echtler et al. 1997).

4.4.2.9. Ragedara, Sri Lanka, Asia

There are several active mining operations extracting graphite in Sri Lanka. The mined graphite in Sri Lanka is consistently of the inorganic lump graphite type. In addition to the Ragedara mine, from which the sample material originates, the Bogala Mine, the Queens Mine, and the Karasnagala Mine are currently extracting lump graphite. The occurrence of vein graphite deposits in Sri Lanka is concentrated in the Wannu Complex in the Central Highlands underlain by supracrustal rocks (Dissanayake et al. 1988). The Wannu Complex is a Precambrian high-grade metamorphic terrain and consists of 770 - 1100Ma granitoids, metagabbro, charnockite, gneisses and migmatites (Touret et al. 2019).

The Ragedara Graphite Mine operated by Ragedara Mines Ceylon Private Ltd., held the distinction of being the world's largest graphite producer until the end of World War II. This deposit, located in the southeast of Sri Lanka is renowned for its exceptionally pure graphite content, ranging from 97 % to 99 %. The veins in this deposit typically measure 30 to 50 cm in width, with some extending up to 2 meters and an average vein length of about 25 meters. Veins are tabular or lenticular in shape. The largest one observed at Ragedara was close to 2 meters wide. The massive crystalline graphite shows uniformity throughout the veins and is composed of massive to millimeter size closely packed black graphite (Filion 2012).

There are several explanations for the formation of vein graphite in Sri Lanka (Hewathilake et al. 2018):

- Carbon-rich fluids resulting from decarbonation reactions in calcareous environments could be a source for the vein graphite formation (Hapuarachchi 1997)
- An organic origin for the carbon source, with subsequent deformation and magmatic activities mobilizing carbonaceous material (Dissanayake, 1981 and 1986).
- The role of CO₂-rich fluids in promoting vein graphite formation through hydraulic fracturing and crack-seal processes under low oxygen conditions (Katz, 1987).
- Hydrothermal activity in fracture zones of metasedimentary rocks as a contributor to vein graphite formation through decarbonation processes (Silva, 1987).

4.4.2.10. Zavallia, Ukraine, Europe

The Zavalja mine is the only active graphite mining operation in Ukraine. Despite Russia's war with Ukraine, production at the mine continues, production amount however dropped significantly (- 95 %) (Reichl & Schatz 2024). The mining operation is managed by Volt, an Australian mining company that owns 70 % of the company, with the remaining 30 % held by Ukrainian authorities. The mine and its processing facilities have been in operation since 1934. Flotation, concentration, and purification of the graphite material are carried out directly at the mine site. In the future, purity levels of up to 99.95 % are targeted through on-site processing (Zavaliievsky Graphite Operation – Volt Resources Limited 2024).

Graphite deposits in Ukraine are primarily situated within the Ukrainian Shield. Extending from the northwest to the southeast of the country, the Ukrainian Shield comprises folded Precambrian basement and overlying Phanerozoic sedimentary cover, reaching a cumulative thickness of up to 120 meters (Mykhailov 2020). The concentration of graphite deposits is notable within four distinct ore districts: Pobuzhzhia (Zavallivske in Kirovograd Oblast), Kryvyj Rig (Petrivske and Balakhivske in Kirovograd Oblast), Volyn (Burtynske in Khmelnytskyi Oblast), and Near Azov (Troitske in Zaporizhzhia Oblast and Mariupolske in Donetsk Oblast). Presently active mining operations are limited to the Zavallivske/Zavaljevsky deposit.

The Zavaljevsky deposit is confined within the biotite-graphite gneisses of the Hashchevato-Zavallya Archean suite. This deposit encompasses approximately 40 tabular ore bodies, varying in length from 200 to 1200 meters and thickness from 3.5 to 80 meters. Mineralogically, the ore-bearing gneisses exhibit variations, including biotite, amphibole-biotite, biotite-chlorite, chlorite-sericite, chlorite-talc, and chlorite varieties. Graphite within these ores is characterized by a relatively irregular distribution in the form of large flakes (2–4 mm), with graphite content ranging from 6% to 14%, averaging to 6.5%. The mining operation is mined through open-pit methods and processing occurs on site. This processing facility operates at an annual rated capacity of 800 thousand tons of graphite ore or 35 thousand t of graphite concentrate. During the early 1990s, production rates exceeded 40 thousand tonnes but subsequently experienced a sharp decline. Presently, the annual production stands between 4 to 8 thousand tons. The rated capacities of both the open pit and the processing plant allow for increased production of graphite concentrate to meet the demand for raw materials (Mykhailov et al. 2023).

5. Methodology - Developed Methodology for Tracing the Origin of Natural Graphite

5.1. Selected Parameters for the Analytical Proof of Origin (APO)

The approach to proof the origin of natural graphite includes multiple parameters, some of which have been already used to analyze graphite or carbon rich materials before, such as $\delta^{13}\text{C}$ or lattice ordering. Specific parameters, such as the chemical composition (e.g. trace elements) have a proven track record in determining the origin of certain raw materials, yet they have not been employed to graphite so far. The highlighted parameters and methods were addressed in the context of this work (Table 8).

Table 8: Potential methods and parameters for the analytical determination of the origin of natural graphite – bold letters indicate that the method/parameter had been considered in this study

Parameter	Method		Conclusion
	Bulk	In-situ	
Chemical composition	XRF	LIBS, LA-QQQ-ICP-MS	Carbon source
$\delta^{13}\text{C}$	gas-MS	LA-MC-ICP-MS	Depositional environment and genesis
$\delta^{34}\text{S}$	--	LA-MC-ICP-MS	Depositional environment and genesis, sulphur source
Lattice ordering	XRD	Raman	Temperature conditions
Grain morphology	--	SEM, EMPA, digital light microscopy	Ore genesis
Mineral paragenesis	XRD	SEM, EMPA, digital light microscopy	Metamorphic history

5.1.1. Structure and Crystallinity

Graphite exhibits a Raman spectrum with several characteristic bands. The most significant Raman band is the G-band (graphitic band) located at around 1580 cm^{-1} . Another important band is the D-band (disorder-induced band D1 and D2), typically appearing at around 1350 cm^{-1} . In highly crystalline graphite, the D-band is relatively weak compared to the G-band, reflecting the ordered and well-structured nature of the material. The intensity ratio of the D-band to the G-band I_D/I_G is often used as a parameter to assess the degree of disorder and defects in graphite. High-quality, crystalline graphite typically has a low I_D/I_G ratio, while more disordered or lower-quality graphite will exhibit a higher ratio (Ferrari & Robertson 2000).

The Raman spectra of graphite originating from both the amphibolite and granulite facies are defined by the presence of a well-defined G band and the minimal presence or complete absence of the D2 band (Rantitsch et al. 2014). Microcrystalline (amorphous) graphite undergoes a transformation into flake graphite at the transition from the amphibolite to granulite facies. This transition encompasses a metamorphic temperature range of $500\text{ }^\circ\text{C}$ to $700\text{ }^\circ\text{C}$, along with a pressure range exhibiting significant variability between 4 and 11 kbar (Rantitsch et al. 2014).

The impact of material preprocessing on Raman spectra was assessed and it was found that differences in Raman parameters between mechanically processed and handpicked/original samples were minor. The processing method of graphite has a limited impact on Raman properties (Rantitsch et al. 2014). Polishing introduces significant, unpredictable changes in the intensity of both D bands relative to the G band. This effect obscures the evaluation of the intrinsic structural order, making this parameter unsuitable for polished carbon materials. The width of the G band appears to remain consistent after polishing (Maslova et al. 2012).

The following factors can influence the structure of graphite:

1. Temperature and Pressure (Graphitization Process): The graphite crystal structure is sensitive to temperature conditions. Changes in these parameters can lead to alterations in the crystal lattice and interlayer spacing of graphite. Graphitization leads to the conversion of structurally disordered carbonaceous material found in anthracite to a more ordered structural state. Semi-graphite serves as an intermediate phase bridging the gap between (meta-) anthracite and graphite (Kwiecińska and Petersen 2004)

2. Time and Geological Processes: The geological history of a graphite deposit plays a significant role in determining its structure and crystallinity. The extent and duration of geological processes, such as metamorphism or regional tectonic events, influence the level of crystallinity and the size of graphite flakes/crystallites. Raman spectroscopy is able to track peak metamorphic conditions of carbon rich materials during regional metamorphism. It had been employed to explore tectono-thermal processes of materials that contain a significant amount of carbon (Lahfid et al. 2010).

5.1.2. Chemical Composition

The chemical composition can provide valuable information about the geological origin of graphite as different deposits may have distinct chemical signatures and trace element profiles. The chemical composition can also serve as an indicator for the quality and purity of the graphite, which is an important factor for industrial applications. Understanding the chemical composition of different graphite materials is essential for selecting the right type of graphite for a particular application. Assessing the chemical composition is also essential to ensure compliance with safety and environmental regulations.

Limited research has been conducted on the geochemical characterization of graphite. A comprehensive study on this topic was conducted by Janda and Schroll in 1960, which served as a valuable reference for this work. Using an emission spectrographic method, the elements Ag, As, B, Ba, Be, Bi, Cd, Co, Cr, Cu, Ga, Ge, Hg, Mn, Mo, Ni, Pb, Sb, Sn, Sr, Ti, Tl, V, W, Zn, and Zr were determined in more than 100 graphite samples and graphitic rock samples. In general, the content of trace elements decreases with increasing crystallinity and carbon content. This trend also applies to biophilic elements such as Ge, V, and B. In amorphous (microcrystalline) graphite, traces of Ge, Be and Cr suggest an origin from coals (humitic), while the presence of Mo, Co and Ni, along with high V contents, point to an origin from bituminous rocks and sapropelites (Janda and Schroll 1960). High B contents are likely associated with graphite formed from paralic coal seams. A conclusion about the source rock of vein graphite is considerably more challenging. All examined graphite samples in the study from Schroll and Janda from 1960 exhibit trace element contents of typical biophilic elements such as V, B, Ni, Co and Mo, which can be taken as evidence that these rocks should be classified as organogenic sediments. In the analyzed Ceylon samples, B and V were absent, which corresponds to the non-organic origin of the carbon in these graphites. During graphitization, all elements bound to the organic matter migrate into associated minerals, resulting in a dependence on the degree of crystallinity and ash content. Therefore, a genetic correlation is mostly possible in microcrystalline and anthracitic graphite. The questions concerning the behavior of trace elements during metamorphism of such rocks, however, require further comprehensive investigations (Janda and Schroll, 1960 ; Schroll 1976).

To summarize, the chemical composition of graphite, especially the presence of trace elements, is influenced by various geological processes:

1. **Metamorphism:** Metamorphic processes, such as regional and contact metamorphism significantly impact on the trace element composition of graphite. During metamorphism, the precursor materials undergo recrystallization and mineral reactions, which can introduce or remove trace elements from the graphite and the other way round.
2. **Host Rock Composition:** The trace element composition of the host rock can influence the trace elements found in the graphite. The interaction between these rocks and the carbon source can lead to the incorporation of trace elements into graphite.
3. **Fluid Infiltration:** The movement of fluids through the geological environment can introduce or leach out trace elements from the graphite.

4. Mining and Processing: The mining and processing of graphite ore can introduce trace elements through contamination during extraction and beneficiation processes into the concentrate, but also leach out certain elements through chemical processing.

5. Geological History: The overall geological history of a graphite deposit, including its formation, geological events, and subsequent alterations can leave a fingerprint on the trace element composition of the graphite.

5.1.3. Carbon Isotope Ratios

Carbon isotopes in graphite are crucial to identify the origin of carbon. They provide information about the source material and the geological processes that led to the formation of graphite. The interpretation of the source of carbon in graphite relies on the distinct isotopic ranges characterizing the primary carbon origins, including organic matter, carbonates, and the Earth mantle. The average $\delta^{13}\text{C}$ value for organic matter typically is around -25 ‰, whereas marine carbonates spanning from the Cambrian to the Cenozoic exhibit higher $\delta^{13}\text{C}$ values, ranging from -2 ‰ to 0 ‰. In contrast, the isotopic signatures of diamonds and mid-oceanic ridge basalts (MORB) suggest that mantle-derived carbon is notably heavier, with $\delta^{13}\text{C}$ approximately -7 ‰, compared to biogenic carbon (Figure 9). Considering these variables, graphite of biogenic origin that has undergone extensive high-temperature metamorphism and isotopic exchange with a carbonate phase would exhibit an isotopic signature significantly heavier than its original source. Graphite of biogenic origin that has been surrounded by graphite precipitated from a CO₂-rich fluid resulting from decarbonation reactions or mantle-derived CO₂ would also display a bulk isotopic ratio heavier than the typically lower $\delta^{13}\text{C}$ value associated with biogenic graphite (Luque et al. 2014).

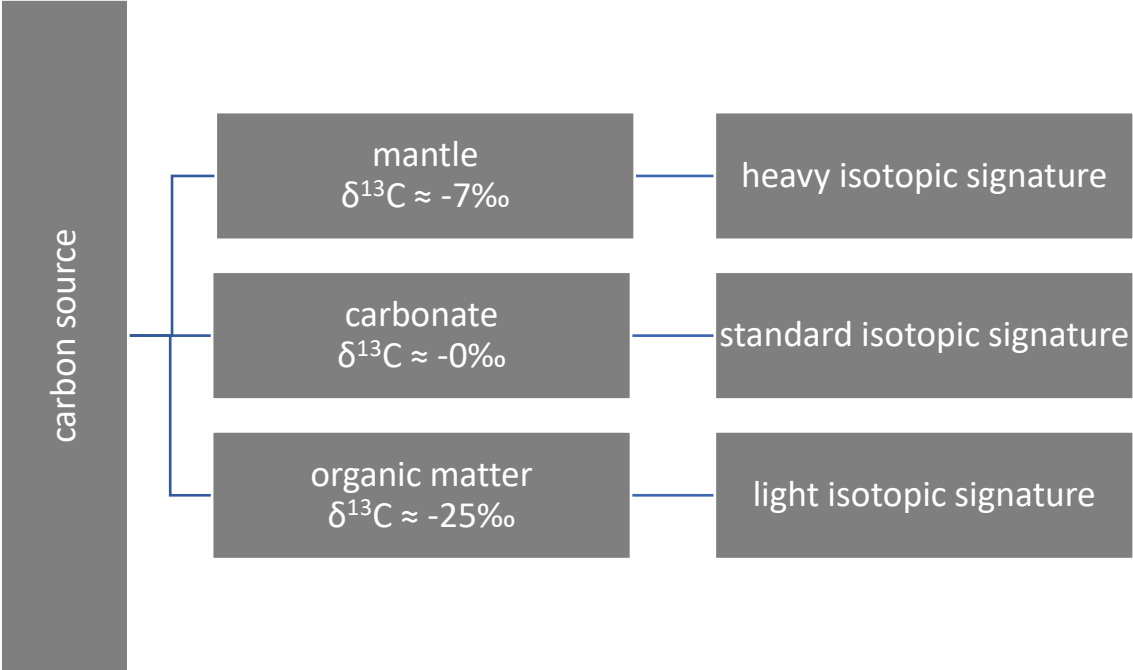


Figure 9: Relation between carbon source and carbon isotope composition (mod. From Luque et al. 2014)

The isotopic difference between disseminated and vein graphite shows that they likely come from different sources. As the carbon in vein graphite is not of biogenic origin, it could have two other sources to account for, the heavy isotopic signature: (a) CO₂ originating from the mantle and (b) CO₂ arising from decarbonation reactions within carbonate-bearing lithologies (Luque et al. 2014).

A straightforward relationship between the carbon isotopic signature of graphite and its source is challenging. It is often altered due to fractionation effects, influenced by factors such as temperature and the presence of other mineral phases, including carbonates. Identifying the origin of carbon in fluid-deposited graphite is even more difficult, given the potential for the mixing of diverse carbon reservoirs before or during graphite precipitation from the fluid, as well as the fractionation of carbon between the different species in the fluid and graphite during the fluid evolution (Luque et al. 2014).

Terrestrial carbon originating from the earth's mantle (diamonds), as well as carbonatites and certain natural carbonates, generally exhibit values similar to meteorites. The significant fractionation of the two isotopes (^{12}C and ^{13}C) makes it challenging to determine the original composition of carbon in the Earth's crust. This fractionation results from two processes:

1. The establishment of a chemical equilibrium between CO_2 - $[\text{HCO}_3]^{-1}$ or $[\text{CO}_3]^{-2}$, wherein the heavier ^{13}C isotope is incorporated into dissolved bicarbonate and precipitated carbonate, while the lighter ^{12}C is enriched in atmospheric CO_2 . Carbonate formed inorganically from seawater typically has a $\delta^{13}\text{C}$ value approximately 10 per mil higher than atmospheric CO_2 .
2. A kinetic effect in the assimilation process of plants, particularly terrestrial plants, which concentrates the lighter ^{12}C in the biosphere. Carbon of biogenic origin tends to have a $\delta^{13}\text{C}$ value roughly 20‰ lower than atmospheric carbon dioxide. The distinct CO_2 regimes on land and in water result in a differentiated isotope distribution in both areas, with terrestrial plants having a higher ^{12}C content than marine plants.

In the case of highly metamorphosed graphite occurrences in marble-bearing metamorphic rocks, the carbon isotopic composition alone does not distinguish whether the graphite was formed from organic matter (as observed in Kropfmühl Passau). The difference between the $\delta^{13}\text{C}$ values of the graphite (-20 to -26 ‰) and the marble (-2 to -22 ‰) is not particularly distinctive (Schroll 1976).

The carbon isotopic composition of graphite falls into two groups: $\delta^{13}\text{C}$ -4 ‰ to -11 ‰ and -19 ‰ to -34 ‰. Graphite derived from coal (e.g., Kaisersberg, 24,4 ‰) fall into the second group. Magmatic graphites fall in the range of -2 ‰ to -12 ‰, while metamorphic graphite exhibits a wide range, varying from -1 ‰ to -45 ‰ (Schroll 1976).

5.2. Sample Treatment and Sample Preparation Procedure

5.2.1. Sample Material

The available sample material included flake, amorphous and vein type of graphite. Flake and amorphous graphite were available as concentrates and vein graphite as lumps/chunks.

The flake dimensions within the concentrates are influenced partly by the processing methods employed and by the inherent flake size of the graphite ore and its genesis. The flakes and corresponding dimensions are presented in chapter 6.1. Optical Properties. Figure 10 illustrates a flake (left image) and amorphous (right image) graphite concentrate for a visual reference.



Figure 10: Flake (left) and amorphous (right) graphite concentrate

The graphite ore from Sri Lanka exists in the form of lumps and chunks (Figure 11), as they are extracted and then crushed. Graphite concentrate from Sri Lanka is not available for the current study; however, high-purity chunky graphite was investigated.



Figure 11: Vein graphite from Sri Lanka, Ragedara mine

5.2.2. Sample Preparation

Each graphite sample was subsequently homogenized and then split, with approximately one-third of it being used as the working sample, while the remaining two-thirds were retained as back-up samples for future analysis and archival purposes. The ultimate goal was to obtain samples or concentrates from different years at the respective mining sites, as this reflects various extraction areas within the mine, thus allowing to test the natural heterogeneity within a deposit. This sampling approach also takes into consideration different mined sectors within the deposit.

The samples have been prepared for further analyses, which will be discussed in more detail in the following subchapter. The subsequent analyses required tailored sample preparation for each applied method.

5.2.3. Powder Pressed Pellets

The majority of samples in this study existed in concentrate or powdered forms, therefore it was necessary to transform them into solid states for specific analyses, notably LA-ICP-MS. This transformation was achieved through a pressing process. Initially, adhesives/binders such as high-purity laboratory wax were employed for this purpose. However, through later, more targeted mechanical press applications, the need for adhesives was not necessary any more. This strategic refinement ensured the avoidance of further dilution or contamination of the already highly pure concentrates by laboratory wax. Two laboratory presses, conventionally employed for fabricating powder pellets for XRF were available. One possessed a press diameter of 11 mm and the other a 31mm press diameter (Figure 12).

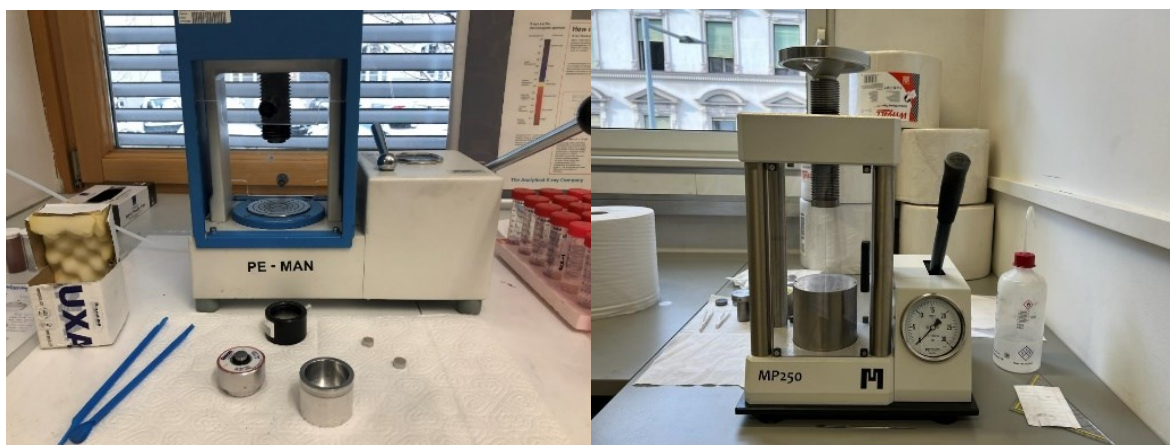


Figure 12: Different presses for the pellet preparation: 11 mm pressing diameter (left image) and 31 mm pressing diameter (right image).

The larger press (MP 250; Maasen Spektroskopie) with 31 mm pressing element diameter, featured films that could be affixed to the upper and lower press components to avoid adherence of the pressed graphite to the metallic surfaces, as seen in

Figure 13. Attempts without these films as separating elements between the pressing part and the material were unsuccessful, as the material adhered to the metal pressing element. While reducing the pressure could prevent material adhesion, it resulted in insufficient stability.

Several pressing experiments revealed ideal pressing parameters for the 31 mm diameter press as 4.5 g of graphite material, 12 t contact pressure and approx. 150 – 180 sec time. It is noteworthy that the pressing dynamics are influenced by grain/flake size and grain/flake morphology, with each material showing distinctly different behaviors during the pressing process. Flake graphite, notably, has better pressing characteristics when compared to the exceedingly fine-grained amorphous (semi-graphite) material, but also the certified reference material BAM S009. Small powder pellets were adopted for LA-ICP-MS analysis due to their capacity to accommodate a greater number of samples within the sample chamber, thereby optimizing analytical efficiency. A gradual and slow release of pressure after the pressing process is necessary, as rapid decompression can result in surface chipping of the pellet. For analytical purposes, only perfectly flat and smooth surfaces are suitable.

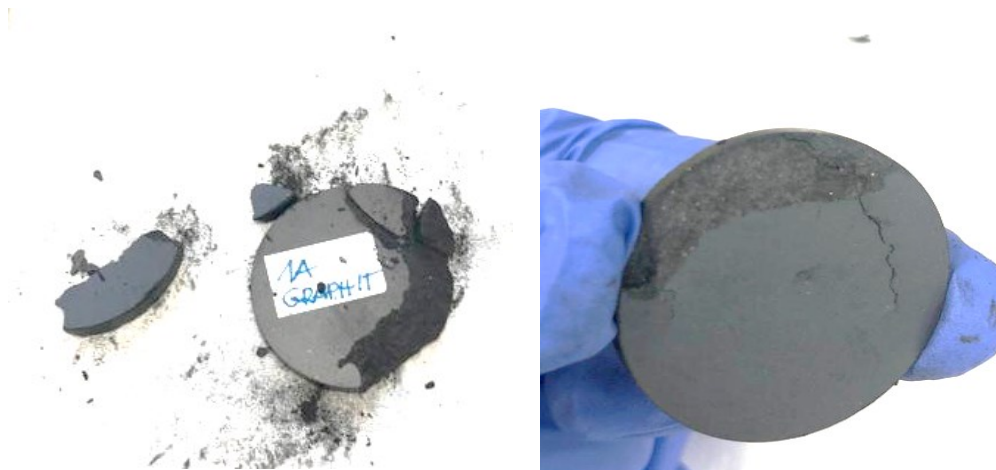


Figure 13: Surface chipping or breaking apart of amorphous graphite material pressed into pellets

5.2.4. Water suspension and Glass Plates

To facilitate Raman analysis, it was necessary to attach the graphite sample onto a smooth surface while ensuring a straightforward sample preparation process, given the need to analyze a substantial quantity of samples. The smooth, optically transparent surfaces of glass plates or glass slides were moistened with high purity water to establish a water interface. Subsequently, individual graphite flakes were dispersed onto this surface and allowed to air dry for a minimum of 24 hours. As the water evaporated, the graphite flakes adhered to the surface of the glass plate, eliminating their loose attachment.

For raw ores, graphite was selectively extracted from the ore using tweezers and spatulas, after which it was directly applied to the glass plate with the aid of high purity water, as described above. Further manipulation, such as fragmenting or milling the flakes, had the potential to modify the inherent structure of the graphite. Given that all the graphite flakes examined were sourced from processed flotation concentrates, that were milled and ground before, the resulting data remained comparable.

5.2.5. Preparation for Carbon Isotope Determination

The standard sample preparation method for measuring carbon isotope ratios involves placing the material within small tin capsules, which are subsequently subjected to combustion during the analytical procedure to liberate the material for analysis. It is inherent that the material is completely carbonate-free, as the presence of carbonates could potentially introduce variations in the carbon signature.

Highly pure graphite, either in the form of concentrates, or manually extracted from raw ores using tweezers or spatulas, is weighed into these pure tin capsules. The quantity of material typically falls within the range of 0.4 to 0.6 milligrams. Since individual flakes do not consistently possess the same weight, and each sample may not contain an identical quantity of material, any deviation from the prescribed amount of material (0.4 – 0.6 mg) is strictly prohibited.

Tin capsules, sample holders, a high-precision balance, spatula and tweezers, as well as an additional sample holder for the filled capsules, were the necessary tools and materials for sample preparation (Figure 14).

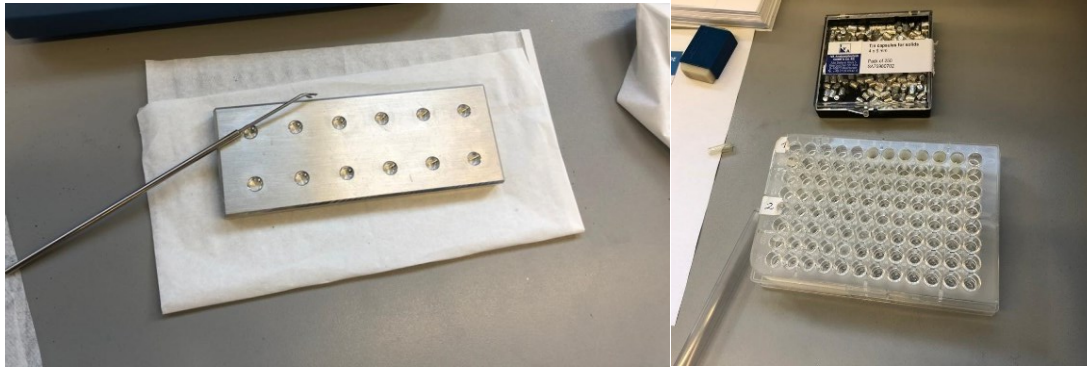


Figure 14: left: Sample holder and a special spatula for the filling process and right: Necessary tools for the preparation as tin capsules and sample holder for final preparations

5.2.6. SEM Specimen Pin Stubs

For the analysis of graphite concentrates using SEM, loose concentrate was applied to stub holders, to which a type of self-adhesive sticker had been previously affixed. This allowed the concentrate to adhere to the holder. In order to facilitate imaging through scanning electron microscopy and the associated visualization, the material on the specimen holder needs to be rendered conductive. This is achieved through a carbon coating, although it did not work effectively for graphite. An attempt was made to capture images and perform analysis without additional coating (conductivity), as graphite is a conductive material already. However, the resulting imaging was insufficient and blurred. Consequently, a shift was made to gold coating with a gold sputter vacuum coater (Agar Sputter Coater) at the Department of Functional Ceramics (Figure 15).

The gold coating parameters were defined as follows - working distance: 3 cm, electric current: 30 mA, time: 30 sec.



Figure 15: Gold sputter vacuum coaters for SEM

5.2.7. Polished Sections/Mounts

In the context of graphite imaging via light microscopy and its application for LA-ICP-MS, the creation of polished sections (Figure 16) was necessary and presented a notable challenge due to the softness of graphite, which contrasts with the considerably harder minerals present in the raw ore samples. The procedural approach involved the following steps assuming the material already being completely dry:

- ✓ Embedding samples in epoxy resin (epoxy casting resin "water-clear" and hardener W300 in a ratio of 100:30); curing time minimum 3 days
- ✓ Removing air from samples and resin in a vacuum chamber (Heraeus)
- ✓ Grinding with a 125 μm diamond grinding wheel

- ✓ Grinding with a 40 μm diamond grinding wheel
- ✓ Grinding with a 10 μm diamond grinding wheel
- ✓ Lapping: ATM Alpha polishing cloth and 120 mesh (approximately 9 μm) aluminum oxide abrasive
- ✓ Polishing Step 1: DP-Pan polishing cloth, 350 mm diameter, with 3 μm diamond suspension
- ✓ Polishing Step 2: MD-Dur polishing cloth, 350 mm diameter, with 1 μm diamond suspension
- ✓ Polishing Step 3: Buehler Metaserv polishing machine, Mambo Chen polishing cloth, 200 mm diameter, with 0.05 μm aluminum oxide polishing abrasive

Due to the softness of graphite, the polishing process was demanding, resulting in frequent incidents of breakage and relief occurrences and not achieving a perfectly flat and uniform polish, as typically attained with conventional rock samples.

In some cases, graphite flakes exhibited such low density that they failed to settle during the thick section preparation, remaining suspended and thus not emerging on the surface of the final thick section. Careful resin pouring ensured that the pre-introduced concentrates remained at the bottom and subsequently became visible on surface of the mount.



Figure 16: Graphite concentrates (left image) and raw ore (right image) embedded into epoxy resin

5.2.8. Sample Digestion

For the solution-based-ICP-MS analysis the ions of interest in the graphite samples had to be brought into a solution. Consequently, the development of a sample preparation and measurement procedure for ICP-MS measurement emerged as a pivotal component of this research study. The high resistance of graphite to digestion was a primary consideration and resulted in the main challenge for multi-element analysis. The aim of extensive laboratory experiments was to develop a complete digestion procedure method, for the natural, geogenic graphite samples. Understanding the elemental composition of carbon-rich samples, whether of geogenic, biogenic, or technological origin, is crucial for advancing technical applications and identifying their origin through elemental fingerprinting. The methods used for digestion are both microwave digestion procedures and both convert C to CO_2 . In the MIC digestion procedure, the material is combusted in pressed form under oxygen in quartz glass vessels. In the MW methodology, the digestion procedure is performed in PTFE vessels with the sample material in loose form without the addition of oxygen.

High purity water (HPW) water was sourced from a Milli-Q element module (18.2 $\text{M}\Omega\text{ cm}$; Merck Millipore, Darmstadt, Germany). Nitric acid (HNO_3 , w = 65 %, p.a. grade; Carl Roth GmbH, Karlsruhe, Germany) was purified in perfluoroalkoxy-polymer (PFA) sub-boiling units (DST-1000 and DST-4000, Savillex, Eden Prairie, MN, USA). Hydrogen peroxide solution (H_2O_2 , w = 30 %; Merck KGaA, Darmstadt, Germany) for digestion assistance were purchased in ultra-pure quality. All plastic

consumables underwent cleaning by vaporization in HNO₃ (w= 65 %), followed by thorough rinsing with ultra-pure water.

5.2.8.1. Literature Review on Graphite Digestion Methods

As part of the preparation process for experiments aimed at achieving complete digestions of graphite material and to gather insights on how to tackle this challenge, a literature review on graphite sample preparation for various analytical methods was conducted. So far, graphite has not been dissolved for the purpose of element analysis for proof of origin methods. However, it has been addressed in the context of various technical and technological inquiries related to carbon-rich materials.

Synthetic graphite powders with varying purities were thoroughly analyzed by Bögershausen in 1997 using a range of techniques. ICP-MS was employed for the determination of elements including Al, Co, Cu, Cr, and Pb. The analysis involved a microwave digestion process and the addition of specific acids. The study aimed to establish reference values rather than compare methods and laboratories. Notably, techniques capable of direct solid analysis (INAA, XRF, and DC-OES) demonstrated superior consistency and fewer outliers compared to methods such as ICP-MS, ICP-OES, FAAS (flame atomic absorption spectroscopy), and ETAAS (electrothermal atomic absorption spectroscopy), which required various wet chemical sample preparation steps. The quality of the complicated sample preparation significantly influenced the overall accuracy and precision of the results. Among the acid digestion techniques, using water-free nitric acid and concentrated sulfuric acid produced the clearest solutions, while other methods left some solid particles behind (Bögershausen et al. 1997).

A digestion technique based on microwave-induced combustion, tailored for organic samples such as bovine liver, pig kidney, and skim milk was established and tested in 2004. The sample digestion was described as a vulnerable point in analytical processes, susceptible to errors, especially in trace element determinations, with potential systematic errors as high as 100%. Closed-system microwave digestion has gained widespread acceptance as an efficient sample preparation method. It reduces digestion time and reagent usage, minimizes contamination and loss of volatile substances and provides safety. The use of concentrated mineral acids in these systems may elevate blank values due to contaminant elements. High acid concentrations may also pose challenges for certain analytical techniques, necessitating a subsequent step to remove or dilute excess acid (Flores et al. 2004).

A review from 2007 focuses on combustion, oxygen flask, and microwave-induced combustion (MIC) techniques, discussing their operational conditions, characteristics, and limitations. MIC techniques are highlighted, particularly for samples challenging to digest using traditional microwave-assisted wet digestion, such as coal, and for subsequent halogen determination. Traditional techniques based on microwave-assisted acid digestion have certain limitations for specific applications, primarily due to the stability of organic materials like graphite, which hinders efficient digestion. In such cases, combustion methods emerge as the most effective and convenient approach to decompose organic materials. Combustion techniques, driven by high temperatures, can efficiently decompose almost all organic matrices, minimizing interference with subsequent analytical determinations. Moreover, there is a growing need for non-metal determination in various samples, where conventional acid digestion techniques are less suitable. In summary, sample digestion techniques can be broadly categorized into two major groups: wet digestion and combustion techniques (Flores et al. 2007).

The challenges associated with digesting graphite and the critical need for impurity control in industrial applications was assessed by Cruz et al. in 2015. Microwave-induced combustion as a method for graphite digestion and the subsequent determination of multiple elements was proposed. This approach involved burning a relatively high mass of graphite (400 mg) using a pressurized system with oxygen and diluted nitric acid solution. Remarkably, it achieved a high digestion efficiency (over 99%) and

excellent recovery rates for various elements, even with relatively low concentrations of nitric acid. The results were confirmed through both inductively coupled plasma optical emission spectrometry (ICP-OES) and inductively coupled plasma mass spectrometry (ICP-MS). Traditional combustion methods using sample pellets and filter paper led to inconsistent combustion and suspended solid residues, highlighting the unique challenges posed by graphite's resistance to oxidation, particularly at atmospheric pressure (Cruz et al. 2015).

Microwave digestion advantages were reported in another publication in terms of efficiency, reagent economy, and reduced risk of contamination and elemental loss. However, it is acknowledged that microwave digestion remains time-consuming despite these benefits (Botelho et al. 2001).

The MIC digestion method was identified as a suitable method for digesting up to 100 mg of the graphite sample. However, microwave-assisted digestion and dry-ashing methods resulted in insoluble residues and analyte losses. To ensure complete oxidation of the graphite material matrix, cellulose should be utilized as a combustion aid during MIC digestion. For MIC digestion, two independent absorbing solutions (4 mol L⁻¹ HNO₃ and inversed aqua regia) were necessary for the quantitative recovery of embrittling elements. The limits of detection achieved through MIC, coupled with ICP-OES and FI-CVG-AAS, were satisfactory for quantifying all potentially embrittling elements (Enders et al. 2016).


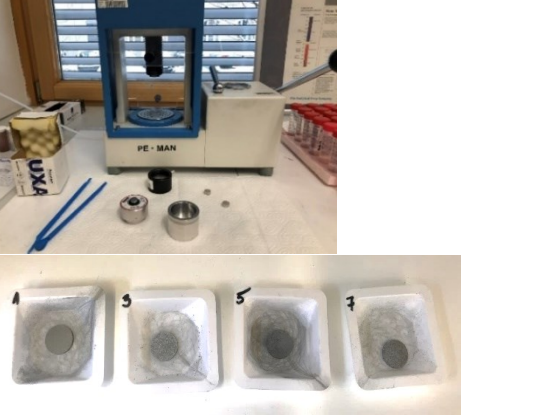
Nuclear-grade graphite is the core structural material for the high-temperature gas-cooled reactor systems. The quantification of trace and ultra-trace elements in nuclear grade manufactured graphites by fast flow glow discharge mass spectrometry and by inductively coupled plasma mass spectrometry after microwave induced combustion digestion have been successfully tested (Wang et al. 2010).

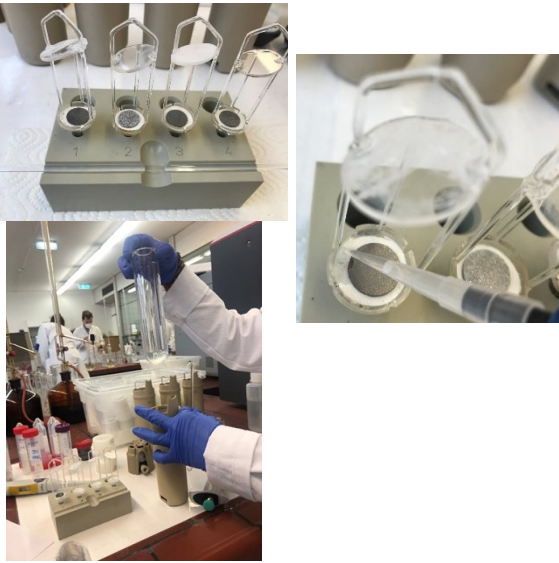

The analysis methods presented in the literature, such as FTIR and TGA, are indeed applied to graphite materials. However, they primarily serve the purpose of identifying the carbon content rather than the trace or minor elements in graphite (Zethof et al. 2019), and thus, they are not relevant for this study.



In 1960, Janda and Schroll analyzed various elements in graphite and graphite-containing rocks using an emission spectrographic method. They found that with increasing crystallization degree and generally higher carbon content, the concentration of trace elements decreases (Janda and Schroll 1960).

5.2.8.2. Microwave Induced Combustion Digestion Procedure

Table 9: Combustion digestion procedure

1	Sample material	For the systematic comparison of different digestion procedures four samples had been chosen for method development. Also, the certified reference material BAM-S009 (<i>“Medium Purity Graphite Powder”</i>) was used for method validation in this study. All graphite concentrate samples were available as loose powders.	
2	Pellet Pressing	To place the samples in the quartz glass sample holder, the samples needed to be formed into a pellet shape and mixed beforehand with high-purity cellulose as a combustion aid in a 1:1 ratio. The pellet had to be pressed firmly enough with the cellulose to be stable in the MIC sample holder. If the pellet experienced too much pressure and became too firm, the complete combustion process would no longer work. Pressing was performed at 5 t contact pressure for max. 60 sec with a mixture of 100 mg powdered sample and 100 mg cellulose.	

3	<p>Preparing the sample holder and vessel</p>	<p>The sample holders made of quartz glass were covered with cellulose filter paper (Macherey-Nagel, Grade MN 1640 de), which was precisely cut to the size of the sample tray. The samples were placed in the quartz glass tray on the filter paper. The filter paper around the sample was moistened with with 50 μl NH_4NO_3 solution (50 % w/v). Ammonium nitrate was used as a combustion aid (oxidizer).</p> <p>The sample holder was placed into a quartz vessel containing 6 mL of 5 % nitric acid (or 8 mL 50% v/v).</p>	
4	<p>Gas-loading</p>	<p>The vessels were sealed with a PTFE TFM lid and placed into the rotor (Rotor 8NXQ80, Multiwave Pro, Anton Paar). The vessels were prior filled with 20 bar oxygen (5.0) using a four-way valve station. When microwave irradiation is applied, the impregnated filter paper in the vessels, and thus the samples, are ignited by the microwave energy.</p>	

5	Digestion process	<p>The microwave program was set to a maximum temperature of 280 ° and 80 bar, which was reached within a ramp time of 10 min and then held for 14 min. The maximum microwave power was 1800 W. After completing the digestion procedure and cooling down to 55°C, the vessels were vented, opened and their contents were transferred into 50 mL PP tubes. Successful digestion runs resulted in completely digested samples or an incomplete digestion leaving residues in the form of solid particles at the sample holder or in the solution. The operational conditions and the heating program followed the recommendations from the microwave oven manufacturer (Anton Paar, Graz, Austria).</p>	
6	ICP-MS analysis	<p>Multi-element analysis using a 7500cx Agilent ICP-MS for the measurement of the diluted digestion solutions.</p>	

Following numerous tests aiming to achieve a complete dissolution of graphite material and subsequent analysis of the produced solutions, it has become apparent that the combustion process is not reproducible. It cannot be excluded that elements may be lost during the digestion procedure and degassing of the vessels, however, there was no way to verify this. The process does not facilitate the capture of the escaped gases.

During the MIC combustion digestion procedure, the following observations were made:

- *The digestion efficiency depends on the crystallinity and the grain sizes (amorphous (semi-graphites) are easier to digest, as well as the Reference Material BAM S009, which is a semi-graphite)*
- *Microwave induced combustion leads to a more complete digestion, however digestion results were not reproducible and could not be successfully applied to each graphite concentrate (because of crystallinity and flake size differences)*

Hypothesis: Digestion rate depends on the structure and morphology of the carbon (Bandoniene et al. 2023)

5.2.8.3. Conventional Microwave Digestion Procedure

An Anton Paar Multiwave PRO closed-vessel digestion system, equipped with a 24HVT50 rotor (Anton Paar, Graz, Austria) and 30 mL PTFE vessels, was employed for the microwave digestion of natural graphite concentrates. Approximately 0.2 g of graphite sample was weighed into PTFE digestion vessels directly. After adding the chemicals, the microwave program was set to a maximum temperature of 200 °C, reached within a ramp time of 10 min, and held for 15 min. The maximum microwave power was 1500 W (Trimmel et al. 2023). After cooling down to 55 °C, the program concluded, and the vessels were vented and opened. The contents were transferred into 50 mL PP tubes, rinsed with ultrapure water to obtain a total digest volume of about 40 mL. The tubes were weighed empty and after adding the digest solution to determine exact masses.

The used reagent mixture was as follows: 5 mL HNO₃ and 1 mL H₂O₂ applied to each sample material. To determine the reproducibility of the digestions and eliminate outliers, three digestions of each sample were prepared in different digestion sessions. Alongside each batch, two procedural blanks were processed in the microwave using the same procedure and the same reagents. One cleaning digestion run with 5 mL of HNO₃ in each vessel was conducted between batches.

5.2.8.4. Comparison of the digestion procedures

Both digestion procedures were compared for five graphite samples and the certified reference material (BAMRM) for two elements (Figure 17). The digestions generated by conventional microwave procedure (MW) show higher precisions in general, compared to digestions from the microwave induced combustion digestion procedure (MIC). To ensure reproducible digestions and analyses, all samples must ultimately be processed with the same method. Since this was not feasible with the MIC method, the sample preparation for solution-based ICP-MS via MW was chosen.

BAM S009 (BAMRM) was digested using both methods. Results from MW and MIC for the elements Cr and Zn are quite close to the certified value for the respective element (Cr and Zn). Element concentrations for other elements with lower concentrations (in BAMRM) could not be reproduced properly (due to blank values higher than certified element concentrations).

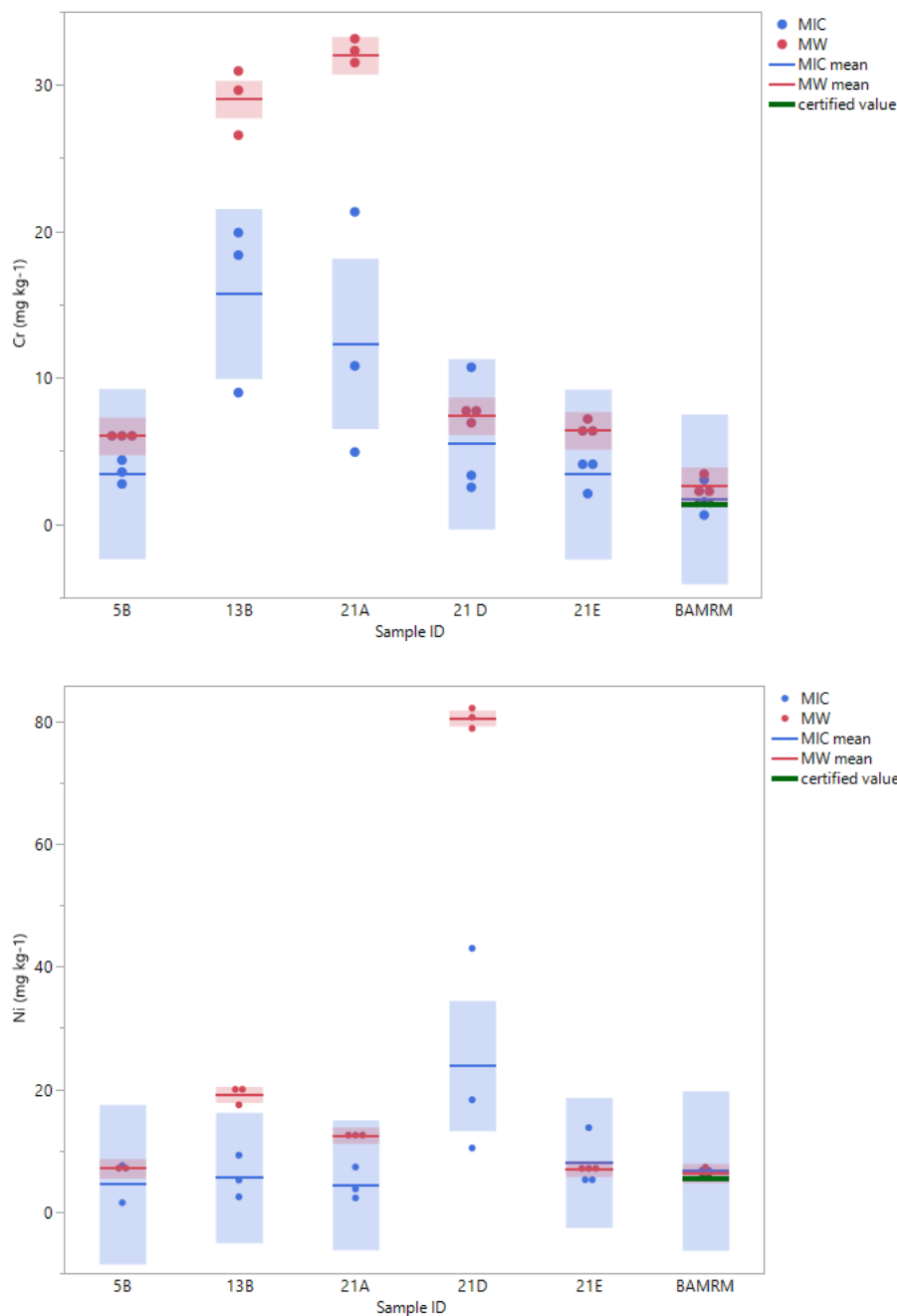


Figure 17: Mass fractions ($n=3$) of selected trace elements (Cr and Ni) in graphite concentrate samples and in certified reference material (BAMRM) BAM-S009 determined using solution-based-ICP-MS analysis after different digestion methods (conventional MW – red, MIC – blue)

5.3. Analytical Methods and Instrumental Procedure

5.3.1. Overview of the Analytical Methods

A broad range of analytical methods are used in natural sciences such as chemistry, metallurgy, archaeometry and geosciences. A common application in the latter is quantifying the composition of minerals and rocks, spanning from major elements in the percentage range to trace elements in the parts per million range. Isotope compositions and ratios often play an important role in these analyses. Simultaneously, diffraction techniques are essential for phase analysis and determining the geometric arrangement of atoms, ions, or molecules in minerals. Conventional microscopic methods are complemented by high-resolution scanning and electron microscopy. A wide array of spectroscopic

methods, employing the full electromagnetic spectrum, are deployed for investigating chemical bonding in minerals and crystals, as well as resolving structural issues. In this chapter, the analytical instruments applied for graphite are listed, in five groups, which are based on the excitation energy.

Table 10: Overview of the analytical methods used for the Analytical Proof of Origin of natural graphite divided into 5 groups based on their excitation energy

Group 1: Emission and mass spectrometric techniques	Group 2: Electron beam methods	Group 3: X-ray methods	Group 4: laser induced methods	Group 5: Imaging and visual analysis
Inductively coupled plasma-mass spectrometry (ICP-MS)	Scanning electron microscopy (SEM)	Handheld XRF	Laser-ablation inductively coupled plasma-mass spectrometry (LA-ICP-MS)	(Digital) light microscopy
Gas isotope ratio mass spectrometry (Gas-IR-MS)			Raman spectroscopy	

5.3.2. LA-ICP-MS (Laser-Ablation Single Collector Inductively Coupled Plasma Mass Spectrometry)

Laser ablation is used for quantitative analysis of major, minor and trace elements in solid samples without the necessity of dissolving the sample. Performance and application type depend on the choice of laser and mass spectrometer. Available options include quadrupole (Q), sector-field (SF), and multi-collector (MC) mass spectrometers. Additionally, time-of-flight (TOF) instruments are becoming increasingly important.

Laser ablation single collector ICP-MS analyses of natural graphite ore and concentrates were performed at the Geological Survey of Finland (GTK), using a Nu AttoM SC-ICP-MS (Nu Instruments Ltd., Wrexham, UK) coupled with an Excite 193nm ArF laser-ablation system (Photon Machines, San Diego, USA). The mass spectrometer was tuned with NIST612 glass using a 40 % output on 5mJ, a repetition rate of 10 Hz, a circle spot of 50 μm , yielding around 500,000 to 600,000 cps. The laser was run at a pulse frequency of 10 Hz and a pulse energy of 5 mJ to produce an energy flux of about 10-15 J/cm² on the sample surface in a line raster mode of 135 μm circle spot. Prior to analysis, each line was preablated with a rastering at 30 $\mu\text{m/s}$. With these instrument parameters, about 13,000,000 cps can be obtained on a baseline of 5,000,000 cps. Each analysis was initiated with a 20 second baseline measurement followed by switching on the laser for 60-90 seconds for signal acquisition. These analyses were made using time resolved analysis (TRA) with continuous acquisition of data for each set of lines (generally following the scheme of 2 primary standards and 15 unknowns). An in-house synthetic pressed nanopellet of graphite standard Gh20220218 was used for external standardization (the preparation process is described separately below). The isotope ¹³C has been used as an internal standard assuming a content of 98 wt % carbon. The measurements were performed on 68 isotopes covering 61 elements at low resolution ($\Delta M/M = 300$) using the Fastscan mode. Data reduction was handled using the software GLITTER™ (Van Achterbergh et al. 2001) which allows the baseline subtraction, the integration of the signal over a selected time resolve area and the quantification using known concentrations of the external and internal standards. Due to unavailability of a second good in-house pressed pellet as a quality control, the analytical precision and accuracy was monitored by treating Gh20220218 as an unknown. The results suggested a relative standard deviation (RSD) of 10-40 % for all elements except Li, Ni, Ba, Bi and an accuracy of ± 5 -10 % for most elements except for Li, Fe, Ni, Ba, and Bi.

The standard was prepared by spiking a high purity (≤ 0.0002 % total impurities) natural graphite powder (200 mesh) with multielement solution from LabKing. The spiked powder was then soaked with MilliQ water to make it slurry, which was then dried on a hot plate at 70 °C. The dried material was then grinded and pressed to a pellet. This process can produce relatively homogeneous element distributions.

The LOD values for the LA-ICP-MS methodology were determined for graphite concentrates and the corresponding pressed pellets. LOD values for graphite raw ore is not reported due to the highly variable grain sizes of the graphite in raw ore which required different spot sizes for each analysis/raw ore. LOD values were determined as the point where signal from the analyte becomes differentiable from inherent noise/background in the system and is defined as the lowest concentration of an analyte in a sample that can be detected reliably. The LOD value for the LA-ICP-MS methodology is composed of the mean value of all LOD values reported for each respective element/analyte. Element concentrations below detection limit were replaced by 0.5 times the detection limit for the respective element.

Table 11: LOD values for LA-ICP-MS analysis for graphite concentrates

LA-ICP-MS	concentrate
Analyte	LOD $\mu\text{g/g-1}$
Mg	0.0019
Al	0.0119
Ca	0.7314
V	0.0054
Cr	0.1059
Mn	0.1446
Fe	0.8656
Co	0.0403
Cu	0.0369
Zn	0.0891
Ga	0.0050
As	0.0384
Rb	0.0122
Sr	0.0014
Y	0.0010
Zr	0.0004
Ce	0.0003
Th	0.0006
U	0.0002

5.3.3. ICP-MS (Inductively Coupled Plasma Mass Spectrometry)

The trace element concentrations in the graphite concentrates were determined using an Agilent 7500cx ICP-QMS (Agilent Technologies, Tokyo, Japan) equipped with a 100 μL PFA nebulizer and a quartz glass spray chamber cooled to 2 °C. Isotopes at given m/z which were affected by spectral interferences (V, Cr, Mn, Co, Ni, Cu, Zn, As) were measured with He as collision gas, where most isotopes could be measured without significant interferences. Signal intensities on m/z 24 (Mg), 27 (Al), 43 (Ca), 51 (V), 53 (Cr), 55 (Mn), 57 (Fe), 59 (Co), 62 (Ni), 65 (Cu), 66 (Zn), 71 (Ga), 75 (As), 82 (Se), 85 (Rb), 88 (Sr), 97 (Mo), 107 (Ag), 111 (Cd), 137 (Ba), 205 (Tl), 207 (Pb), 238 (U), 89 (Y), 139 (La), 140 (Ce), 141 (Pr), 145 (Nd), 146 (Nd), 147 (Sm), 149 (Sm), 151 (Eu), 160 (Gd), 159 (Tb), 161 (Dy), 163 (Dy), 165 (Ho), 166 (Er), 167(Er) 169 (Tm), 172 (Yb), 173 (Yb), 175 (Lu), 232 (Th) were measured.

The establishment of an external calibration curve for the quantification of ICP-MS signals offers an extensive range of calibration solutions available in different concentrations for all elements, encompassing the REE. This versatility enables the calibration of the method, even with the absence of dedicated reference or calibration materials tailored for graphite itself. General calibration solutions designed for ICP-MS applications are universally adaptable and applicable also to this study.

The ICP-MS multi element standard solution Merck VI (Merck Certipur, Darmstadt, Germany) featured the following element concentrations: Ag, Al, Ba, Bi, Cd, Co, Cr, Cu, Ga, In, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Rb, Sr, Te, Tl, U and V, with 10.0 mgL^{-1} , As, B, Be, Fe, Se, Zn with 100 mgL^{-1} , Ca with 1000 mgL^{-1} effective certified concentrations diluted in HNO_3 ($w=6 \%$). 40 ng mL^{-1} stock solution for Ag, Al, Ba, Bi, Cd, Co, Cr, Cu, Ga, In, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Rb, Sr, Te, Tl, U, V, 400 ng mL^{-1} for As, B, Be, Fe, Se, Zn and 4000 ng mL^{-1} for Ca of ICP multi-element standard solution Merck VI in HNO_3 ($w=1 \%$) was used for a calibration. Calibration ranges were diluted accordingly from $0-1 \text{ } \mu\text{g g}^{-1}$, $0-10 \text{ } \mu\text{g g}^{-1}$ and $0-100 \text{ } \mu\text{g g}^{-1}$ for the respective groups of elements.

Given the uneven distribution of Rare Earth Element (REE) concentrations, with higher concentrations of light REE compared to heavier REE, and higher concentrations of even-numbered elements compared to odd-numbered elements as per the Oddo–Harkins rule, a custom-made calibration solution was used. This solution (REE multi-element standard AHF-CAL-7, Inorganic Ventures, NJ, USA) featured the following REE concentrations: $1000 \text{ } \mu\text{g mL}^{-1}$ Ce, $500 \text{ } \mu\text{g mL}^{-1}$ La, Nd, Y, $150 \text{ } \mu\text{g mL}^{-1}$ Th, $100 \text{ } \mu\text{g mL}^{-1}$ Pr, $50 \text{ } \mu\text{g mL}^{-1}$ Dy, Gd, Sm, U, $20 \text{ } \mu\text{g mL}^{-1}$ Er, Eu, Yb, $10 \text{ } \mu\text{g mL}^{-1}$ Ho, Tb, and $5 \text{ } \mu\text{g mL}^{-1}$ Lu, Tm in 7% (m/v) HNO_3 . A 40 ng mL^{-1} stock solution (calculated for Ce) was prepared and diluted with 1% HNO_3 to establish a calibration range from 0 to 40 ng mL^{-1} .

$100 \mu\text{L}$ portions of a 100 ng mL^{-1} In and Re internal standard solutions (CertiPur, Merck Darmstadt, Germany) were added to 5.0 mL of each sample solution and to the calibration solutions to correct instrumental drift. Three independent digestion replicates were analyzed for each sample. Additionally, total procedural blanks, containing the same amount of concentrated HNO_3 and H_2O_2 used for sample decomposition, prepared in the same manner as the samples, were analyzed. The calibration for this analytical method offers an extensive range of calibration solutions available in various concentrations for all elements, including the REE. This versatility enables the calibration of the method, even when working with graphite digests, despite the absence of dedicated reference or calibration materials tailored for graphite. Calibration solutions designed for ICP-MS applications are universally adaptable and applicable.

Two independent analysis runs were conducted, one for the analysis of non-Rare Earth Elements (Mg, Al, Ca, V, Cr, Mn, Fe, Co, Cu, Zn, Ga, As, Se, Rb, Sr, Mo, Ag, Cd, Ba, Tl, Pb, U) and the other for REE (Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Th, U). The mean of mass fraction of two isotopes was calculated for Nd (145, 146), Sm (147, 149), Dy (161, 163), Er (166, 167) and Yb (172, 173). The most commonly used REE mass fractions for normalization are derived from chondrite data (for bulk Earth, Earth's mantle rocks, and most magmatic rocks) and shale composite values, such as PAAS (post-Archean Australian shale, representing the average present-day continental crust), for normalizing crustal sediments and soils. In this study, the European shale composite (EUS) was used for normalization (Bau et al. 2018b).

The LOD and LOQ values were determined using six procedural blanks with LOD three times the standard deviation and LOQ ten times the standard deviation of the mean procedural blank concentrations.

Table 12: LOD and LOQ values for solution-based-ICP-MS analysis for graphite concentrates for each analyte

ICP-MS Analyte	LOD $\mu\text{g/g-1}$	LOQ $\mu\text{g/g-1}$
Mg	0.922	3.07
Al	1.37	4.56
Fe	2.16	7.21
Cu	0.366	1.22
Zn	1.82	6.05
As	0.431	1.44
Se	0.399	1.33
Mo	0.625	2.08
ICP-MS Analyte	LOD ng/g-1	LOQ ng/g-1
Mn	194	647
V	5.52	18.40
Cr	22	74
Ni	107	356
Ga	4.71	15.70
Rb	41	135
Sr	18	60
Ag	8.10	27
Cd	10	32
Ba	44	147
Tl	18	60
Pb	52	174
Y	3.98	13
La	20	67
Ce	27	89
Pr	0.656	2.19
Nd	2.625	8.75
Sm	0.703	2.34
Eu	0.158	0.526
Gd	0.342	1.140
Tb	0.0633	0.211
Dy	0.361	1.210
Ho	0.087	0.289
Er	0.248	0.828
Tm	0.0491	0.164
Yb	0.213	0.711
Lu	0.0015	0.005
U	2.57	8.58

5.3.4. Raman Spectroscopy

Raman spectroscopy is used to evaluate the lattice ordering of graphite. Raman spectra were obtained using a Horiba Labram HR Evolution instrument at the Chair of Resource Mineralogy equipped with a 100mW Nd:Yag (532nm) laser, a confocal microscope (hole aperture=100 μm), a 1800 g/mm grating and a Peltier cooled CCD detector. Ca. 5-10 % of the full laser power of 100 mW was applied in order to avoid sample damage. For laser focusing and sample observation, an Olympus BX 40 microscope with a 100x objective lens was employed. In order to enhance signal-to-noise ratios, two scans were accumulated with an acquisition duration of 20 seconds within two distinct spectral regions: 700–2000 cm^{-1} (first-order) and 2200–3200 cm^{-1} (second-order). In total, 10–20 spectra were recorded for each sample and the calibration of band positions was achieved using a silicon wafer standard.

5.3.5. Microscopy

5.3.5.1. Digital Light Microscopy

Graphite concentrates were analyzed and documented with a KEYENCE VHX 6000 digital light microscope.

Samples from raw ores were analyzed using a Keyence VHX-7000 equipped with an EA300 LIBS analyzer at GTK in Espoo, Finland. This allowed for the direct determination of mineral phases in the raw ore samples.

5.3.5.2. Scanning Electron Microscopy

This microscopical method is mainly used for the assessment of microstructural characteristics. Non-destructive electron beam-based methods such as SEM offer the choice of simultaneously imaging and analyzing the chemical composition of points and areas in graphite samples. The ZEISS EVO10 SEM at the Chair of Geology and Economic Geology was operated in standardless mode at 15 kV acceleration voltage and 8 seconds measurement time per spectrum. Detection limits using this setup are in the order of 0.1 wt%.

5.3.6. Gas-IR-MS (Gas Isotope Ratio Mass Spectrometry)

0.5 mg of graphite samples were placed into tin foil containers and subjected to combustion using an elemental analyzer (Flash EA 1112) at a temperature of 1020 °C, while ensuring an excess of oxygen. The resulting CO₂, meticulously isolated through column chromatography, underwent real-time analysis employing a ThermoFisher DELTA-V isotope ratio mass spectrometer. The isotope ratios (¹³C/¹²C) of the CO₂ were subsequently cross-referenced with those of a monitoring gas, calibrated against the Vienna-Pee Dee Belemnite (VPDB) standard. The overall analytical procedure is reproducible within the range of 0.2–0.4 ‰.

5.3.7. Standard and Reference Materials

Reference materials are used as benchmarks to validate methods and ensure measurement accuracy, whereas standard materials, also known as calibration materials, are used for instrument calibration and measurement standardization. For the present study both were required (Tab. 13).

Table 13: Various standard and reference materials used for this study

Name	Description	Type	Parameter	Method	Comment
AHF CAL 7	REE multi-element standard, Inorganic Ventures, NJ, USA	Custom made standard material	REE	ICP-MS	For REE in solutions only
BAM S009	Medium purity graphite powder, Bundesanstalt für Materialforschung, Germany	Certified reference material	Trace elements	ICP-MS, LA-ICP-MS	TE values are too low and below LOD
Gh20220218	Spiked high purity graphite powder	Reference Material (in-house from GTK)	Trace elements	LA-ICP-MS	In-house material for TE analysis
MERCK IV	Multi-element standard solution	Standard Material	Trace elements	ICP-MS	For Digestion analysis only
NIST 610/612	Multi-element glasses	Standard Material	(Trace) elements	LA-ICP-MS	Instrument Calibration LA-ICP-MS
Si wafer	Artificial Si-monocrystal	Standard material	Crystallinity	Raman spectroscopy	Instrument Calibration

5.4. Statistical Methods

For the present study, the workflow proposed by (Frenzel 2023) was used as a guideline starting from the conceptual preparation of the work, followed by data preparation and data analysis. The general workflow proposed is illustrated below (Figure 18).

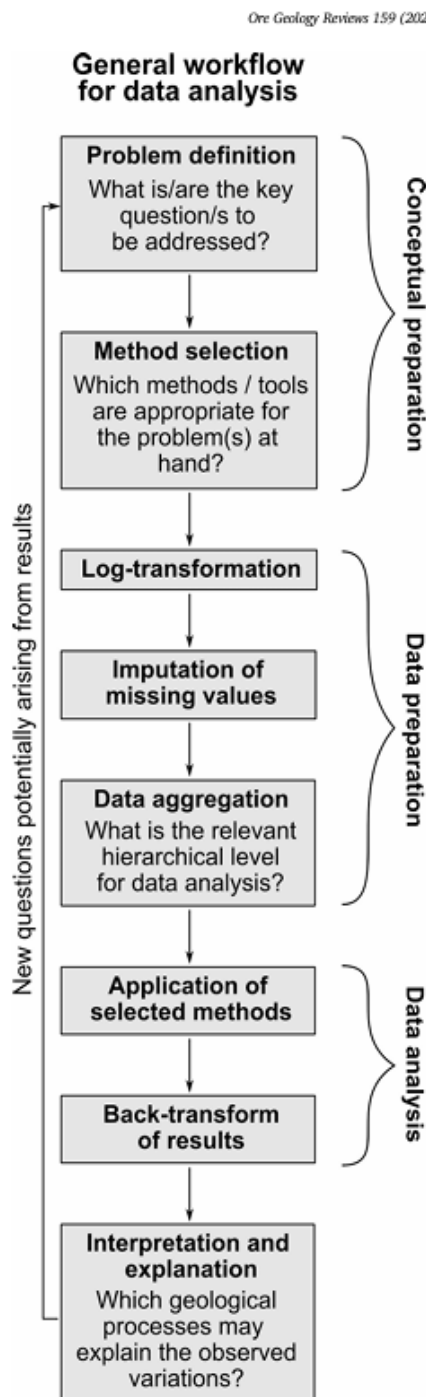


Figure 18: General workflow for data analysis (Frenzel 2023)

I. CONCEPTUAL PREPARATION

Problem definition:

- (How) Can natural graphite concentrates be differentiated based on analytical methods? What are the parameters that allow differentiation?
- Can the analytical methods, that are usually applied to mineral phases also be applied to graphite concentrates?
- How do material properties influence the applicability and feasibility of the method?

Method selection:

- Which analytical methods can be used to identify parameters based on which graphite can be differentiated?
- Is the analytical method developed for the application in graphite?
- What needs to be done to apply the analytical methods to the graphite concentrates in form of loose powders?
- What is the respective sample preparation?
- Are there any standard, reference or calibration materials available?

II. DATA PREPARATION

Log transformation:

Within the data types present in this study, the log transformation only had to be done for trace element data. The explanation and an example about data distribution before and after log transformation is provided in chapter 5.4.1.

Imputation of missing values:

The imputation of missing values was only relevant for the trace element data. Missing values in LA-ICP-MS data were supplemented by an average measurement value from the value before and after in the measurement series of the same sample. Elements with more than just individual values missing were excluded from this study and are not reported. Solution ICP-MS data for the selected elements showed no individual missing values. Elements that have not provided measurement results at all were excluded from this study.

Data aggregation and relevant hierarchical level:

As the aim of this work is the geographical differentiation of deposits, the main interest in the hierarchical level of the data is the mine/province level of the data. Most samples could be assigned to

one individual mine, however for some locations material aggregates could only be obtained. This is quite common because processing plants aggregate material from different mines in one place. Samples from China, Madagascar and Brazil are affected. All other samples refer to the mine site directly. Other hierarchical levels in this study are: continent and country of origin, organic vs. non-organic precursor material of the graphite (organic vs. hydrothermal), as well as vein, flake and amorphous graphite.

III. DATA ANALYSIS

Application of the selected methods for data analysis:

Data analysis included describing the trace element data by reporting median values and standard deviations, as well as using scatter and box plots. For trace element data also PCA was applied. Data classification was conducted using LDA. For prediction purposes, the database still needs to be enlarged, as this study aimed at method development. More data for each parameter, as well as more samples per mine/province need to be obtained to enable reliable predictions of the geographic location for unknown samples. An outlook on data prediction is provided in the chapter on carbon isotopes. Log-transformed data is used for multivariate data analysis of trace element data.

IV. INTERPRETATION AND EXPLANATION

Obtained data from the analytical methods were primarily used to identify certain patterns for each location to differentiate within the individual graphite samples and find common features for each location (mine/province).

5.4.1. Data Types and Structure

The choice of the applied statistical methods depends on the dataset, the number of samples and the specific features of the data (e.g. trace elements, isotope ratios and Raman spectra). Different types of data need to be treated separately from each other. Data preparation after Frenzel (2023) requires the log transformation of compositional. Compositional data (e.g. trace element data) refers to data that represents the relative proportions or a composition of parts within a sum and they are constrained in that the sum of the parts always equals a constant or a fixed whole (J. Aitchison 1982). There is a significant difference between unconstrained data in real Euclidean space and compositional data in sample space. Using the wrong data pre-processing methods can lead to serious misinterpretations (John Aitchison 2002; Filzmoser et al. 2010; Raič et al. 2022). To extract the relevant information from the elements being analyzed and convert the original data into Euclidean space, compositional data is treated by log transformation. Through the log transformation, the data becomes normally distributed (Figure 19).

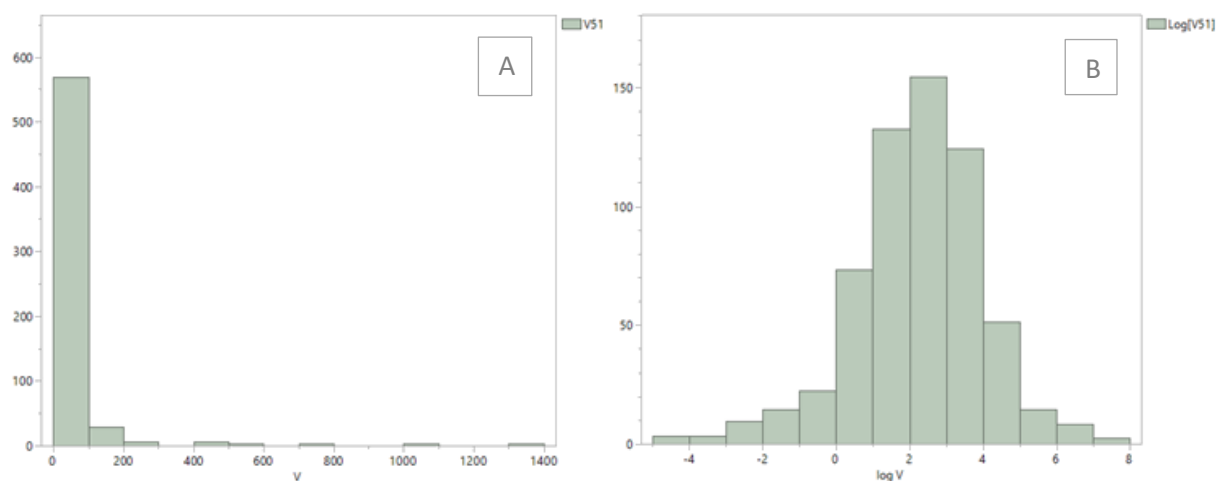


Figure 19: An example for LA-ICP-MS trace element for V - data non-log transformed (A) and LA-ICP-MS log-transformed trace element data for V (normally distributed) (B)

Carbon isotope ratios are not considered as compositional data, as they do not represent parts of a whole, but only ratios of an element or compound.

Raman spectra refer to spectral data. The information of the spectra is related to the interaction of light across different wavelengths with a sample. The relevant information in Raman spectra is dispersed across a spectrum (Hideyuki Shinzawa, Kimie Awa, Wataru Kanematsu, Yukihiro Ozaki 2009a, 2009b).

Another important part of data preparation is the treatment of missing values. Ignoring these would result in biased results (Frenzel 2023). For this study values below detection limit and values missing at random need to be differentiated. Missing individual values were observed in the LA-ICP-MS data. These missing values do not adhere with any discernible patterns associated to the sample and its properties, rather than occurring randomly. Such occurrences are typically attributed to external factors, such as human or machine errors leading to the absence of specific values during measurement (Frenzel 2023). Missing values at random for this study mainly occurred within the LA-ICP-MS dataset and were replaced by mean values from the measurement before and after from the same sample. Another category of missing values was observed in this study, which represents the below detection limit values. These were replaced by 0.5x of the detection limit value for LA-ICP-MS data. All trace element data obtained from solution-based-ICP-MS data was above detection limit, except the element Lu, which is not reported, however included in the patterns for REE for completeness.

5.4.2. Tools and Methods for the Evaluation of the Dataset

After the selection and development of the appropriate analytical method, obtaining data and pre-processing it, relevant tools and methods for the statistical evaluation had to be identified. Most research questions related to the study of mineral/raw materials composition fall into one (or more) of the following categories, which have been proposed by Frenzel (2023): Description, Classification, Prediction, Simple hypothesis and explanation/interpretation. The following categories had been assessed for the respective study.

5.4.2.1. Data Description

This category focuses on providing a descriptive data presentation approach without interpreting the data. Standard deviations and mean values of the dataset are reported, as well as displaying the individual data as histograms, scatter- and boxplots. Variables are also displayed as correlation matrix, which describes relationships between variables (Frenzel 2023). PCA is another method of data description which allows data presentation in new coordinates (Anderson 2003; Frenzel 2023).

Histograms are a graphical representation of the distribution of numerical values of a dataset, visualizing frequencies of values within different ranges. The horizontal axis of the histogram commonly represents the range of values and the vertical axis represents the frequency of observations falling within each range.

Boxplots, also known as box and whisker plots, show central tendencies, spreads and the overall distribution of the data. They are less affected by extreme values or outliers and provide a robust visualization of important statistical information, such as:

- Median: The line inside the box represents the median, which is the middle value of the dataset when it is ordered
- Quartiles (Q1 and Q3): The box itself represents the interquartile range (IQR), with the lower and upper edges representing the first quartile (Q1) and the third quartile (Q3). The IQR contains the central 50 % of the data
- Whiskers: The whiskers extend from the edges of the box to the minimum and maximum values within a specified range, typically 1.5 times the IQR

- Outliers: Individual data points beyond the whiskers are often plotted individually as dots and are considered as potential outliers

Trace element values displayed in box plots in this study show non-transformed data.

Principal Component Analysis (PCA) is a method for simplifying complex data. It reduces a large set of possibly interconnected variables to a smaller set of uncorrelated variables, helping to reveal relationships in elements (Macheyeki, Athanas S., Kafumu, Dalaly Peter et al. 2020) This technique simplifies the data but retains most of its variability (Reimann et al. 2009). The first principal component (PC1) delineates the direction of unit length that encapsulates the highest variance in the dataset, while the second principal component (PC2), orthogonal to PC1, encapsulates the maximum variance remaining within the dataset. This process can be extended to further components, each capturing maximal variance while remaining orthogonal to the other components. PCA methodology constructs an orthogonal basis that imposes a specific geometric structure on the set of variables (Dyer and Kording 2023).

5.4.2.2. Data Classification

Linear Discriminant Analysis (LDA) is widely employed for addressing dimensionality reduction challenges in the context of machine learning and pattern classification applications. For optimal separation between distinct classes, LDA finds linear combinations of features, which is achieved by minimizing variance within each class, all the while incorporating class labels as crucial information during the dimensionality reduction process (Tharwat et al. 2017).

Canonicals or LD1 and LD2 as LDA axes refer to the primary and secondary transformed space that can maximize the separation between different groups (mines/provinces) in the data. Scoring coefficients represent most relevant information from the dataset to describe and classify data (e.g. key elements).

Multivariate statistical methods analyze multiple variables (the whole dataset) simultaneously. The diagrams in chapter 6.3.1.3. and 6.3.2.3. demonstrate the limitations of bivariate statistical methods. Even ratios considering four variables simultaneously are only partially suitable for distinguishing between all geographic origins. Some reasons for the application of multivariate statistical methods include the complexity of the data. The variables are interrelated and their relationships within the group (mine/province) are complex. Additionally, multivariate methods allow for dimensionality reduction. The interpretation of numerous variables, as well as analyzing and interpreting each variable individually without multivariate methods is highly challenging and for the number of individual variables for the different methods simply not feasible. The application of multivariate statistics was useful to identify patterns within the dataset and data visualization helped to reveal patterns that may be less visible/understandable in a tabular form or based on bivariate statistics only.

LDA serves as the first application of data classification for the underlying dataset on discriminating geographic origins of natural graphite deposits. More complex statistical data classification methods (such as support vector machines (SVM), random forests (RF), gaussian mixture models (GMM), partials least squares discriminant analysis (PLSDA), as proposed by Frenzel in 2023) are not applied within the frame of this work.

Digression - PCA vs. LDA: LDA is inapplicable to unlabelled data and is a supervised learning algorithm; it classifies labelled data. PCA in contrast, ignores class labels and preserves variance across the dataset.

5.4.2.3. Data Prediction

Data prediction was applied to the carbon isotope dataset. Further analysis needs to be done, to enlarge the database and perform plausible data prediction on all parameters, as the dataset need to be split into a test and a training dataset with the respective amount of data for each group.

A logistic regression model is mainly used for prediction. The model is capable of predicting the value variables, such as the C isotopic values in the regions of Sri Lanka, Ukraine, or the Rest of the World. This is particularly useful when new data is introduced, and the corresponding output needs to be forecasted. Additionally, logistic regression models can be employed for outlier detection, quickly identifying data points that deviate from the general dataset.

5.4.2.4. Explanation and Interpretation

A separate chapter is dedicated to the discussion, explanation, and interpretation of the data in this study (Chapter 7 Discussion).

5.4.3. Data Processing Software

The following software products were used for data processing and data presentation. This includes a mix of open-source programs and licensed programs.

ioGAS-64-8.1. is a software tool designed for geoscientists, especially those working in the fields of geochemistry and geostatistics. ioGAS was primarily used for the analysis and visualization of geochemical and geological data. The program allows users to import and manage various types of geochemical and geoscientific data, such as elemental concentrations, isotopic data, and geological information. The software offers a wide range of visualization options, including scatter plots, ternary diagrams, histograms, and more, which helps to gain insights into their data by visualizing the data. It is commonly used in mineral exploration, environmental monitoring, and other geoscientific applications and is an effective tool to make data-driven decisions and to communicate and visualize the findings.

GLITTER4.5. is a data reduction software tool designed for the analysis and visualization and to provide a simple and more consistent and faster reduction of LA-ICP-MS data. It represents a tool for real-time interactive data reduction for LA-ICP-MS analysis. One of the significant advantages of this software is the capacity to identify the optimal time intervals for background and signal measurement, which is achieved through the presentation of time-resolved data and allows a rapid identification of irregularities and data spikes.

JMP (student version) is a statistical data analysis software. It is designed to explore, analyze, and visualize data to make data-driven decisions in research, business, and various other industries. It provides a user-friendly interface for data visualization, such as the generation of graphs, histograms and scatter plots. It supports predictive modeling and (unsupervised) machine learning techniques.

Most of the programs mentioned above are complementary to each other. As a significant amount of this work focuses on data processing and analysis, various programs were tested and applied to the data. Apart from the more sophisticated applications and software products for data processing, reduction and displaying, Microsoft Excel was used as the basic program for data reduction and data pre-processing.

6. Results

6.1. Optical Properties

Microscopic as well as scanning electron microscopic images were taken from all graphite samples in this study to analyze the optical properties of the graphite concentrates. Mainly scanning electron microscopic images of the concentrates are presented in this chapter, as well as digital light microscopy images of the graphite raw ores, if available for the respective mine/deposit.

Kaisersberg, Austria

Sample 1A from Kaisersberg shows an irregular grain size distribution of an amorphous graphite concentrate with a notable presence of foreign components/impurities (quartz and mica). Sample 21G is a raw ore from Kaisersberg, with sub-parallel graphite (beige-grey-yellowish) embedded in a matrix of quartz and mica (Fig. 20).

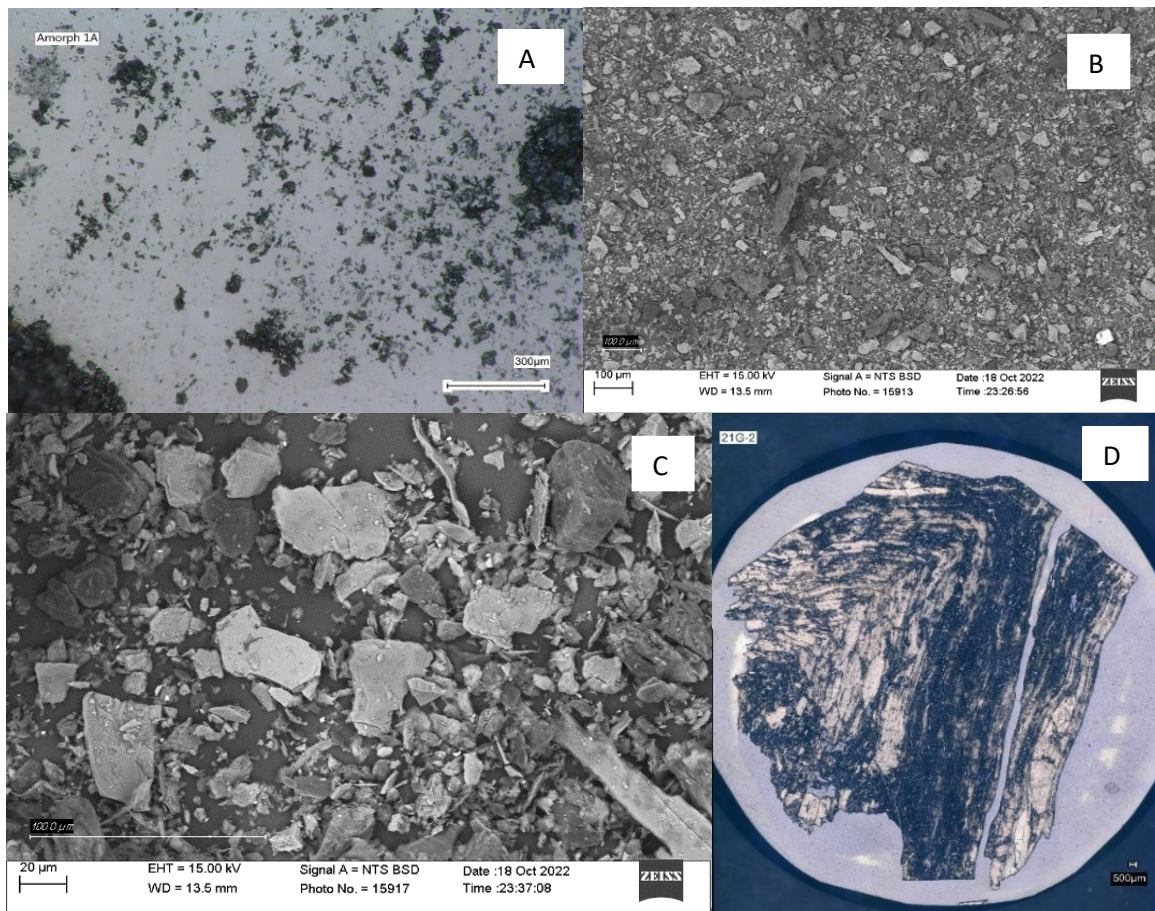


Figure 20: A: digital light microscope image sample 1A, B: SEM image sample 1A 200x magnification C: SEM image sample 1A 1000x magnification, D: digital microscope image sample 21G showing disseminated graphite raw ore from Kaisersberg

Minas Gerais (Pedra Azul, Itapecerica, Salto da Divisa), Brazil, South America

Samples from Minas Gerais province in Brazil are mixtures of material from the mines Pedra Azul, Itapecerica and Salto da Divisa. The material shows homogeneous grain size distributions with distinct flakes. Concentrates from Minas Gerais province show a relatively homogeneous grain size distribution ranging from 100 – 200 μm (Fig. 21).

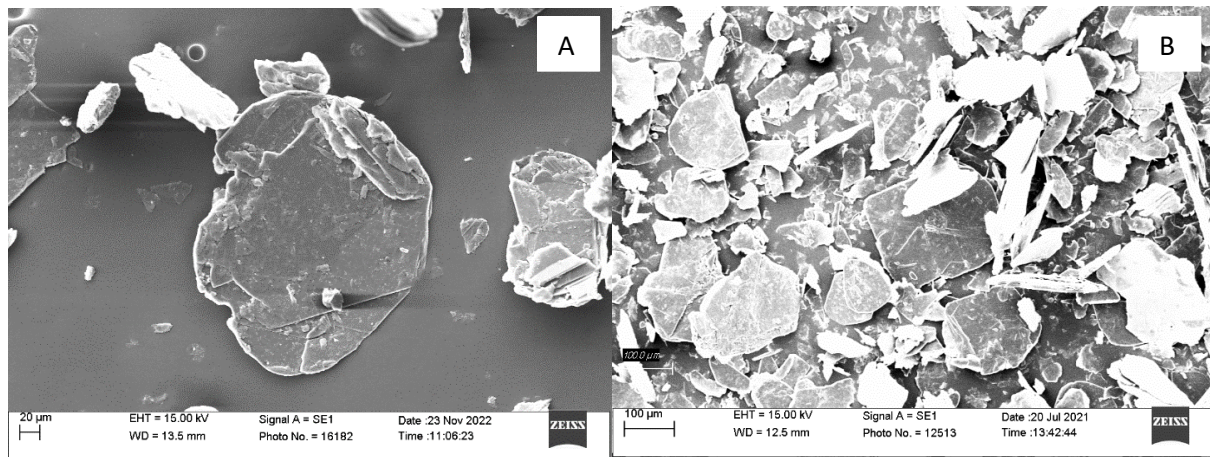


Figure 21: A: Flake with 200 μm diameter in sample 3B, 250 x magnification, B: overview of flake distribution, SEM image from sample 21C, 50x magnification

China, Asia

Heilongjian

Sample 7B exhibits significantly small, less idiomorphic flakes, partly aggregated and distributed inhomogeneously in terms of size. Individual flakes show sizes of up to 150 μm (Fig. 22).

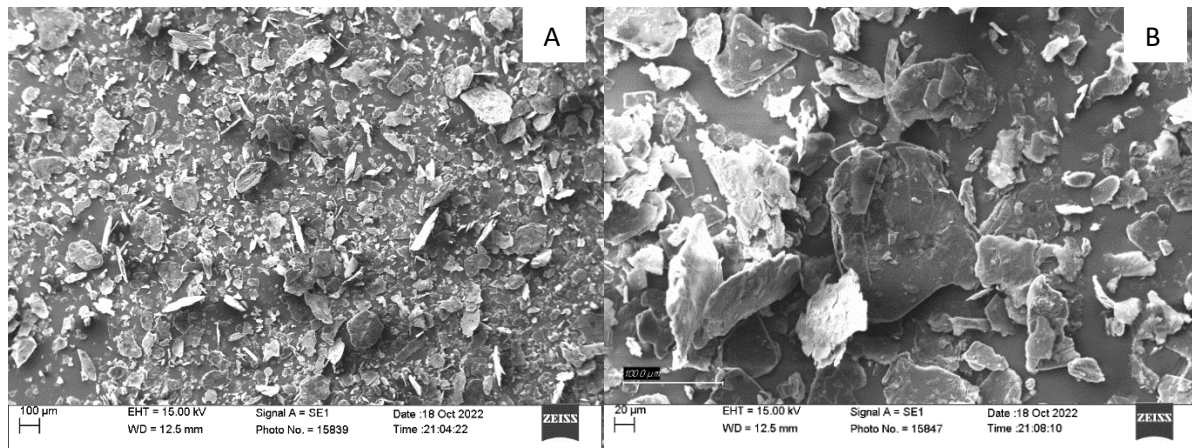


Figure 22: A: sample 7B overview, 100x magnification, B: Flake sample 7B 500x magnification with flake sizes up to 250 μm and smaller flakes and fragments of 100 μm particles and smaller

Hunan Lutang

Sample 4A (amorphous graphite sample) lacks a flake-like structure and is rather a loose aggregation of components without individual grain sizes (Fig. 23).

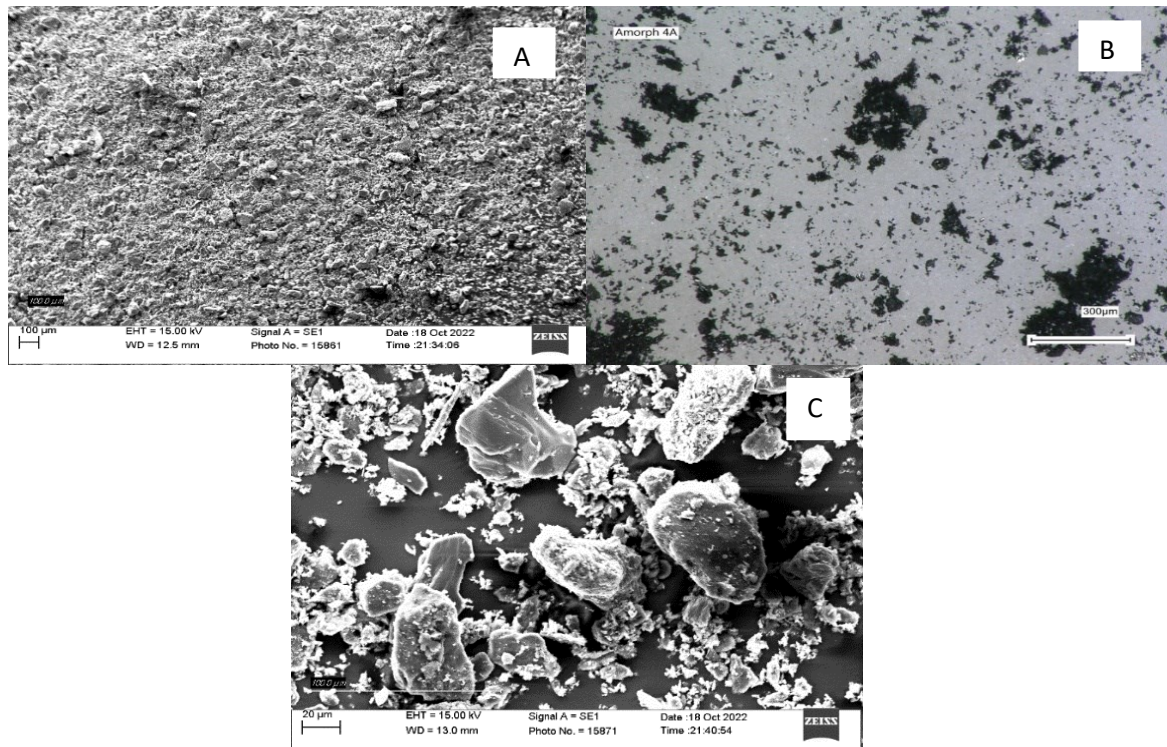


Figure 23: A: sample 4A, SEM image overview, 100x magnification, B: Overview image sample 4A, digital light microscope without visible individual flakes/grains, C: detailed SEM image sample 4A 1000x magnification with grain sizes up to 80 µm and smaller aggregates

Inner Mongolia

Sample 6B (Inner Mongolia, China) exhibits distinctly pronounced idiomorphic flakes with a size ranging from 350 to 450 μm and a very homogeneous grain size distribution. Even in digital light microscopy images, the idiomorphic flakes are identifiable (Fig. 24).

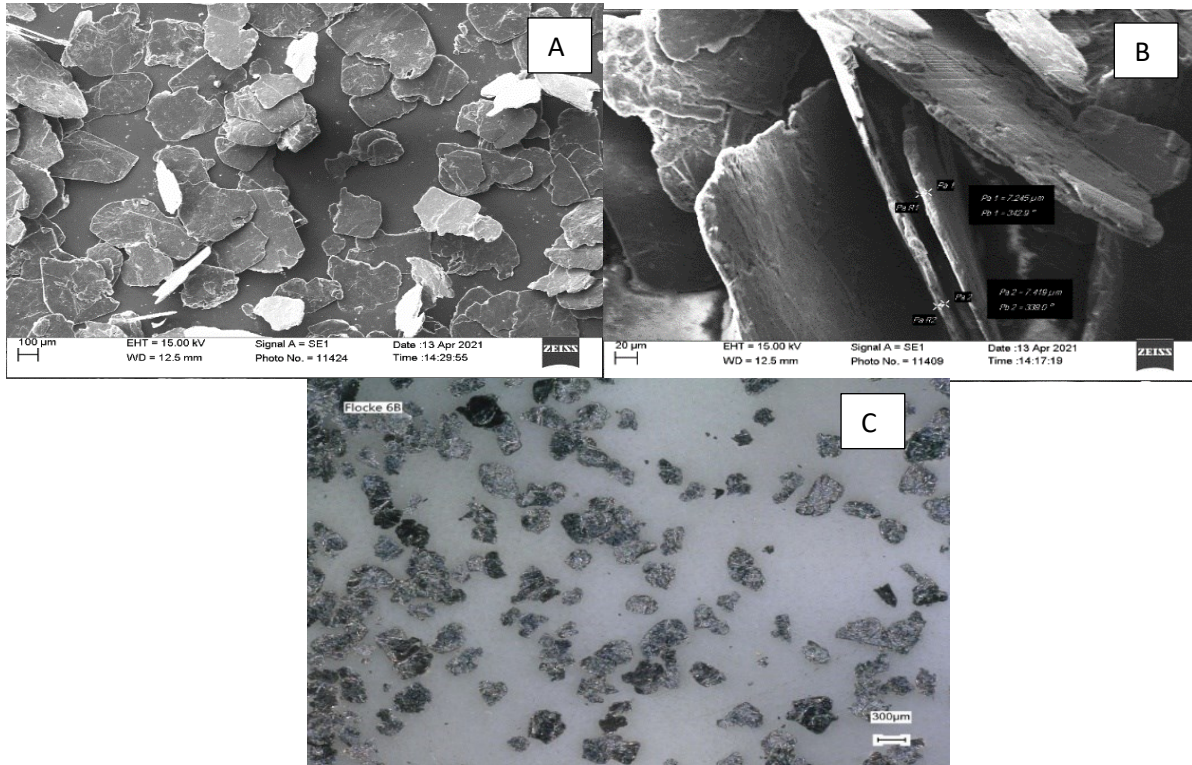


Figure 24: A: sample 6B SEM image overview, 100x magnification, B: detailed SEM image sample 6B 550x magnification with a measurable flake thickness of almost 8 μm ; C: overview digital light microscopy image sample 6B

Shandong

Samples 21A and 22A from Shandong, China exhibit idiomorphic flakes with a maximum grain size of approximately 250 μm down to small flakes and fragments of only 20 μm . The grain size distribution in this concentrate is relatively heterogeneous compared to other graphite concentrates (Fig. 25).

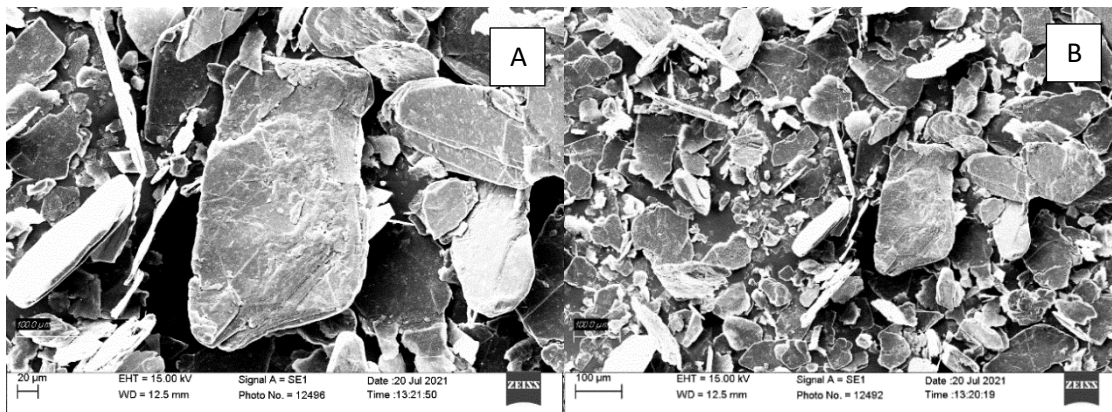


Figure 25: A: SEM image, graphite flake in sample 21A, 550x magnification, B: sample 21A SEM image overview, 100x magnification

Kropfmühl, Germany, Europe

Sample 13B from Kropfmühl, Germany exhibits idiomorphic flakes; however, the grain size distribution is very heterogeneous, ranging from a maximum grain size of approximately 400 μm down to a minimum grain size of 100 μm and below. Raw ore sample 14B shows disseminated graphite flakes (greyish-yellow) in a matrix of quartz (grey) and individual grains of pyrrhotite (white – light greyish) (Fig. 26).

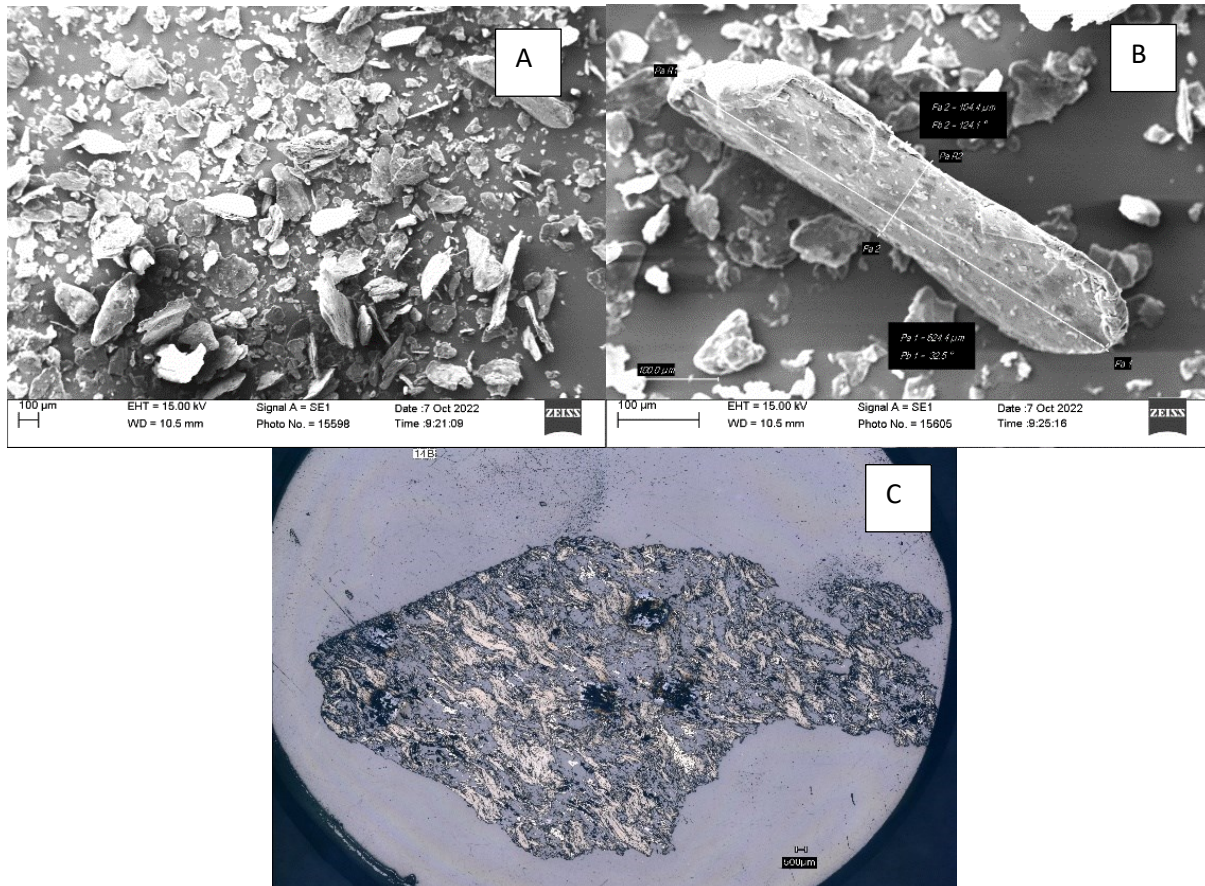


Figure 26: A: Sample 13B overview SEM image with 100x magnification, B: sample 13B detailed SEM image sample 13B individual flake with 600 μm length and 400x magnification, C: raw ore sample 14B overview digital light microscope showing graphite flakes and pyrrhotite

Brickaville (Vatomina und Sahamamy Sahaso), Madagascar, Africa

Samples 11B, 21D and 22D from Brickaville province consist of idiomorphic graphite flakes with a maximum grain size of approximately 650 μm down to small flakes and fragments of only 20 μm . The grain size distribution in this concentrate is homogeneous and the flake size is among the largest in this study with flake diameter of up to 650 μm (Fig. 27).

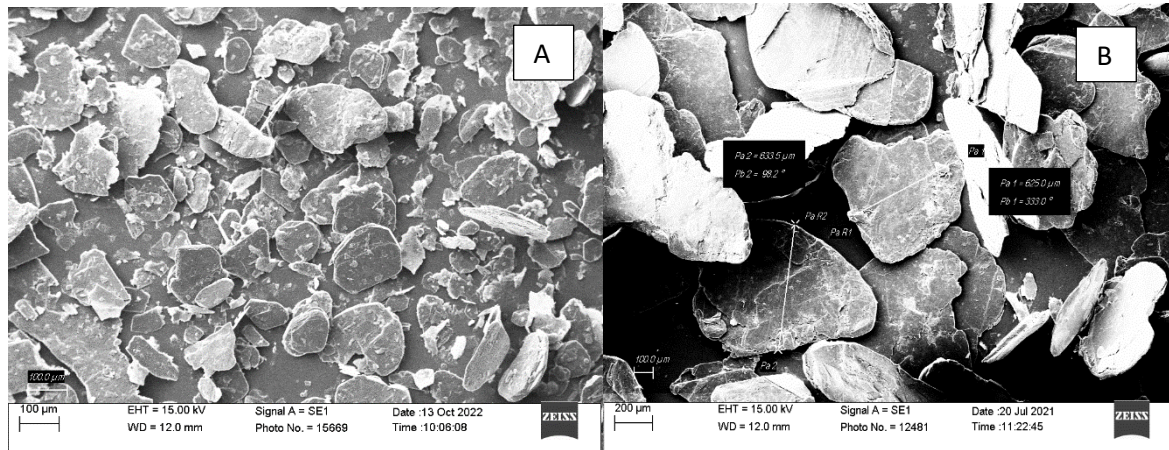


Figure 27: A: sample 22D, overview SEM image 200x magnification, B: sample 21D, SEM image 1000x magnification and flake diameters of 500 – 600 μm

Balama, Cabo Delgado Province Mozambique, Africa

Samples 1B, 21B, 21J, 21K, 21L and 22B from Balama mine consist of distinctly pronounced idiomorphic graphite flakes with a maximum identified flake size of 600 μm and average flake sizes of 200 μm and a flake thickness of up to 5 μm . Raw ore sample 22H from Balama mine shows disseminated graphite flake in a matrix consisting of quartz (grey), mica (light grey) and isolated titanite grains (bright white) (Fig. 28).

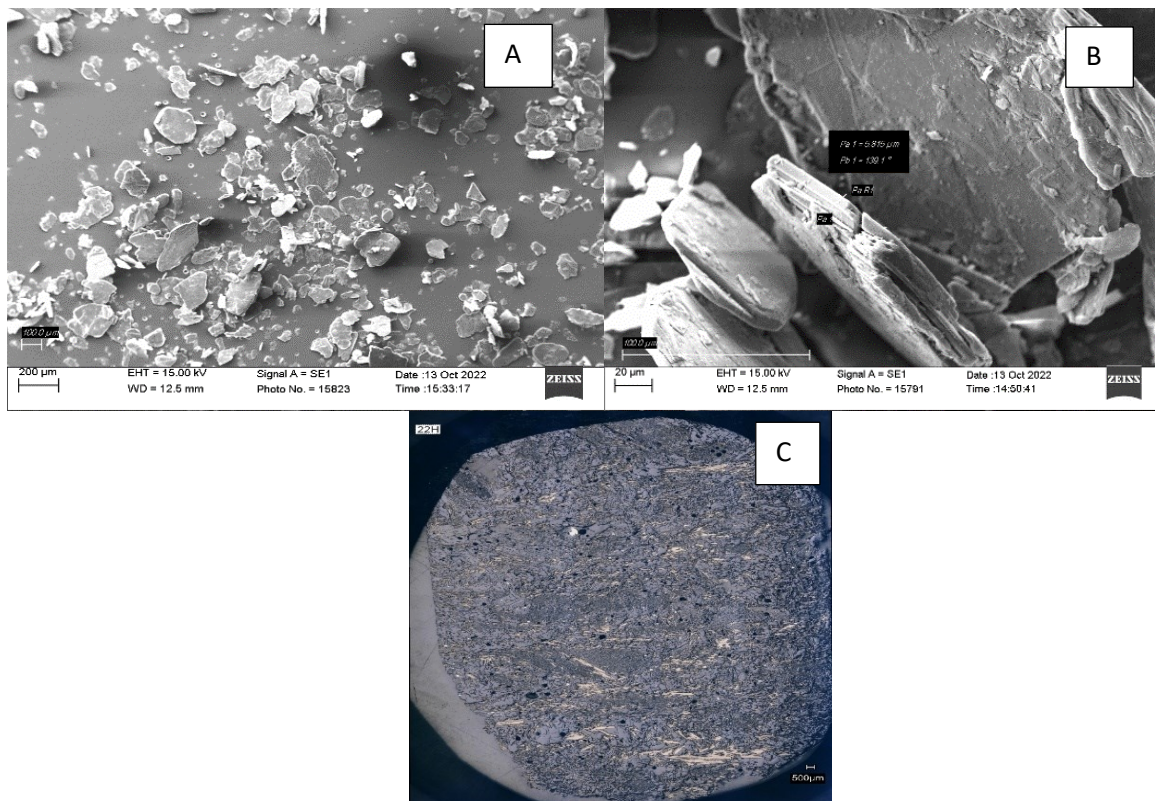


Figure 28: A: sample 21J overview, 100x magnification, B: sample 21L detailed flake structure of individual components, C: 22H disseminated graphite raw ore sample from Balama

Skaland Norway

Sample 2B, 15B and 16B, and 22G from Skaland mine in Norway exhibit well-defined graphite flakes with sizes ranging from 250 to 350 μm and fragments. Sample 16B (Skaland, Norway) presents a raw ore sample, predominantly composed of graphite, including pyrite, albite, and quartz (Fig. 29).

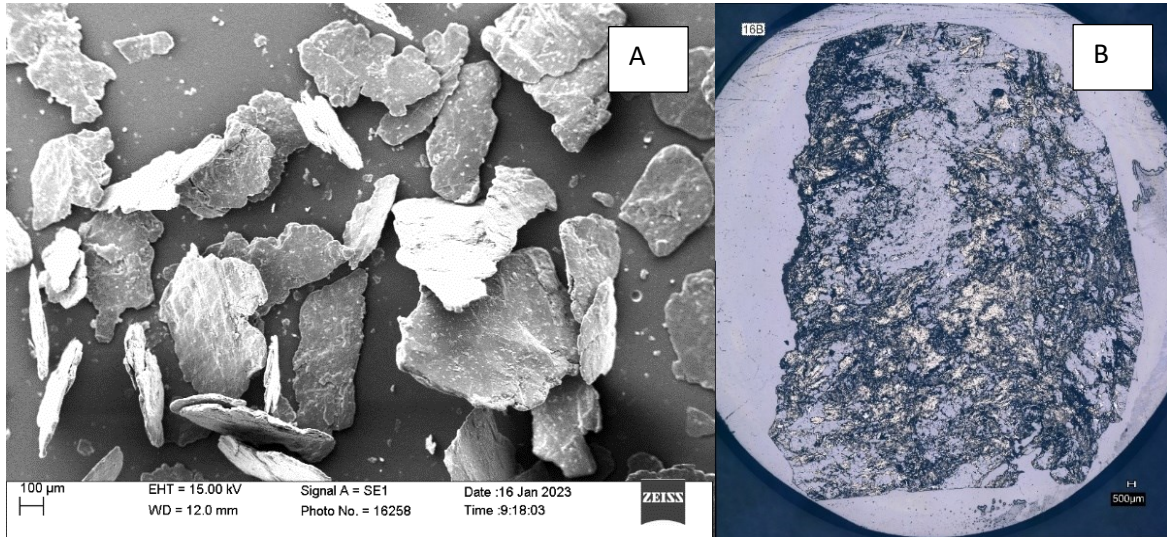


Figure 29: A: individual flakes in sample 15B, magnification 100x, B: 16B disseminated graphite raw ore digital light microscope image (graphite particles too small for laser-ablation)

Taiginka, Chelyabinsk region, Russia, Asia

Samples 5B, 21E and 22E from Tayginka exhibit idiomorphic flakes, with a relatively homogeneous grain size distribution, ranging from a maximum grain size of approximately 200 μm down to a minimum grain size of 100 μm and below (Fig. 30).

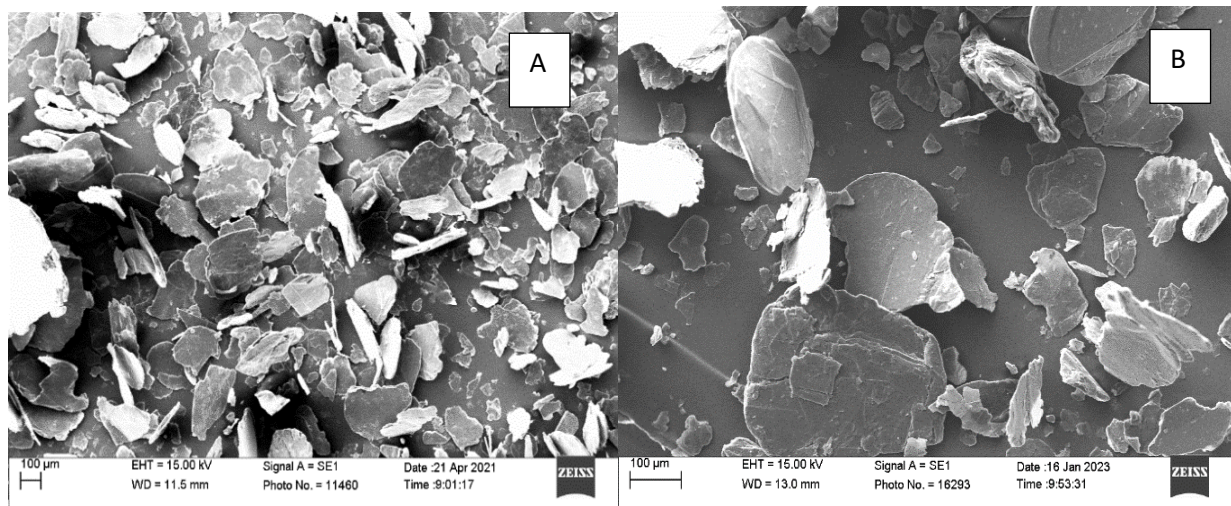


Figure 30: A: SEM image sample 5B, 100x magnification, B: SEM image sample 21E, 250 x magnification

Ragedara, Sri Lanka, Asia

Samples from Ragedara, Sri Lanka represent vein graphite and show highly pure graphite raw ore. Identified mineral phases are exclusively graphite. Black spots/areas on the sample only show relief phenomena due to polishing or uneven surfaces and laser ablation patterns from analysis (Fig. 31).



Figure 31: Highly pure vein graphite (lump ore) (yellowish-grey) sample 10B

Zavallia, Ukraine, Europe

Samples from Zavallia (UKR) show idiomorphic flakes with a relatively homogeneous grain size distribution and distinct idiomorphic flakes with sizes ranging from 100 µm to 200 µm. Even at lower magnification individual flakes can be identified. Through some flakes standing normal to the scanning surface, the flake thickness can be measured and is about 3 µm. Disseminated graphite flakes (yellowish-grey) in a raw ore (D) from Zavallia are embedded mainly in a quartz (grey) and mica (light grey) matrix (Fig. 32).

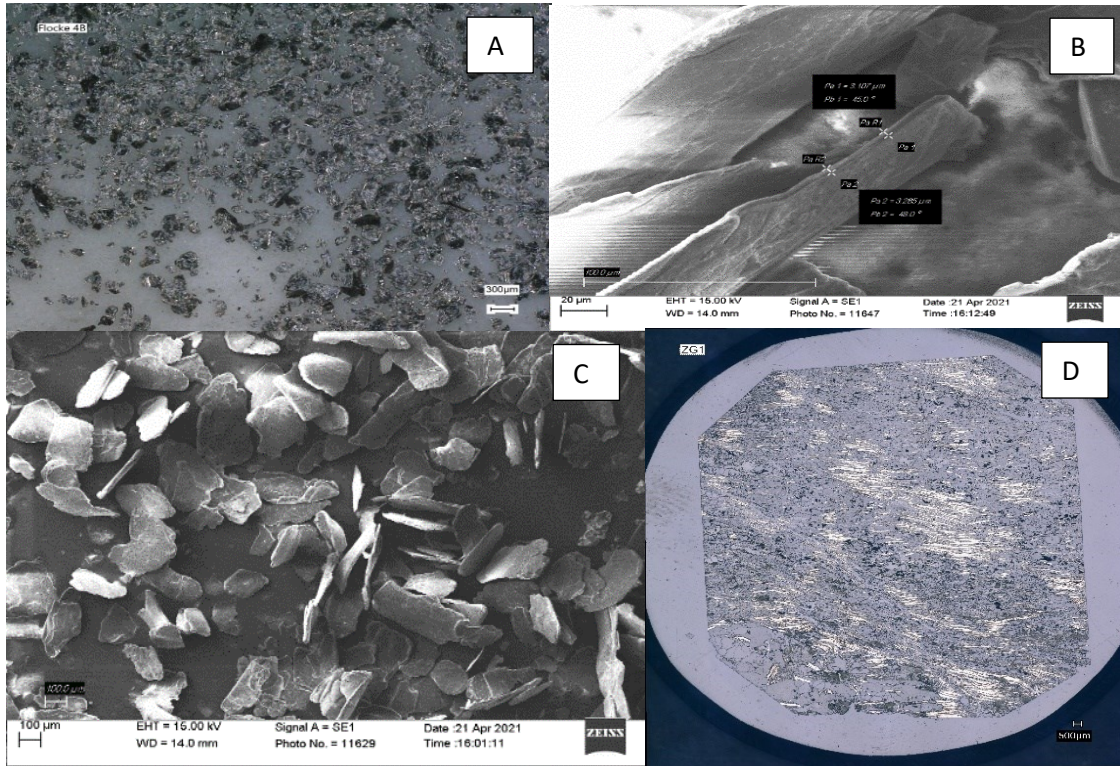


Figure 32: A: digital light microscope image sample 4B, B: SEM image sample 4B 1200x magnification and flake thickness of 3 µm, C: overview SEM image sample 21E 100x magnification, D: digital light microscope image sample 22K-S disseminated graphite raw ore (graphite particles too small for laser-ablation)

Summary table:

The optical properties of the samples are summarized in Table 14. All graphite concentrates under consideration are processing products, containing between 92-96+ % C, with the majority of impurities already removed by on-site processing. The flake size is mechanically manipulated and does not completely reflect the naturally occurring flake size. Accompanying mineral phases as residues and impurities in the concentrates represent mainly quartz and mica particles. Some deposits however are specified by their large flake sizes. Specific details are summarized in the table below.

Table 14: Summary of optical properties of graphite concentrates assessed by SEM

mine/province	country	graphite type	av. flake size (µm)	concentrate properties
Kaisersberg	Austria	amorphous	50	no prominent flake-like structure, inhomogeneous
Minas Gerais	Brazil	flake	150	relatively homogeneous size distribution
Heilongjian	China	flake	120	inhomogeneous, less idiomorphic flakes
Hunan Lutang	China	amorphous	30	no prominent flake-like structure, inhomogeneous
Inner Mongolia	China	flake	450	very homogeneous size distribution, largest average flake sizes, thick flakes
Shandong	China	flake	180	inhomogeneous flake size distribution
Kropfmühl	Germany	flake	200	inhomogeneous flake size distribution
Brickaville	Madagascar	flake	450	homogeneous flake size distribution, large flakes
Balama	Mozambique	flake	200	rel. homogeneous flake size distribution
Skaland	Norway	flake	300	homogeneous flake size distribution
Taiginka	Russia	flake	200	inhomogeneous flake size distribution
Ragedara	Sri Lanka	vein/lump	--	large chunks of pure graphite
Zavallia	Ukraine	flake	200	homogeneous flake size distribution

6.2. Stable Carbon Isotope Ratios ($\delta^{13}\text{C}$)

There are three naturally occurring isotopes of carbon, namely ^{12}C , ^{13}C and ^{14}C . The latter is the radioactive isotope, with a half-life of 5730 years. It is generated by thermal neutrons from cosmic radiation in the upper atmosphere and is subsequently transported to Earth, where it is absorbed by living biological material. ^{12}C predominates as the most abundant isotope at 98.89 %, while the ^{13}C isotope occurs with a frequency of only 1.11 % (O'Leary et al. 1992). The isotopic ratios of ^{12}C and ^{13}C are quantified and expressed using the notation $\delta^{13}\text{C}$ in parts per thousand, called per mil (‰) and are calculated as follows:

$$\delta^{13}\text{C} = \left(\frac{\left(\frac{^{13}\text{C}}{^{12}\text{C}} \right)_{\text{sample}}}{\left(\frac{^{13}\text{C}}{^{12}\text{C}} \right)_{\text{standard}}} - 1 \right) \times 1000 \text{ ‰} \quad (\text{O'Leary 1981})$$

The globally accepted standard is based upon carbon dioxide sourced from limestone within the Pee Dee formation in South Carolina and is denoted as PDB (Craig 1957).

A clear demarcation of the carbon signature is observed in graphite, indicating significantly heavier values for hydrothermal/vein graphite (Weis 1981). Within organic graphite (flake vs. amorphous), there are two trends, attributed to different source materials. Amorphous graphite is formed through regional or contact metamorphism of coal seams, while the flake type of graphite undergoes graphitization of organic precursors through regional metamorphism. Coals tend to include material enriched in ^{13}C (Weis 1981). The precursor material for coals is higher land plants, whereas the precursor material in disseminated graphite flakes is more likely composed of more primitive organisms (e.g. bacteria, algae).

6.2.1. Worldwide Database of Carbon Isotope Values ($\delta^{13}\text{C}$)

A database consisting of graphite stable carbon isotope data ($n=433$) was developed in the frame of this project in collaboration with researchers from GTK, Finland. The database contains stable carbon isotope data obtained from samples within this study, but also literature data from various global graphite occurrences. The corresponding data table for Figure 33 can be found in the appendix, including references. Only values that could be assigned to a graphite type (amorphous, flake, vein) were included in the database. Potential outliers in the data may have been caused by mixture analyses, for example, by mixing/contaminating graphite material with carbonates, which have a $\delta^{13}\text{C}$ signature of ≈ 0 ‰. The figure presented below (Figure 33) illustrates the $\delta^{13}\text{C}$ values from global graphite occurrences. The values are associated with their respective mines/provinces, as well as the corresponding countries. Further differentiation could be made based on the graphite type (amorphous, flake, vein).

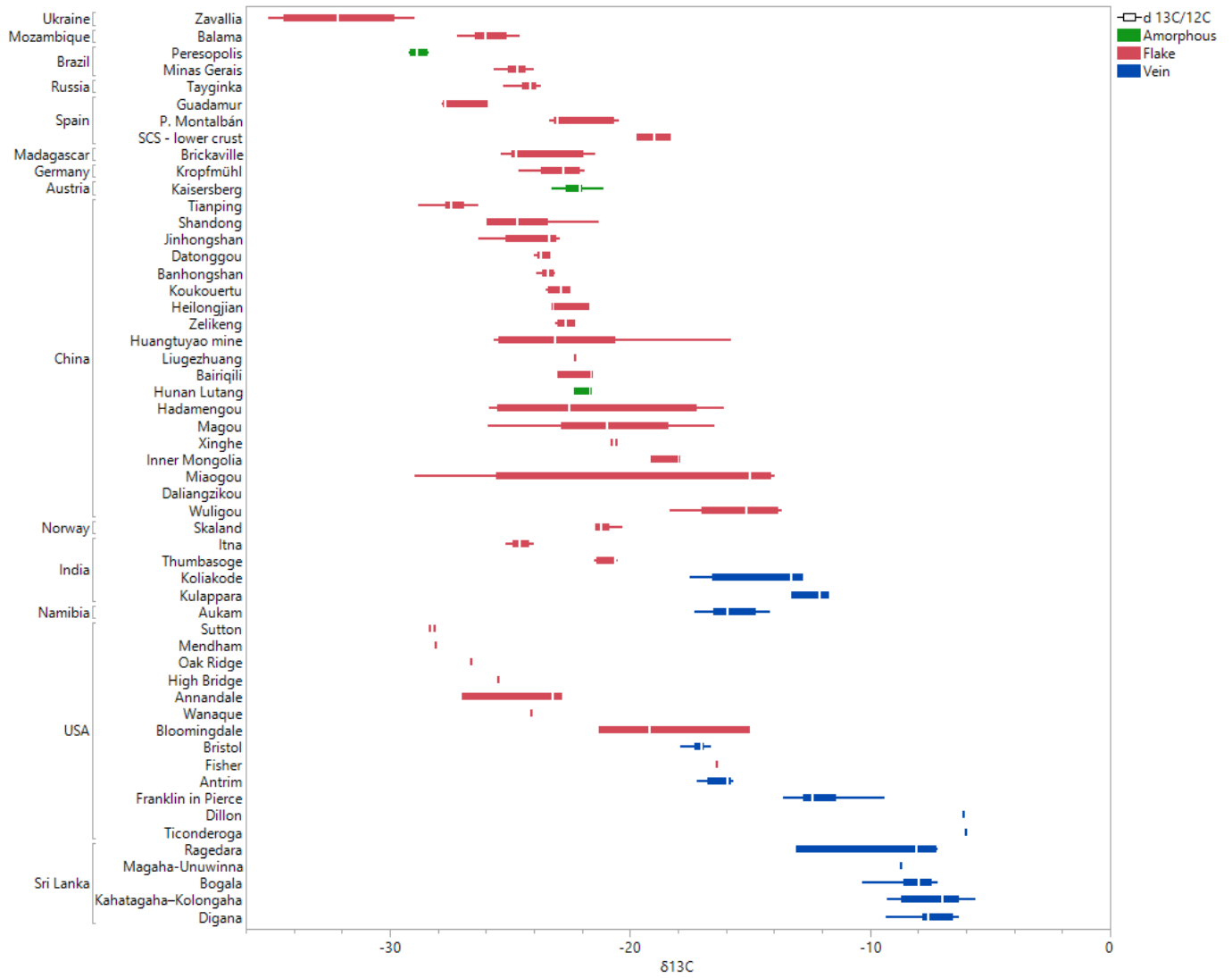


Figure 33: Worldwide database of stable carbon isotope signatures in graphite (Data table and references can be found in the Appendix)

A representation of the $\delta^{13}\text{C}$ values sorted by continent and country is illustrated in Figure 34. Amorphous graphite in this study and in the database are limited to the continents of South America (Brazil), Europe (Austria), and Asia (China). Flake graphite is globally distributed. Vein graphite refers to North America (USA), Asia (Sri Lanka), and Africa (Namibia) in this study. The most common graphite type worldwide is flake graphite. The values of vein graphite are distinguishable (-5 to -15 ‰), as are those of the Zavallia deposit in Ukraine, with a very light $\delta^{13}\text{C}$ value (-28 to -35 ‰). The $\delta^{13}\text{C}$ signatures of the remaining flake graphite overlap, and a distinction based on countries is not possible.

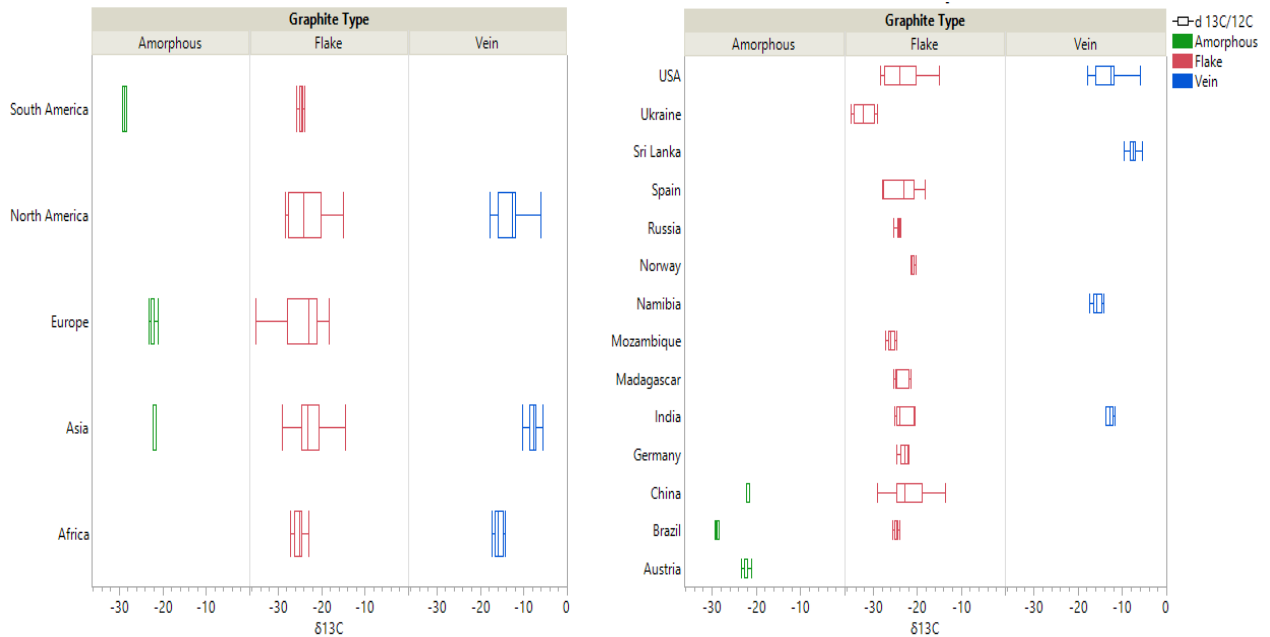


Figure 34: Data distribution of $\delta^{13}\text{C}$ values sorted by continent (left) and countries (right) and the respective graphite type (amorphous, flake, vein)

6.2.2. Carbon Isotope Values from Sample Collection in this Project

The following data refers to $\delta^{13}\text{C}$ values from measurements of graphite concentrates, conducted in in-house laboratories at MUL. The data collection covers key countries in global graphite production, spanning South America, Europe, Africa and Asia. These measurements have also been associated with additional attributes.

Most of the isotope values correspond to flake graphite. Hydrothermal (vein) graphite was sampled from Asia (Sri Lanka) only and amorphous graphite was collected from Europe (Austria) and Asia (China). The analyzed graphite in Africa and South America can be assigned to as flake graphite. A geographical differentiation of continents based on carbon isotope values is not feasible, as carbon isotope signatures of the continents overlap (Fig. 35).

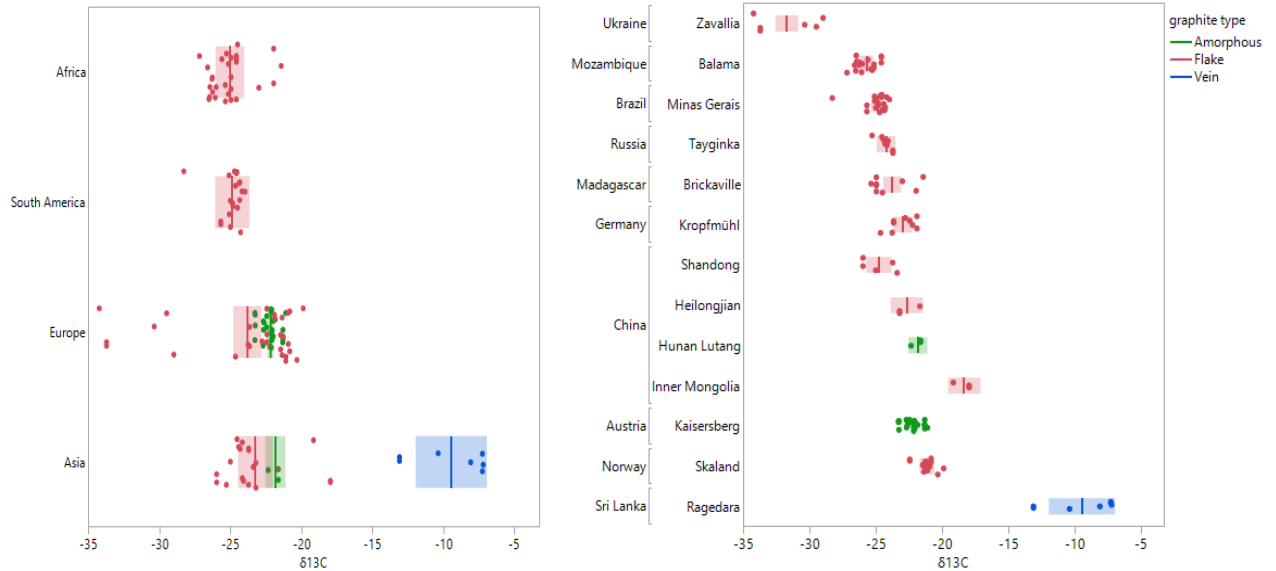


Figure 35: Carbon isotope ratios categorized by graphite type (amorphous, flake, vein) and geographical origin across continents hierarchy (left) and mine/province and country hierarchy (right), vertical line in box represents mean value

Another differentiation of $\delta^{13}\text{C}$ values was made based on geological ages (Fig. 36). The graphite samples from Africa and South America are classified as Proterozoic. The group of the oldest graphite (Archean) can be assigned to Ukraine. Geological ages ranging from Archean to Proterozoic and Paleozoic can be found in Europe. The graphites from Asia are classified as Proterozoic and Paleozoic. The measurement values across all continents overlap. The only clear distinction is observed in the Archean graphite in Europe (Ukraine).

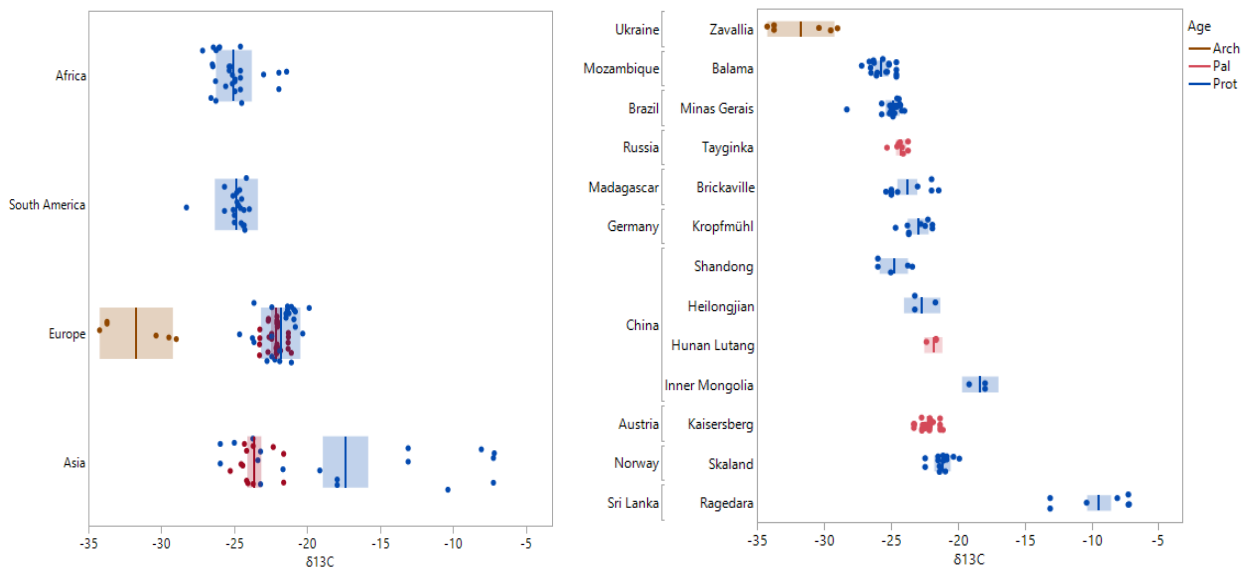


Figure 36: Carbon isotope ratios categorized by geological age (Archean, Palaeozoic, Proterozoic) and geographical origin across continents hierarchy (left) and mine/province and country hierarchy (right); vertical line in box represents mean value

Variability of the data

The data distribution of the $\delta^{13}\text{C}$ values shown below (Fig. 37) can be classified as follows: Vein graphite is categorized in a significantly heavier $\delta^{13}\text{C}$ range (-7 ‰ to -13 ‰) and can be separated from the other two groups, which is flake type (-17 ‰ to -34 ‰) and amorphous type (-21 ‰ to -24 ‰). Amorphous and flake type overlap and cannot be differentiated.

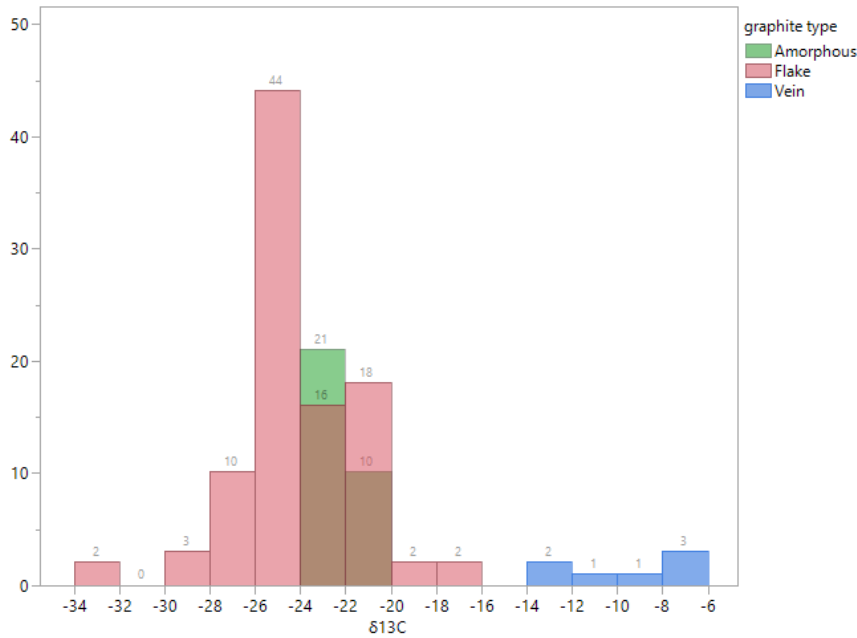


Figure 37: General data distribution divided into three groups (amorphous, flake, and vein graphite).

The geological age allows the distinction of Archean graphite, as shown in Fig 38. Paleozoic and Proterozoic $\delta^{13}\text{C}$ values overlap. Archean graphite shows a $\delta^{13}\text{C}$ range of -28 ‰ to -34 ‰, whereas Paleozoic graphite falls within a $\delta^{13}\text{C}$ range of -21 to -26 ‰ and the Proterozoic graphite lies within the values -7 ‰ to -28 ‰).

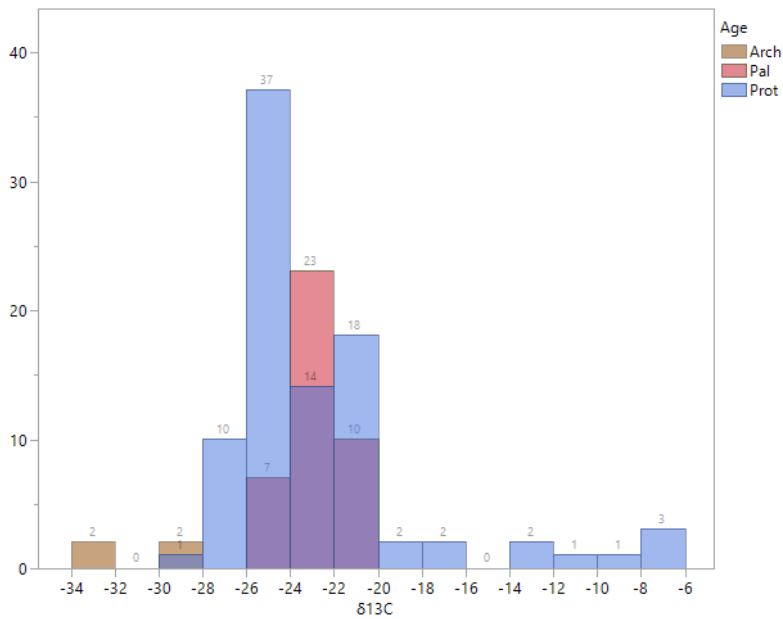


Figure 38: General data distribution divided into three groups (Archean, Paleozoic and Proterozoic).

6.2.3. Differentiation between country groups

The available data could be divided into three subgroups (Sri Lanka, Ukraine and the rest of the world (RoW)). A more detailed distinction within the RoW group is not feasible based on this parameter only.

The differentiation of the above-mentioned groups can be explained by additional attributes and is further illustrated (Fig. 39).

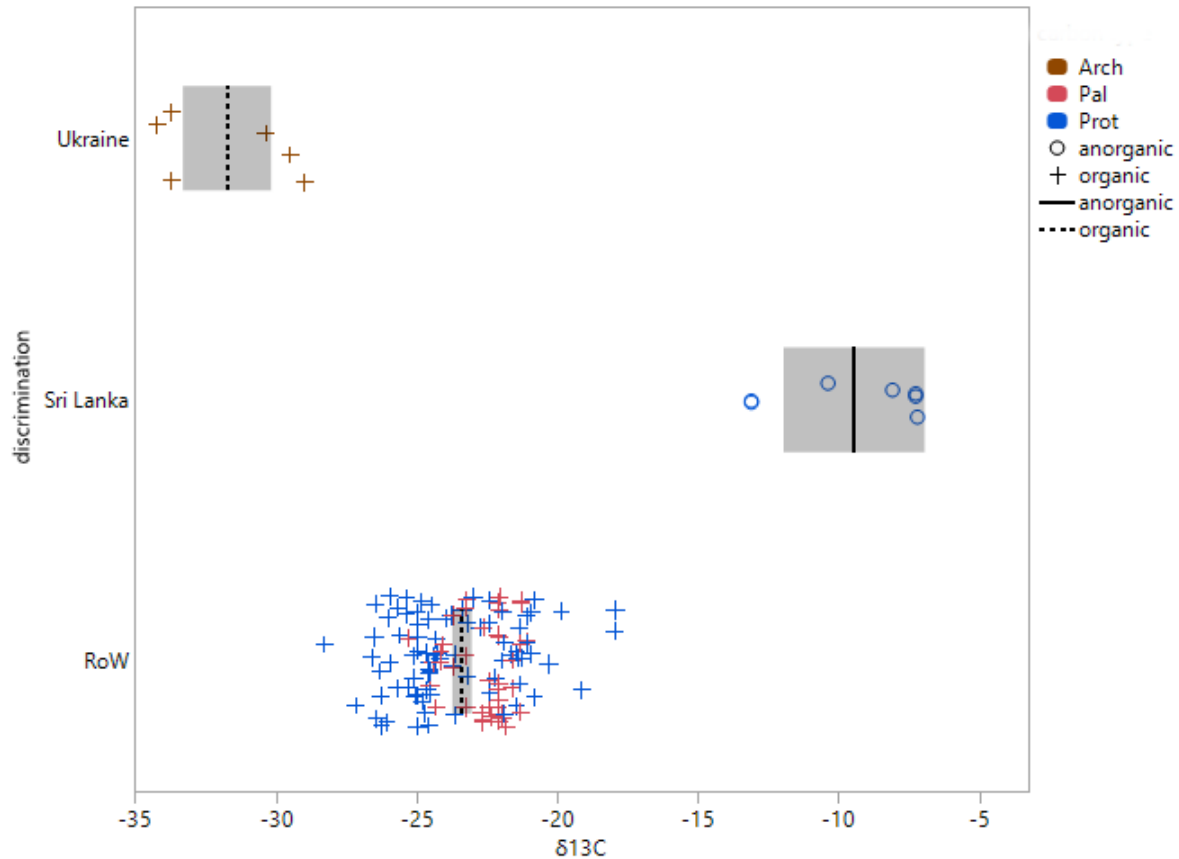


Figure 39: Discrimination of samples into groups (Sri Lanka, Ukraine and rest of the world (RoW) with additional attributes related to the carbon source (organic or inorganic); vertical line in box represents mean value

The data distribution of carbon isotopes within these three groups exhibits a significantly denser dataset for the RoW group. The Sri Lanka and Ukraine groups consequently display fewer data points; however, the existing data points fall within the defined groups and are therefore clearly distinguishable. Graphite within the group Ukraine is exclusively referred to as Archean organic graphite. The group RoW represents a mixed sample set, consisting of organic graphite, showing Paleozoic and Proterozoic ages and cannot be differentiated further. The group Sri Lanka consists only of anorganic (vein) graphite.

6.2.4. Supply Chain Consistency

To which extent carbon isotope values remain constant throughout the processing chain and what effect graphite processing has on the stable carbon isotope on the material was analyzed using samples from Finnish occurrences (Aitolampi, Emäs, Koivuniemi and Raisjoki). The samples originate from the sample repository from GTK, as well as processing and intermediate materials. The stable carbon isotope data were analysed in the course of this study at MUL (Fig. 40).

Analyzing further products along the supply chain from the raw ore or flotation concentrates to the purified graphite is an advanced step in the analytical proof of origin study, focusing on the stability of individual parameters along the supply chain. Processing and intermediate products often represent

mixtures. The exact origin is often unclear, as processing steps may be carried out by different companies, possibly even at different locations.

Samples from the Aitolampi deposit were processed directly at the on-site pilot processing plant (Beowulf Mining) and at the GTK in-house facilities. Core and crushed core samples, flotation concentrates, by-products and residues, alkaline purified material and purified spherical graphite were analyzed. Samples from other Finnish occurrences (Emas, Raisjoki, and Koivuniemi) were processed by GTK in their in-house pilot processing plant. These samples include raw ore (flotation feed), flotation (rougher and cleaner) concentrates and the product (purified graphite).

The Aitolampi deposit exhibits a wider range of $\delta^{13}\text{C}$ values in the flotation feed, attributed to the increased (spatial) variability within the mine and the analyzed core samples from two different areas within the deposit (eastern and western zone), whereas the purified concentrates consistently show a significantly smaller range of isotopic values. A clear change in carbon isotopic composition is not observable throughout the graphite processing. Stable carbon isotope signatures from flotation concentrates and products typically overlap within a wide range within the same deposit. The majority of variability can be attributed to the (spatial) variability within the deposit, which may translate into temporal variability over the life of the mine and can be seen in flotation feed or crushed core material.

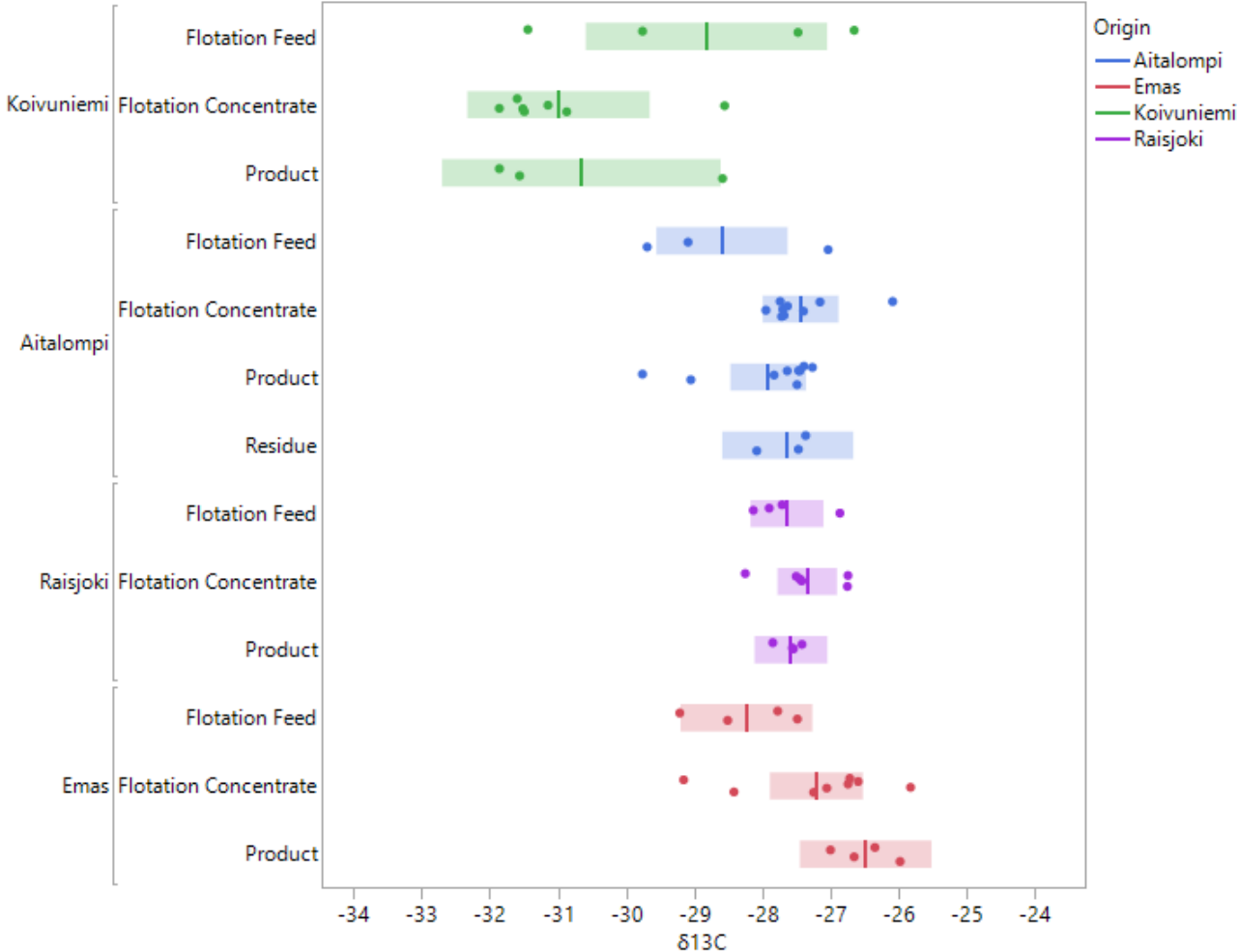


Figure 40: Carbon isotope values from different occurrences in Finland and different intermediate products along the value chain; vertical line in box represents mean value

6.3. Crystallinity and Spectral Data

The samples investigated are mainly graphite concentrates, with some graphite raw ore samples. The type of sample is important for the application of Raman spectroscopy. The concentrates have been ground and floated, with their Raman spectra representing a mixed signature of natural processes (organic maturation) and structural alteration through processing in the (processing) plant on the mine site itself. Previous work on Raman data quality showed that mechanical stress from shearing or polishing may influence the Raman spectra (Rantitsch et al. 2016b, Beyssac et al. 2004, Lünsdorf and Lünsdorf 2016), which however cannot be compared to the sample processing on the mine site. A study comparing Raman data from unprocessed, handpicked raw ore samples that suffered no mechanical stress and processed graphite concentrate both from the same mine (Kaisersberg (AUT)) showed that the discrepancies within the mean Raman parameters between these two samples types are lower compared to the variability of the within-sample data and also smaller than the variability observed among samples from different mines. These observations provide evidence that the graphite processing has an insignificant impact on the ordering properties (Rantitsch et al. 2016b). Evidence is provided with own data from this study. Figure 41 displays Raman spectra from Kropfmühl (GER). Spectrum 1 (red) shows a characteristic Raman spectrum of a graphite concentrate from this mine, while spectrum 2 (blue) represents a characteristic spectrum of a raw ore sample from the same mine. The two spectra do not differ from each other, apart from the varying intensity of the red (concentrate) spectrum. On-site sample processing does not induce significant spectral alterations into the sample.

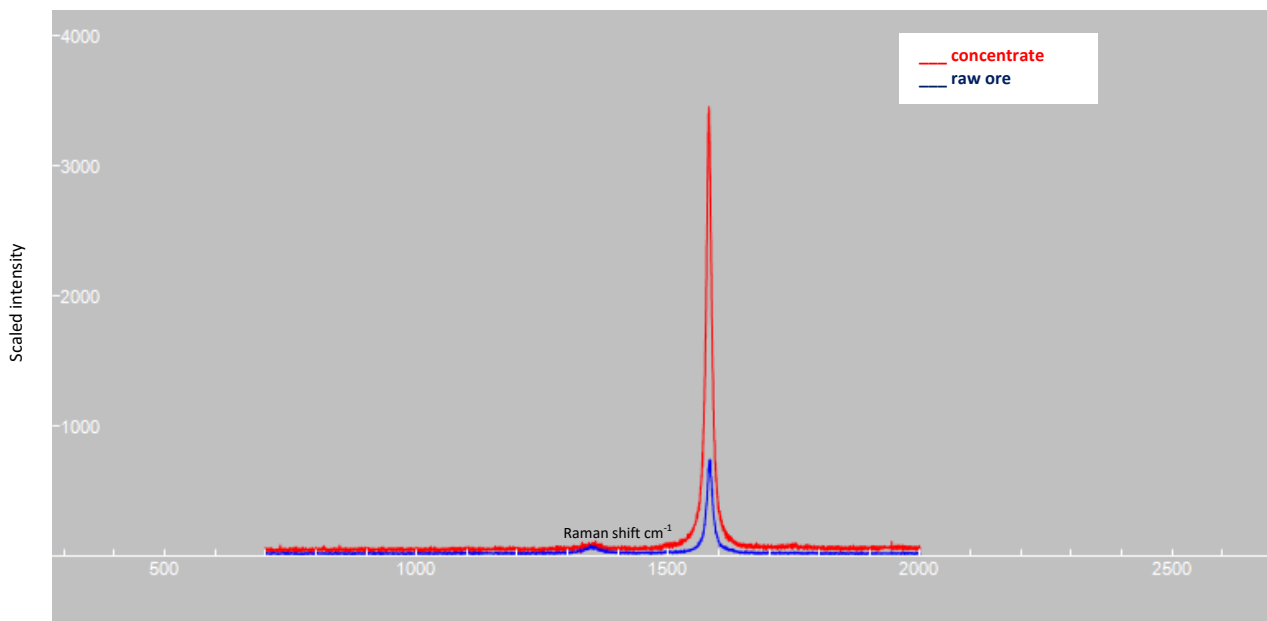


Figure 41: Raman spectra from Passau (GER) - raw ore spectrum in blue and concentrate spectrum in red

In order to avoid alteration of the graphite structure by sample preparation (polishing or shearing), samples were prepared as a water-suspension on a glass slide for the analysis. A variable number of spectra (10-20 spectra per sample) was recorded on different points (flakes). Each sample is assigned to a mine/province. Fig. 42 illustrates the total number of spectra per location. The Balama mine is overrepresented due to the presence of numerous individual samples. For the Chinese occurrences, mostly only one sample is available, hence the number of corresponding spectra is lower.

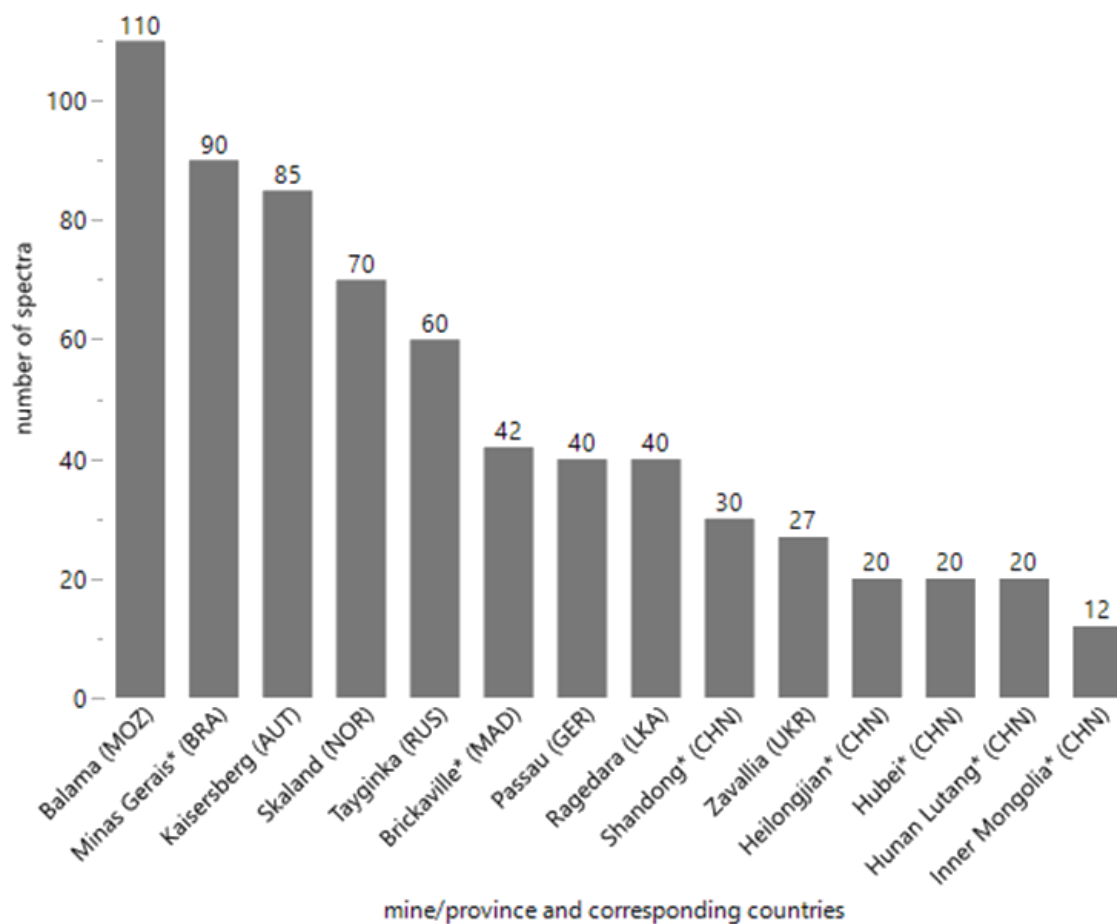


Figure 42: Number of spectra per location (mine/province)

Important parameters describing the spectra, were extracted using iFORS (iterative Fitting of Raman Spectra) (Rantitsch et al. 2014) by Dr. Ròbert Aratò. The extracted parameters, following (Lünsdorf et al. 2017) terminology, comprise the following features:

- D-band (disorder band): is associated with disordering or defects in the crystal lattice structure
- G-band (graphitic band): is associated with the crystalline structure of graphite and often used to describe the degree of graphitization
- Dmax and Gmax: D-band and G-band regions exhibit a central Raman band containing Dmax, with the G-band region characterized by the Raman band containing Gmax
- D_STA and G_STA: representing the area beneath the D-band/G-band; STA, an acronym for Scaled Total Area. D_STA is used to describe the evolution of organic matter during graphitization.
- G_shape_factor: indicating a mathematical descriptor of the fitted curve, the factor identifies the terminus of the transition zone, facilitating the connection between G_STA and D_STA.

Three Raman spectra are presented below (Figure 43), each of them representing one type of graphite (amorphous, flake, vein/hydrothermal) investigated in this study.

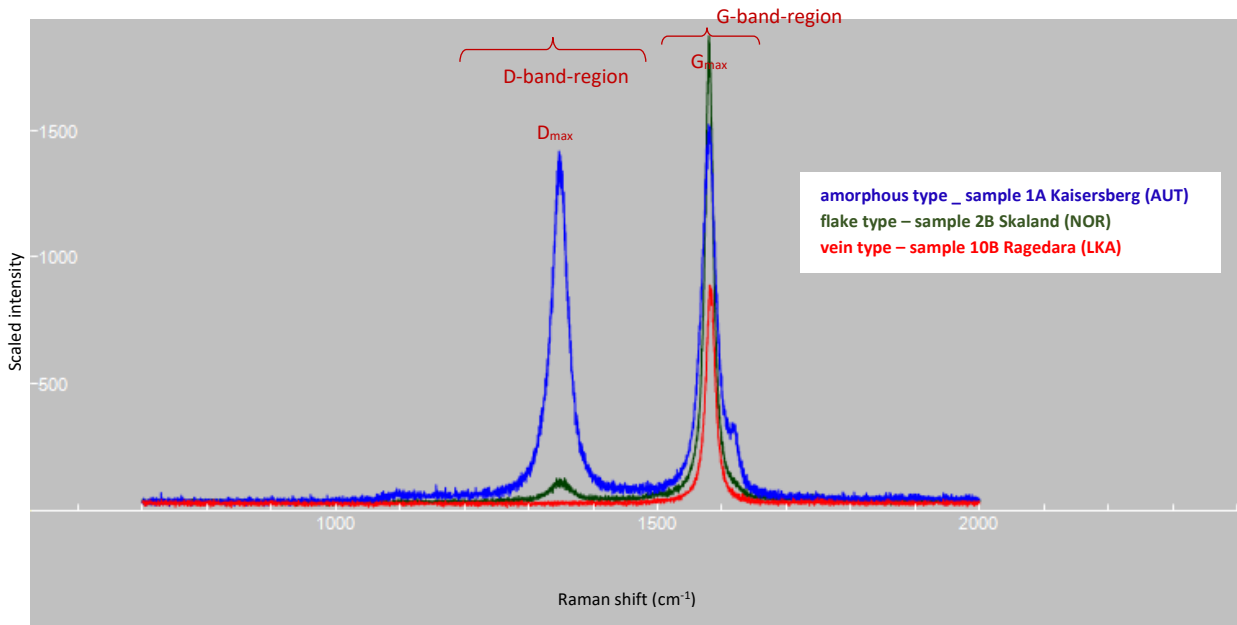


Figure 43: Raman spectra of sample 1A from Kaisersberg (AUT, amorphous, green), sample 2B from Skaland (NOR, flake, blue) and sample 10B from Ragedara (LKA, hydrothermal, orange)

The first order spectrum from sample 1A Kaisersberg (AUT) (green) reveals a prominent D-band and the G-band. First order spectrum from sample 2B from Skaland (NOR) (blue) reveals a sharp G-band with a high intensity and a weak D-band. Spectra from sample 10B from Ragedara (LKA) represent hydrothermal graphite with a pronounced G-band and missing D-band. Spectra from this sample hardly ever show any occurrence of a D-band.

The relevant parameters according to Lünsdorf et al (2017) were collected and analyzed in a Linear Discriminant Analysis (LDA). The first model (Figure 44) demonstrates the differentiation based on mine/province. Hunan Lutang (CHN) and Kaisersberg (AUT), representing amorphous (semi-) graphite can be distinguished from the rest of the samples. Two classes can be identified which represent the amorphous graphite type (Hunan Lutang and Kaisersberg) and the rest of the samples (flake and hydrothermal type), that overlap significantly. Further discrimination into individual mines/provinces is not feasible. Within the flake and vein graphite types, no further separation into subclasses (e.g. mine/province) is possible.

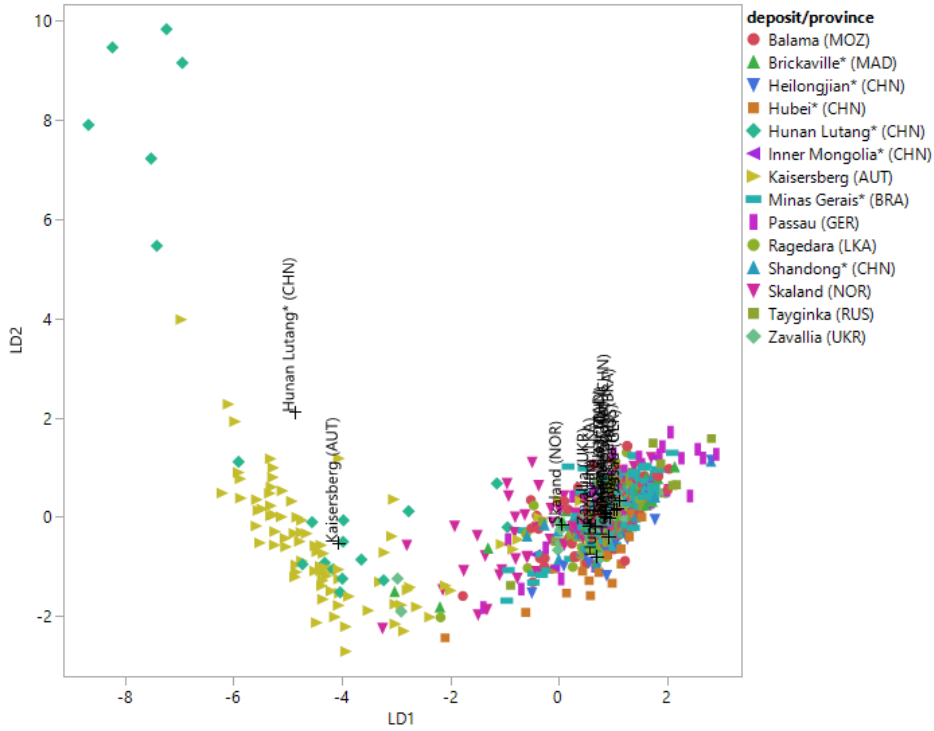


Figure 44: LDA of the extracted parameters of graphite Raman spectra categorized into mines/provinces

The same parameters were analyzed using Linear Discriminant Analysis (LDA), but this time classified by carbon type (Fig. 45). Discrimination based on the graphite type (amorphous – flake – hydrothermal) shows a clearer result. The group of amorphous graphite can be differentiated. However, the hydro and flake graphite groups overlap and cannot be separated from each other.

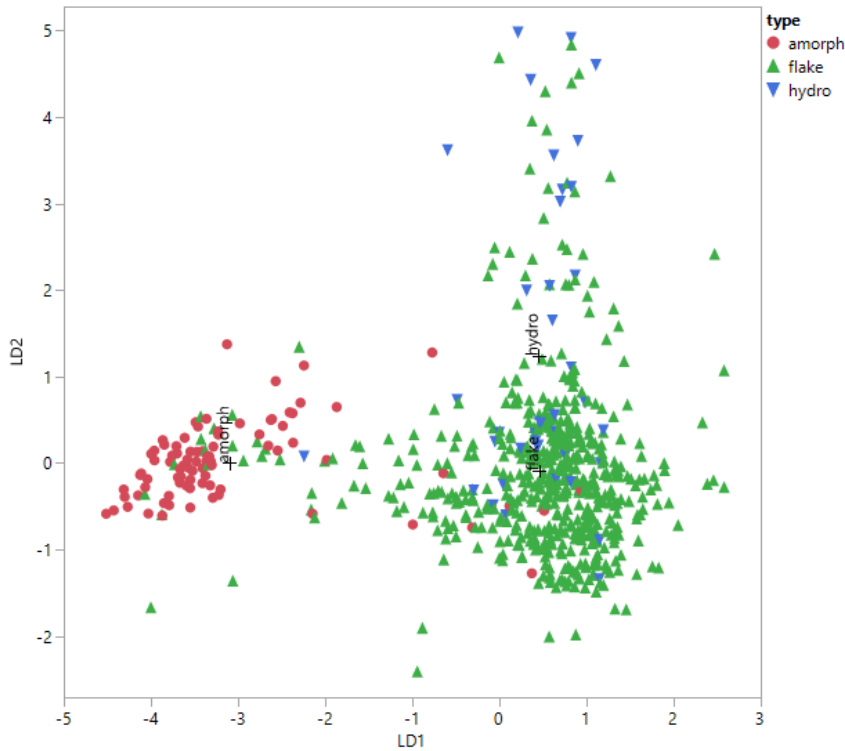


Figure 45: LDA of the extracted parameters of graphite Raman spectra categorized into mines/provinces

Identification of important parameters for the discrimination (scoring coefficients):

To interpret the result of the linear discriminant analysis (LDA) and to identify the contribution of each independent variable for the formation of the discriminant functions, the scoring coefficients of the canonicals were extracted from LDA, with LD1 and LD2 having the highest impact and class separability. Larger coefficients indicate stronger influence on the classes. G shape-factor and G_STA are the variables that are most important for discriminating between the graphite types (Tab. 15). Canonicals 1 and 2 (expressed as LD1 and LD2) refer to linear combinations of the original variables that maximize the separation between groups. Canonical 1 of LD1 represents the linear combination of variables that provide the maximum discrimination between the graphite types (groups) and LD2 the most discriminative variable combination orthogonal to LD1 related to the same groups. In the matrix, there are further canonicals, but their significance gradually diminishes.

Table 15: Standardized scoring coefficients from LDA (Fig. 60)

	<i>D_STA</i>	<i>G_STA</i>	<i>G_shape_factor</i>	<i>Dmax_pos</i>	<i>Gmax_pos</i>	<i>Dmax/Gmax-ratio</i>	<i>D_shape_factor</i>
<i>LD1</i>	-0.3777	-0.3227	0.9987	0.0158	0.3254	0.0098	0.0025
<i>LD2</i>	-0.6420	1.6246	0.8693	0.0235	-0.4583	-0.6565	0.2312
<i>LD3</i>	0.1446	-0.5074	-0.2807	0.6562	-0.1466	0.4171	0.7157
<i>LD4</i>	0.9268	0.9521	-0.1353	-0.5172	0.5315	-0.8023	-0.2705
<i>LD5</i>	-0.6569	-0.3681	0.1847	0.2794	0.7349	0.5887	0.1465
<i>LD6</i>	-0.6382	-2.7770	-0.0446	-0.4761	-0.2646	2.7767	1.2758
<i>LD7</i>	0.6463	-1.1580	0.3262	0.1956	-0.0314	1.7492	-0.7205

6.4. Multi-Element-Analysis

6.4.1. Trace Element Concentrations Assessed by LA-ICP-MS

For each graphite concentrate a powder pressed pellet was prepared and approximately 15 to 20 line scans per pellet and five to ten points per raw ore, depending on the graphite flake size were carried out using LA-ICP-MS analysis. A total of 13 graphite mines/provinces are available for this study, divided into 37 individual graphite samples, which results into 17316 trace element data for this method. In some cases, distinguishing down to the level of the mine was not possible; therefore, samples marked with * will only be discussed at the province level as the material is likely a mixture from multiple mines from one province. In China, the exact mines are unknown and can partly be artisanal and small-scale mines. Brickaville* (MAD) samples are from the mines Vatomina and Sahamamy Sahasoa. In the province Minas Gerais* (BRA), samples originate from the mines Pedra Azul, Itapecerica and Salto da Divisa. All localities in China are referred to as provinces. For each mine/province, there was a varying number of samples available. The subsequent tables (Tab. 16 to 28) list the available samples for each location (concentrate/raw ore) and show median values for each element.

Table 16: Median values Kaisersberg (AUT): 1A = concentrate; 21G, H and I are raw ore samples

Kaisersberg (AUT)	Analyte ($\mu\text{g g}^{-1}$)																
sample ID	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	U
1A median	396.04	5296.52	287.10	137.71	88.20	362.91	5084.70	4.45	26.34	52.00	101.21	19.76	149.71	112.08	11.64	57.28	3.80
21G median	44.97	1479.72	45.17	15.42	35.14	6.30	101.94	0.20	0.36	5.11	11.34	3.37	11.34	16.06	5.12	1.60	0.68
21H median	22.87	1777.98	23.22	17.01	27.52	3.36	58.63	0.06	0.62	3.85	10.66	4.08	17.38	14.87	5.45	1.51	0.48
21I median	2.89	152.82	15.28	2.47	22.89	1.51	11.53	1.09	0.86	2.33	1.50	12.89	1.03	4.02	1.34	0.95	0.17

Table 17: Median values Minas Gerais (BRA): all samples are concentrates

Minas Gerais (BRA)	Analyte ($\mu\text{g g}^{-1}$)																
sample ID	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	U
21C median	1.17	281.85	3.50	3.80	30.63	6.19	120.52	0.18	5.37	1.41	0.61	0.17	0.03	0.29	6.27	1.91	0.04
21N median	0.93	317.90	3.74	3.81	29.10	5.04	128.26	0.19	5.10	1.30	0.53	0.17	0.02	0.29	6.53	1.95	0.04
3B median	0.69	330.86	6.70	6.29	35.27	15.86	171.70	0.83	4.89	1.04	1.01	0.16	0.04	0.30	1.83	1.08	0.12
21M median	1.32	361.06	5.27	4.85	30.17	8.27	158.62	0.34	5.68	1.60	0.90	0.13	0.04	0.53	7.08	2.39	0.06
22C median	1.38	410.90	4.27	3.95	26.88	7.52	109.16	0.32	3.31	2.00	0.85	0.18	0.05	0.50	14.83	3.01	0.05

Table 18: Median values Shandong (CHN): all samples are concentrates

Shandong (CHN)	Analyte ($\mu\text{g g}^{-1}$)																
sample ID	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	U
21A median	1.47	397.79	13.77	34.85	45.12	2.41	461.04	0.16	8.76	9.90	1.53	0.28	0.10	1.71	1.64	0.85	0.31
22A median	4.20	16.86	5.05	0.70	21.92	0.75	14.56	0.08	0.53	0.34	0.08	0.27	0.19	0.12	0.34	0.27	0.02

Table 19: Median values Hunan Lutang (CHN): concentrate

Hunan Lutang* (CHN)	Analyte ($\mu\text{g g}^{-1}$)																
sample ID	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	U
4A median	37.47	977.16	51.64	16.96	27.79	41.35	496.97	2.66	7.65	15.98	6.35	36.72	13.54	16.41	21.41	4.60	0.59

Table 20: Median values Inner Mongolia (CHN): concentrate

Inner Mongolia* (CHN)	Analyte ($\mu\text{g g}^{-1}$)																
Sample ID	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	U
6B median	2.42	2.76	14.85	0.14	30.73	0.75	41.25	0.13	0.37	0.31	0.04	0.17	0.08	0.41	1.59	0.89	0.02

Table 21: Median values Heilongjian (CHN): concentrate

Heilongjian* (CHN)	Analyte ($\mu\text{g g}^{-1}$)																
sample ID	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	U
7B median	33.16	271.43	68.51	29.23	33.58	13.58	393.92	1.92	6.51	17.63	1.92	0.47	2.03	2.94	1.55	1.94	0.94

Table 22: Median values Kropfmühl (GER): 14B is a raw ore, 13B a concentrate

Kropfmühl (GER)	Analyte ($\mu\text{g g}^{-1}$)																
sample ID	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	U
13B median	101.18	359.74	21.51	19.81	29.74	33.68	464.22	0.89	17.66	1.81	1.12	0.14	3.44	0.90	0.50	0.71	0.16
14B median	136.80	538.99	20.94	63.30	63.18	35.46	697.79	0.49	7.56	7.73	2.80	0.43	21.02	1.05	0.59	0.14	0.12

Table 23: Median values Brickaville (MAD): all samples are concentrates

Brickaville (MAD)	Analyte ($\mu\text{g g}^{-1}$)																
sample ID	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	U
21D median	2.01	194.76	2.67	13.08	22.58	19.06	134.60	0.61	5.04	5.15	0.39	0.36	0.07	0.06	0.03	2.55	0.11
11B median	2.16	988.56	6.02	10.24	47.70	2.69	465.94	0.54	6.49	2.52	3.07	1.42	0.41	0.51	2.77	3.23	0.18
22D median	7.08	345.51	8.56	8.04	25.46	16.05	397.01	0.79	5.72	6.36	0.80	0.41	0.78	0.45	0.74	11.62	0.22

Table 24: Median values Balama (MOZ): sample 22H is a raw ore, rest of the samples are concentrates

Balama (MOZ)	Analyte ($\mu\text{g g}^{-1}$)																
sample ID	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	U
1B median	2.16	51.87	7.15	27.00	28.41	4.46	62.33	0.17	4.42	13.55	1.35	0.25	0.42	1.24	10.29	0.51	0.72
21B median	0.93	143.39	9.35	11.84	34.54	6.21	200.28	0.28	4.56	9.60	1.06	0.24	0.16	0.75	2.12	0.91	0.25
21J median	1.51	341.78	10.93	35.84	48.40	5.23	248.29	0.24	9.74	13.46	2.15	0.33	0.21	2.17	2.42	1.72	0.53
21K median	1.07	396.58	13.27	44.48	36.95	4.03	202.35	0.15	6.70	11.26	3.41	0.30	0.17	4.15	4.93	1.90	0.42
21L median	1.34	343.64	9.53	26.58	28.99	1.25	147.90	0.09	6.74	9.94	1.57	0.17	0.09	1.95	1.72	0.41	0.35
22B median	0.99	341.60	10.50	45.89	36.31	4.87	181.91	0.15	6.08	14.07	1.44	0.28	0.14	1.20	4.12	1.61	0.41
22H median	0.62	17.64	4.01	0.50	33.81	0.16	2.24	0.04	0.45	0.81	0.24	0.13	0.02	0.14	0.14	0.03	0.00

Table 25: Median values Skaland (NOR): all samples are concentrates

Skaland (NOR)	Analyte ($\mu\text{g g}^{-1}$)																
sample ID	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	U
15B median	16.94	95.84	16.31	2.50	35.74	11.61	132.31	0.37	4.42	1.91	0.76	0.10	1.85	0.53	0.06	0.27	0.07
2B median	37.97	213.82	38.77	16.91	26.45	17.81	267.89	0.76	3.94	6.12	1.05	0.57	2.07	3.13	0.57	2.04	0.49
22G median	38.30	163.39	22.92	5.69	26.75	10.81	174.94	0.55	6.84	1.03	0.61	0.11	1.38	1.09	0.10	0.65	0.19

Table 26: Median values Tayginka (RUS): all samples are concentrates

Tayginka (RUS)	Analyte ($\mu\text{g g}^{-1}$)																
sample ID	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	U
21E median	23.98	239.06	34.03	4.29	27.19	16.01	245.17	0.29	9.43	2.50	1.32	0.18	1.10	1.59	0.93	0.43	0.14
22E median	11.24	65.48	15.77	1.49	25.77	9.36	138.66	0.23	3.92	1.98	0.34	0.13	0.28	0.58	0.37	0.44	0.07
5B median	18.04	264.68	18.55	3.03	24.69	12.09	154.05	0.26	6.11	3.13	0.87	0.16	0.72	0.67	0.66	0.09	0.09

Table 27: Median values Ragedara (LKA): all samples are raw ore (lumps/chunks)

Ragedara (LKA)	Analyte ($\mu\text{g g}^{-1}$)																
sample ID	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	U
10B median	9.23	43.06	20.91	1.90	37.71	14.21	502.60	2.85	55.83	16.41	2.20	0.82	0.09	1.50	0.08	0.44	0.20
21F median	2.46	43.19	5.91	0.17	61.59	1.34	12.53	0.08	0.82	9.26	0.40	1.23	0.17	0.59	0.30	0.38	0.02

Table 28: Median values Zavallia (UKR): sample 22K-S is a raw ore sample, rest of the samples are concentrates

Zavallia (UKR)	Analyte ($\mu\text{g g}^{-1}$)																
sample ID	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	U
22F median	94.70	355.38	23.50	13.78	27.25	14.92	831.34	0.55	7.80	26.01	6.04	1.22	1.99	1.94	3.23	0.28	0.15
4B median	8.61	218.62	10.95	3.26	32.95	3.66	162.02	0.15	3.91	5.48	0.41	0.32	0.20	0.54	0.89	0.02	0.02

6.4.1.1. Laser-Ablation Signals and Ablation Marks

The ablation of graphite material using LA-ICP-MS is challenging. The properties of the graphite material (fine-grained/powdery, difficult to press into a stable form, generally unstable even as a powder pellet, partly heterogeneous grain sizes, conductivity of the material, etc.) complicated this application. To avoid the heterogeneity within the concentrates, lines with approximately 150 μm diameter were ablated for 72-75 seconds. This allowed for more stable signals (Figure 46) (compare signal Cr concentrate). Individual peaks in the intensities may also be attributed to accompanying mineral grains. Therefore, areas reflecting the graphite ablation signal only, were selected during data reduction. Data reduction was particularly important to eliminate outliers and choose stable areas of the signal for further data processing.

The analysis of graphite in raw ore was limited by the sometimes very small grain or flake sizes. The laser diameter for ablation was limited to about 50 μm ; for smaller graphite grain sizes the analysis could not be performed.

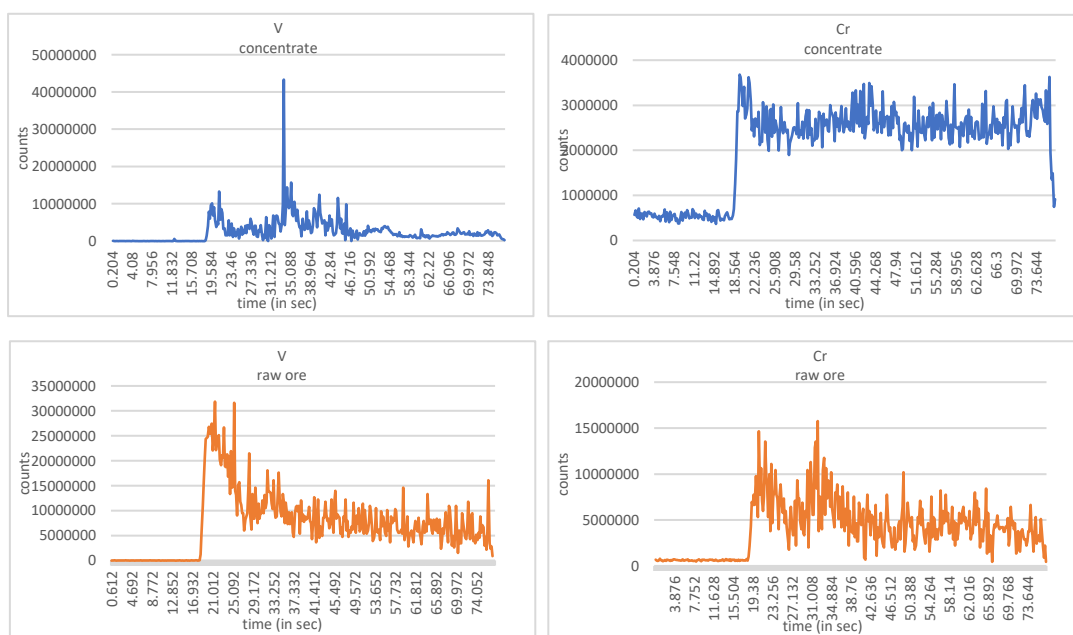


Figure 46: Laser ablation signals for V and Cr for graphite concentrates (blue) and raw ore (orange)

The remaining craters from the ablation were clearly visible after performing the analytical method. Figure 47 on the left shows the ablation of the lines in a powder pellet. The ablation lines are not directly visible, only the dark area of the ablation with the outer brownish area where the material burns out. Figure 47 right image shows the ablation crater in a raw ore graphite sample as remaining dark circle.

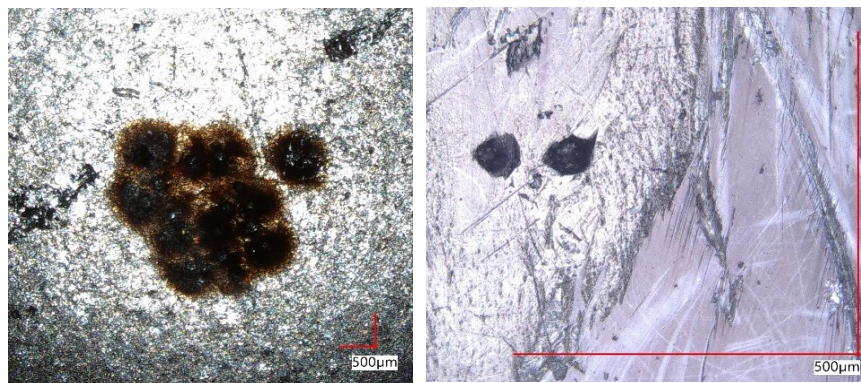


Figure 47: Laser ablation craters in graphite concentrate, sample 15B (left) and raw ore sample 10B (right)

6.4.1.2. Within Element Variability

Each of the following diagrams displays the variability of the respective element across all mines/provinces, allowing for the initial recognition of differences and the possibility of discriminations. A grouping of the individual graphite deposits for each element is not possible with the LA-ICP-MS data, as they show too few differences within the element.

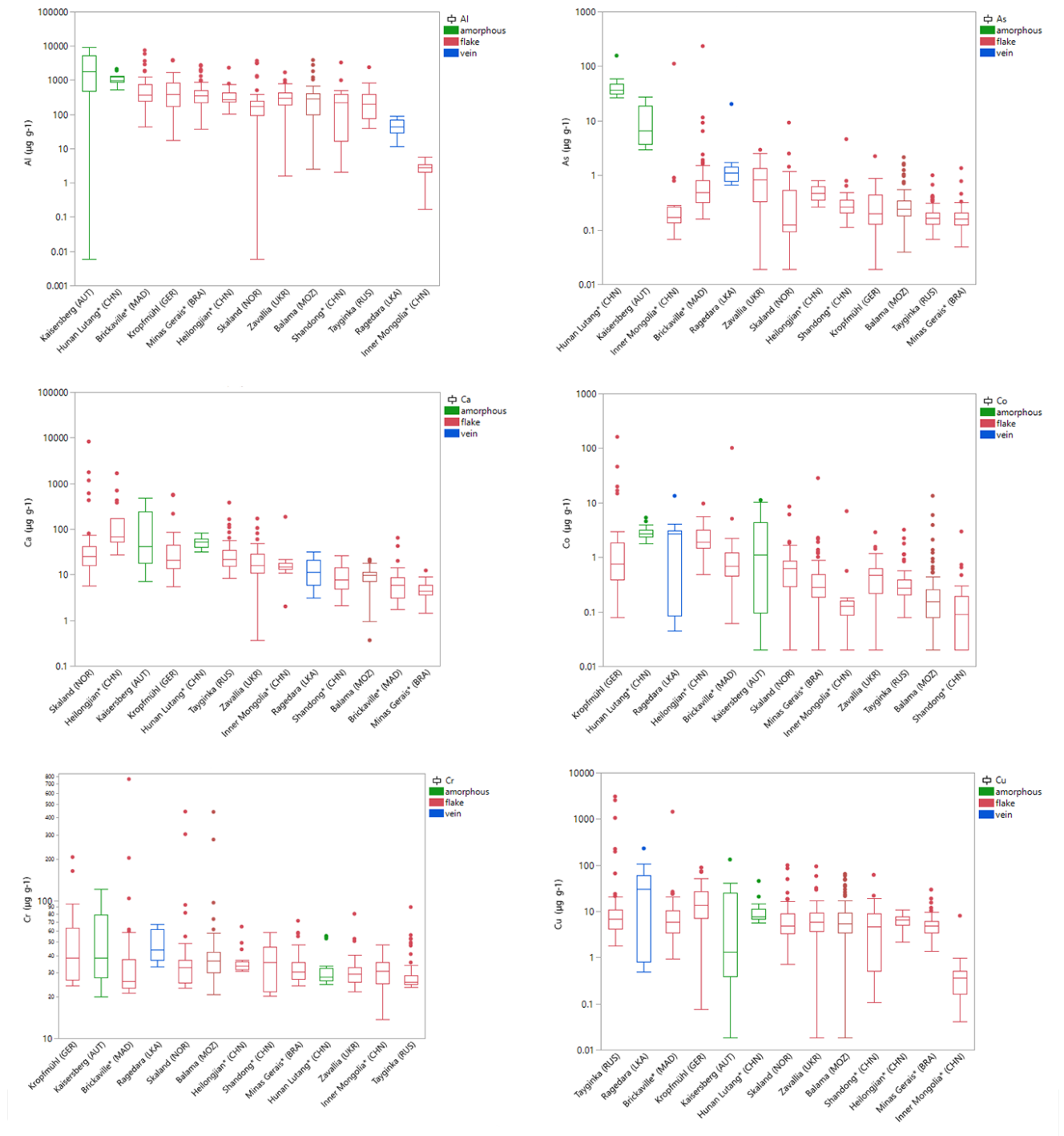


Figure 48: Within element variability for Al, As, Ca, Co, Cr, Cu (LA-ICP-MS data) in $\mu\text{g g}^{-1}$

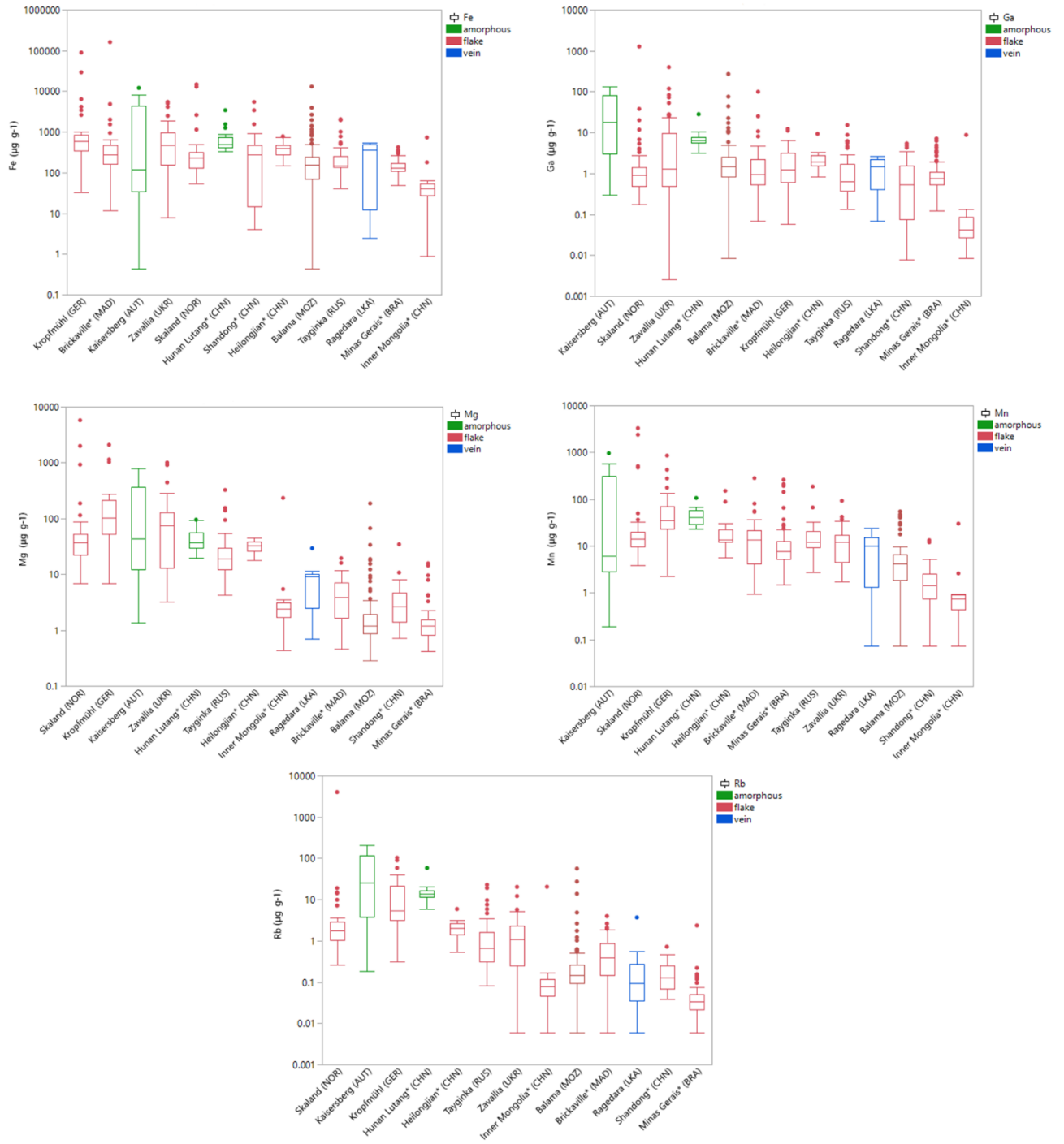


Figure 49: Within element variability for Fe, Ga, Mg, Mn, Rb (LA-ICP-MS data) in $\mu\text{g g}^{-1}$

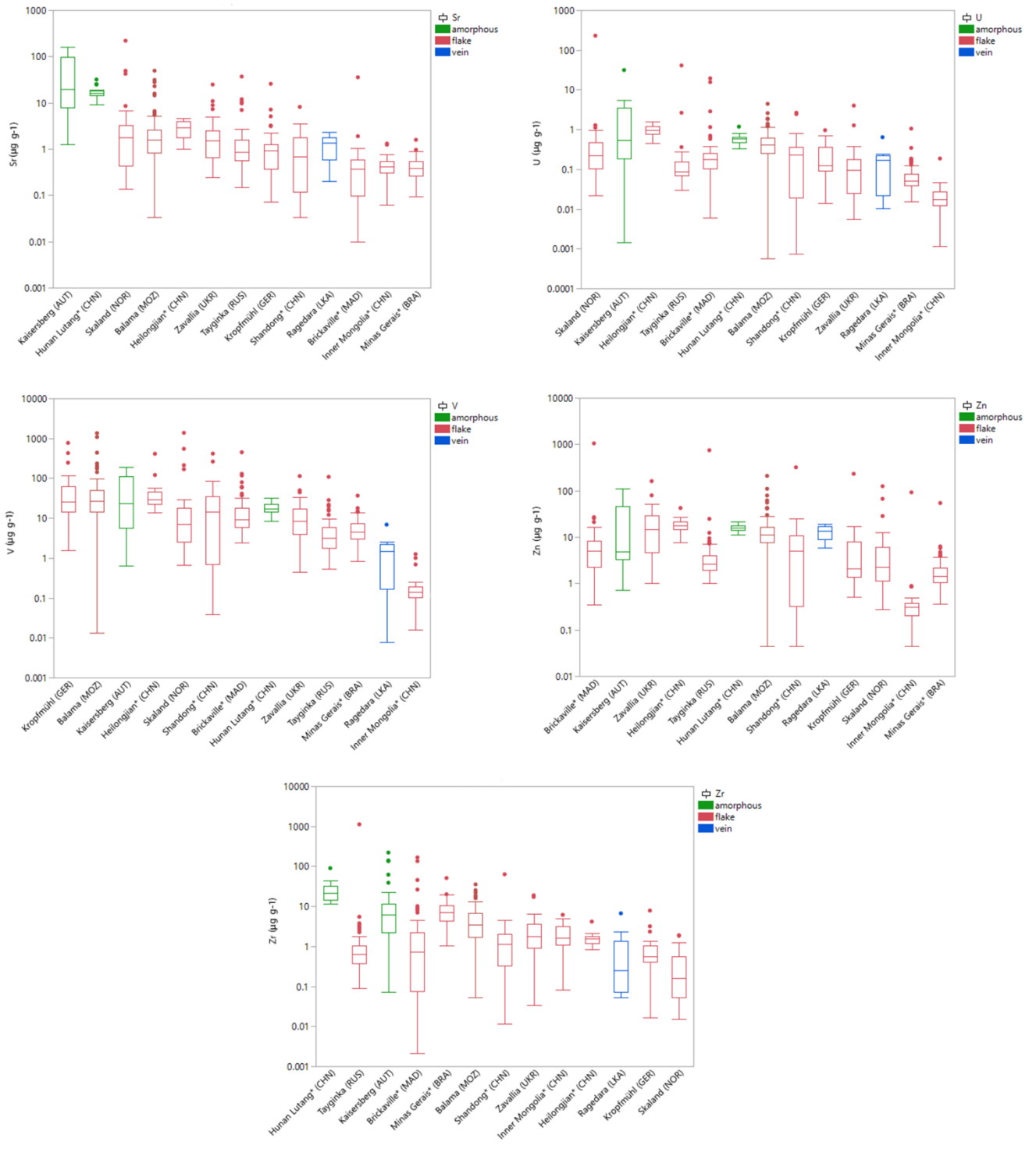


Figure 50: Within element variability for Sr, U, V, Zn, Zr (LA-ICP-MS data) in $\mu\text{g g}^{-1}$

6.4.1.3. Scatter Plots

The linear Al and Ga correlation is displayed in the scatter plot (Figure 51). A group of data points with higher Al and Ga is related to Kaisersberg. One group is associated to low Al and Ga levels, representing Inner Mongolia. Intermediate Ga and Al levels can be attributed to Ragedara, but interfere with other deposits. The rest of the samples cannot be differentiated based on these two elements. Aluminum (Al) and gallium (Ga) substitute for each other in silicate structures. High aluminum contents are usually associated with high gallium contents.

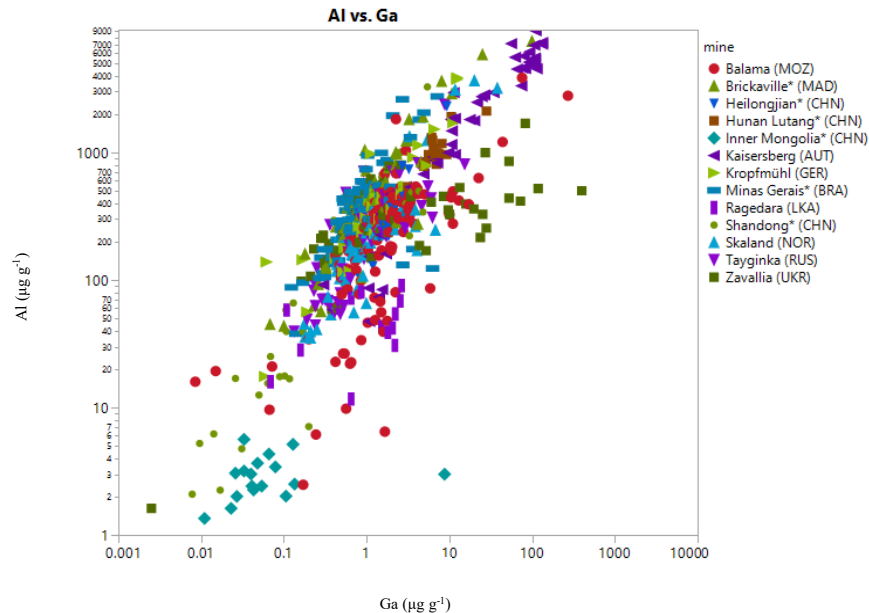


Figure 51: Scatter plot of Al/Ga (log scale, LA-ICP-MS data) in $\mu\text{g g}^{-1}$

Scatter plot (Figure 52) shows the Al/Cr variation with overlapping data points for each deposit. Data from Kaisersberg show a group at higher Al and Cr levels, with outlying points at very low Al levels. Inner Mongolia also shows a set of data points located at lower Al levels, compared to other deposits. Overall, data points for Al/Cr do not allow a discrimination of individual deposits. There is no substitution between Al and Cr. Chromium might be incorporated into iron oxides, which in turn can occur as accompanying minerals in the graphite concentrate.

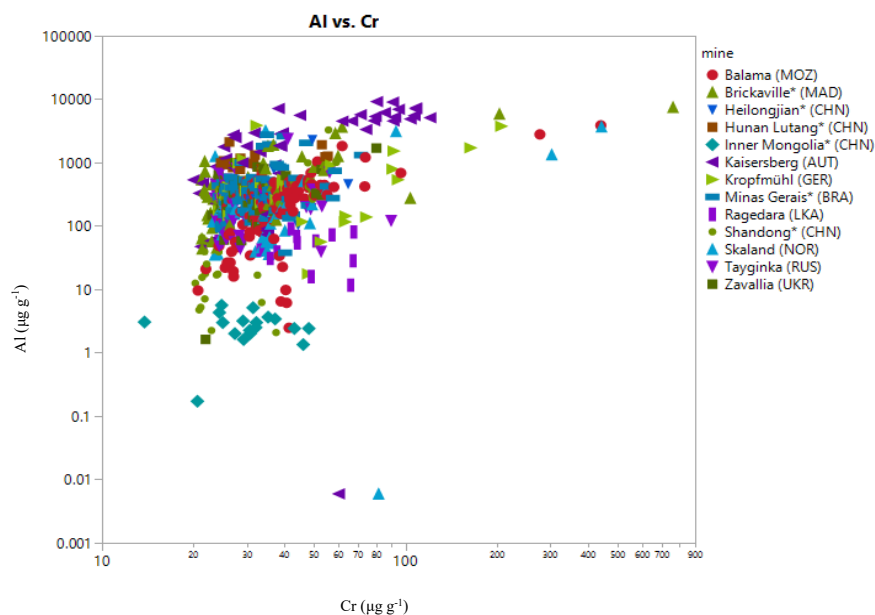


Figure 52: Scatter plot of Al/Cr (log scale, LA-ICP-MS data) in $\mu\text{g g}^{-1}$

Al/Sr variation shows most data points are distributed across the graph (Fig. 53) with some data points at significant lower Al and Sr levels (Inner Mongolia), as well as some higher Al and Sr values for Kaisersberg. Most locations (data points) are located at intermediate Al and Sr levels. Strontium (Sr) can be incorporated into carbonates and feldspars, whereas aluminum (Al) can only be incorporated into feldspars.

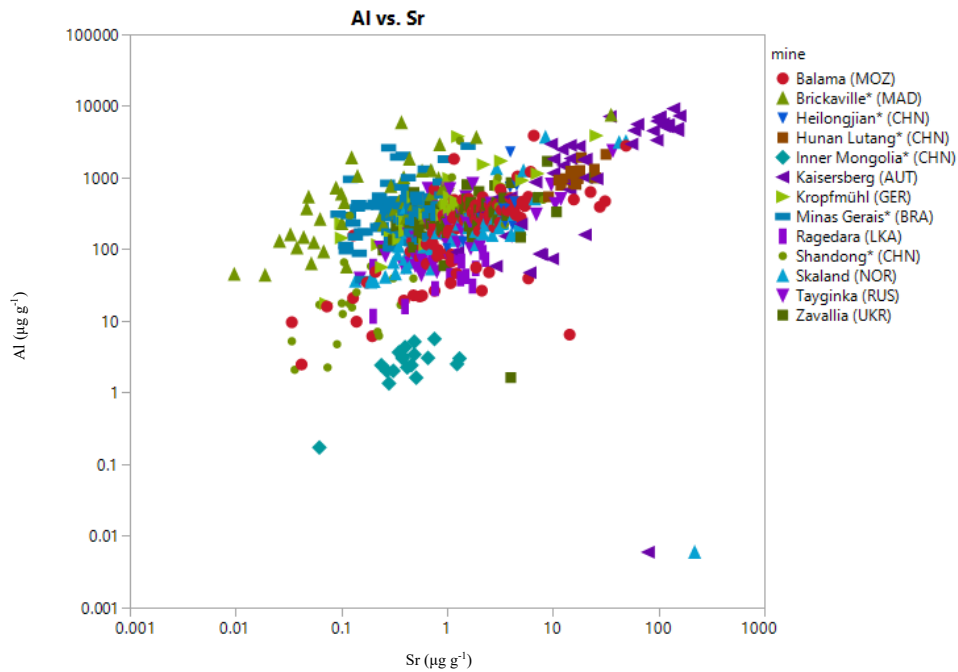


Figure 53: Scatter plot of Al/Sr (log scale, LA-ICP-MS data) in $\mu\text{g g}^{-1}$

V/Ga element concentrations (Figure 54) demonstrate a linear correlation. Graphite deposits scatter across a large range of Ga and V levels. A differentiation of mines/provinces based on these elements is not feasible. Inner Mongolia can be characterized by lower Ga and V levels, as well as Kaisersberg to higher Ga and V levels. Zavallia shows higher Ga levels, compared to other deposits.

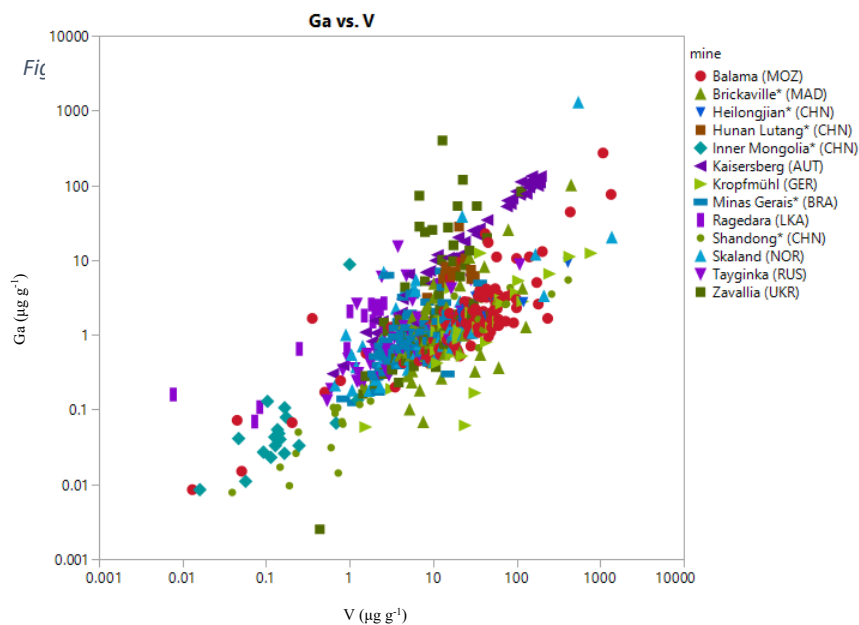


Figure 54: Scatter plot of Ga/V (log scale, LA-ICP-MS data) in $\mu\text{g g}^{-1}$

As/U (Figure 55) are not linear correlated. Data points of this element ratio allow a discrimination of some deposits, with Hunan Lutang at higher As levels compared to other data points. Kaisersberg is also associated with higher As levels, however the grouping is not as precise as with Hunan Lutang province. The rest of the data points do not allow discrimination.

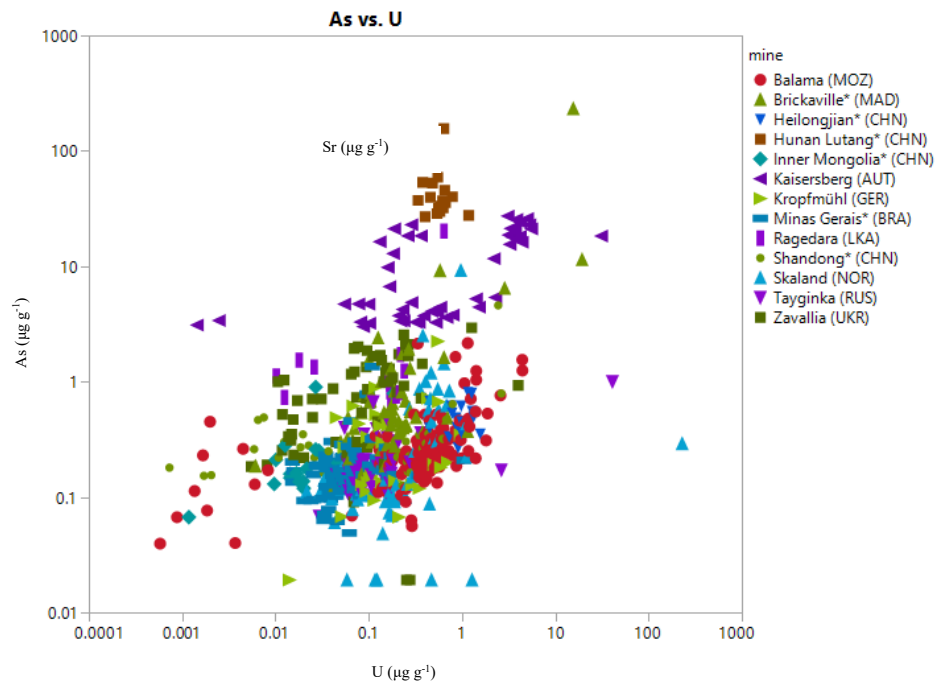


Figure 55: Scatter plot As/U (log scale, LA-ICP-MS data)

Rb/Sr (Figure 56) shows very densely distributed data points at intermediate Rb and Sr levels, with Kropfmühl at higher Rb levels and Kaisersberg at higher Sr levels, however scattered among data points from other deposits and therefore not clearly distinguishable. High rubidium (Rb) contents indicate the presence of silicates, while strontium (Sr) is incorporated into carbonates or feldspars.

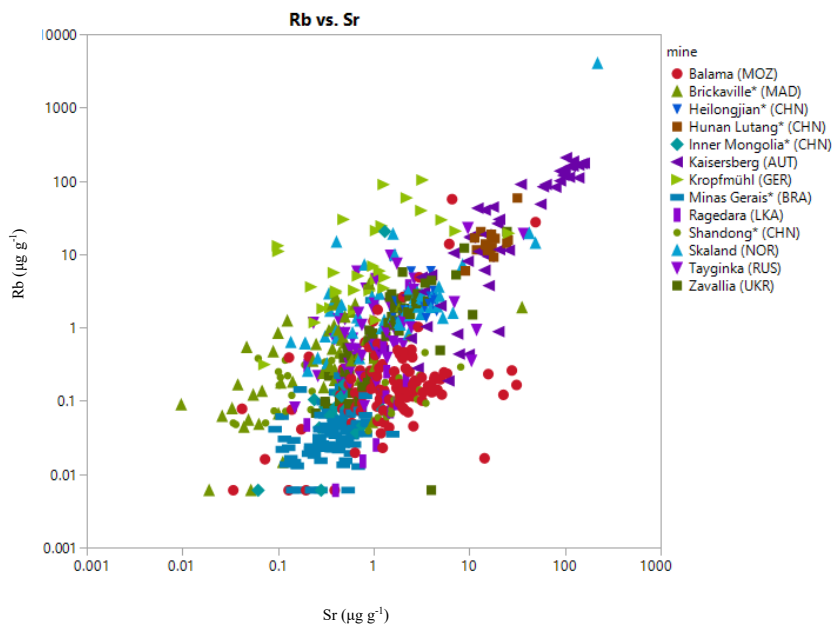


Figure 56: Scatter plot Rb/Sr (log scale, LA-ICP-MS data) in $\mu\text{g g}^{-1}$

6.4.1.4. Factor Analysis

A factor analysis was performed using the trace element data from the LA-ICP-MS analysis to reduce a large number of variables (trace elements) to a smaller number of underlying factors and to better understand the structure within the dataset. These factors were then interpreted as component groups with potential mineral phases. The loadings (L %) assist in interpreting the data structure and understanding which variables are strongly associated with which factors. The table below (Tab. 28) provides an overview of the results of the factor analysis using the LA-ICP-MS trace element data. From the dataset, five component groups were interpreted and identified: K-Al silicates (Rb as a K proxy, plus Ga or Al), K-free Ca-Al silicates (Ca plus Ga/Al), Ca plus Ce (allanite), Al/Ga with Mg/Fe (chlorite), and Ca, Sr with Mn-Mg-Fe (carbonates).

Table 28: Factor Analysis from LA-ICP-MS data; individual factors interpreted as component groups

mine/province	K-Al-Silicates (Rb as K proxy, plus Ga or Al)	L (%)	K-free Ca-Al-Silicates (Ca plus Ga/Al)	L (%)	Ca plus Ce (allanite?)	L (%)	Al/Ga mit Mg/Fe (chlorite?)	L (%)	Ca, Sr with Mn-Mg-Fe (carbonates?)	L (%)
Kaisersberg (AUT)	Sr, Ga, Rb, V, Ce, Ca, Cu, Fe, Mg, Mn, Zn, Cr	34			Sr, Ce, Ca, Cu, Mn, Zn, As, Co, Cr,	20				
Minas Gerais (BRA)			Mn, Co, Ga, V, Fe, Ca	16						
Shandong (CHN)			Cu, Al, Fe, V, Ga, Sr, Zn, Ca, Cr, Mn, Ce	54						
Hunan Lutang (CHN)	Rb, Ga, Al, Sr, Ce, Mg, V	24								
Inner Mongolia (CHN)	Rb, Ga, Zn, As, Mg, Cu, Ca, Mn, Co, Sr, V	36			Ca, Al, Sr, V, Ce	15			As, Mg, Ca, Mn, Co, Sr, V	19
Heilongjian (CHN)	Al, V, Ga, Rb, Mg, Fe, Ce, Zn	18	Mn, Ca, Al, V, Ga, Mg, Sr, Co, Zn	22	Ca, Al, Ga, Mg, Sr, Fe, Ce	12				
Kropfmühl (GER)	V, Mg, Mn, Rb, Cr, Ga, Al, Zn, AS	32					Mg, Rb, Sr, Ga, Al, Fe, As	16		
Brickaville (MAD)	Rb, Mg, Al, Ce, Sr, Ca	16					V, Cr, As, Cu, Fe, Co, Al, Ga	19		
Balama (MOZ)	V, Al, Mg, Rb, Cr, Ga	18	Zn, Fe, Cu, Al, Ca, Ce, Sr, Mn	18						
Skaland (NOR)	Mn, Mg, Fe, V, Ca, Rb, Ga, Sr	20	Cr, Mn, Mg, Fe, V, Ca, Ga, Zn, As	23						
Tayginka (RUS)	Ga, Rb, Al, Sr, V	22	V, Mg, Mn, Ca, Co, Fe	14					Sr, Mn, Ca	7
Ragedara (LKA)	Cu, Co, V, Sr, Zn, Ce, Al, Rb, As, Mg	24	Ga, Cu, Co, Mn, Ca, V, Cr, Sr, Zn, Al, Fe	40						
Zavallia (UKR)	As, Zn, Fe, Ce, Mn, Rb, Ga, V, Cu	25	Zn, Co, Mg, Sr, Mn, Rb, V, Ca	22	Zn, Fe, Ce, Sr, Al, Rb, Ga, V, Cu	20			Sr, Ca	7

6.4.1.5. Multivariate Statistical Analysis

Multivariate statistical methods analyze multiple variables simultaneously. In the diagrams in the previous chapters, the limitations of bivariate statistical methods are demonstrated. Reasons favoring the application of multivariate statistical methods include the complexity of the data. The variables are interrelated, and their relationships within the groups (mine/province) are complex. Data visualization in two or three dimensions helps to uncover patterns that may be less apparent in tabular form or uni-/bivariate statistics.

6.4.1.5.1. Linear Discriminant Analysis (LDA)

Two models using LDA were created for the LA-ICP-MS trace element dataset. The data shows significantly more dispersion within each group/deposit, compared to the solution-based-ICP-MS trace element dataset, but also more available data points. The models are based on continents and mines/provinces and allow classification of the data. The input data consist of the trace element data as well as the categories on which the classification is based.

The first LDA (Fig. 57) demonstrates the discrimination of graphite samples based on continents. Through the multivariate analysis of trace element data obtained from LA-ICP-MS, data points from South America and Africa can be differentiated from each other, whereas data points related to Europe and Asia overlap. Confidence ellipses display 95 % confidence for the mean value of each group in the diagram and assume a normal distribution of the data.

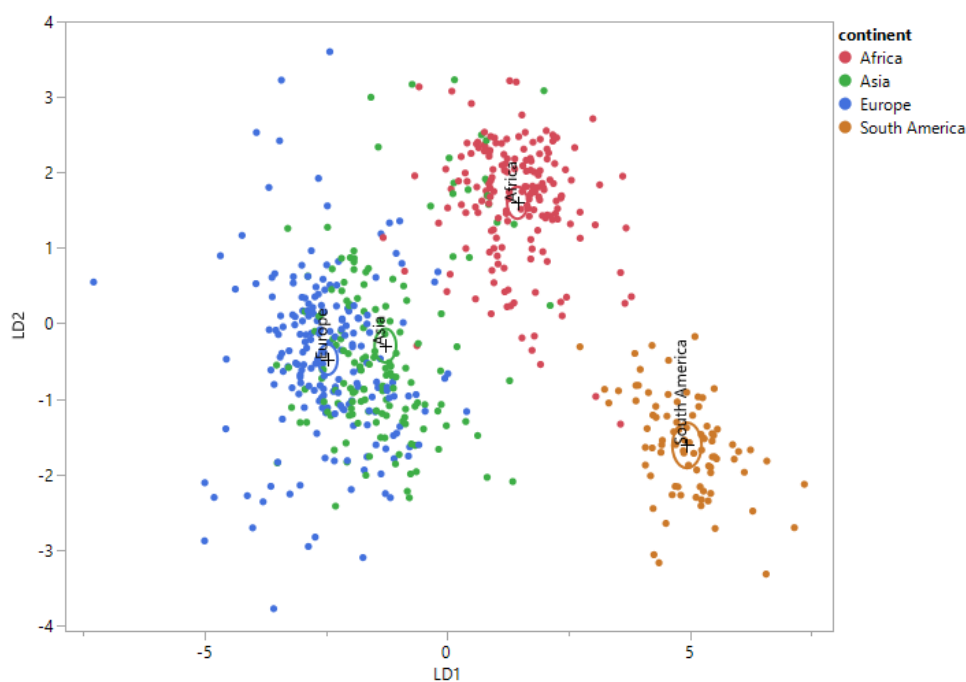


Figure 57: LDA of LA-ICP-MS trace element data based on continents

To identify **key elements** for differentiation, the scoring coefficients of the LDA were extracted and are presented in Table 29. The elements **V, Cr, Mn, Ga, Zn, Zr, Ce** contribute most for the classification of individual graphite samples based on continents (based on the scoring coefficients in Tab. 29).

Table 29: Scoring coefficients from LDA based on continents

	Log[Mg]	Log[Al]	Log[Ca]	Log[V]	Log[Cr]	Log[Mn]	Log[Fe]	Log[Co]	Log[Cu]	Log[Zn]	Log[Ga]	Log[As]	Log[Rb]	Log[Sr]	Log[Zr]	Log[Ce]	Log[U]
LD1	-0.48	0.05	-0.43	0.28	0.27	0.49	-0.34	0.24	0.08	-0.09	0.37	-0.32	-0.51	-0.25	0.51	0.41	-0.41
LD2	-0.47	-0.17	-0.14	0.37	0.22	-0.39	-0.09	-0.06	-0.08	0.58	-0.10	-0.01	0.35	-0.12	-0.34	0.01	0.35
LD3	-0.31	-0.15	0.70	-0.41	-0.05	-0.32	0.07	0.04	0.37	0.14	-0.38	0.09	0.15	-0.18	0.17	0.04	0.10

A LDA based on the classification of LA-ICP-MS trace element data according to individual mines/provinces is displayed in Fig. 58. Minas Gerais can be clearly differentiated from the rest of the dataset, as well as amorphous graphite from Kaisersberg and Hunan Lutang. The rest of the data is scattered and no further discrimination can be done.

Confidence ellipses display 95 % confidence for the mean value of each group in the diagram.

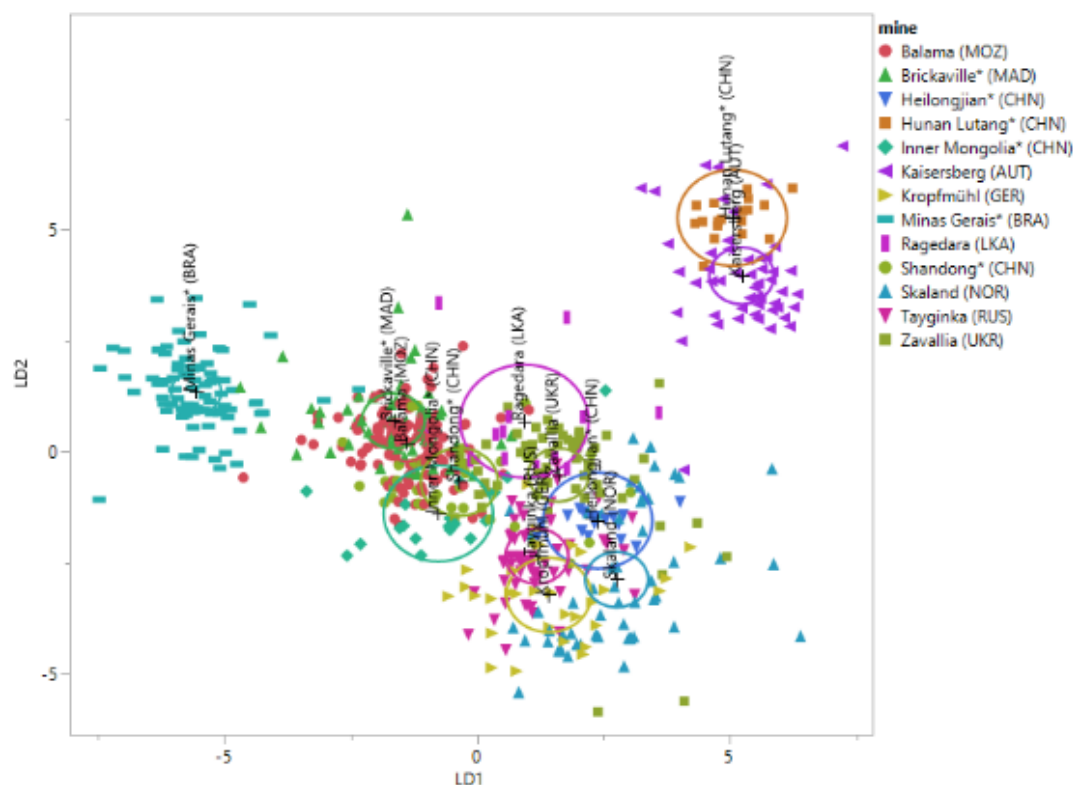


Figure 58: LDA of LA-ICP-MS trace element data based on mines

To identify **key elements** for differentiation, the scoring coefficients of this LDA were extracted and displayed in Table 30. The elements **Al, Ca, Mn, Zn, As, Rb, Sr** and **U** contribute most for the classification of individual graphite samples based on individual mines/provinces (based on the scoring coefficients in Tab. 30).

Table 30: Scoring coefficients from LDA based on all mines/provinces

	Log[Mg]	Log[Al]	Log[Ca]	Log[V]	Log[Cr]	Log[Mn]	Log[Fe]	Log[Co]	Log[Cu]	Log[Zn]	Log[Ga]	Log[As]	Log[Rb]	Log[Sr]	Log[Ce]	Log[U]
LD1	0.32	0.08	0.46	-0.43	-0.86	-0.05	0.08	-0.16	-0.19	0.11	-0.38	0.61	0.50	0.41	-0.51	0.25
LD2	-0.36	0.29	-0.56	0.03	-0.17	0.26	-0.35	0.08	-0.36	0.00	0.14	0.93	-0.13	0.35	0.44	-0.21
LD3	-0.32	-0.38	0.12	0.16	0.28	-0.33	-0.21	-0.19	-0.19	0.86	0.17	-0.19	-0.15	0.53	-0.42	0.25
LD4	-0.52	-0.19	0.59	0.81	0.27	-0.42	0.07	0.16	-0.32	-0.30	-0.60	-0.37	0.73	-0.14	0.41	0.02
LD5	-0.10	0.41	-0.21	0.70	-1.05	0.34	-0.34	-0.04	0.14	0.02	-0.09	0.00	0.17	-0.14	-0.30	-0.02
LD6	-0.38	0.16	0.11	-0.38	0.24	0.77	-0.42	-0.13	0.06	-0.27	0.10	-0.54	0.04	0.83	-0.03	0.00
LD7	-0.13	0.13	0.40	-0.69	0.27	0.00	-0.60	0.40	0.17	0.65	0.24	-0.26	0.13	-0.60	0.11	0.57
LD8	-0.53	-0.06	-0.61	-0.15	0.49	0.25	-0.02	-0.22	0.77	-0.14	-0.74	0.27	0.62	0.42	-0.20	0.02
LD9	0.43	-0.07	-0.90	-0.39	1.93	-0.07	-0.13	-0.15	-0.11	0.09	0.40	-0.26	0.15	-0.11	0.12	0.29
LD10	-0.06	-0.32	-0.33	0.46	0.94	0.11	-0.04	0.69	-0.03	-0.03	-0.25	-0.06	-0.11	0.58	-0.07	-0.49
LD11	-0.48	-0.38	-0.39	-0.28	-0.96	0.66	0.42	-0.09	-0.54	0.29	0.21	-0.18	0.37	-0.02	-0.12	0.15
LD12	0.72	-0.22	-0.75	0.19	-0.75	0.00	-0.16	0.05	0.10	-0.32	-0.16	0.12	-0.48	0.35	0.06	0.62

6.4.1.5.2. Principal Component Analysis (PCA)

Based on the elements that have been identified as significant (key elements) in the LDA, a Principal Component Analysis (PCA) was performed, which refers to the same hierarchies (continents and mines/province). PCA is a method of dimension-reduced data projection, where data are summarized into new, simplified variables and expressed as PC1 and PC2.

PC1 (50 %) and PC2 (10.6 %) for the hierarchy of continents account most of the variance in the data. They are presented in a two-dimensional graph with corresponding elements displayed (Figure 59). Data points corresponding to South America (orange) are densely distributed, whereas the datapoints of the other continents are highly dispersed, with a subgroup of data for European samples.

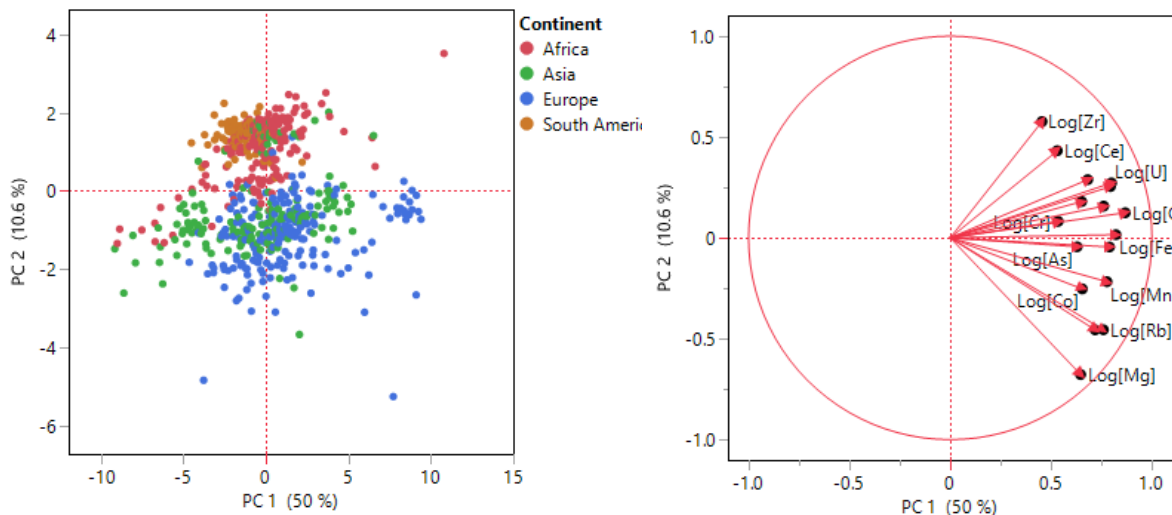


Figure 59: PCA of LA-ICP-MS trace element data at continent level (left) with the indicated elements (right)

PC1 (50 %) and PC2 (10 %) for the hierarchy of mines/provinces account for most of the variance in the data and are presented in a two-dimensional graph (Figure 60). The subgroup for European samples can be attributed to Kaisersberg, Inner Mongolia shows a densely distributed dataset, also Brickaville. The remaining data is not distinguishable based on the LA-trace element dataset only.

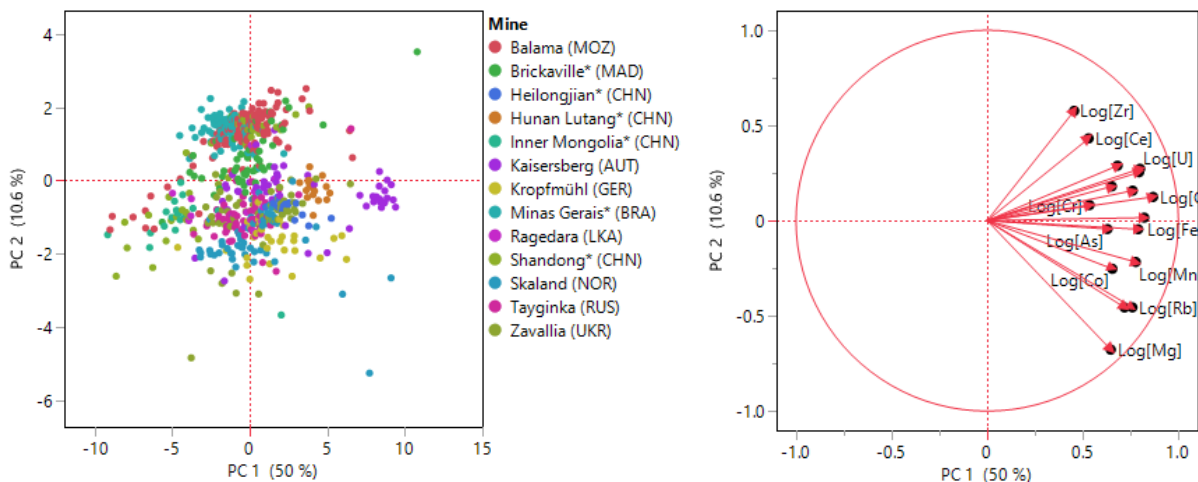


Figure 60: PCA of LA-ICP-MS trace element data at mine level (left) with the indicated elements (right)

6.4.2. Trace Element Concentrations Assessed by ICP-MS

For each available graphite sample three individual digestions were analyzed by solution-based-ICP-MS analysis. A total of 13 graphite mines/provinces are available for this study, divided into 38 individual samples, which results into 3990 trace element data for this method/parameter. In some cases, distinguishing down to the level of the mine was not possible; therefore, samples marked with * will only be discussed at the province level as the material is likely a mixture from multiple mines from one province. In China, the exact mines are unknown and can partly be artisanal and small-scale mines. Brickaville* (MAD) samples are from the mines Vatomina and Sahamamy Sahasoa. In the province Minas Gerais* (BRA), samples originate from the mines Pedra Azul, Itapecerica and Salto da Divisa. All localities in China are referred to as provinces. For each mine/province, there was a varying number of samples available. The subsequent tables (Tab. 31 to 43) list the number of samples available for each location and show median values of the measurements for each element.

Table 31: Trace element concentrations Kaisersberg (Austria) with median, mean and standard deviation

Kaisersberg (AUT)	Analyte ($\mu\text{g g}^{-1}$)																				
sample ID	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
1A	2235	28809	24.70	21.59	196.45	11993	7.80	16.13	31.48	8.45	10.51	1.92	22.37	47.87	3.40	0.13	0.07	77.34	0.18	29.46	3.22
1A	2335	29475	25.21	22.31	207.05	12583	8.21	16.93	33.93	8.70	10.77	2.15	22.93	48.55	3.89	0.15	0.07	77.69	0.18	29.39	3.38
1A	2257	29517	25.52	21.70	201.08	12029	8.27	16.32	33.18	8.61	10.97	1.88	23.23	48.72	3.44	0.14	0.08	80.12	0.18	29.63	3.28
2A	1892	16091	18.95	20.82	128.57	10471	21.13	59.03	40.87	4.88	5.82	2.07	13.67	19.41	1.96	0.21	0.14	56.40	0.10	25.62	2.00
2A	1995	14502	16.65	18.65	122.82	9999	21.19	45.30	47.16	4.34	7.41	1.82	11.74	17.28	1.98	0.16	0.17	51.93	0.10	26.97	1.88
2A	1744	14088	17.19	19.60	113.74	8592	17.63	101.10	63.01	4.34	6.03	1.88	12.35	16.82	1.88	0.24	0.27	50.76	0.09	26.62	1.72
median	2115	22450	21.82	21.21	162.51	11232	12.95	31.12	37.40	6.66	8.96	1.90	18.02	33.64	2.69	0.15	0.11	66.87	0.14	28.18	2.61
mean	2076	22080	21.37	20.78	161.62	10945	14.04	42.47	41.60	6.55	8.59	1.95	17.72	33.11	2.76	0.17	0.13	65.71	0.14	27.95	2.58
SD	214	7216	3.84	1.28	40.26	1391	6.06	30.96	10.97	2.04	2.23	0.12	5.17	15.29	0.83	0.04	0.07	12.82	0.04	1.60	0.72

Table 32: Trace element concentrations Minas Gerais (BRA) with median, mean and standard deviation

Minas Gerais (BRA)	Analyte ($\mu\text{g g}^{-1}$)																				
sample ID	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
21M	22.71	3435	10.91	9.63	23.07	1797	3.06	12.53	3.75	1.08	0.05	1.76	0.15	1.41	3.61	0.12	0.02	5.54	0.02	0.30	0.13
21M	19.43	3018	9.92	8.83	20.98	1669	2.89	11.72	3.48	0.96	0.06	1.36	0.14	1.20	2.51	0.11	0.02	5.07	0.01	0.26	0.12
21M	18.58	2865	9.48	8.46	21.16	1678	2.65	11.43	3.53	0.92	0.04	1.57	0.13	1.16	3.02	0.11	0.02	5.08	0.01	0.26	0.13
21N	24.10	2534	8.84	13.23	19.20	1409	2.64	13.86	3.24	0.84	0.04	1.28	0.10	1.19	2.28	0.10	0.02	4.60	0.02	0.20	0.12
21N	24.45	2773	9.18	13.59	20.13	1420	2.88	13.93	4.54	0.89	0.03	1.01	0.10	1.12	2.17	0.09	0.02	4.86	0.02	0.21	0.13
21N	22.85	2408	8.58	12.98	18.79	1415	2.53	13.75	4.01	0.80	0.04	1.06	0.10	1.10	2.61	0.09	0.02	4.67	0.02	0.19	0.11
22C	21.64	3254	6.69	6.07	32.58	1311	2.43	8.96	4.01	0.95	0.06	0.30	0.11	1.29	1.80	0.08	0.01	7.31	0.03	0.31	0.11
22C	21.33	3134	6.57	5.97	32.02	1286	2.47	9.02	4.39	0.92	0.04	0.41	0.10	1.35	1.51	0.08	0.01	7.13	0.03	0.30	0.10
3B	17.56	4839	33.89	17.78	135.85	2897	3.22	13.66	2.80	1.27	0.19	0.65	0.10	1.61	6.01	0.08	0.03	34.71	0.08	0.60	0.55
3B	15.01	3721	33.93	17.10	144.93	3016	3.29	13.85	3.93	1.04	0.19	0.57	0.09	1.50	5.62	0.08	0.03	43.86	0.09	0.64	0.55
3B	16.12	4234	38.28	17.41	124.79	2999	3.45	13.70	3.77	1.15	0.19	0.60	0.10	1.76	5.95	0.08	0.03	33.48	0.08	0.59	0.58
21C	25.78	2560	8.88	13.45	21.64	1482	2.68	14.11	2.90	0.84	0.03	1.08	0.10	1.83	2.55	0.09	0.02	4.96	0.02	0.19	0.12
21C	25.37	2682	9.34	13.94	20.89	1480	2.71	15.03	3.14	0.87	0.07	1.30	0.11	1.88	2.59	0.09	0.02	4.91	0.02	0.18	0.12
21C	25.13	2493	8.92	13.80	20.61	1464	2.61	14.63	2.92	0.82	0.05	1.16	0.10	2.19	2.25	0.10	0.02	4.74	0.02	0.33	0.13
median	22.17	2941	9.26	13.34	21.40	1481	2.70	13.72	3.64	0.92	0.05	1.07	0.10	1.38	2.57	0.09	0.02	5.08	0.02	0.28	0.13
mean	21.43	3139	14.53	12.30	46.90	1809	2.82	12.87	3.60	0.95	0.08	1.01	0.11	1.47	3.18	0.09	0.02	12.21	0.03	0.33	0.21
SD	3.43	687	10.98	3.79	46.44	622	0.31	1.85	0.53	0.13	0.06	0.43	0.02	0.32	1.48	0.01	0.01	13.33	0.03	0.16	0.18

Table 33: Trace element concentrations Hunan Lutang (CHN) with median, mean and standard deviation

Hunan Lutang (CHN)	Analyte ($\mu\text{g g}^{-1}$)																				
sample ID	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
4A	524	7891	20.42	5.55	92.25	4186	13.02	24.30	33.14	3.06	64.29	1.82	18.59	24.78	3.46	0.25	0.29	33.89	0.28	37.64	0.64
4A	523	7498	20.73	5.26	94.62	4213	13.77	25.54	31.22	2.94	65.06	1.77	17.80	25.01	3.29	0.26	0.29	33.35	0.28	40.80	0.68
4A	533	7885	21.54	5.45	96.02	4281	13.52	26.07	31.79	3.06	65.76	1.65	18.56	24.46	3.17	0.29	0.29	34.26	0.29	38.85	0.68
median	524	7885	20.73	5.45	94.62	4213	13.52	25.54	31.79	3.06	65.06	1.77	18.56	24.78	3.29	0.26	0.29	33.89	0.28	38.85	0.68
mean	526.79	7758	20.89	5.42	94.30	4227	13.44	25.30	32.05	3.02	65.04	1.75	18.32	24.75	3.31	0.27	0.29	33.83	0.28	39.09	0.66
SD	4.71	184	0.47	0.12	1.56	40	0.31	0.74	0.80	0.06	0.60	0.07	0.37	0.23	0.12	0.01	0.001	0.37	0.002	1.30	0.02

Table 34: Trace element concentrations Inner Mongolia (CHN) with median, mean and standard deviation

Inner Mongolia (CHN)	Analyte ($\mu\text{g g}^{-1}$)																				
sample ID	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
6B	60.22	53.91	0.62	21.11	4.19	891	6.54	1.17	1.57	0.08	0.17	0.05	0.18	0.97	1.76	0.08	0.01	3.41	0.01	0.33	0.06
6B	56.94	49.93	0.50	16.31	4.23	829	5.39	0.98	0.84	0.08	0.10	0.01	0.16	0.91	1.08	0.07	0.01	0.98	0.02	0.31	0.06
6B	57.81	52.38	0.54	16.91	4.20	1256	5.26	0.98	13.17	0.09	0.12	0.00	0.16	0.97	1.13	0.08	0.01	1.01	0.02	0.25	0.06
median	57.81	52.38	0.54	16.91	4.20	891	5.39	0.98	1.57	0.08	0.12	0.01	0.16	0.97	1.13	0.08	0.01	1.01	0.02	0.31	0.06
mean	58.33	52.07	0.55	18.11	4.21	992	5.73	1.04	5.20	0.08	0.13	0.02	0.17	0.95	1.32	0.08	0.01	1.80	0.02	0.29	0.06
SD	1.39	1.64	0.05	2.14	0.02	189	0.58	0.09	5.65	0.01	0.03	0.02	0.01	0.03	0.31	0.00	0.00	1.14	0.004	0.03	0.001

Table 35: Trace element concentrations Heilongjian (CHN) with median, mean and standard deviation

Heilongjian (CHN)	Analyte ($\mu\text{g g}^{-1}$)																				
sample ID	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
7B	650.14	3686.97	69.78	9.07	38.08	3728.04	16.78	19.33	51.96	1.31	0.48	4.48	4.11	10.79	28.65	0.23	0.73	18.61	0.12	7.18	2.40
7B	642.97	3745.60	69.12	8.97	37.50	3694.67	16.67	19.04	51.67	1.34	0.49	4.86	4.27	10.80	28.07	0.23	0.74	18.47	0.12	6.71	2.45
7B	627.25	3595.49	68.70	8.95	36.68	3597.35	16.58	18.16	50.25	1.29	0.51	4.59	4.02	10.30	26.45	0.22	0.70	18.50	0.12	6.58	2.65
median	642.97	3686.97	69.12	8.97	37.50	3694.67	16.67	19.04	51.67	1.31	0.49	4.59	4.11	10.79	28.07	0.23	0.73	18.50	0.12	6.71	2.45
mean	640.12	3676.02	69.20	9.00	37.42	3673.36	16.68	18.84	51.29	1.32	0.49	4.64	4.13	10.63	27.73	0.23	0.72	18.53	0.12	6.82	2
SD	9.56	61.77	0.44	0.05	0.57	55.44	0.08	0.50	0.74	0.02	0.01	0.16	0.10	0.23	0.93	0.01	0.02	0.06	0.00	0.26	0.11

Table 36: Trace element concentrations Shandong (CHN) with median, mean and standard deviation

Shandong (CHN)	Analyte ($\mu\text{g g}^{-1}$)																				
sample ID	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
22A	208	564.1	11.60	0.93	4.73	387	1.43	2.27	1.55	0.17	0.08	0.17	1.07	2.66	6.61	0.12	0.01	3.16	0.05	0.64	0.11
22A	193	609.4	7.06	0.99	5.47	503	1.64	2.17	1.51	0.17	0.07	0.20	1.15	2.46	5.30	0.13	0.01	4.31	0.05	0.71	0.10
21A	16.52	3298.9	70.20	27.80	5.34	5827	11.40	24.87	20.39	0.77	0.35	4.39	0.24	5.94	29.61	0.33	0.57	18.44	0.02	0.49	1.54
21A	15.79	3464.5	70.54	28.26	4.27	6109	11.07	23.52	20.52	0.79	0.37	4.57	0.23	6.27	30.47	0.34	0.83	19.13	0.02	0.47	0.86
21A	17.79	3377.7	70.41	28.11	4.28	5969	11.25	23.67	21.23	0.78	0.38	4.64	0.24	6.23	28.54	0.33	0.52	18.55	0.01	0.45	0.80
median	17.79	3299	70.20	27.80	4.73	5827	11.07	23.52	20.39	0.77	0.35	4.39	0.24	5.94	28.54	0.33	0.52	18.44	0.02	0.49	0.80
mean	90.23	2263	45.96	17.22	4.82	3759	7.36	15.30	13.04	0.54	0.25	2.79	0.59	4.71	20.11	0.25	0.39	12.72	0.03	0.55	0.68
SD	90.18	1370	29.95	13.27	0.51	2708	4.76	10.69	9.40	0.30	0.15	2.13	0.43	1.76	11.58	0.10	0.33	7.35	0.02	0.11	0.54

Table 37: Trace element concentrations Kropfmühl (GER) with median, mean and standard deviation

Kropfmühl (GER)	Analyte ($\mu\text{g g}^{-1}$)																				
sample ID	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
13B	4145	4331	82.05	23.92	186.48	10738	16.53	293.78	23.07	2.12	0.78	1.49	21.39	4.22	29.97	0.13	0.21	8.83	0.20	1.58	1.62
13B	4271	4413	84.18	24.86	190.89	10841	15.90	305.17	16.81	2.12	0.50	1.14	22.20	3.95	26.11	0.13	0.19	8.93	0.21	1.41	1.41
13B	4217	4331	85.53	25.06	191.27	10823	16.56	307.16	19.05	2.16	1.28	1.24	21.64	3.26	39.28	0.13	0.16	8.64	0.20	1.57	1.70
median	4217	4331	84.18	24.86	190.89	10823	16.53	305.17	19.05	2.12	0.78	1.24	21.64	3.95	29.97	0.13	0.19	8.83	0.20	1.57	1.62
mean	4211	4358	83.92	24.61	189.55	10801	16.33	302.04	19.64	2.13	0.85	1.29	21.74	3.81	31.79	0.13	0.19	8.80	0.21	1.52	1.58
SD	51.74	38.93	1.43	0.50	2.17	44.84	0.30	5.90	2.59	0.02	0.32	0.15	0.34	0.40	5.53	0.00	0.02	0.12	0.00	0.07	0.12

Table 38: Trace element concentrations Brickaville (MAD) with median, mean and standard deviation

Brickaville (MAD)	Analyte ($\mu\text{g g}^{-1}$)																				
sample ID	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
11B	34.65	21663	54.31	57.51	5.61	24561	7.76	45.87	4.73	11.42	15.00	6.71	1.57	4.18	15.57	0.14	0.03	15.19	0.02	12.81	1.53
11B	35.51	21028	52.01	60.13	5.69	23321	7.70	48.06	6.43	11.53	15.49	4.78	1.59	4.04	15.41	0.12	0.03	15.19	0.02	12.67	1.43
11B	37.44	21774	54.39	58.44	5.45	23398	8.49	52.13	6.10	10.95	15.38	4.85	1.68	5.49	15.32	0.11	0.03	14.91	0.02	13.32	1.53
22D	128	4813	19.30	3.63	63.15	4883	9.24	21.11	28.76	1.59	0.79	0.43	3.42	1.54	4.39	0.09	0.46	9.07	0.14	6.28	0.50
22D	119	4475	18.48	3.39	59.92	4638	8.58	21.80	28.51	1.48	0.82	0.25	3.18	1.46	4.87	0.09	0.44	8.30	0.13	5.88	0.49
21D	160	3857	129.29	5.17	259.95	3989	60.20	143.38	29.61	1.55	2.48	0.66	2.65	0.71	1.91	0.02	0.11	19.09	0.17	2.42	0.90
21D	140	3730	135.36	5.26	253.62	4130	55.56	34.88	27.89	1.52	2.64	0.50	2.57	0.72	1.78	0.02	0.10	14.02	0.18	2.46	0.89
21D	174	3799	135.04	5.03	281.87	4277	64.68	39.24	31.16	1.55	3.58	0.75	3.34	0.99	2.03	0.02	0.13	12.25	0.20	2.66	0.94
median	124	4644	54.35	5.21	61.54	4761	8.91	42.55	28.20	1.57	3.11	0.70	2.61	1.50	4.63	0.09	0.11	14.46	0.13	6.08	0.92
mean	104	10642	74.77	24.82	116.91	11650	27.78	50.81	20.40	5.20	7.02	2.37	2.50	2.39	7.66	0.08	0.17	13.50	0.11	7.31	1.03
SD	54.85	8410	47.27	26.25	117.09	9391	25.18	36.60	11.39	4.73	6.46	2.45	0.74	1.76	6.11	0.05	0.17	3.31	0.07	4.57	0.40

Table 39: Trace element concentrations Balama (MOZ) with median, mean and standard deviation

Balama (MOZ)	Analyte ($\mu\text{g g}^{-1}$)																				
sample ID	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
21K	17.56	4371	102.10	30.91	13.75	3269	13.96	19.83	29.28	1.90	0.38	5.59	0.44	12.14	24.11	0.34	0.76	39.80	0.03	1.14	1.25
21K	16.75	4148	99.95	30.45	12.43	3148	13.96	19.32	30.15	1.86	0.36	5.42	0.43	11.78	22.24	0.33	0.70	39.09	0.02	1.10	1.28
21K	15.07	4122	101.20	30.97	12.23	3145	14.27	19.49	29.52	1.85	0.37	5.51	0.42	11.69	22.26	0.34	0.66	38.28	0.02	1.07	1.19
21L	18.77	4806	74.66	18.46	4.39	2319	14.87	20.47	29.44	1.32	0.13	6.11	0.25	6.60	48.41	0.49	1.13	21.18	0.03	0.67	1.00
21L	17.55	4714	73.52	18.26	4.51	2354	14.78	20.49	27.85	1.31	0.16	6.05	0.23	6.91	44.04	0.46	1.16	21.99	0.03	0.69	1.01
21L	18.68	4802	75.45	19.03	5.04	2407	14.68	21.27	30.22	1.34	0.13	6.12	0.24	6.91	50.24	0.51	1.14	22.18	0.03	0.76	1.07
1B	31.85	363	38.82	8.92	11.84	995	9.45	13.32	30.21	0.35	0.39	1.43	0.58	3.50	10.95	0.51	0.33	11.29	0.05	2.39	2.50
1B	30.00	339	37.95	8.78	11.65	958	9.21	12.35	30.40	0.36	0.46	1.39	0.53	3.47	10.28	0.51	0.33	10.33	0.05	2.54	2.37
1B	30.60	345	36.81	8.61	11.88	965	8.99	12.77	31.03	0.34	0.33	1.48	0.52	3.79	10.17	0.48	0.38	10.69	0.05	2.48	2.25
21B	15.13	1980	53.26	16.45	29.83	4339	10.26	17.22	27.98	0.59	0.40	2.21	0.37	4.19	14.71	0.17	0.25	17.62	0.04	0.60	1.13
21B	18.72	2030	54.74	16.91	31.81	4351	11.36	20.84	30.15	0.61	0.46	2.04	0.39	6.35	15.15	0.19	0.20	16.71	0.05	0.62	1.12
21B	18.46	2010	58.82	17.36	29.00	4468	11.28	18.00	29.78	0.63	0.50	1.82	0.39	6.30	14.72	0.15	0.26	16.12	0.05	0.61	1.15
21J	17.64	4169	79.96	37.06	10.61	2643	14.80	18.78	26.73	1.01	0.25	4.23	0.29	6.53	34.09	0.33	0.57	22.08	0.03	0.55	1.08
21J	16.00	3982	77.95	36.28	10.57	2564	14.48	17.82	28.55	0.96	0.31	4.38	0.27	6.23	33.72	0.33	0.62	20.71	0.03	0.49	0.98
21J	15.73	4136	80.32	37.13	10.45	2636	14.76	18.24	26.59	1.00	0.27	4.73	0.28	6.19	32.95	0.31	0.54	20.96	0.03	0.49	1.14
22B	15.59	4052	102.71	28.76	17.72	2977	15.17	20.10	29.52	1.33	0.33	4.48	0.38	5.49	25.92	0.41	1.51	20.29	0.03	0.88	1.24
22B	13.66	3794	100.02	26.84	16.31	2914	15.34	19.56	28.66	1.26	0.31	4.47	0.34	5.53	24.51	0.38	0.52	22.99	0.03	0.78	1.19
median	17.56	4052	75.45	19.03	11.88	2643	14.27	19.32	29.52	1.01	0.33	4.47	0.38	6.30	24.11	0.34	0.57	20.96	0.03	0.76	1.15
mean	19.28	3186	73.43	23.01	14.35	2732	13.04	18.23	29.18	1.06	0.33	3.97	0.37	6.68	25.79	0.37	0.65	21.90	0.03	1.05	1.35
SD	5.54	1585	22.68	9.65	8.15	1049	2.27	2.73	1.24	0.51	0.11	1.76	0.10	2.65	12.65	0.11	0.37	8.88	0.01	0.69	0.48

Table 40: Trace element concentrations Skaland (NOR) with median, mean and standard deviation

Skaland (NOR)	Analyte ($\mu\text{g g}^{-1}$)																				
sample ID	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
2B	816	2084	37.72	7.71	46.95	3488	19.97	12.68	15.89	0.75	0.94	3.11	5.71	8.96	52.41	0.48	0.19	10.42	0.08	19.49	1.89
2B	774	1871	35.60	7.07	44.51	3396	19.19	12.23	14.69	0.71	0.89	3.10	4.88	8.56	48.84	0.47	0.18	8.03	0.07	19.39	1.75
2B	776	1883	35.44	7.06	43.07	3407	18.70	12.18	14.01	0.72	0.91	3.31	5.02	8.60	51.25	0.47	0.18	7.65	0.07	19.05	1.97
15B	560	1059	7.25	5.98	24.18	1857	5.70	23.50	5.01	0.56	0.11	0.80	3.68	1.95	4.63	0.08	0.01	6.08	0.02	0.51	0.21
15B	514	962	7.01	5.56	22.78	2062	6.58	19.86	5.42	0.53	0.04	0.95	3.38	1.81	1.27	0.07	0.01	5.27	0.02	0.51	0.22
15B	614	1089	7.89	6.35	25.95	1927	5.56	22.20	5.12	0.59	0.04	0.73	4.16	3.85	10.06	0.06	0.02	7.26	0.02	0.49	0.21
22G	829	1560	11.02	7.11	23.11	3119	15.46	33.65	3.01	0.75	0.13	1.24	5.01	3.80	16.77	0.05	0.02	9.94	0.02	0.57	0.59
22G	849	1518	11.11	7.29	23.24	3108	14.76	35.05	2.83	0.74	0.10	1.49	5.13	3.63	23.75	0.05	0.02	9.71	0.03	0.58	0.59
median	775	1539	11.06	7.07	25.06	3114	15.11	21.03	5.27	0.71	0.12	1.36	4.94	3.83	20.26	0.07	0.02	7.84	0.03	0.58	0.59
mean	717	1503	19.13	6.77	31.72	2796	13.24	21.42	8.25	0.67	0.40	1.84	4.62	5.15	26.12	0.22	0.08	8.04	0.04	7.58	0.93
SD	124	400	13.36	0.68	10.25	670	5.89	8.58	5.22	0.09	0.40	1.06	0.74	2.86	20.21	0.20	0.08	1.74	0.03	9.09	0.74

Table 41: Trace element concentrations Tayginka (RUS) with median, mean and standard deviation

Tayginka (RUS)	Analyte (µg g ⁻¹)																				
sample ID	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
5B	726	3189	14.90	4.91	52.14	2623	6.23	203.54	34.58	1.03	0.34	1.15	2.96	2.25	75.66	0.57	0.48	6.22	0.02	7.08	0.65
5B	717	3331	15.22	5.01	51.53	2703	6.57	233.58	36.19	1.06	0.19	1.02	2.90	2.17	90.81	0.53	0.66	6.06	0.02	6.92	0.62
5B	760	3520	15.72	5.05	52.31	2767	6.71	224.91	38.58	1.11	0.16	1.28	3.08	2.39	76.54	0.57	0.54	6.10	0.02	6.55	0.41
21E	734	2107	14.94	4.82	77.42	3701	6.42	286.52	19.80	0.88	0.30	0.40	4.08	7.32	94.52	0.46	0.17	10.50	0.02	2.80	0.85
21E	747	2194	15.39	5.11	77.13	3738	6.18	291.74	20.50	0.91	0.20	0.34	4.18	6.67	107.23	0.49	0.16	10.37	0.02	3.26	0.79
21E	776	2350	16.28	5.25	81.39	3928	5.96	297.33	26.05	0.98	0.23	0.44	4.38	7.50	100.65	0.47	0.17	11.11	0.02	3.00	0.65
22E	634	1797	13.93	6.21	45.62	2497	4.56	97.55	21.34	0.69	0.11	0.19	2.97	3.47	42.62	0.62	0.21	11.67	0.02	1.13	0.52
22E	632	1769	13.88	5.80	45.77	2480	4.63	105.41	25.89	0.68	0.04	0.18	2.99	3.34	47.17	0.57	0.21	11.51	0.02	1.11	0.41
median	730	2272	15.08	5.08	52.23	2735	6.21	229.25	25.97	0.95	0.19	0.42	3.04	3.41	83.67	0.55	0.21	10.44	0.02	3.13	0.63
mean	716	2532	15.03	5.27	60.41	3055	5.91	217.57	27.87	0.92	0.20	0.63	3.44	4.39	79.40	0.54	0.32	9.19	0.02	3.98	0.61
SD	51	661	0.77	0.45	14.38	579	0.79	74.25	7.05	0.15	0.09	0.42	0.60	2.20	22.36	0.05	0.19	2.41	0.00	2.35	0.15

Table 42: Trace element concentrations Ragedara (LKA) with median, mean and standard deviation

Ragedara (LKA)	Analyte (µg g ⁻¹)																				
sample ID	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
10B	808	559	0.35	0.42	41.83	1909	1.41	1.63	1.43	0.42	0.08	0.04	0.06	1.97	0.00	0.01	0.00	0.30	0.01	0.59	0.04
10B	162	158	0.13	0.36	64.12	1213	8.78	197.62	5.95	0.12	0.08	0.03	0.14	1.62	42.52	0.05	0.06	0.90	0.02	7.22	0.01
10B	234	508	0.06	0.54	12.53	2416	3.87	33.03	3.81	0.24	0.05	0.05	0.08	0.36	0.21	0.04	0.01	0.39	0.02	2.24	0.02
21F	121	102	0.12	0.34	57.29	891	8.39	186.65	7.71	0.09	0.07	0.10	0.16	1.64	56.73	0.05	0.07	0.94	0.02	6.82	0.04
21F	146	42.15	0.05	0.19	226.07	9610	22.53	711.71	2.90	0.70	0.05	0.10	0.02	4.26	5.94	0.11	0.02	0.56	0.53	13.17	0.01
median	162	158	0.12	0.36	57.29	1909	8.39	186.65	3.81	0.24	0.07	0.05	0.08	1.64	5.94	0.05	0.02	0.56	0.02	6.82	0.02
mean	294	274	0.14	0.37	80.37	3208	9.00	226.13	4.36	0.32	0.07	0.06	0.09	1.97	21.08	0.05	0.03	0.62	0.12	6.01	0.02
SD	259	216	0.11	0.11	74.99	3245	7.31	255.28	2.22	0.23	0.01	0.03	0.05	1.27	23.83	0.03	0.03	0.26	0.20	4.40	0.01

Table 43: Trace element concentrations Zavallia (UKR) with median, mean and standard deviation

Zavallia (UKR)	Analyte ($\mu\text{g g}^{-1}$)																				
sample ID	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
4B	284	3950	16.17	7.86	30.19	3179	4.86	15.05	16.39	1.20	1.08	0.49	1.25	2.42	3.26	1.17	0.06	7.28	0.02	1.08	0.11
4B	275	4204	16.12	8.09	29.48	3041	4.71	14.40	18.37	1.27	0.58	0.51	1.17	2.17	1.57	1.11	0.06	5.65	0.02	1.10	0.10
4B	278	3971	16.02	7.90	30.29	3012	4.37	15.22	16.48	1.22	0.49	0.39	1.17	2.20	1.56	1.10	0.06	6.37	0.02	1.05	0.11
22F	1656	3076	29.31	9.22	48.36	9516	15.04	43.65	59.45	1.18	1.14	0.71	4.41	6.01	73.52	0.87	1.78	120.12	0.04	2.89	0.26
22F	1510	2826	26.72	8.39	43.95	8805	14.31	39.99	54.63	1.10	1.02	0.71	4.01	7.09	65.84	0.89	1.53	106.39	0.03	2.70	0.28
median	284	3950	16.17	8.09	30.29	3179	4.86	15.22	18.37	1.20	1.02	0.51	1.25	2.42	3.26	1.10	0.06	7.28	0.02	1.10	0.11
mean	801	3606	20.87	8.29	36.45	5511	8.66	25.66	33.06	1.19	0.86	0.56	2.40	3.98	29.15	1.03	0.70	49.16	0.03	1.76	0.17
SD	640	547	5.89	0.50	8.05	2989	4.92	13.25	19.65	0.06	0.27	0.13	1.48	2.13	33.19	0.12	0.79	52.51	0.01	0.84	0.08

6.4.2.1. Within Element Variability

Each of the following diagrams (Fig. 61 – 63) displays the variability of the respective element across all mines/provinces, allowing for the recognition of differences and the possibility of discriminations. Trace element data obtained from solution-based-ICP-MS analysis allows a better discrimination of locations, even when only one variable (element) is used, due to smaller dispersion of the data resulting from the higher reproducibility of the analyses.

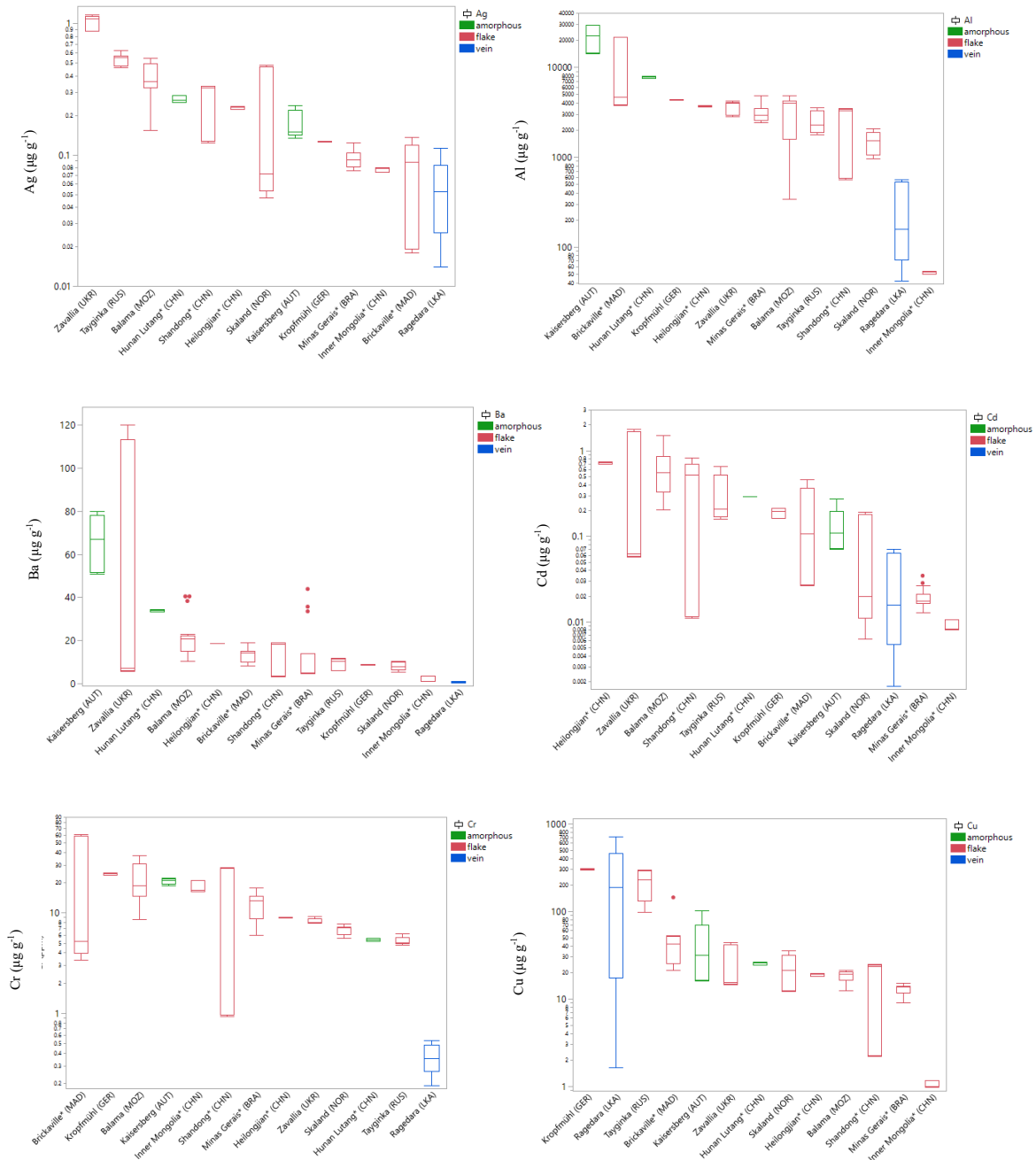


Figure 61: Within element variability for Ag, Al, Ba Cd, Cr, Cu (solution-ICP-MS data) in $\mu\text{g g}^{-1}$

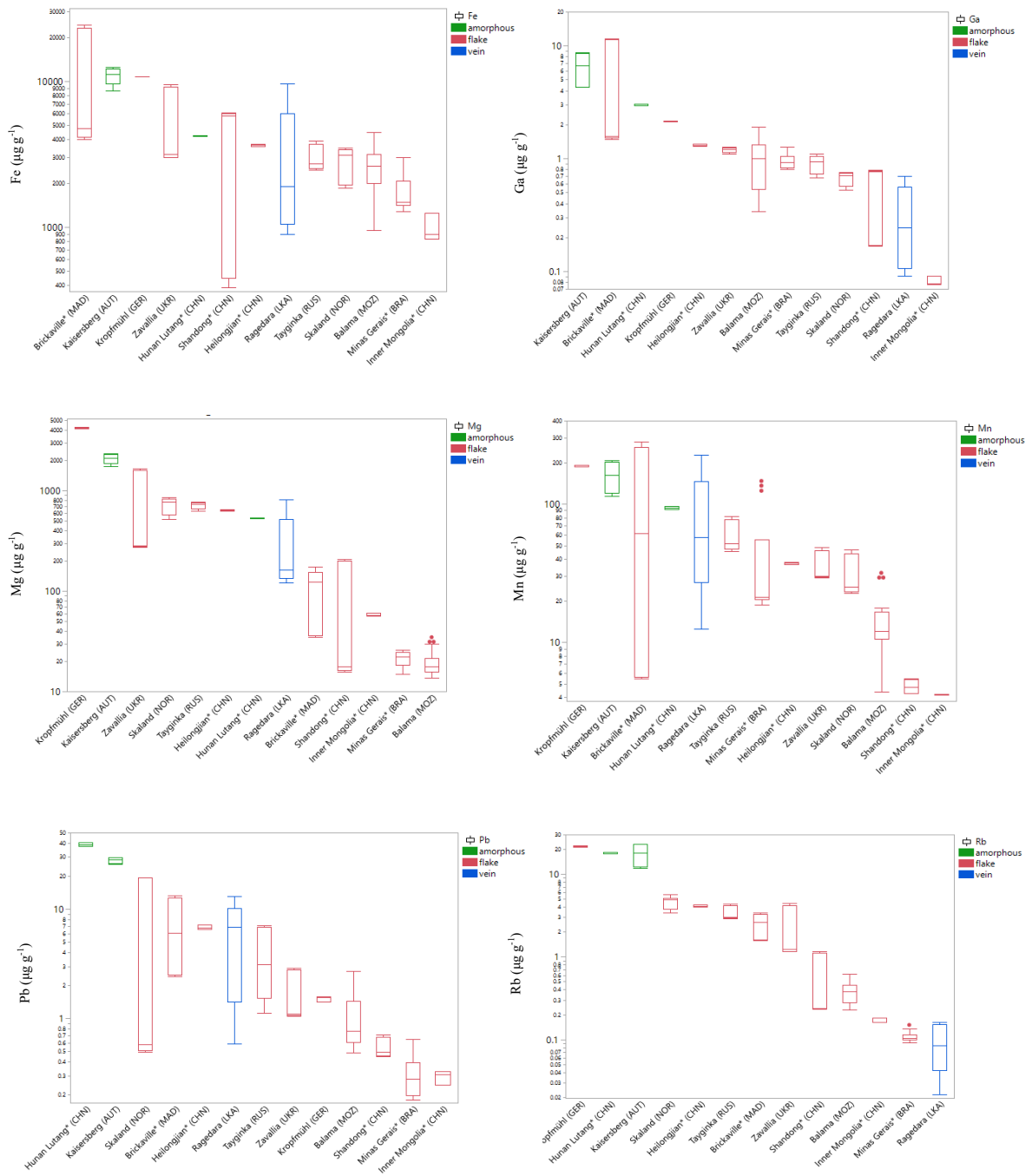


Figure 62: Within element variability for Fe, Ga, Mg, Mn, Pb, Rb (solution-ICP-MS data) in $\mu\text{g g}^{-1}$

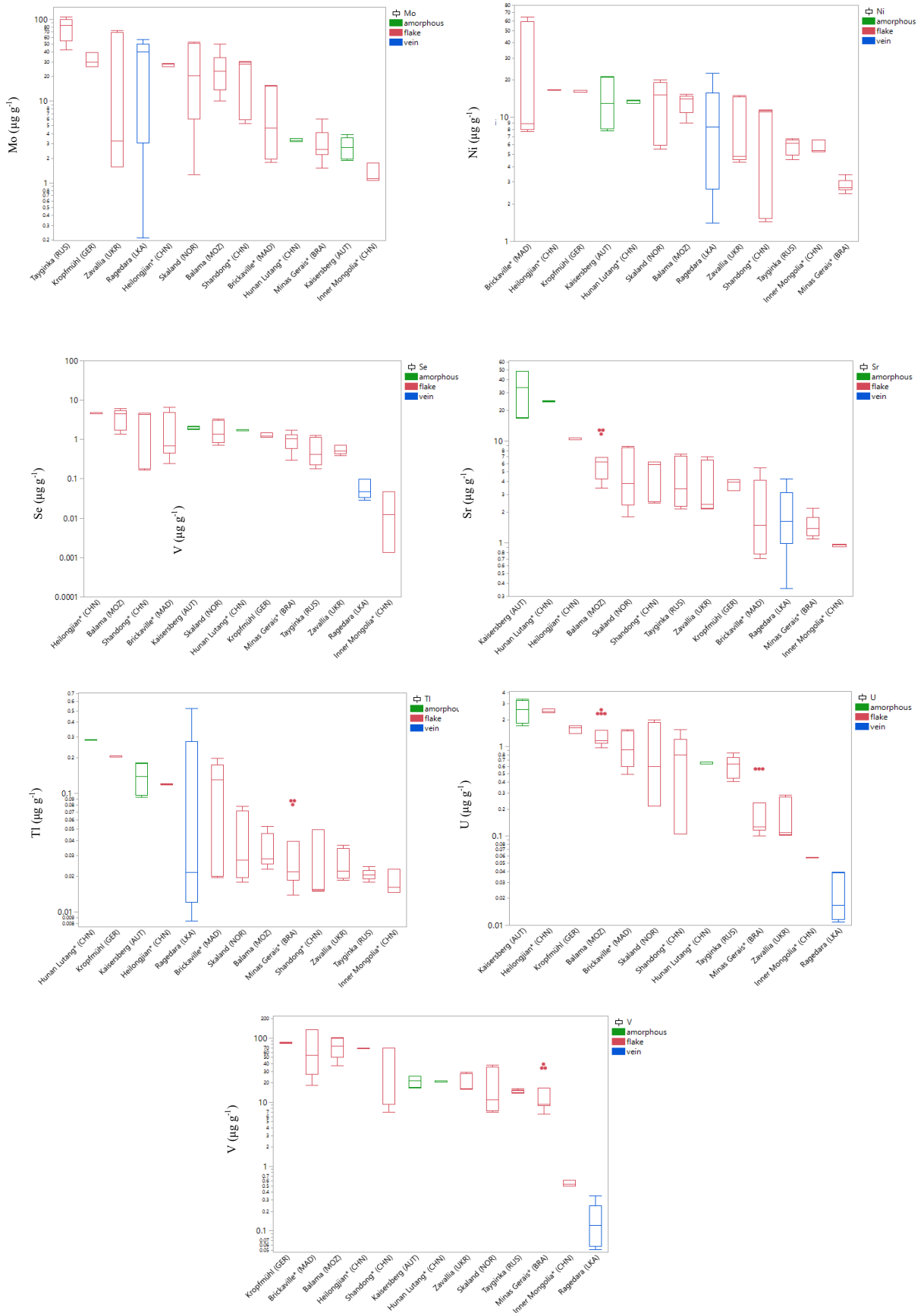


Figure 63: Within element variability for Mn, Ni, Se, Sr, Ti, U, V (solution-ICP-MS data) in $\mu\text{g g}^{-1}$

Diagrams displayed above (Fig. 61 to Fig. 63) were used for the initial qualitative differentiation of the individual locations. The mean element concentrations were divided into four groups per element, ranging from higher mean element concentrations (group 1) to lower mean element concentrations (group 4) (Tab. 44).

Kaisersberg (AUT) and Hunan Lutang* (CHN) as amorphous graphite stand out due to their high trace element content, which is attributed to their high impurity content and less precise processing.

Similarly, Kropfmühl (GER) is characterized by high trace element contents, despite being a flake graphite, which could be due to less processing. Balama (MOZ), Tayginka (RUS), and Zavallia (UKR) are characterized by their association with group 1 for the element Ag. Heilongjian* (CHN) stands out due to its higher Cd content related to the other mines/provinces. Skaland (NOR), Tayginka (RUS), Ragedara (LKA) and Zavallia (UKR) are characterized by their low trace element content. Samples from Shandong* (CHN) and Inner Mongolia* (CHN), Skaland (NOR) and Minas Gerais* (BRA) are not associated with group 1 at all. Inner Mongolia* (CHN) is predominantly associated with Group 4 in most cases, characterized by low trace element contents.

Table 44: Grouping of elements and individual mines/provinces based on mean element concentrations, as seen in Fig. 62 - 64

Location	Ag	Al	Ba	Cd	Cr	Cu	Fe	Ga	Mg	Mn	Pb	Rb	Mo	Ni	Se	Sr	Tl	U	V
Kaisersberg (AUT)	3	1	1	3	2	2	1	1	1	1	1	1	3	2	3	1	1	1	2
Minas Gerais* (BRA)	3	2	3	4	2	3	2	3	4	3	4	4	3	4	3	3	3	3	3
Hunan Lutang* (CHN)	2	2	2	3	3	3	2	2	2	2	1	1	3	2	3	1	1	2	2
Heilongjian* (CHN)	2	2	2	1	2	3	2	2	2	3	2	2	2	2	1	2	1	1	1
Shandong* (CHN)	2	3	3	2	2	3	2	3	3	4	3	3	2	2	2	2	4	2	2
Inner Mongolia* (CHN)	3	4	4	4	2	4	4	4	3	4	4	3	4	3	4	4	4	3	4
Kropfmühl (GER)	3	2	3	3	1	1	1	2	1	1	3	1	1	2	3	2	1	1	1
Brickaville* (MAD)	3	2	3	3	1	2	1	1	3	2	2	2	3	1	3	3	2	2	1
Balama (MOZ)	1	2	2	2	2	3	3	3	4	4	3	3	2	2	1	2	2	1	1
Skaland (NOR)	4	3	3	4	3	3	3	3	2	3	3	2	2	2	3	2	2	2	3
Tayginka (RUS)	1	2	3	3	3	1	3	3	2	2	2	2	1	3	3	2	4	2	2
Ragedara (LKA)	4	4	4	4	4	1	3	3	3	2	2	4	1	3	4	3	2	4	4
Zavallia (UKR)	1	2	2	3	3	3	2	3	2	3	3	3	3	3	3	2	3	3	2

6.4.2.2. Scatter Plots

Al/Ga element correlations are displayed in the graph below (Figure 64). There is a group of data points associated to higher Al and Ga levels related to Kaisersberg and Brickaville. One group is associated to low Al and Ga levels, representing Inner Mongolia. The rest of the samples cannot be differentiated based on these two elements only. Aluminum (Al) and gallium (Ga) substitute for each other in silicate structures. High aluminum contents are usually associated with high gallium contents.

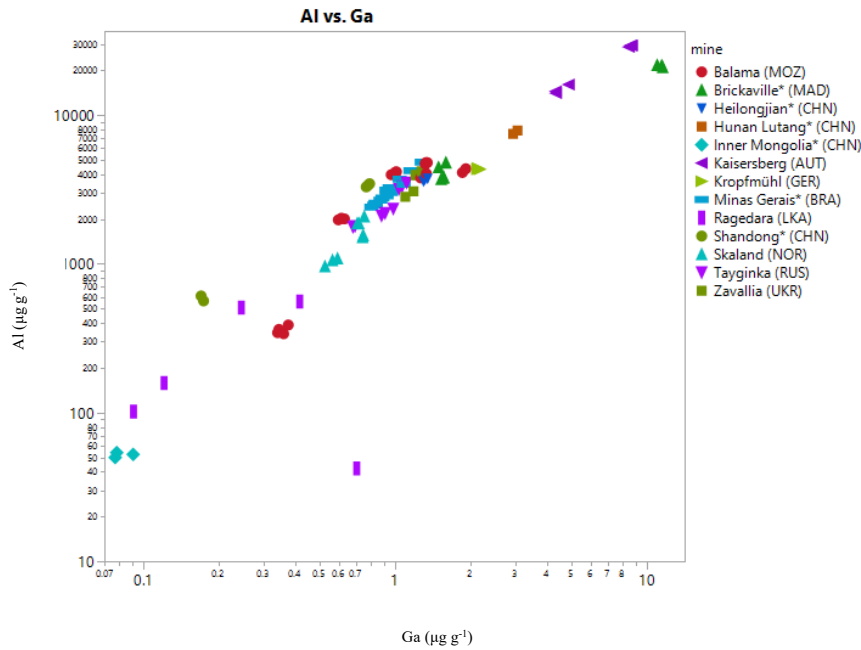


Figure 64: Scatter plot of Al/Ga (log scale, solution-ICP-MS data) in $\mu\text{g g}^{-1}$

The scatter plot (Figure 65) shows Al/Cr correlations and individual data points for Ragedara, Inner Mongolia, Kaisersberg and Hunan Lutang. They can clearly be distinguished from other provinces. The rest of the mines/provinces show similar ratios, that do not offer a distinct differentiation. Chromium might be incorporated into iron oxides, which in turn can occur as accompanying minerals in the graphite concentrate.

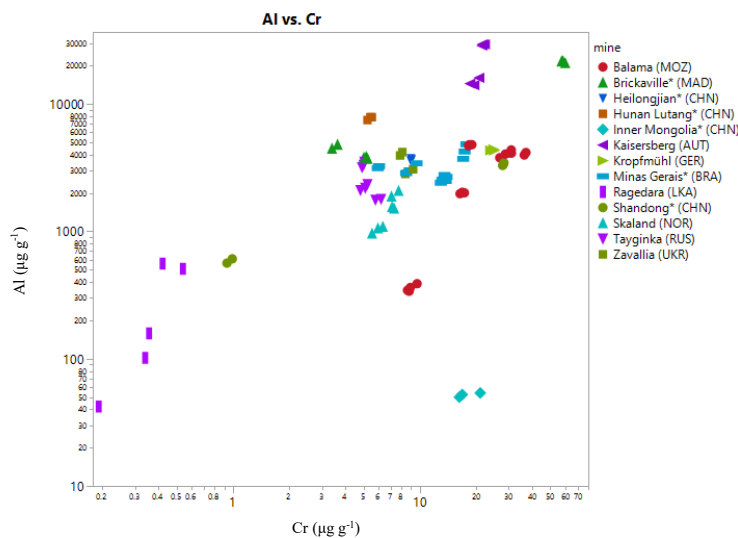


Figure 65: Scatter plot of Al/Cr (log scale, solution-ICP-MS data) in $\mu\text{g g}^{-1}$

Al/Sr correlations shows most data points scattered across the graph (Figure 66), with some data points at significant lower Al and Sr levels (Ragedara) as well as some higher Al and Sr values for Kaisersberg. Most locations (data points) are located at intermediate Al and Sr levels. Strontium can be incorporated into carbonates and feldspars, whereas aluminum (Al) can only be incorporated into feldspars.

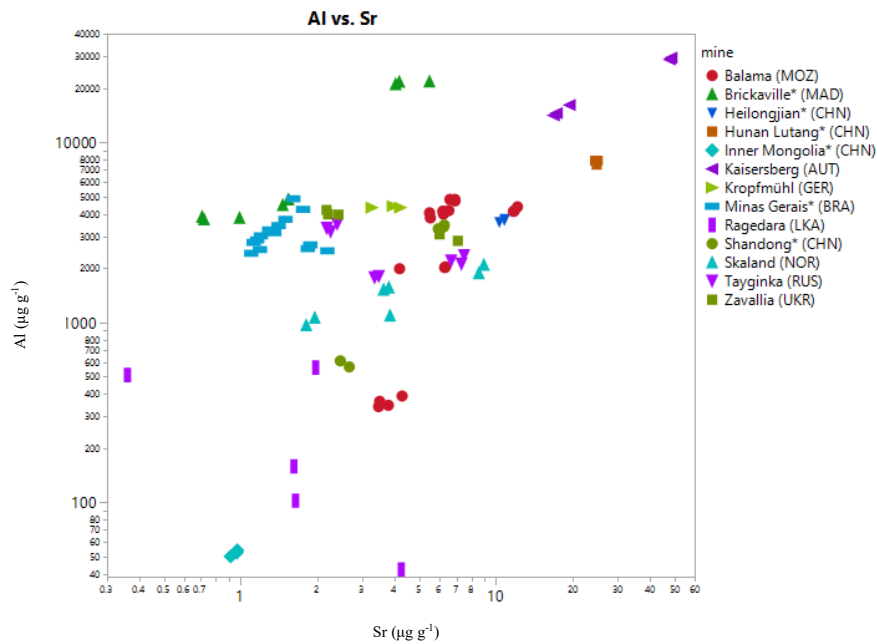


Figure 66: Scatter plot of Al/Sr (log scale, solution-ICP-MS data) in $\mu\text{g g}^{-1}$

V/Ga element concentrations (Figure 67) demonstrate a differentiation of some mines/provinces. Ragedara can be completely separated from other occurrences, as well as Inner Mongolia, along with Brickaville, which exhibits high V and Ga contents. In the range of intermediate Ga and high V content, a large number of data points are accumulated, leading to an overlap of the individual occurrences. Data points for Balama (MOZ) show a distinct correlation, which might be addressed to different V-bearing mica.

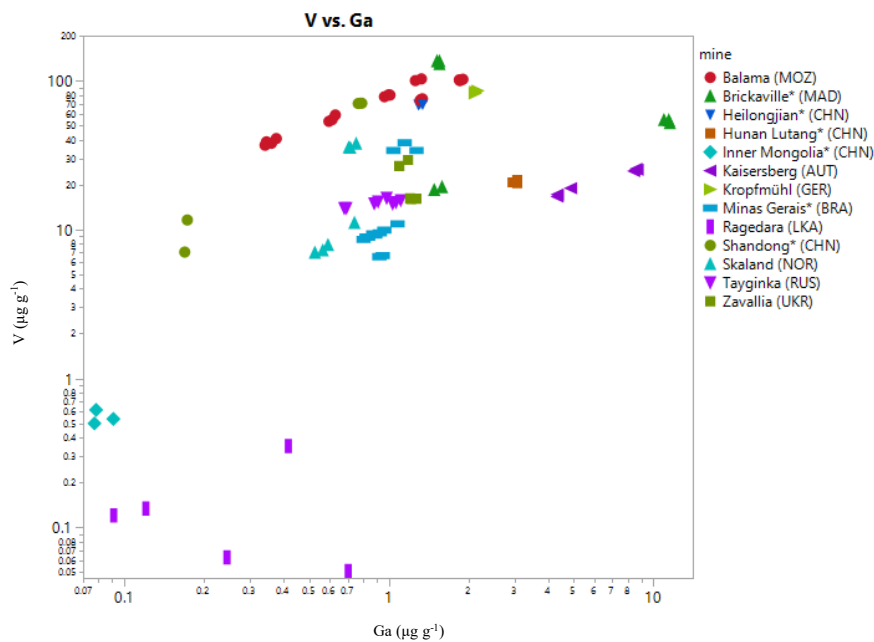


Figure 67: Scatter plot of V/Ga (log scale, solution-ICP-MS data) in $\mu\text{g g}^{-1}$

Fe/Ni element correlations (Figure 68) offer another parameter to differentiate between individual locations. Minas Gerais is characterized by lower Ni and intermediate Fe values. Some locations show two trends within the same location (e.g. Brickaville) at intermediate and high Fe values. Shandong province at low Ni and Fe concentrations scattered across datapoints from other locations. Kaisersberg shows two different trends, associated with different Ni levels at constant Fe levels.

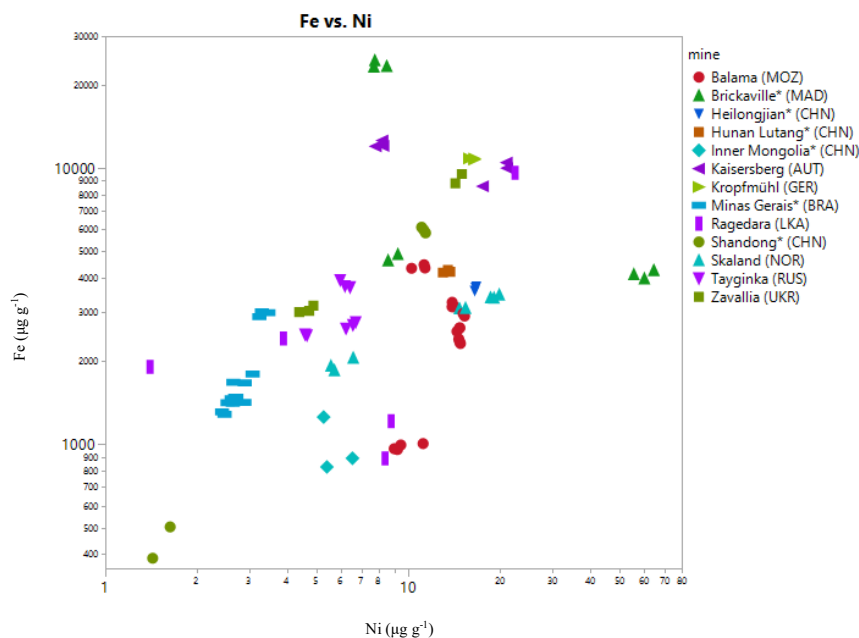


Figure 68: Scatter plot of Fe/Ni (log scale, solution-ICP-MS data) in $\mu\text{g g}^{-1}$

A potential for distinguishing individual mines/provinces is evident in the element correlation Rb/Sr (fig. 69). Samples from Minas Gerais form a distinctive group. Balama exhibits a relatively homogeneous grouping however influenced by some data points from Shandong.

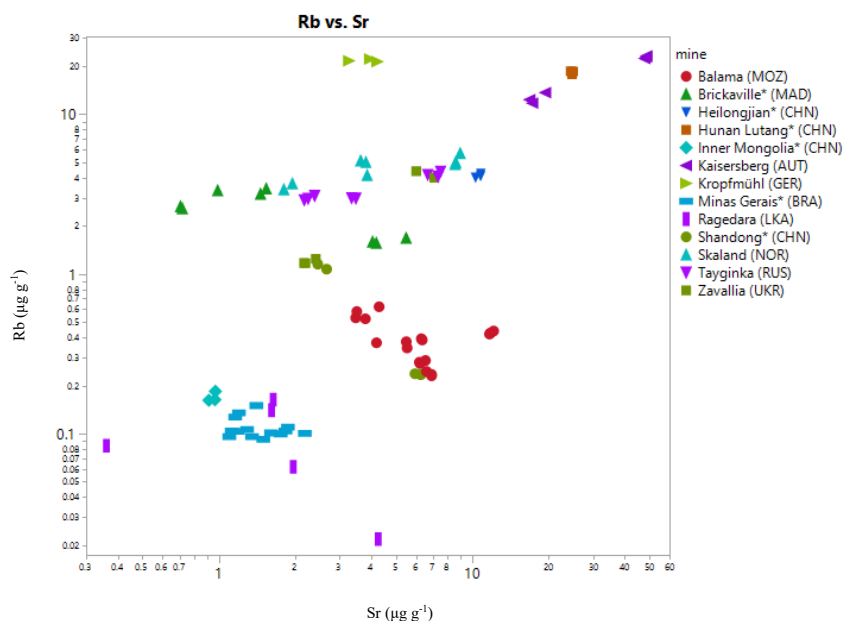


Figure 69: Scatter plot of Rb/Sr (log scale, solution-ICP-MS data) in $\mu\text{g g}^{-1}$

6.4.2.3. REE Patterns

REE concentrations from solution-based-ICP-MS analysis are presented below, due to the potential of reliably measuring all REE with this analytical method. The REE values were normalized to the EUS (Tab. 45). EUS refers to European Shale and serves as a standard reference material frequently employed to normalize REE concentrations across various samples to align with the compositional characteristics of European material analogous to Australian or North American shales (Bau et al. 2018). The diagrams below show log-normalized REE distributions of each mine/province individually and are later averaged and presented in a graph to compare the individual REE patterns of each mine/province with the others in this study. La-Eu can be indicated as LREE, Gd-Ho as MREE and Er-Lu as HREE.

Table 45: REE concentrations used for REE normalisation for European Shale (EUS)

Recommended data set for European Shale (EUS) for REY normalisation (Bau et al. 2018) in mg kg ⁻¹														
Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
31.9	44.3	88.5	10.6	39.5	7.30	1.48	6.34	0.944	5.86	1.17	3.43	0.492	3.26	0.485

The REE pattern (Fig. 70) from Kaisersberg shows a negative Eu anomaly and a depletion in heavy REE, as well as increasing LREE and decreasing MREE. The two patterns of samples 1A and 2A from Kaisersberg are similar and can be combined into a single curve without losing information, allowing for comparison with the REE patterns of other mines/provinces.

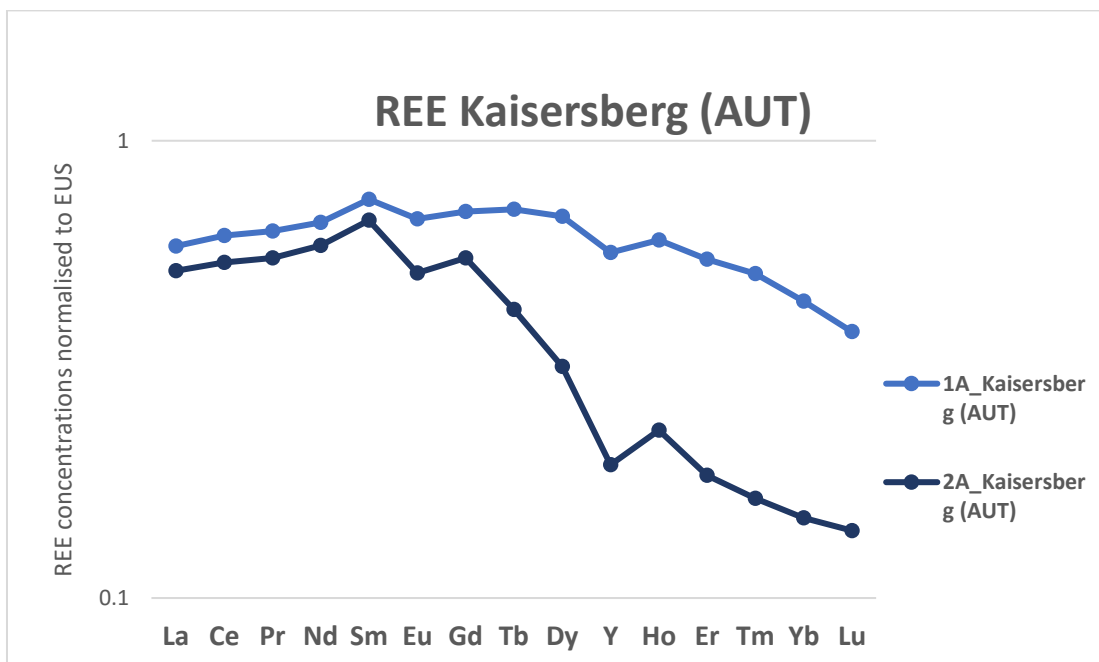


Figure 70: REE patterns of sample 1A and 2A from Kaisersberg (AUT)

REE patterns from the Minas Gerais (Fig. 71) province, including samples 21M, 21N, 22C, 3B and 21C, exhibit very homogeneous REE patterns, which can be combined for the comparison with REE patterns from other mines/provinces. The REE patterns shows a positive Ce anomaly, as well as enriched MREE and depleted HREE.

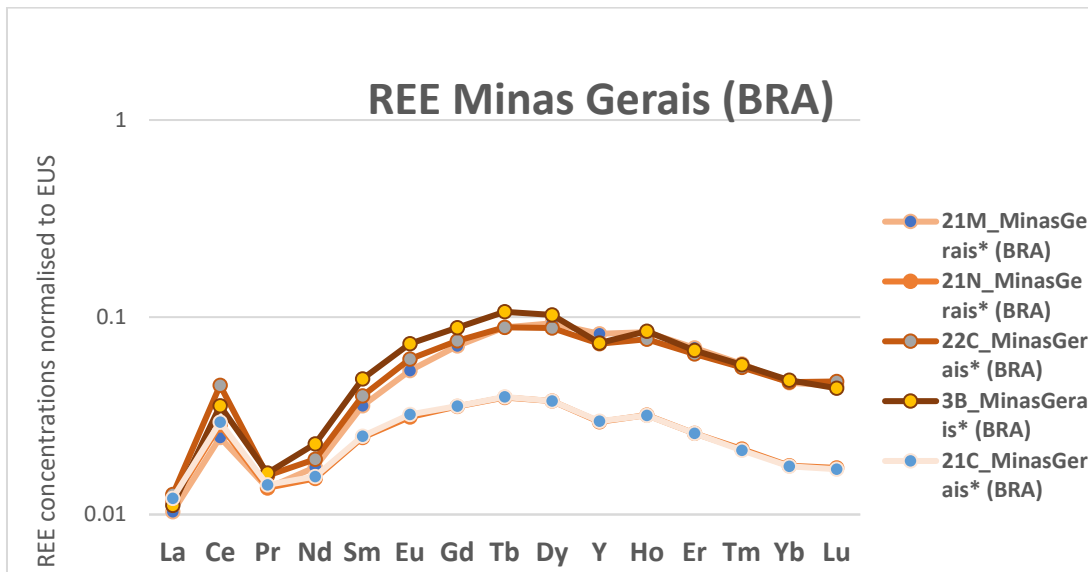


Figure 71: REE patterns of sample 21M, 21N, 22C, 3B and 21C from the Minas Gerais province (BRA)

From province Hunan Lutang (CHN) (Fig. 72) only one REE pattern was analyzed. The REE pattern shows a negative Eu anomaly, as well as depleted HREE. Positive outliers of Nd and Dy might be due to analytical errors and not related to natural processes.

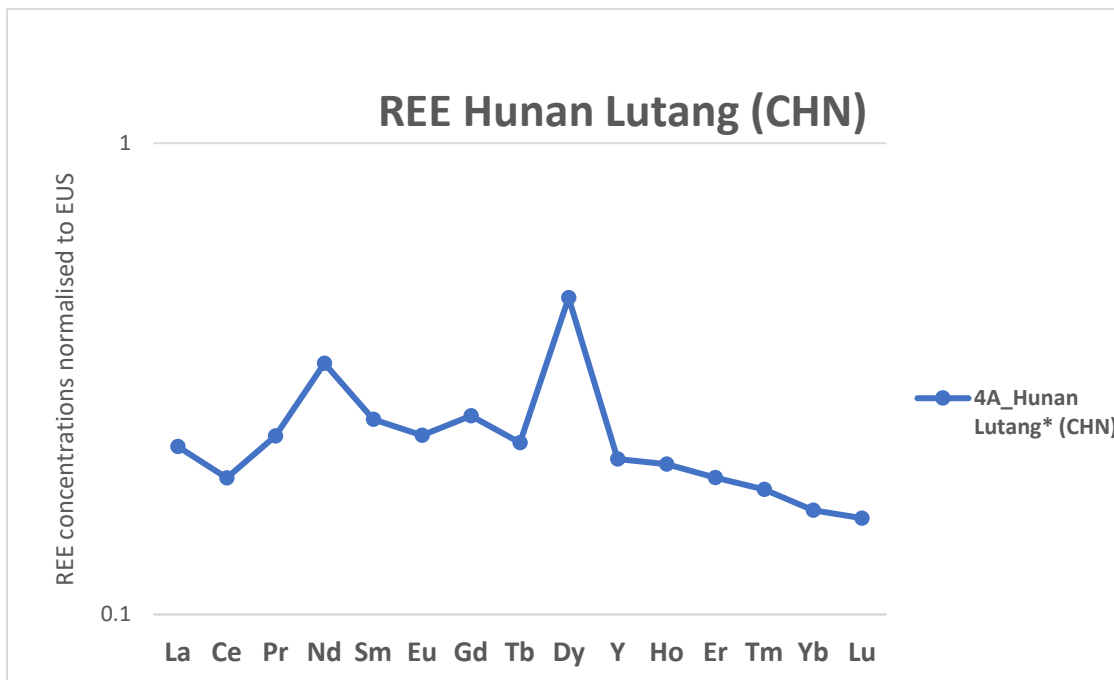


Figure 72: REE pattern of sample 4A from the Hunan Lutang province (CHN)

The REE pattern from sample 7B from Heilongjian(Fig. 73) province (CHN) shows a positive Ce anomaly, as well as increasing LREE and MREE and decreasing HREE.

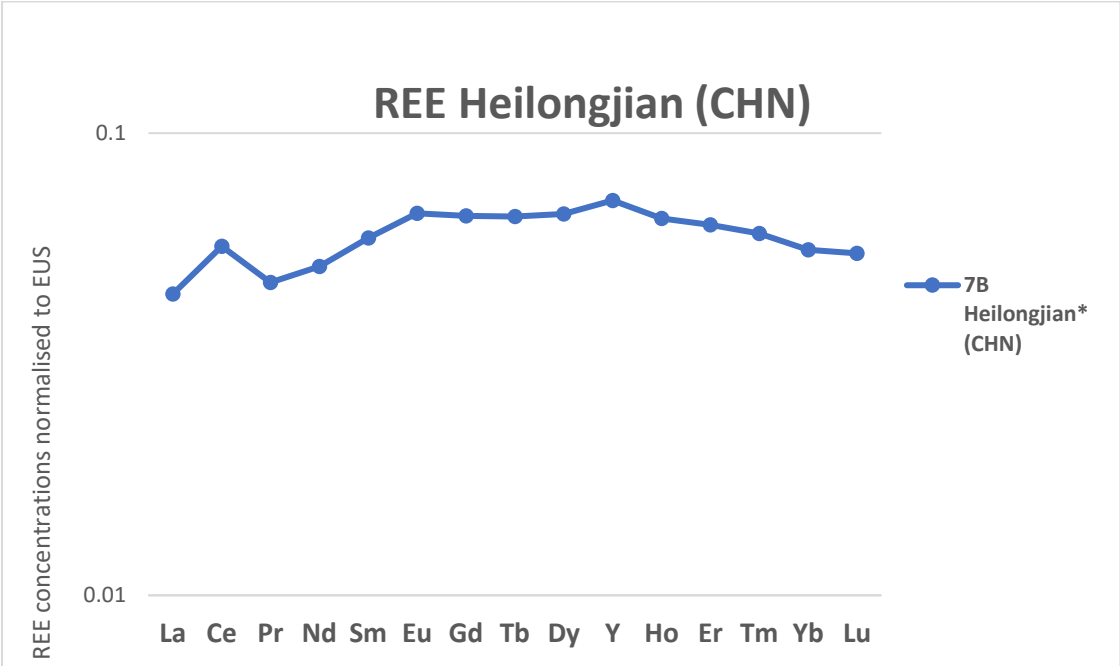


Figure 73: REE pattern of sample 7B from the Heilongjian province (CHN)

There are two available samples (21A and 22A) from Shandong province. Both samples exhibit different REE patterns (Fig. 74). Sample 21A shows a distinct negative Ce anomaly and positive Eu anomaly, with decreasing MREE and HREE. Sample 22A shows a positive Ce anomaly with decreasing LREE.

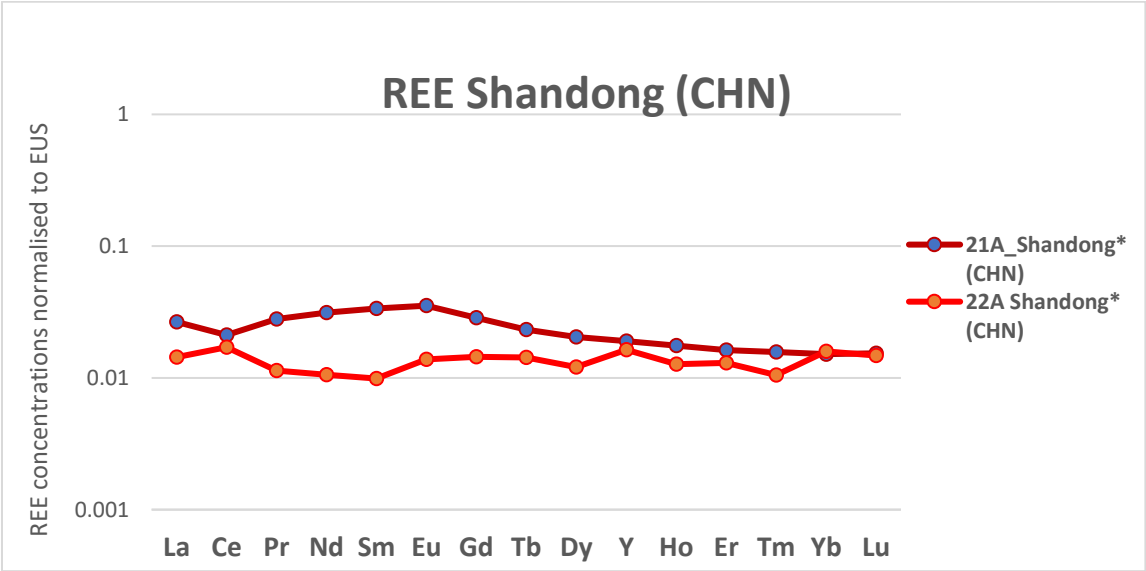


Figure 74: REE patterns of sample 21A and 22A from the Shandong province (CHN)

The REE pattern from sample 13B from Kropfmühl mine (GER) shows increasing LREE, a distinct negative Eu anomaly, as well as a depletion in MREE and HREE (Fig. 75).

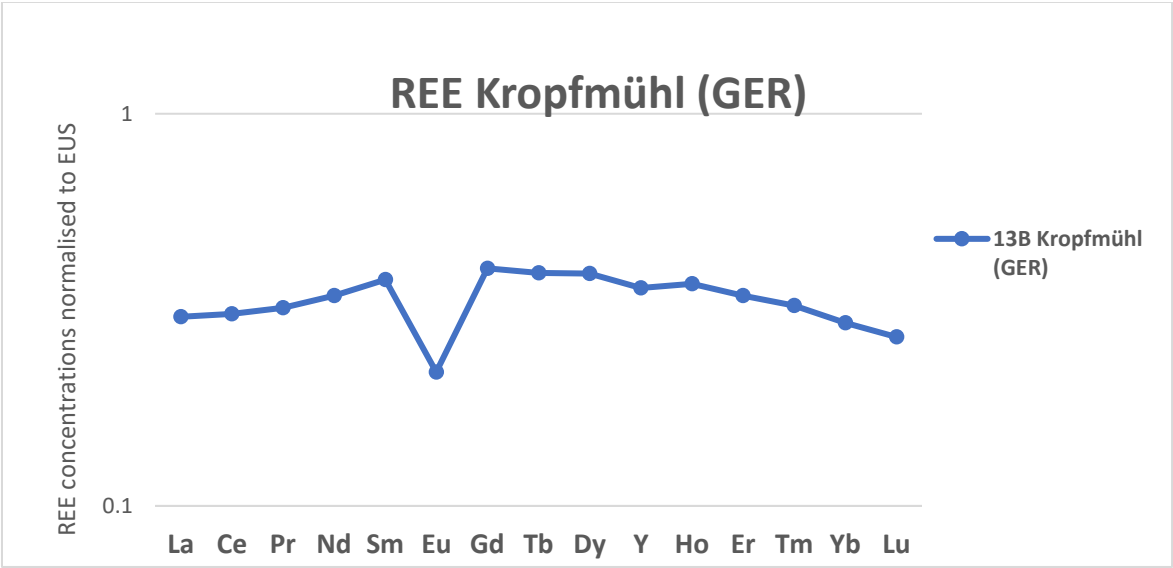


Figure 75: REE pattern of sample 13B from Kropfmühl (GER)

The REE pattern of sample 6B from Inner Mongolia (CHN) shows a distinct positive Ce anomaly with slightly decreasing LREE and MREE= HREE (Fig. 76).

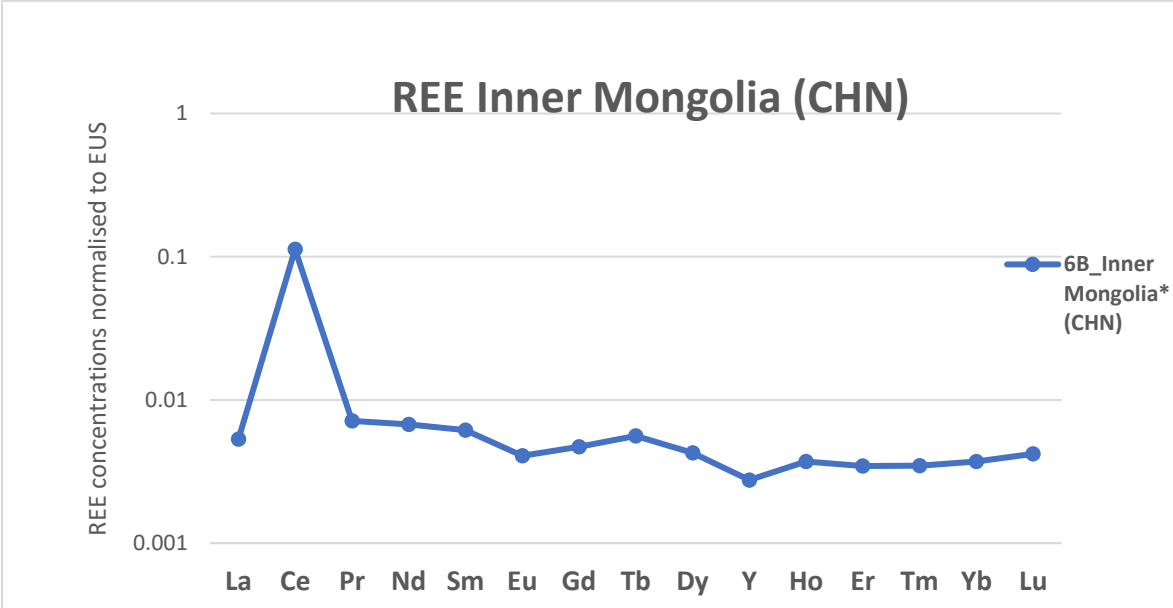


Figure 76: REE pattern of sample 6B from Inner Mongolia province (CHN)

From the Balama mine (MOZ), six samples were analyzed (21K, 21L, 1B, 21B, 21J, 22B), which exhibit relatively homogeneous REE patterns (Fig. 77). All samples, except for sample 21B, show a distinctly negative Ce anomaly. All samples from Balama mine show increasing LREE, as well as decreasing MREE and HREE. Sample 1B exhibits a slightly negative Eu anomaly in comparison to the other samples.

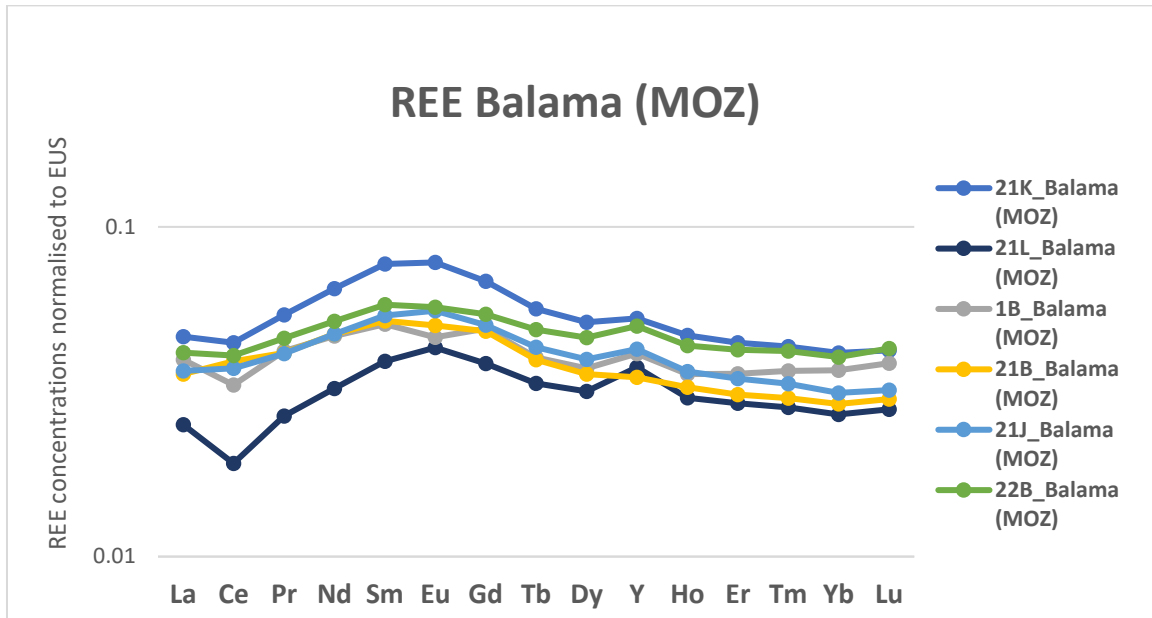


Figure 77: REE patterns of sample 21K, 21L, 1B, 21B, 21J, 22B from Balama mine (MOZ)

All three samples from the Brickaville deposit (MAD) exhibit a strongly positive Ce anomaly. The MREE and HREE are enriched in sample 21D, whereas they are relatively depleted in the other two samples, 11B and 22D (Fig. 78).

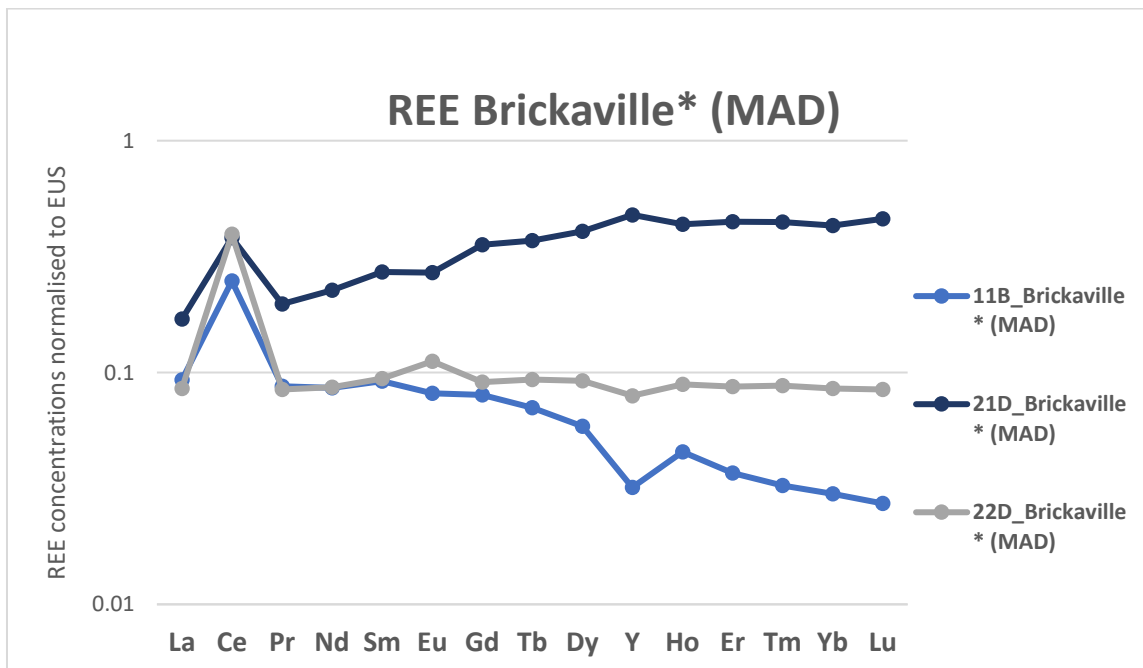


Figure 78: REE patterns of sample 11B, 21D, 22D from Brickaville province (MAD)

For the Skaland mine in Norway, three samples were analyzed (2B, 15B, and 22G), with samples 15B and 22G exhibiting similar REE patterns, displaying a distinct positive Eu anomaly and LREE = MREE = HREE. Sample 2B, in comparison, shows a distinctly positive Ce anomaly increasing LREE, MREE and HREE (Fig. 79).

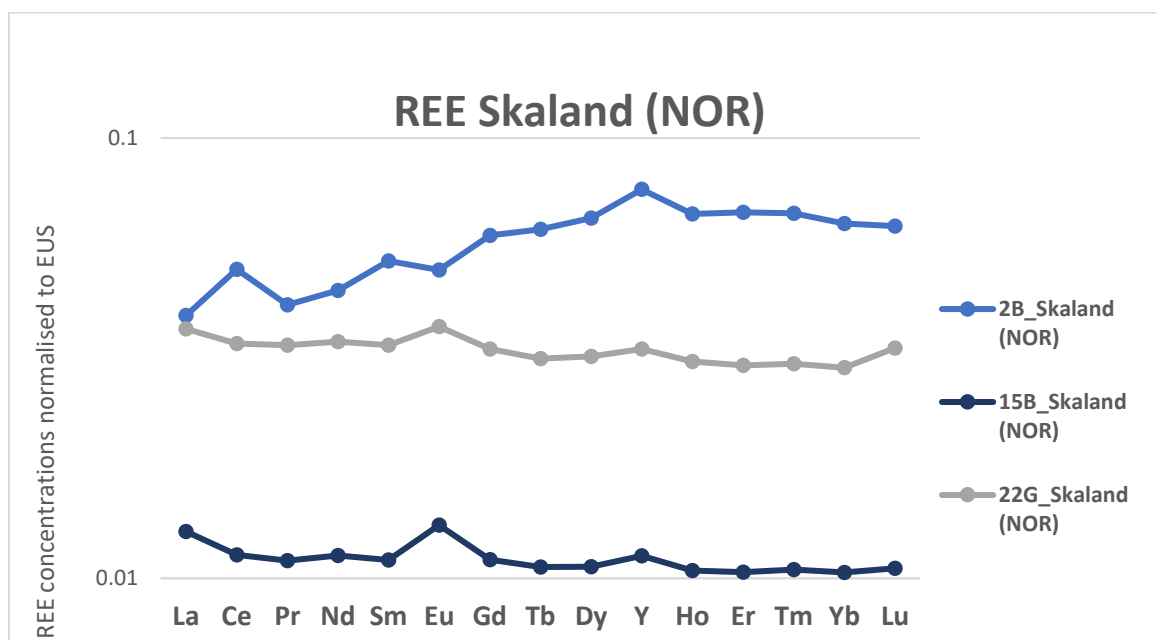


Figure 79: REE patterns of sample 2B, 15B and 22G from Skaland (NOR)

Both available samples from Zavallia mine (UKR) show slightly decreasing LREE, an enrichment in MREE and HREE with sample 22F showing a positive Eu anomaly compared to sample 4B (Fig. 80).

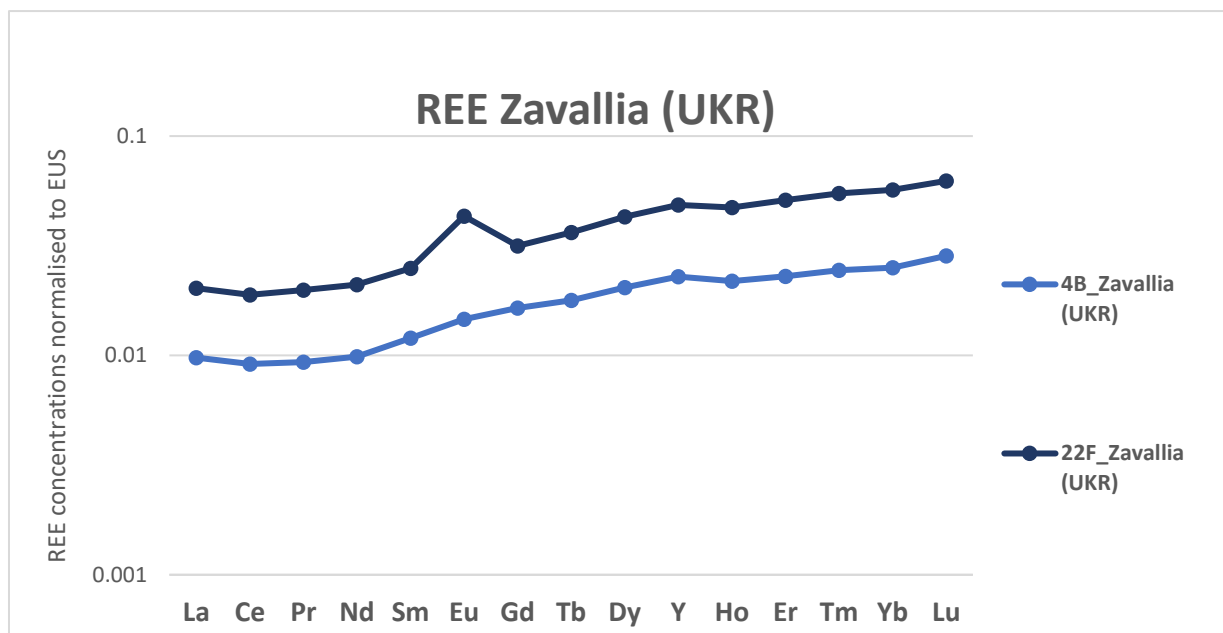


Figure 80: REE patterns of sample 4B and 22F from Zavallia mine (UKR)

Both samples from the Ragedara province (LKA) exhibit a distinct negative Eu anomaly, as well as decreasing HREE (Fig. 81). Sample 21F shows increasing MREE, whereas sample 10 B shows decreasing MREE.

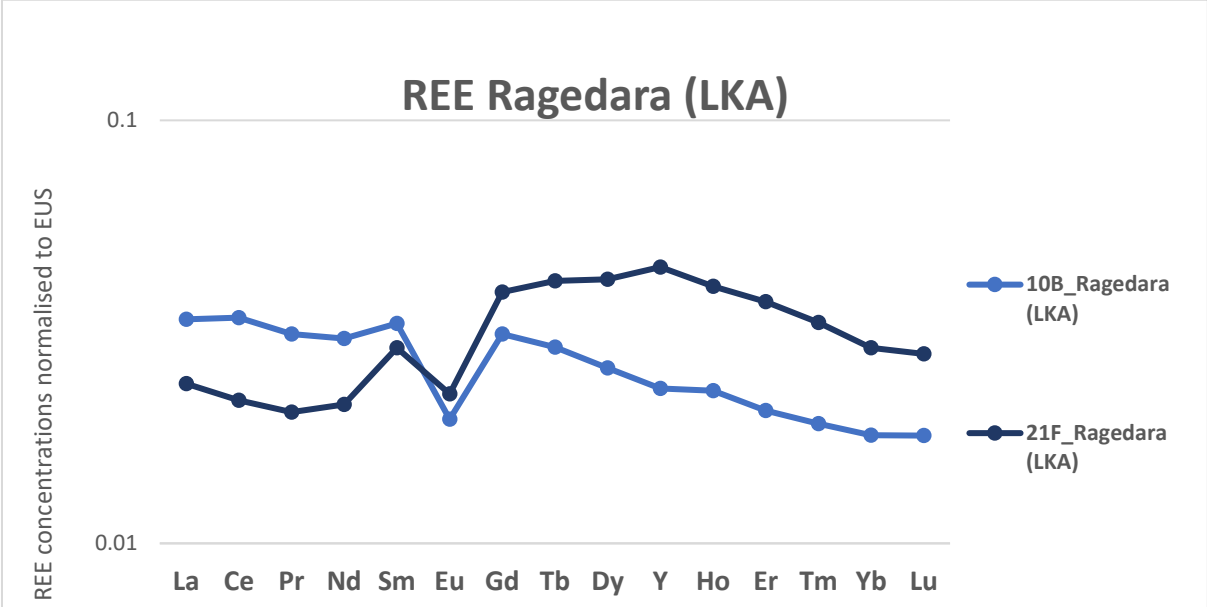


Figure 81: REE patterns of sample 10B and 21F from Zavallia mine (UKR)

Samples 5B and 21E from Tayginka mine (RUS) exhibit a distinct positive Eu anomaly. Samples 21E and 22E both show decreasing MREE and HREE. Sample 22E displays a negative Eu anomaly but otherwise follows a pattern very similar to that of sample 21E with depletion of MREE and HREE (Fig. 82).

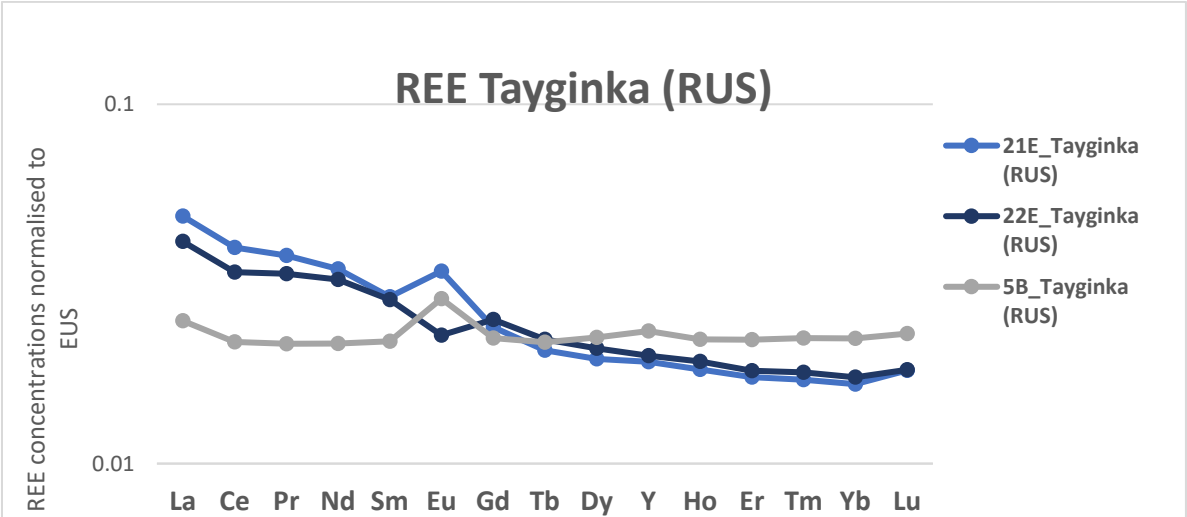


Figure 82: REE patterns of sample 21E, 22E and 5B from Tayginka (RUS)

Comparison of REE patterns from all locations:

Each individual location (mine/province) exhibits differences in the REE composition, even when represented as mean REE values from each location (Fig. 83). Brickaville (MAD), Inner Mongolia (CHN) and Minas Gerais (BRA) show positive Ce anomalies as a common feature. Kropfmühl (GER), Brickaville (MAD), Ragedara (LKA) and Inner Mongolia (CHN) show negative Eu anomalies. However REE patterns differ. Shandong REE patterns were different from sample 21A and 22A and could not be averaged into one pattern to represent the REE data from this province. The following figure (Fig. 84) shows two lines for Shandong, one representing sample 21A, the other sample 22A. Overall, we observe flat shale-normalized REE patterns, some with distinct to small Ce and Eu anomalies. In some samples, LREE prevail over HREE, but opposite patterns also occur (e.g., Zavallia).

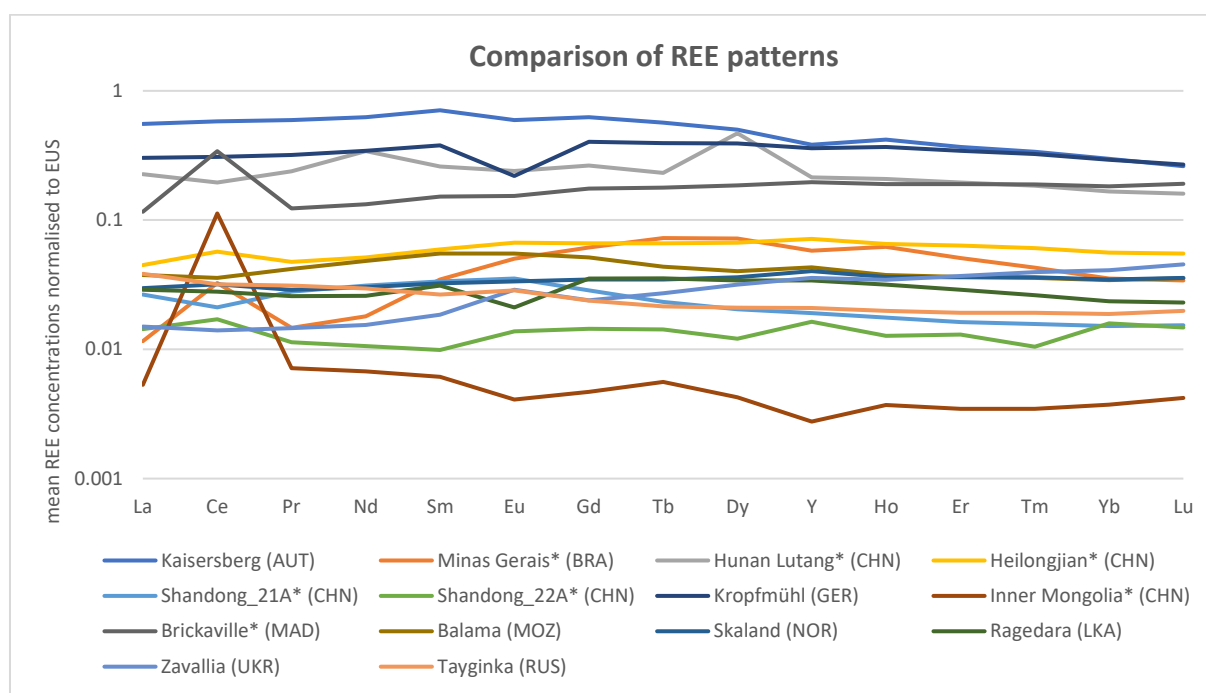


Figure 83: Comparison of mean REE patterns across individual locations

6.4.2.4. Multivariate Statistical Analysis and Key Elements

6.4.2.4.1. Linear Discriminant Analysis (LDA)

Two models using LDA were calculated using the solution-based-ICP-MS trace element data. The data shows significantly less dispersion within each group, as the measurements of the digestions vary little, producing almost identical values. The models are based on continents and on mines/provinces and allow classification of the data. The input data consist of the trace element data as well as the categories on which the classification is based.

The first LDA (Fig. 84) demonstrates the discrimination of graphite samples based on continents. Through the multivariate analysis of trace element data obtained from solution-ICP-MS, four continents can be clearly distinguished from each other. The mines/provinces from Asia and Europe are closer to each other, but there is minimal overlap in individual data points. Confidence ellipses display 95% confidence for the mean value of each group in the diagram assuming a normal distribution of the data. To identify **key elements** for differentiation, the scoring coefficients of this LDA were extracted (Table 46). The elements **Mg, Al, V, Fe, Ga, Sr, Ag, Cd, and U** contribute most to the classification of individual graphite samples (based on continents).

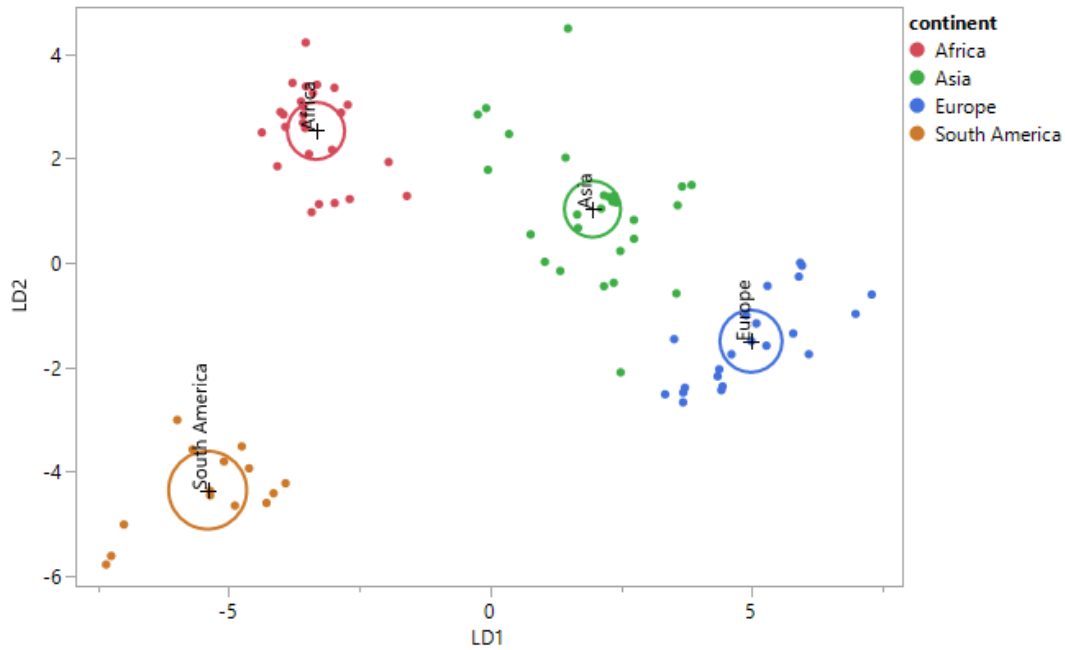


Figure 84: LDA of solution-based-ICP-MS trace element data based on continents

Table 46: Scoring coefficients from LDA based on continents

	Log[Mg]	Log[Al]	Log[V]	Log[Cr]	Log[Mn]	Log[Fe]	Log[Ni]	Log[Cu]	Log[Zn]	Log[Ga]	Log[Se]	Log[Rb]	Log[Sr]	Log[Mo]	Log[Ag]	Log[Cd]	Log[Ba]	Log[Tl]	Log[Pb]	Log[U]
LD1	1.84	1.10	0.32	-0.19	-0.42	1.73	0.76	-0.04	-0.61	-3.22	-0.02	0.07	1.34	-0.79	1.01	-0.45	-0.18	-0.63	-0.06	-0.38
LD2	-1.04	-1.16	1.05	-1.47	-1.32	0.39	0.48	0.16	0.20	1.42	-1.82	0.97	0.88	-0.05	-0.69	1.13	-1.00	-0.69	0.09	0.83
LD3	-0.45	-0.30	-1.50	-0.36	0.48	0.16	1.05	-1.06	0.55	1.23	0.90	0.95	-1.15	0.47	1.06	-1.32	0.51	0.06	-1.24	0.72

A second LDA, based on mines/provinces is displayed in Fig. 85. All occurrences can be differentiated from each other. Balama mine and Shandong province show a similar pattern in close proximity to each other. Ragedara mine is situated at the greatest distance from the other occurrences and differs most significantly, likely due to its hydrothermal graphite type and the non-organic origin of carbon in this mine.

Confidence ellipses display 95 % confidence for the mean value of each group in the diagram and assume a normal distribution of the data.

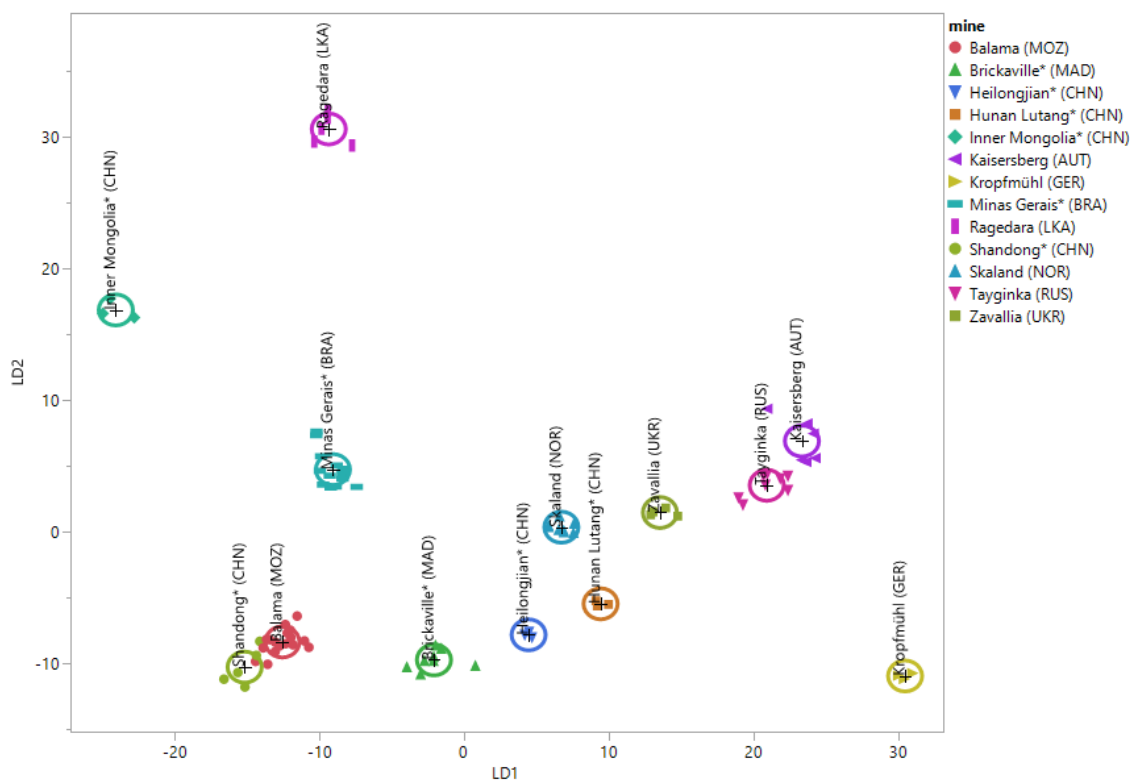


Figure 85: LDA of solution-based-ICP-MS trace element data based on mines/provinces

To identify **key elements** for differentiation, the scoring coefficients of this LDA were extracted and are displayed in Table 47. The elements **Mg, Al, Cr, Mn, Ni, Cu, Ga, Rb, Ag and U** contribute most for the classification of individual graphite samples based on individual mines/provinces.

Table 47: Scoring coefficients from LDA based on mines/provinces

	Log[Mg]	Log[Al]	Log[V]	Log[Cr]	Log[Mn]	Log[Fe]	Log[Ni]	Log[Cu]	Log[Zn]	Log[Ga]	Log[Se]	Log[Rb]	Log[Sr]	Log[Mo]	Log[Ag]	Log[Cd]	Log[Ba]	Log[Tl]	Log[Pb]	Log[U]
LD1	2.04	1.55	-1.08	0.95	2.42	-0.69	-0.56	1.86	0.26	0.95	-0.05	0.77	-0.74	0.03	2.36	-1.11	-0.43	-0.55	-1.23	1.00
LD2	1.38	1.24	-3.28	1.49	2.93	-1.19	1.48	-0.48	-0.31	-0.32	-0.52	-1.67	0.31	0.27	0.25	-0.46	0.60	-2.28	1.24	0.61
LD3	-0.82	-0.31	-1.62	2.00	-0.55	-0.59	2.00	-1.69	0.27	1.20	-0.36	1.08	-0.13	0.23	-1.33	-0.01	-0.40	1.07	0.72	-0.14
LD4	0.22	1.34	-1.13	-0.73	0.27	-1.74	-0.44	0.29	1.08	-0.51	1.29	-0.75	1.45	-0.98	-0.26	-0.87	1.08	0.21	-0.12	0.98
LD5	-0.45	0.24	0.65	-1.84	-0.70	0.45	-0.92	1.11	-0.11	0.67	-0.21	-0.13	-0.08	0.03	-1.71	0.85	-0.88	-0.10	0.90	0.64
LD6	0.04	0.25	0.48	-1.91	-0.85	1.31	0.81	-0.10	-0.11	-1.24	-0.08	0.02	0.99	-0.49	0.14	-0.16	0.05	-0.92	0.10	0.12
LD7	0.59	0.19	-0.43	1.10	-0.47	-0.69	-0.56	0.73	0.10	-0.67	0.32	-0.34	0.64	0.26	-0.71	0.36	-1.00	1.14	-0.59	0.98
LD8	-0.53	-0.41	-0.62	0.09	0.38	-1.07	0.44	0.08	0.19	1.58	-1.12	0.22	-0.01	0.30	0.24	0.42	-0.17	-0.64	-0.27	0.83
LD9	0.72	-0.17	1.35	0.53	-0.93	0.53	-1.40	0.72	-0.07	-0.57	-0.85	-0.64	0.91	-1.10	-0.83	1.51	-0.34	0.38	0.81	-0.83
LD10	0.28	-0.79	-0.29	0.21	0.10	-0.07	0.12	-0.66	1.17	0.98	0.90	-0.19	-0.68	0.73	0.05	-0.51	0.02	0.59	-0.23	-0.46
LD11	-0.71	-0.22	-0.26	0.28	0.57	-0.40	-0.07	-0.12	-0.15	0.29	0.31	0.64	0.24	0.45	0.16	0.33	-0.46	0.48	-0.22	-0.77
LD12	0.27	-0.20	0.18	0.43	-0.31	-0.67	-0.65	-0.20	0.55	0.07	0.27	-0.10	-0.46	0.49	-0.41	-0.14	0.23	0.22	0.76	-0.37

6.4.2.4.2. Principal Component Analysis (PCA)

Based on the elements that have been identified as significant in the LDA shown in chapter 6.4.2.4.1., a Principal Component Analysis (PCA) was performed, which refers to the same hierarchies as above (continents and mines/province). PCA is a method of dimension-reduced data projection, where data are summarized into new, simplified variables and expressed as PC1 and PC2.

PC1 (48.3 %) and PC2 (15.6 %) for the hierarchy of continents account for most of the variance in the data and are presented in a two-dimensional graph with corresponding elements displayed in Fig. 86. Data points corresponding to South America (orange) are densely distributed, datapoints referring to Asia (green) show two sets of samples. A clear distinction of the data points based on the continents is therefore not entirely possible.

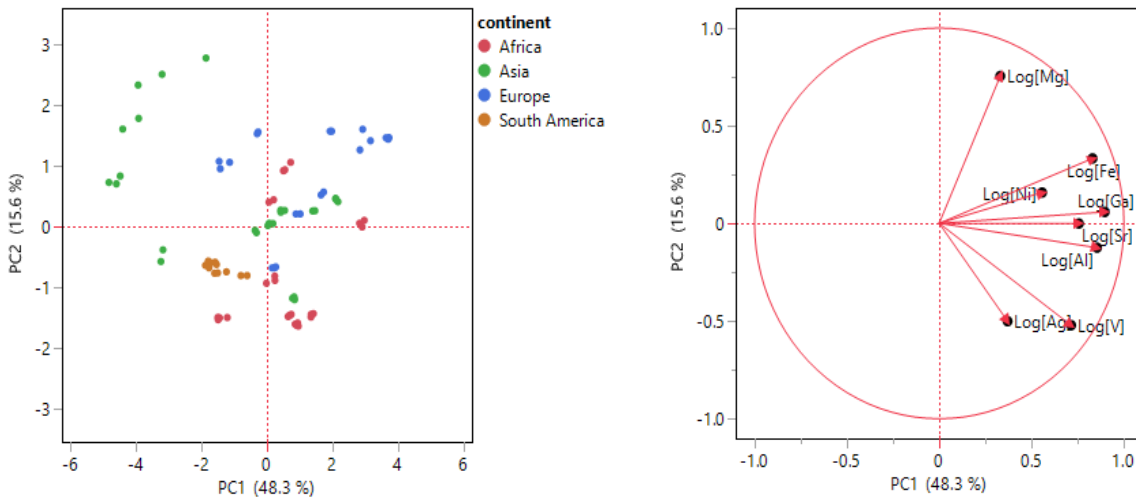


Figure 86: PCA of solution-ICP-MS trace element data at continent level (left) with the indicated elements (right)

PC1 (41.2 %) and PC2 (21.7 %) for the hierarchy of mines/provinces account for most of the variance in the data and are presented in a two-dimensional graph with corresponding elements displayed in Fig. 87. Data points referring to Ragedara (LKA) form a distinct group, as well as datapoints for Kaisersberg (AUT), Minas Gerais (BRA) and Inner Mongolia (CHN). Shandong (CHN) shows two sets of datapoints, which correlates to the REE pattern of this province, where two different patterns for the two samples from the same province were identified (see 6.4.2.3.)

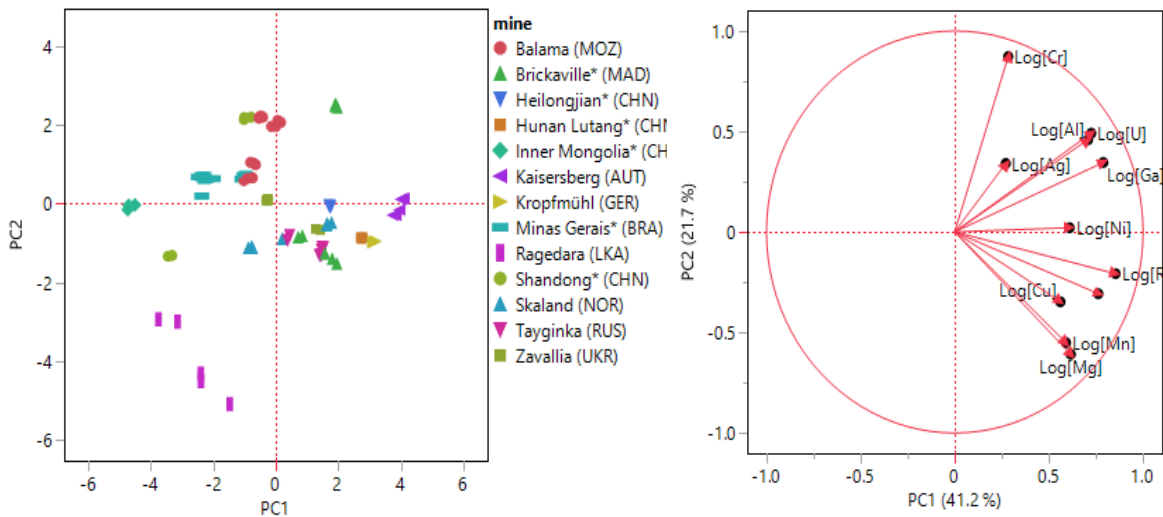


Figure 87: PCA of solution-ICP-MS trace element data at mine/province level (left) with the indicated elements (right)

6.5. Multi-Parameter Approach

The data generated in this study consist of various types, including compositional trace element data, stable carbon isotope ratios and dimensionless Raman data. For the multi-parameter approach, all data needed to be merged. Since a different number of measurements per sample was available for each method, standardization was necessary. Therefore, a 1:1:1 data fusion strategy was adopted, resulting in one value per sample for each parameter. The carbon isotope mean values, based on at least three measurements per sample, were used and the 10-20 Raman spectra, from which relevant information was extracted, as Raman parameters, were also averaged. Median values from the trace element data were used for each analytical method. This data fusion strategy was utilized for multivariate data analysis, which now includes multiple parameters. In the previous chapters, multidimensional data analyses only considered trace element or Raman data separately. This is an attempt to combine all parameters in a data analysis, since a single parameter, when used individually, could not achieve complete differentiation between the graphite deposits.

Linear discriminant model 1 (Fig. 88) uses the combination of trace element data obtained from solution-based-ICP-MS data with Raman parameters and $\delta^{13}\text{C}$ data. All analyzed trace elements have been selected for this purpose, as well as the Raman parameters related to the G-band (G_STA, G-shape-factor, Gmax, Dmax/Gmax ratio) and the stable carbon isotope ratios. The combination of solution-based-ICP-MS trace element data, selected Raman parameters and $\delta^{13}\text{C}$ clearly discriminates locations. Moreover, none of the locations overlap with each other. Balama and Heilongjian are in close proximity to each other and with a greater dispersion of the data, there might be an overlap between these two locations. The results of the trace element analysis using solution-based-ICP-MS show a greater difference in the elemental content between different locations (see grouping 6.4.2.1) and therefore also a larger difference between the individual deposits. Including all three parameters, the individual graphite deposits are clearly distinguishable.

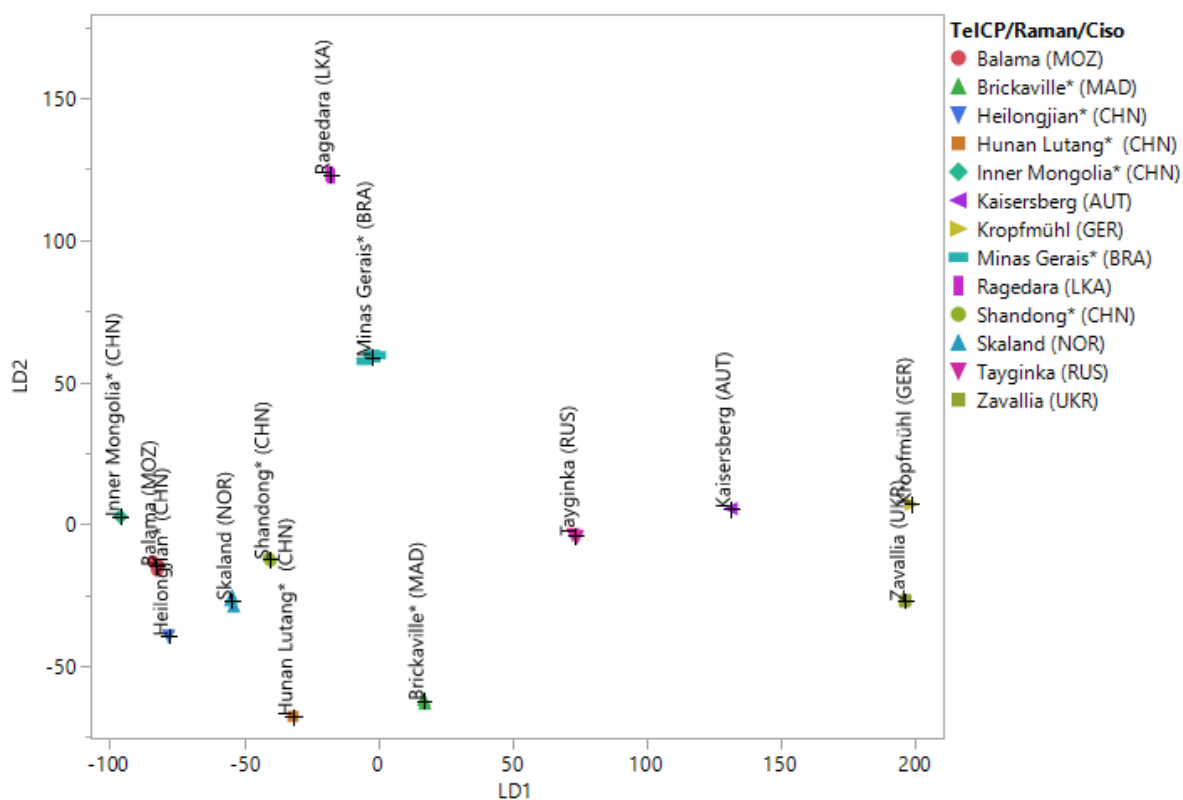


Figure 88: LDA of trace element data from solution-based-ICP-MS analysis, Raman parameters and $\delta^{13}\text{C}$

Model 2 (Figure 89) uses trace element data obtained from LA-ICP-MS analysis combined with Raman parameters and $\delta^{13}\text{C}$ data. The combination of trace element data from LA-ICP-MS analysis with Raman and $\delta^{13}\text{C}$ does not allow a clear discrimination of all locations. Ragedara and Minas Gerais are in a close proximity to each other, as well as Brickaville, Inner Mongolia and Skaland, but also Kropfmühl, Tayginka, Balama and Shandong.

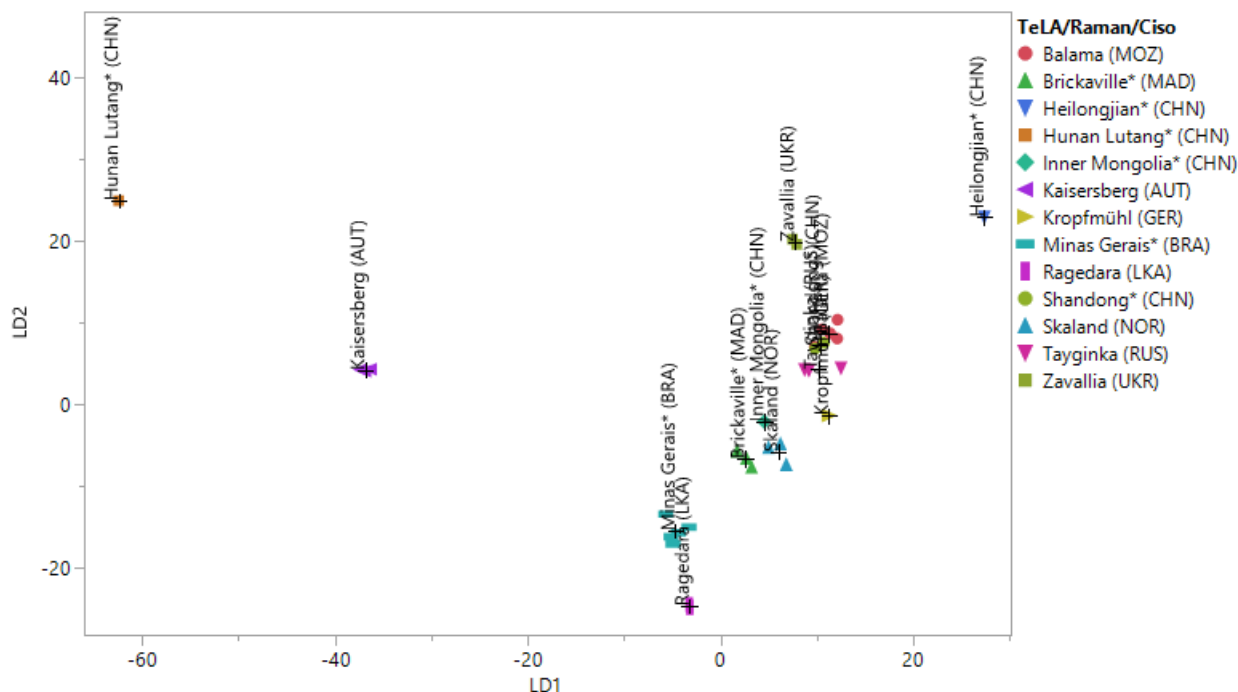


Figure 89: LDA of trace element data from LA - ICP-MS analysis, Raman parameters and $\delta^{13}\text{C}$

7. Discussion

7.1. Stable Carbon Isotope Ratios ($\delta^{13}\text{C}$)

The $\delta^{13}\text{C}$ values were divided into three major groups (related to the geographic origin): Ukraine, Sri Lanka and RoW (all occurrences except Ukraine and Sri Lanka). The $\delta^{13}\text{C}$ values within the RoW group overlap. The significant difference in the heavy $\delta^{13}\text{C}$ values from Sri Lanka is attributed to the inorganic origin of carbon. Hydrothermal graphite from Sri Lanka typically exhibits a significantly heavier signature (Luque et al. 2014a), than graphite deposits in other countries. Carbon isotope signatures of hydrothermal graphite from Sri Lanka are documented in literature and displayed in Fig. 90.

Reported $\delta^{13}\text{C}$ values for Bogala mine reach from -7.76 ‰ to -10 ‰ (Touzain 2010, Weis et al, 1981, Dobner, 1978), for Digana mine between -7.6 ‰ and -8.5 ‰ (Binu-Lal et al. 2003; Touzain et al. (2010)) and $\delta^{13}\text{C}$ values for Kahatagaha–Kolongaha mine ranging from -5.61 ‰ to -9.29 ‰ (Weis et al. (1981); Touzain et al. (2010)).

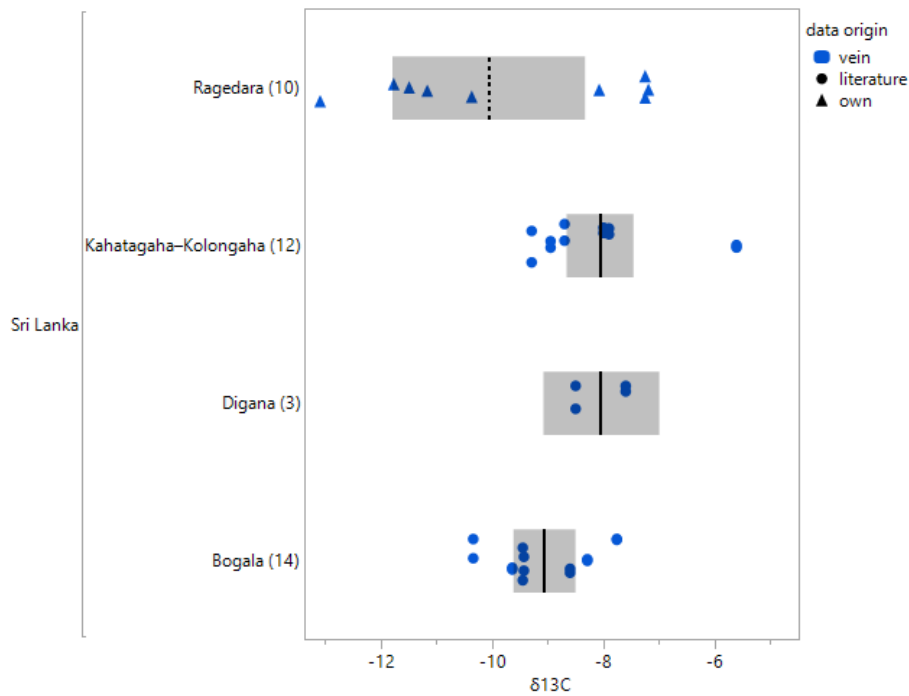


Figure 90: Carbon isotope values from Sri Lankan graphite deposits, numbers in brackets represent number of available data; vertical line in box represents mean value

Hydrothermal (vein) graphite from Sri Lanka (Ragedara) thus forms a distinct group for discrimination and is clearly distinguishable.

Within the database and worldwide collection of $\delta^{13}\text{C}$ values, more data could be referred to as hydrothermal (vein) graphite. Fig. 91 demonstrates $\delta^{13}\text{C}$ values from various hydrothermal graphite types worldwide which all fall within the range of -4.5‰ to -17.5‰ . (references for $\delta^{13}\text{C}$ values can be found in the worldwide database table in the appendix)

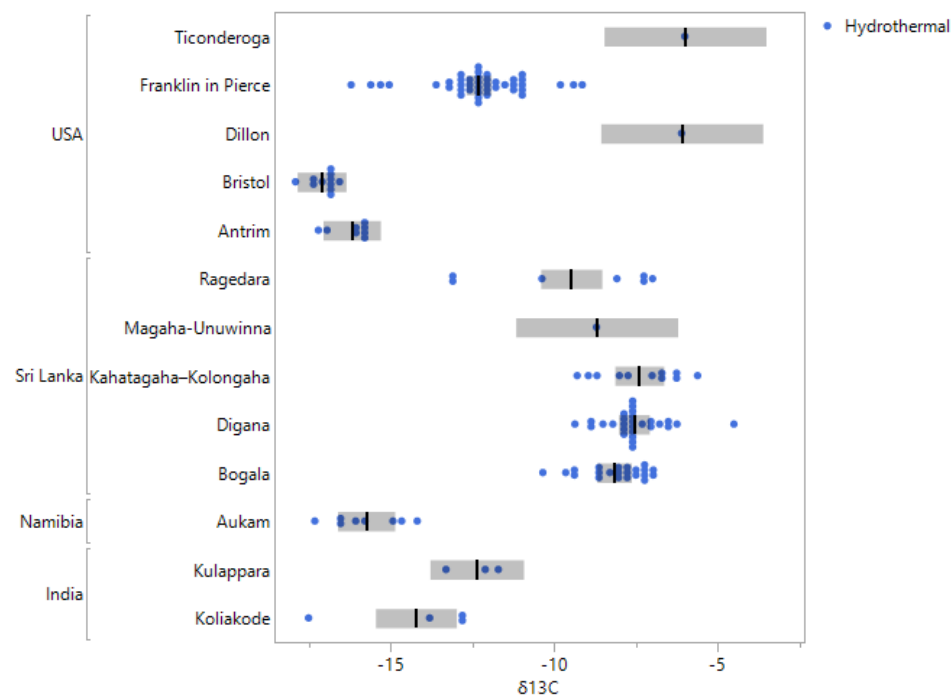


Figure 91: Stable Carbon isotope values from worldwide hydrothermal graphite deposits (references see worldwide database table in appendix); vertical line in box represents mean value

Another group that can be clearly discriminated using $\delta^{13}\text{C}$ values is Ukraine. These values are consistently very light and are well distinguishable from both RoW (rest of the world) and Sri Lanka. Graphite from Ukraine is considered the geologically oldest graphite (Archean) (Mykhailov, 2020). The source material for this graphite was exclusively organic. However, during this geological era, only very primitive organisms (Prokarya) with a simple metabolism prevailed. These organisms tend to absorb the more abundant ^{12}C , explaining the light signature of this graphite material. (O Leary, 1992)

The group of samples from Ukraine differs significantly from the other deposits within the own sample collection. Literature $\delta^{13}\text{C}$ values for Zavallia, Ukraine range from -35,08 ‰ to -30,50‰ (Lyzhachenko, 2020).

Within the RoW group, more detailed distinctions could not be made. Available $\delta^{13}\text{C}$ values from literature confirm the data obtained in this study. All graphite samples from the RoW group are considered as organic material (both amorphous and flake). Within the RoW group, a trend can be observed that can be attributed to different source materials of the graphite (coal- amorphous) and thus different depositional environments.

Biological isotope fractionation

Organisms discriminate ^{12}C and ^{13}C uptake during photosynthetic processes, which reflect metabolic plant reactions, diffusion processes or other biochemical mechanisms and environment (O. Leary, 1992). More developed plants, such as higher land plants, e.g. leaves, roots and plant fragments can take up more ^{13}C (because they have a more advanced structure and metabolism) and are therefore more depleted in ^{12}C . This influences the ratio towards heavier carbon isotope ratios (e.g. amorphous graphite from Kaisersberg originating from coal seams). More primitive organisms, such as prokaryotes and algae do not take up the heavier and less abundant ^{13}C , but incorporate a higher amount of the more abundant ^{12}C . Thus, they are enriched in ^{12}C ($\delta^{13}\text{C}$ towards -35 ‰). This process is also known as biological isotope fractionation, occurring primarily during the photosynthetic process (O Leary, 1992). Vein graphite is clearly distinguishable due to the inorganic source of the material (Fig. 92).

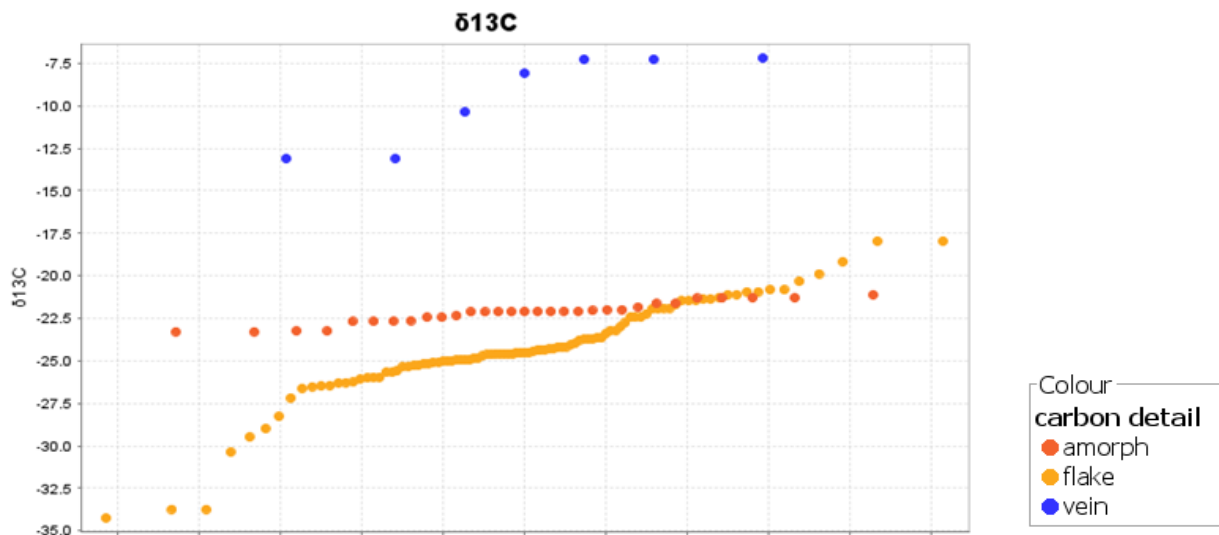


Figure 92: Probability diagram of carbon isotope ratios divided into three groups based on the information about the graphite type (flake, vein, amorphous)

Depositional environment and geological ages

More advanced life forms occur in a terrestrial environment, while more primitive life forms such as prokaryotes or algae are prevalent in aquatic environments. Where the signatures begin to overlap (at

approximately -22 ‰), a mixed environment can be assumed (e.g., river or limnic). The downward trend is an indicator of a marine deposition environment for the organic source material, while towards higher signatures, especially in amorphous graphite, a terrestrial depositional environment can be inferred. Carbon in non-organic graphite (vein graphite) exhibits a significantly heavier signature, clearly indicating this non-organic carbon and allowing for distinct separation.

The rapid evolution of organisms observed during the Proterozoic resulted in the initial deposition of carbonaceous material in sediments. Most significant carbon-containing material could form within this geological context (Parnell, 2021, Weber 2023). Most samples in this study could be attributed to the Proterozoic. The Great Oxidation Event led to an increase in the concentration of oxygen in the atmosphere and water at the Archean-Proterozoic boundary (Blaustein 2016), which explains the deposition of carbonaceous material in the Proterozoic era and thus assigns most graphite deposits to the Proterozoic.

Fig. 93 shows $\delta^{13}\text{C}$ signatures divided into geological ages. The group in the top right corner indicates the inorganic origin of carbon in the samples from Sri Lanka, which are, regardless their geological age, distinguishable. Values in the lower range of the diagram (-28 to -32 ‰) are labelled as Archean, thus representing the oldest graphites in this study. The rest of the data points are attributed to Paleozoic and Proterozoic ages. The Paleozoic graphites in this study are from Kaisersberg. The point in the diagram where Paleozoic values overlap with Proterozoic values can be seen as a change in depositional environments (e.g., marine - limnic - terrestrial) towards heavier values or as a result of the Great Oxidation Event.

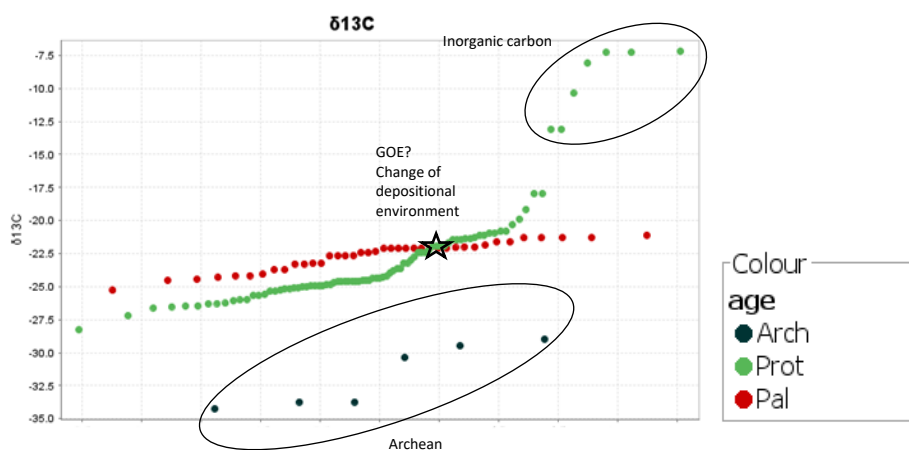


Figure 93: Probability diagram of carbon isotope values divided into four groups based on their geological ages (Archean – Paleozoic and Proterozoic) and the carbon source (organic vs. inorganic)

Carbon isotope analysis provides insights into the origin and precursor materials of graphite and consequently, the depositional environment. The parameter only is suitable to a limited extent for identifying the origin of graphite.

The study on graphite samples from Finnish deposits (Aitolampi, Raisjoki, Emas, and Koivuniemi) aimed to demonstrate that carbon isotope values do not undergo significant changes throughout the processing stages from raw ore to concentrates or purified graphite. The signatures do not change along the processing chain. Most of the variability is due to the (spatial) variability within the deposit which may translate into temporal variability over the life of the mine (different areas are mined within the years). The $\delta^{13}\text{C}$ values of the Finnish samples range from -25‰ to -32‰, which could overlap with the

$\delta^{13}\text{C}$ signature of the Zavallia deposit in Ukraine. The Ukrainian graphite is classified as Archean (Weber 2023), while the Finnish occurrences are classified as Paleoproterozoic (Weber 2023).

7.2. Crystallinity and Spectral Data

Raman spectroscopy serves as a standard application for the analysis of graphite, however it can only be used to a very limited extent for the determination of the geographic origin of graphite. A detailed discrimination based on mines/provinces is not feasible. Kaisersberg (Austria) and Hunan Lutang (China) stand out in this regard, both being identified as semi-graphite (amorphous graphite) with a more pronounced D-band and less intense G-band, indicating lower organic maturity of the material and associated lower crystallinity at lower formation temperatures. Other samples exhibit a weaker or almost non-existent D-band, indicating the higher maturity of organic material and the associated higher crystallinity and associated metamorphic temperatures. The crystallinity, especially in flake graphite, is crucial for the specific inherent properties that make this raw material highly demanded, particularly as anode material in the battery production. Hydrothermal graphite from Sri Lanka also shows very weak to no D-band at all and cannot be distinguished from other occurrences (flake graphites) by using Raman spectra only.

The G_STA and G_shape factors had been identified as the most important variables for discrimination between amorphous graphite and the rest (flake and vein).

This method is not suitable to determine the geographic origin of graphite, but serves as a method to characterize the material and provides information about the type of the graphite material.

No temperature data were derived from the Raman data obtained within this study. The subsequent temperature data are literature values (Tab. 48) and serve as a comparison of the temperature conditions of the occurrences among each other.

Table 48: Mines/provinces included in this study with respective formation temperatures

<i>mine/province</i>	<i>Temperatures (°C) (from Rantitsch et al., 2016)</i>
<i>Kaisersberg (AUT)</i>	<i>360° - 410°</i>
<i>Hubei (CHN)</i>	<i>700°</i>
<i>Balama (MOZ)</i>	<i>600°-850°</i>
<i>Minas Gerais (BRA)</i>	<i>> 750°</i>
<i>Taiginka (RUS)</i>	<i>650° - 750°</i>
<i>Inner Mongolia (CHN)</i>	<i>860° - 960°</i>
<i>Heilongjian (CHN)</i>	<i>800°- 850°</i>
<i>Brickaville (MAD)</i>	<i>700° - 880 °</i>
<i>Ragedara (LKA)</i>	<i>700° - 830 °</i>
<i>Zavallia (UKR)</i>	<i>640° - 800°</i>
<i>Kropfmühl (GER)</i>	<i>580° – 650°</i>
<i>Hunan Lutang (CHN)</i>	<i>300° – 450°</i>
<i>Skaland (NOR)</i>	<i>> 900°</i>
<i>Shandong (CHN)</i>	<i>< 500°</i>

7.3. Multi-Element Analysis

Comparison of trace element data obtained from LA-ICP-MS and solution-based-ICP-MS:

Trace element data were analyzed using two different analytical methods, yielding non-uniform results. To provide an overview of the comparison of the entire dataset, a summary table was generated (Table 49). This table includes the median element concentrations for each deposit for elements measured by both analytical methods and the comparison factor, which is calculated by dividing the median solution-

based-ICP-MS value by the median LA-ICP-MS value and then collectively summarized to an average factor of comparison for each element. Fig. 94 shows the minimum, maximum and mean comparison factor for each element. The horizontal line indicates the same element concentrations in both analytical methods (e.g. Ga).

Generally, solution ICP-MS data shows higher values than LA-ICP-MS data, except for the values marked in red (in Tab. 49), which include some values for Ga, As, Rb, Zn, and V. Notably, for Cr, the measurements from the LA-ICP-MS analysis are consistently higher than those from solution-based-ICP-MS analysis.

The largest differences in measured values per analytical method and element are observed for Mg, Al, and Fe. For Mg, the average difference from solution-based-ICP-MS values compared to LA-ICP-MS values is 23 times as high, for Al approximately 12 times higher in solution-based-ICP-MS values and for Fe approximately 20 times higher in solution-based-ICP-MS data.

These observations allow the following conclusions for this study:

- Mg, Al and Fe occur in minerals that can be dissolved easily (e.g. silicates)
- Mg, Al and Fe are not distributed homogeneously within the pressed pellet (present in minerals)
- Cr (and Rb) are not soluble
- Ga and As show limited solubility, pointing to different host minerals

Table 49: Comparison of median trace element data (in $\mu\text{g g}^{-1}$) obtained from solution-based-ICP-MS and LA-ICP-MS analysis

mine/province	Mg	Al	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr
S_Kaisersberg (AUT)	2115.00	22449.80	21.82	21.21	162.51	11232.20	5.97	31.12	37.40	6.66	8.96	18.02	33.64
LA_Kaisersberg (AUT)	43.47	1824.35	23.63	38.19	6.20	118.80	1.09	1.33	4.92	17.72	6.67	26.19	19.88
factor	49	12	0.9	0.6	26	95	5	23	8	0.38	1.34	0.69	2
S_Minas Gerais* (BRA)	22.17	2941.37	9.26	13.34	21.40	1481.35	1.02	13.72	3.64	0.92	0.05	0.10	1.38
LA_Minas Gerais* (BRA)	1.18	355.35	4.57	30.35	7.60	128.26	0.28	4.89	1.46	0.77	0.16	0.03	0.39
factor	19	8	2	0.44	3	12	4	3	3	1.2	0.32	3	4
S_Hunan Lutang* (CHN)	523.93	7885.28	20.73	5.45	94.62	4213.17	5.93	25.54	31.79	3.06	65.06	18.56	24.78
LA_Hunan Lutang* (CHN)	37.47	977.16	16.96	27.79	41.35	496.97	2.66	7.65	15.98	6.35	36.72	13.54	16.41
factor	14	8	1.2	0.20	2	8	2	3	2	0.48	2	1.4	2
S_Inner Mongolia* (CHN)	57.81	52.38	0.54	16.91	4.20	891.07	0.63	0.98	1.57	0.08	0.12	0.16	0.97
LA_Inner Mongolia* (CHN)	2.42	2.76	0.14	30.73	0.75	41.25	0.13	0.37	0.31	0.04	0.17	0.08	0.41
factor	24	19	4	0.6	6	22	5	3	5	2	0.70	2	2
S_Heilongjian* (CHN)	642.97	3686.97	69.12	8.97	37.50	3694.67	5.38	19.04	51.67	1.31	0.49	4.11	10.79
LA_Heilongjian* (CHN)	33.16	271.43	29.23	33.58	13.58	393.92	1.92	6.51	17.63	1.92	0.47	2.03	2.94
factor	19	14	2	0.27	3	9	3	3	3	0.68	1.04	2	4
S_Shandong* (CHN)	17.79	3298.89	70.20	27.80	4.73	5827.27	0.26	23.52	20.39	0.77	0.35	0.24	5.94
LA_Shandong* (CHN)	2.62	223.83	14.18	35.53	1.41	281.27	0.09	4.62	5.12	0.54	0.27	0.13	0.68
factor	7	15	5	0.8	3	21	3	5	4	1.43	1.33	2	9
S_Kropfmühl (GER)	4216.92	4331.01	84.18	24.86	190.89	10822.54	6.72	305.17	19.05	2.12	0.78	21.64	3.95
LA_Kropfmühl (GER)	103.79	399.44	26.11	38.35	34.84	581.87	0.77	13.40	2.13	1.26	0.20	5.33	0.94
factor	41	11	3	0.65	5	19	9	23	9	2	4	4	4
S_Brickaville* (MAD)	123.87	4643.94	54.35	5.21	61.54	4760.54	4.56	42.55	28.20	1.57	3.11	2.61	1.50
LA_Brickaville* (MAD)	3.90	368.47	9.11	26.07	13.77	276.72	0.70	5.76	5.06	0.94	0.49	0.39	0.37
factor	32	13	6	0.20	4	17	7	7	6	2	6	7	4
S_Balama (MOZ)	17.60	4016.79	75.05	18.74	12.05	2639.42	0.48	19.05	29.52	1.00	0.34	0.38	6.26
LA_Balama (MOZ)	1.20	290.36	27.07	36.43	4.18	153.96	0.15	5.48	11.25	1.50	0.24	0.15	1.59
factor	15	14	3	0.51	3	17	3	3	3	0.67	1.44	3	4
S_Skaland (NOR)	775.01	1539.23	11.06	7.07	25.06	3113.59	2.94	21.03	5.27	0.71	0.12	4.94	3.83
LA_Skaland (NOR)	36.91	174.92	6.96	32.82	13.95	233.61	0.62	4.82	2.22	0.92	0.12	1.81	1.76
factor	21	9	2	0.22	2	13	5	4	2	0.77	0.98	3	2
S_Tayginka (RUS)	729.98	2272.15	15.08	5.08	52.23	2735.34	1.02	229.25	25.97	0.95	0.19	3.04	3.41
LA_Tayginka (RUS)	18.92	204.57	3.14	25.75	12.27	150.23	0.27	6.80	2.63	0.64	0.16	0.68	0.86
factor	39	11	5	0.20	4	18	4	34	10	1.49	1.17	5	4
S_Ragedara (LKA)	161.99	158.05	0.12	0.36	57.29	1909.18	2.84	186.65	3.81	0.24	0.07	0.08	1.64
LA_Ragedara (LKA)	9.13	43.06	1.50	43.98	9.92	359.95	2.73	29.82	13.55	1.51	1.12	0.09	1.34
factor	18	4	0.08	0.01	6	5	1.0	6	0.28	0.16	0.06	0.89	1.2
S_Zavallia (UKR)	284.29	3950.09	16.17	8.09	30.29	3179.14	0.63	15.22	18.37	1.20	1.02	1.25	2.42
LA_Zavallia (UKR)	74.86	297.77	8.41	29.33	12.30	473.67	0.47	5.81	14.61	1.30	0.84	1.08	1.54
factor	4	13	2	0.28	2	7	1.3	3	1.26	0.93	1.23	1.16	2
mean factor	23	12	3	0.37	5	20	4	9	4	1.03	2	3	3

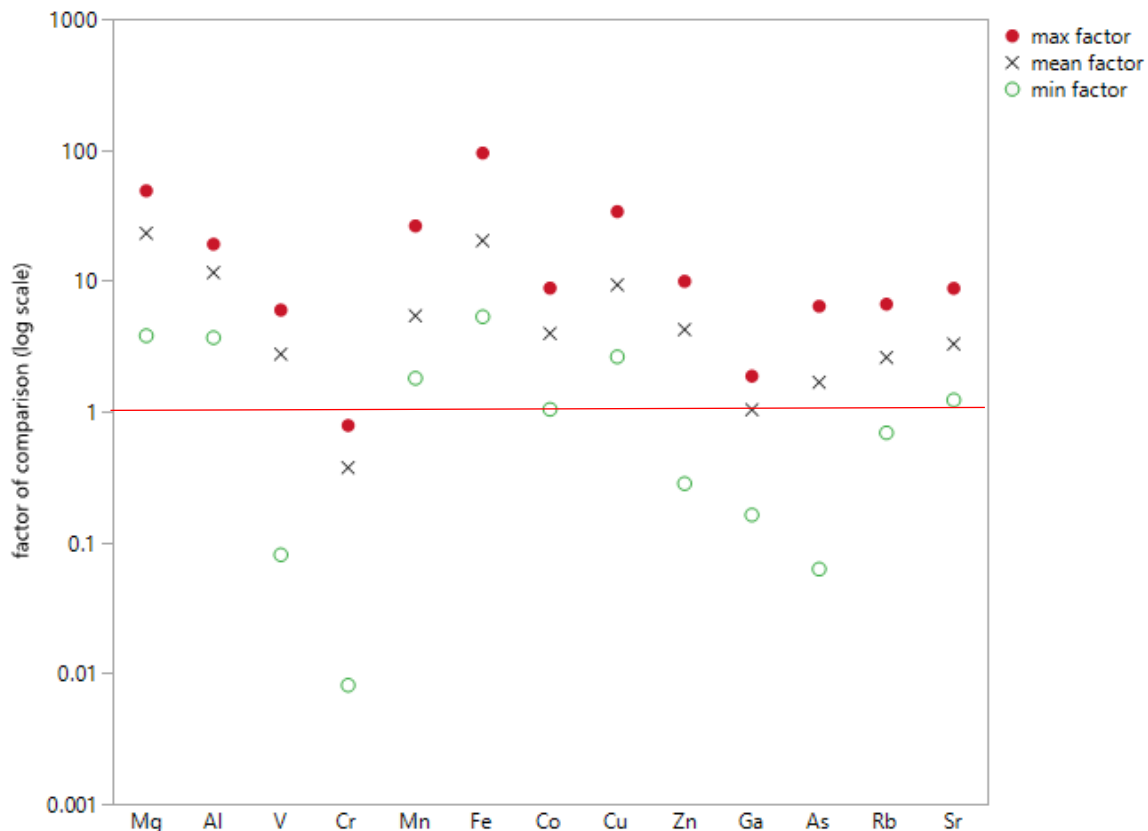


Figure 94: Factor of comparison (log-scale) for solution-based-ICP-MS data and LA-ICP-MS data indicating minimum, maximum and mean comparison factors (data from Tab. 49)

The primary difference between the methods are the sample preparation process and the sample condition during analysis. In the solution-based ICP-MS methodology, the digestions were measured, thus the measured fluid was homogeneous, with all dissolved substances present in the solution. This method also offers significantly lower detection limits. Calibration processes especially in the lower range, were feasible and the calibration in general is a standard method. With the availability of various calibration solutions this is a routine application. However, the sample preparation is considerably more complex, consuming more time and resources (personnel, consumables, acids, other). Another factor that must be considered is the current inability to precisely determine what exactly can be leached from the sample material and which elements/minerals may remain as a residue. A significant advantage of this methodology is the high reproducibility of the data. Measurements from each digestion (three digestions were made per sample) show minimal variation and high reproducibility.

The LA-ICP-MS methodology for trace element analysis in graphite proves to be a more time-efficient method, requiring the samples to be pressed into a stable form only. However, sample homogeneity plays a crucial role. Along the measured lines, impurity particles might be measured, likely contributing to the outliers in the data collection. The limits of detection are significantly higher than those in solution-based-ICP-MS analysis. Calibration, especially in the trace element range, is challenging due to the lack of suitable reference and calibration materials. In-house standard materials (provided by GTK) had to be used which were not certified and definitely need more testing and development of the production process of the material. The data are not reproducible to the same extent as the trace element data from ICP-MS analysis, partly because different areas in the pressed sample are analyzed and each area/line shows varying homogeneity. The graphite concentrates currently cannot be considered as routine material and should not be treated with the same ablation parameters as other minerals, likely due to the thermal conductivity of the material, the reaction (absorption and reflection) of the material

to the laser energy and the softness of the graphite material. Overall advantages and disadvantages of each method are summarized in Table 50.

Table 50: Comparison of solution-based-ICP-MS and LA-ICP-MS method

Solution-based-ICP-MS		LA-ICP-MS	
Pro	Contra	Pro	Contra
Availability of reference and calibration materials	Lack of knowledge what is being dissolved	Time efficient and easy sample preparation, reduced risk of contamination	No suitable reference and calibration materials available
High sensitivity, low LOD/LOQ	Time-consuming digestion process	Rapid analysis, high sample throughput, if method is developed and applicable	Not widely available
Analytical method is a standard application	Potential contamination (through chemicals or other consumables)	In-situ analysis, minimally destructive	Only the surface of the pellet is ablated, the centre of the pellet is not analysed
High precision and accuracy of results	Complex sample preparation	High spatial resolution and detailed mapping possible	Inhomogeneous sample material
Automated sample introduction	Memory effects	Own reference and calibration materials in production	Low precision of results

The analyzed elements for the solution-based-ICP-MS analysis are collectively summarized in Fig. 95 and the analyzed element concentrations from the LA-ICP-MS analysis are collectively presented in Fig. 96. The median element concentrations for each element from each location were summarized to a median graphite element content across all locations for solution-based-ICP-MS and LA-ICP-MS values (Tab. 51). The median element concentrations for each location were then normalized to that mean element concentration listed in Tab. 51.

The two diagrams do not exhibit the same element patterns. However, common features between the two figures include generally higher element concentrations in the Kaisersberg, Hunan Lutang, and Kropfmühl provinces. Ragedara shows low V values in the measurements of both methods. Inner Mongolia generally exhibits very low element concentrations in the data from both methods. Ragedara shows high Cu values in both methods.

Table 51: Average solution-based ICP-MS trace element concentrations (left) and av. LA-ICP-MS trace element concentrations (right) used for normalization of the data for Fig. 95 and Fig. 96

Average solution-based-ICP-MS element concentrations ($\mu\text{g g}^{-1}$)		Average LA-ICP-MS element concentrations ($\mu\text{g g}^{-1}$)	
Mg	745.33	Mg	28.39
Al	4709.69	Al	417.96
V	34.44	Ca	23.10
Cr	12.54	V	13.15
Mn	58.02	Cr	32.99
Fe	4346.11	Mn	13.24
Co	2.95	Fe	283.88
Ni	10.49	Co	0.91
Cu	71.76	Cu	7.48
Zn	21.28	Zn	7.45
Ga	1.58	Ga	2.72
As	6.21	As	3.66
Se	1.73	Rb	3.96
Rb	5.78	Sr	3.78
Sr	7.73	Y	0.98
Mo	19.43	Zr	3.58
Ag	0.27	Ce	1.68
Cd	0.22	Th	0.30
Ba	16.46	U	0.28
Tl	0.08		
Pb	7.30		
Ce	12.42		
Th	1.97		
U	0.91		

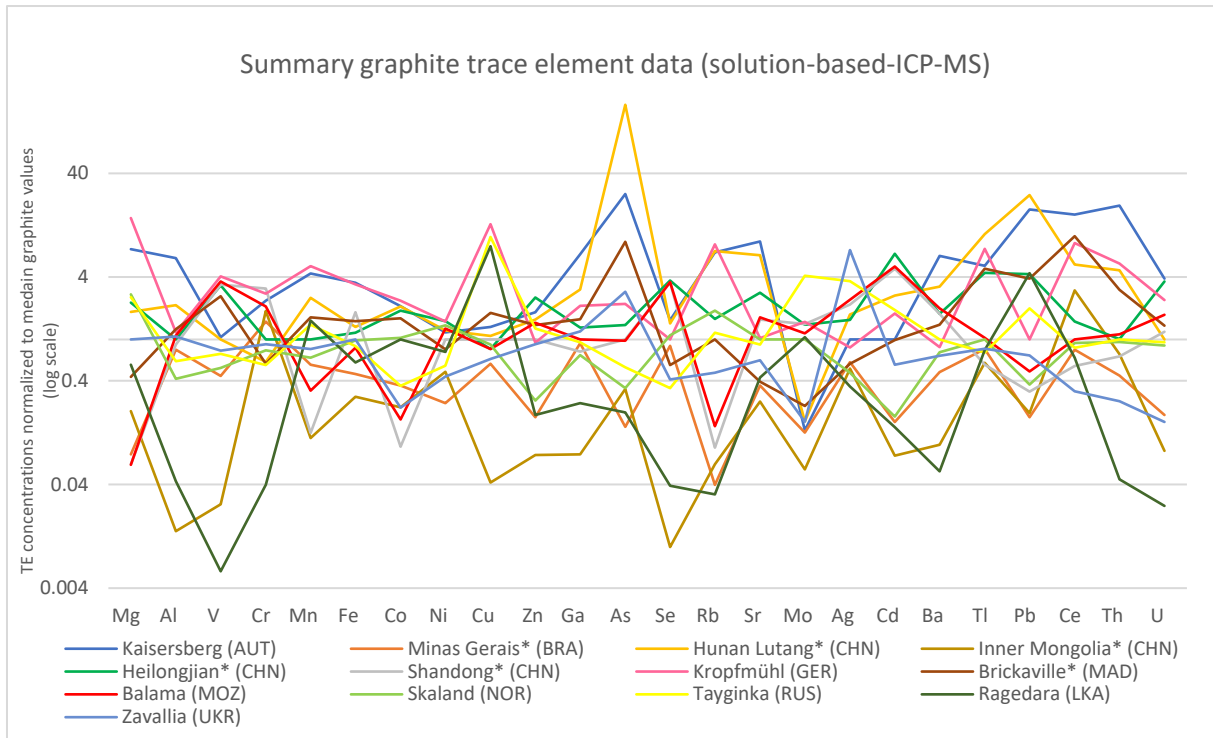


Figure 95: Summary of graphite trace element data obtained from solution-based-ICP-MS analysis

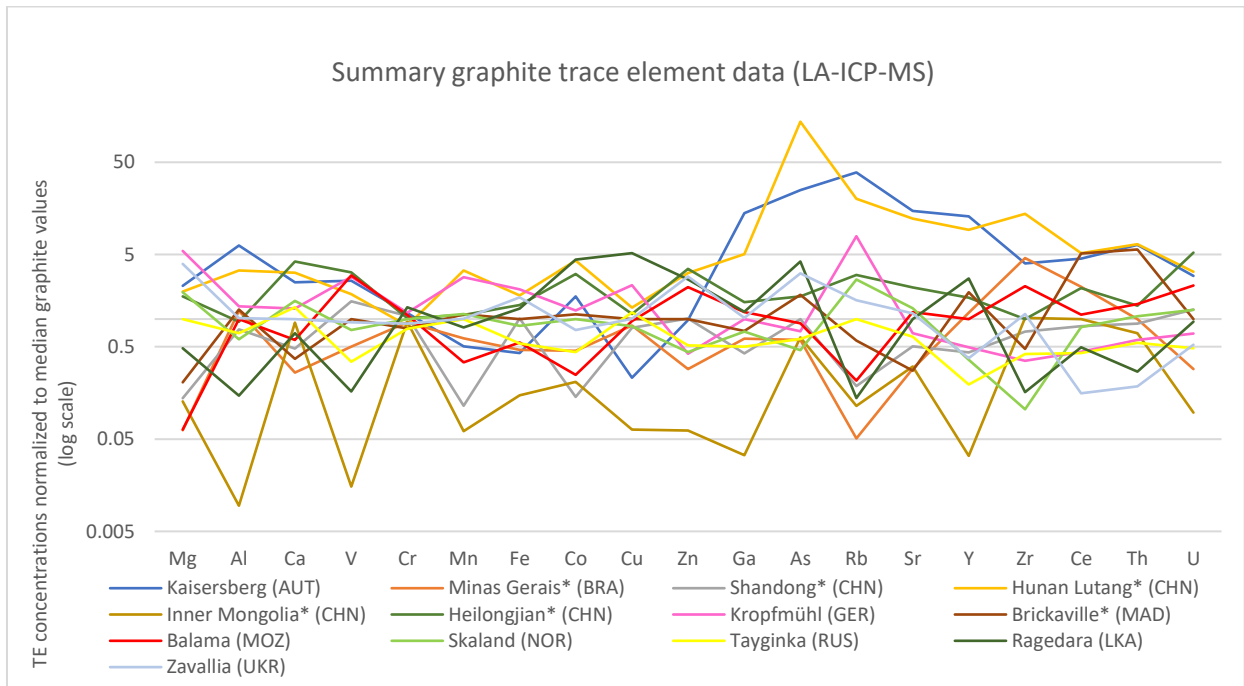


Figure 96: Summary of graphite trace element data obtained from LA-ICP-MS analysis

Comparison of trace elements (based on solution-based-ICP-MS data):

For the description and comparison of trace elements, data from solution-based-ICP-MS analysis was used due to lower dispersion and higher reproducibility of the data.

Magnesium, Al, and Fe are typical elements found in high concentrations and are bound to accessory minerals.

Vanadium: Inner Mongolia and Ragedara differ significantly from the V content of other occurrences, due to their low V content. Vanadium is considered a biophilic element that tends to accumulate in organic source materials (Schroll 1976). The hydrothermal graphite sample from Ragedara (LKA) exhibits the lowest V content ($0.1 \mu\text{g g}^{-1}$). Kropfmühl (GER) ($84 \mu\text{g g}^{-1}$) and Balama (MOZ) ($75 \mu\text{g g}^{-1}$) exhibit the highest values.

Chromium: The Ragedara mine is also distinguishable from the rest of the deposits due to its low Cr content ($0.4 \mu\text{g g}^{-1}$) compared to other locations. Shandong ($27 \mu\text{g g}^{-1}$) and Kropfmühl ($24 \mu\text{g g}^{-1}$) show the highest Cr values. Despite being both amorphous, Kaisersberg and Hunan Lutang exhibit different values of $21 \mu\text{g g}^{-1}$ (Kaisersberg) and only $5 \mu\text{g g}^{-1}$ (Hunan Lutang). According to Schroll (1976), Cr is bound to the lattice of the graphite.

Manganese: Shandong and Inner Mongolia ($4 \mu\text{g g}^{-1}$) have the lowest Mn contents among the locations. Kaisersberg and Hunan Lutang have high Mn contents, which can be attributed to impurities and lower carbon content compared to other samples. However, the sample from Kropfmühl contains even higher Mn contents. Mn might be bound to Mg and therefore associated with carbonates.

Iron: Inner Mongolia ($891 \mu\text{g g}^{-1}$) shows the lowest Fe concentrations, Kaisersberg (1.1 %) and Kropfmühl (1.08 %) the highest concentrations.

Cobalt: Cobalt values reach from very low values in Shandong ($0.26 \mu\text{g g}^{-1}$) to values up to $6.7 \mu\text{g g}^{-1}$ in Kropfmühl.

Nickel: Minas Gerais ($2.6 \mu\text{g g}^{-1}$) and Zavallia ($4.8 \mu\text{g g}^{-1}$) show the lowest Ni content of all graphite deposits in this study. Higher Ni values are associated with amorphous graphite, which generally contain the most impurities, but flake type graphite from Heilongjian ($16.6 \mu\text{g g}^{-1}$) and Kropfmühl ($16.5 \mu\text{g g}^{-1}$) show highest Ni values.

Copper: Inner Mongolia shows very low Cu contents ($1 \mu\text{g g}^{-1}$). Kropfmühl has the highest Cu contents ($305 \mu\text{g g}^{-1}$). The otherwise trace element-poor hydrothermal graphite from Ragedara also has high Cu contents, confirming that ash-free graphites are generally richer in copper (Janda & Schroll 1960).

Zinc: Inner Mongolia has the lowest Zn contents ($1 \mu\text{g g}^{-1}$). Heilongjian has the highest value ($51 \mu\text{g g}^{-1}$), followed by the impure amorphous graphite (Kaisersberg ($37 \mu\text{g g}^{-1}$) and Hunan Lutang ($31 \mu\text{g g}^{-1}$)).

Gallium: Inner Mongolia ($0.08 \mu\text{g g}^{-1}$) and Ragedara ($0.2 \mu\text{g g}^{-1}$) clearly differentiate with the lowest Ga contents among the occurrences. Kaisersberg ($6 \mu\text{g g}^{-1}$) and Hunan Lutang ($3 \mu\text{g g}^{-1}$) show the highest values.

Selenium: The lowest values are found in Inner Mongolia ($0.01 \mu\text{g g}^{-1}$), followed by Ragedara ($0.04 \mu\text{g g}^{-1}$). Balama, Heilongjian and Shandong ($4 \mu\text{g g}^{-1}$) refer to the highest Se values.

Rubidium: Amorphous graphite (Kaisersberg and Hunan Lutang) has the highest values for this element ($18 \mu\text{g g}^{-1}$). Very low values are found in Minas Gerais ($0.1 \mu\text{g g}^{-1}$), Inner Mongolia ($0.16 \mu\text{g g}^{-1}$), Shandong ($0.2 \mu\text{g g}^{-1}$), Ragedara ($0.08 \mu\text{g g}^{-1}$), and Balama ($0.3 \mu\text{g g}^{-1}$).

Strontium: Minas Gerais ($1.5 \mu\text{g g}^{-1}$) and Inner Mongolia ($0.9 \mu\text{g g}^{-1}$) have the lowest contents, while Kaisersberg ($33 \mu\text{g g}^{-1}$) and Hunan Lutang ($24 \mu\text{g g}^{-1}$) show the highest values.

Molybdenum: A reversed pattern is observed for Mo. Tayginka has the highest contents of this element ($84 \mu\text{g g}^{-1}$), followed by Kropfmühl ($29 \mu\text{g g}^{-1}$), Shandong, and Heilongjian ($28 \mu\text{g g}^{-1}$). Schroll (1976) describe Mo as a characteristic trace element for graphite.

Silver: Highest silver values can be found in Zavallia ($1 \mu\text{g g}^{-1}$), compared to very low values in the Ragedara mine ($0.05 \mu\text{g g}^{-1}$).

Cadmium: Heilongjian ($0.7 \mu\text{g g}^{-1}$), Shandong, and Balama ($0.5 \mu\text{g g}^{-1}$) show the highest Cd values, while Inner Mongolia shows the lowest Cd values of all deposits ($0.08 \mu\text{g g}^{-1}$).

Barium: Ragedara mine shows the lowest Ba values ($0.5 \mu\text{g g}^{-1}$), as well as Inner Mongolia with $1 \mu\text{g g}^{-1}$. Highest Ba values can be associated with Kaisersberg ($66 \mu\text{g g}^{-1}$) and Hunan Lutang ($33 \mu\text{g g}^{-1}$), both amorphous graphites.

Thallium: Generally, values for this element are very low with highest Tl values for Hunan Lutang ($0.2 \mu\text{g g}^{-1}$) and lowest values for Shandong ($0.01 \mu\text{g g}^{-1}$).

Lead: Amorphous graphite (Kaisersberg $28 \mu\text{g g}^{-1}$) and Hunan Lutang ($38 \mu\text{g g}^{-1}$) are associated with high Pb levels, whereas Inner Mongolia ($0.3 \mu\text{g g}^{-1}$), Shandong ($0.4 \mu\text{g g}^{-1}$) and Skaland ($0.5 \mu\text{g g}^{-1}$) show low Pb levels.

Yttrium: The highest Y values are associated with Kaisersberg ($12 \mu\text{g g}^{-1}$) and Kropfmühl ($11 \mu\text{g g}^{-1}$). Inner Mongolia ($0.08 \mu\text{g g}^{-1}$) shows the lowest Y values. Most locations show Y values in the range of 1 to $2 \mu\text{g g}^{-1}$.

Thorium: Kaisersberg ($13 \mu\text{g g}^{-1}$) is associated with the highest Th values, followed by Hunan Lutang and Kropfmühl ($3 \mu\text{g g}^{-1}$). Very low Th values ($0.02 \mu\text{g g}^{-1}$) are present in samples from Ragedara mine.

Uranium: Higher U values are associated with Kaisersberg ($2.6 \mu\text{g g}^{-1}$) and Heilongjian ($2.4 \mu\text{g g}^{-1}$) and low U values with Ragedara ($0.01 \mu\text{g g}^{-1}$).

Based on the scoring coefficients from LDA performed in chapter 6.4.2.4.1. and the assessment of the individual element values compared to each deposit the following elements could be identified as key elements for the discrimination of graphite deposits: **Mg, Al, V, Fe, Ga, Sr, Ag, Cd, U.**

Comparison REE patterns:

REE patterns of all locations differ from each other. Only Kaisersberg and Kropfmühl exhibit similarities. The individual patterns of each sample from Kaisersberg, Minas Gerais, Balama, Zavallia, and Ragedara, are very similar within the location, indicating little variability in the deposit, possibly also similar mining areas. The two individual samples from the Shandong province are distinct from each other. Also, one (sample 2B) of three available samples from the Skaland mine shows a different pattern. Sample 2B was collected before 2019, which could be an indication of a different mining area. For the Tayginka deposit, two samples show a positive Eu anomaly and one sample shows a negative Eu anomaly, which could also suggest different mining areas within the deposit. The comparison of REE patterns allows conclusions about the homogeneity of the deposit, meaning they are difficult to contaminate by sample preparation or similar processes. Normalizing the data resulted in smooth patterns that exclusively exhibit positive or negative anomalies. Anomalies could originate from fractionation due to variations in redox conditions during geochemical or biochemical cycles or from anthropogenic influences and contamination (e.g. dust) within the raw material processing.

7.4. Multi-Parameter Approach

Various parameters were tested for the development of an analytical proof of origin for natural graphite deposits. The analytical methods generated individual parameters, which, in a multidimensional model, allow to distinguish individual locations. Utilizing different parameters, such as carbon isotopes, crystallinity and trace elements, increases the number of individual variables for multivariate data analysis. The multi-parameter approach enables greater differentiability of the data, compared to uni- or bivariate data analysis. Crystallinity, measured by Raman spectroscopy mainly refers to the carbon type (amorphous/flake/vein). Carbon isotopes enable differentiation into groups related to the genesis and the geographic origin (Ukraine, Sri Lanka and rest of the world) but are not capable of distinguishing at the level of individual mines/provinces. Trace element data, obtained from both, ICP-MS and LA-ICP-MS does not allow a clear differentiation of mines/provinces, but allows for the highest individual level of differentiability, if combined with crystallinity data. In general, trace element data from ICP-MS shows a clearer differentiation of individual mines/provinces compared to data from LA-ICP-MS. Trace element data obtained from ICP-MS, when combined with the stable carbon isotope ratios, also demonstrates complete differentiation of individual graphite deposits, whereas trace element data obtained from LA-ICP-MS, along with stable carbon isotope data, exhibits overlaps in some occurrences. When all three parameters are combined, the results with trace element data from ICP-MS show a better outcome in terms of differentiation, compared to the multivariate analysis of all parameters, including trace element data from LA-ICP-MS. For the development of an analytical proof of origin (APO) for natural graphite, it made sense to use and analyze all three parameters. For a graphite APO database intended for comparing unknown graphite samples to, all three parameters should be analyzed and taken record of in the database.

7.5. Test Run with Unknown Samples

To verify and test the developed methodology with unknown samples, the attributes related to the origin were removed from three graphite samples in the present study. The identity of the samples only consisted of the Sample ID, along with the trace element data from the solution-based-ICP-MS analysis, stable carbon isotopes and Raman parameters.

Row 3 (in Fig. 97) represents Sample 21N from Minas Gerais (BRA). The study includes four more samples from this deposit. Row 17 (in Fig. 97) represents Sample 21B from Balama (MOZ). The study includes five more samples from this origin. Row 26 (in Fig. 97) represents Sample 4B from Zavallia (UKR). The study only contains one more sample from this deposit.

Through LDA, Sample 21N could be assigned to the correct origin (Minas Gerais, BRA) with 100% probability (Fig. 97 right column and Fig. 98). Sample 21B from Balama (MOZ) could only be assigned to the correct origin with 42 % probability, with 58 % of the probability for the wrong origin (Shandong) (Fig. 97 and 98). Sample 4B (Zavallia) in row 26 was assigned to a wrong origin (Brickaville, MAD) with 100 % probability (Fig. 97). The data basis and multiple samples from one origin (deposit/mine) are thus essential to assign unknown samples to an origin in the database. If a deposit is not stored in the database, it is also impossible to assign an unknown sample to that origin. APO databases constantly need to be enlarged and updated.

Zeile	Beobachtet	Qu.Dist.(Beobachtet)	Wahrsch.(Beobachtet)	-Log(Wahrsch.)	Vorhersage	Wahrsch.(Vorhers.)	Sonstige
1	ICP-MS 1A Kaisersberg (AUT) Austria						
2	ICP-MS 21C Minas Gerais* (BRA) Brazil						
3	ICP-MS 21N						
4	ICP-MS 3B Minas Gerais* (BRA) Brazil						
5	ICP-MS 21M Minas Gerais* (BRA) Brazil						
6	ICP-MS 22C Minas Gerais* (BRA) Brazil						
7	ICP-MS 21A Shandong* (CHN) China						
8	ICP-MS 22A Shandong* (CHN) China						
9	ICP-MS 4A Hunan Lutang* (CHN) China						
10	ICP-MS 6B Inner Mongolia* (CHN) China						
11	ICP-MS 7B Heilongjian* (CHN) China						
12	ICP-MS 13B Kropfmühl (GER) Germany						
13	ICP-MS 21D Brickaville* (MAD) Madagascar						
14	ICP-MS 11B Brickaville* (MAD) Madagascar						
15	ICP-MS 22D Brickaville* (MAD) Madagascar						
16	ICP-MS 1B Balama (MOZ) Mozambique						
17	ICP-MS 21B						
18	ICP-MS 21J Balama (MOZ) Mozambique						
19	ICP-MS 21K Balama (MOZ) Mozambique						
20	ICP-MS 21L Balama (MOZ) Mozambique						
21	ICP-MS 22B Balama (MOZ) Mozambique						
22	ICP-MS 15B Skaland (NOR) Norway						
23	ICP-MS 2B Skaland (NOR) Norway						
24	ICP-MS 22G Skaland (NOR) Norway						
25	ICP-MS 22F Zavallia (UKR) Ukraine						
26	ICP-MS 4B						
27	Tayginka (RUS)						
28	Tayginka (RUS)						
29	Tayginka (RUS)						
30	Ragedara (LKA)						
31	Ragedara (LKA)						

Zeile	Beobachtet	Qu.Dist.(Beobachtet)	Wahrsch.(Beobachtet)	-Log(Wahrsch.)	Vorhersage	Wahrsch.(Vorhers.)	Sonstige
1	Kaisersberg (AUT)	2.274e-13	1.0000	0.000	Kaisersberg (AUT)	1.0000	
2	Minas Gerais* (BRA)	11.2500	1.0000	0.000	Minas Gerais* (BRA)	1.0000	
3					Minas Gerais* (BRA)	1.0000	
4	Minas Gerais* (BRA)	11.2500	1.0000	0.000	Minas Gerais* (BRA)	1.0000	
5	Minas Gerais* (BRA)	11.2500	1.0000	0.000	Minas Gerais* (BRA)	1.0000	
6	Minas Gerais* (BRA)	11.2500	1.0000	0.000	Minas Gerais* (BRA)	1.0000	
7	Shandong* (CHN)	7.5000	0.9805	0.020	Shandong* (CHN)	0.9805	
8	Shandong* (CHN)	7.5000	1.0000	0.000	Shandong* (CHN)	1.0000	
9	Hunan Lutang* (CHN)	9.095e-13	1.0000	0.000	Hunan Lutang* (CHN)	1.0000	
10	Inner Mongolia* (CHN)	-2.27e-13	1.0000	0.000	Inner Mongolia* (CHN)	1.0000	
11	Heilongjian* (CHN)	2.842e-14	1.0000	0.000	Heilongjian* (CHN)	1.0000	
12	Kropfmühl (GER)	2.274e-13	1.0000	0.000	Kropfmühl (GER)	1.0000	
13	Brickaville* (MAD)	10.0000	1.0000	0.000	Brickaville* (MAD)	1.0000	
14	Brickaville* (MAD)	10.0000	1.0000	0.000	Brickaville* (MAD)	1.0000	
15	Brickaville* (MAD)	10.0000	1.0000	0.000	Brickaville* (MAD)	1.0000	
16	Balama (MOZ)	12.0000	1.0000	0.000	Balama (MOZ)	1.0000	
17					Shandong* (CHN)	0.5320	Balama (MOZ) 0.42
18	Balama (MOZ)	12.0000	0.9963	0.004	Balama (MOZ)	0.9963	
19	Balama (MOZ)	12.0000	1.0000	0.000	Balama (MOZ)	1.0000	
20	Balama (MOZ)	12.0000	0.9996	0.000	Balama (MOZ)	0.9996	
21	Balama (MOZ)	12.0000	0.9978	0.002	Balama (MOZ)	0.9978	
22	Skaland (NOR)	10.0000	1.0000	0.000	Skaland (NOR)	1.0000	
23	Skaland (NOR)	10.0000	1.0000	0.000	Skaland (NOR)	1.0000	
24	Skaland (NOR)	10.0000	1.0000	0.000	Skaland (NOR)	1.0000	
25	Zavallia (UKR)	0.0000	1.0000	0.000	Zavallia (UKR)	1.0000	
26					Brickaville* (MAD)	1.0000	
27	Tayginka (RUS)	10.0000	1.0000	0.000	Tayginka (RUS)	1.0000	
28	Tayginka (RUS)	10.0000	1.0000	0.000	Tayginka (RUS)	1.0000	
29	Tayginka (RUS)	10.0000	1.0000	0.000	Tayginka (RUS)	1.0000	
30	Ragedara (LKA)	7.5000	1.0000	0.000	Ragedara (LKA)	1.0000	
31	Ragedara (LKA)	7.5000	1.0000	0.000	Ragedara (LKA)	1.0000	

Figure 97: Test run with unknown samples (row 3, 17 and 26) left and estimated probabilities for the origin right

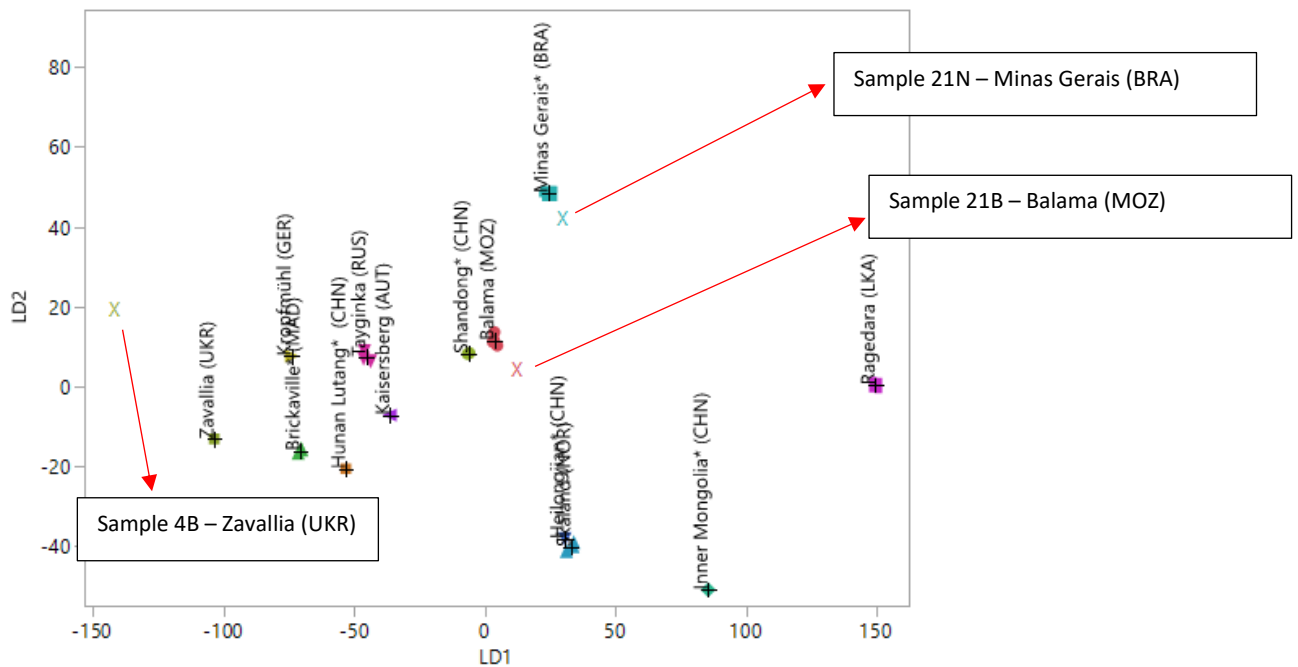


Figure 98: Graphical presentation of unknown samples (X) related to their origin

8. Conclusion and Perspectives

8.1. Analytical Method Evaluation – A Traffic Light System Perspective

The applied analytical methods used for the Analytical Proof of Origin (APO) for graphite are rated using a traffic light system to classify them into three performance levels (green: good/preferred, yellow: moderate/caution, red: poor/not recommended).

The rated categories are the following:

- 1. Differentiating Power:** How meaningful are the results and to what extent can individual locations/origins be distinguished? Can specific deposits be identified, or can only broad categories (e.g. carbon type) be defined?
- 2. Simplicity/Sample Throughput:** How straightforward is the method in its application and how many samples can be processed? How labor-intensive is the sample preparation?
- 3. Accessibility:** How accessible are the laboratory methods or laboratory equipment in general? This needs to be kept in mind for laboratories and analyses especially on processing sites and export hubs in developing countries. Does it require cleanrooms or certified laboratory facilities? Can the analysis also be conducted using portable devices? What about required gases and acids?
- 4. Cost:** How cost-intensive is the methodology? What is the acquisition, maintenance, and operational cost of the laboratory equipment?

The main factors considered are costs and simplicity/sample throughput.

Carbon isotopes			
differentiating power	simplicity/sample throughput	accessibility	cost

Figure 99: Traffic light system for carbon isotopes

The analysis of carbon isotopes is rated yellow for the categories of differentiating power, simplicity/sample throughput and cost, and red for the category of the accessibility (Fig. 99). None of the categories are marked in green. The differentiation power is limited to interpretations about the localities concerning Sri Lanka, Ukraine and “rest of the world”. The methodology is complex, both in sample preparation and measurement, requiring high-end laboratory equipment with gas supply and exhaust. This laboratory equipment is only available in specialized laboratories. The costs are correspondingly high due to the sample preparation and measurement duration.

Structure and Crystallinity			
differentiating power	simplicity/sample throughput	accessibility	cost

Figure 100: Traffic light system for structure and crystallinity

The structural analysis is rated yellow for the category of differentiating power and green for the remaining categories of simplicity/sample throughput, cost and accessibility. (Fig. 100). The differentiating power is limited to the graphite type (amorphous, flake, hydrothermal type). HR Raman Spectroscopy can be performed fast and many samples can be analyzed in a short time. The methodology is available as a portable device also and in terms of handling, it requires no special preparation of the

sample. A high sample throughput is possible and the resulting costs are therefore reasonable and rated as green.

Trace elements			
differentiating power	simplicity/sample throughput	accessibility	cost

Figure 101: Traffic light system for trace elements

Trace element analysis using (LA)-ICP-MS exhibits a different pattern. The differentiating power is rated as green, allowing the differentiation of individual deposits. The rest of the categories remain yellow. The simplicity/sample throughput is moderate and the sample preparation is moderately complex (Fig. 101). The access to this laboratory methodology is limited and requires well-equipped facilities. The resulting costs are moderate due to the relatively large sample throughput. Acquiring trace elements using LA-ICP-MS in general is much less complicated and requires no special sample preparation (only pressing of the material), compared to solution analysis using ICP-MS, making it preferable for the determination of trace elements in graphite.

As a result, trace element analysis is identified as the method with the most significant information regarding the origin, with moderate assessments in the remaining categories of simplicity/sample throughput, accessibility and cost.

8.2. Feasibility and Costs

During the Analytical Proof of Origin (APO) study for graphite, the question of the costs associated and the applicability of this traceability method in the industry arose more frequently. For industrial applications, handheld options are best suited, which are quick and easy to apply directly at the site, without the need for advanced sample preparation, especially without trained personnel and expensive laboratory facilities. Currently it is not feasible to use portable methods for this application. This presented study on method development for an Analytical Proof of Origin (APO) for graphite is only a first step in the application of traceability through laboratory methods.

The APO can only be applied in special cases and does not constitute a routine method for everyday use. However, it is a method directly related to the raw material, making it highly tamper-resistant.

8.3. Conclusion

To conclude this study, a fish bone diagram, or cause and effect diagram or Ishikawa Diagram is used to organize information to see and understand various factors influencing the Analytical Proof of Origin (for graphite) (Fig. 102).

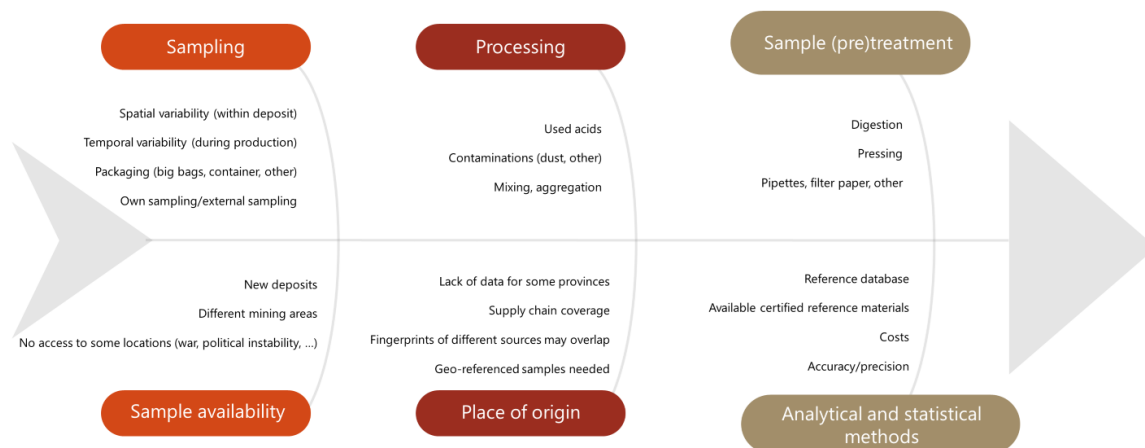


Figure 102: Cause and effect diagram APO Method for Graphite

An essential point at the beginning of the APO process is the availability of samples to conduct fingerprinting studies. Various areas of a deposit need to be covered to represent the spatial variability within the mine/deposit. Also, different sections of a deposit are mined and processed over time (temporal variability). New deposits (currently mostly in Europe and Canada) are also being developed, contributing to the diversification of global production, especially in the coming years due to increasing demand and the desire for a reduced dependence on China. The database needs to be kept up-to-date and new locations must be analyzed and documented. Some production locations are no longer accessible due to war and political instabilities, or production may be halted or exports restricted (see current situation in Ukraine and Russia).

The sampling process heavily relies on the availability of samples and the person taking samples. Efforts should always be made to achieve a representative sampling strategy. In many cases, sampling is conducted externally. Sampling also involves the process of mixing material from mining in the processing plant and determining which concentrate (quality) from the processing plant is filled into a big bag or container.

It is crucial to secure and geo-reference the origin of the samples. For some regions, the origin could only be specified up to the province level (for China, Madagascar and Brazil), not the exact mine site. In the case of China, achieving a more detailed level is not feasible unless samples are collected on-site. Processing plants from locations in Madagascar and Brazil process the material of more than one mine. Resulting concentrates are therefore mixtures of different mines within one province. Mixing and aggregation of material from different sources (countries) might harm the potential use of a geochemical fingerprint and needs to be investigated further.

Various acids are used in the processing process, which can vary between different plants (processing fingerprint!). Contaminations from the processing plant can be introduced, especially if other raw materials are processed in the same plant as well and external inputs (equipment abrasion, dust) can introduce contaminations. The potential supply chain coverage of this methodology needs to be considered and at which stage in the supply chain the geo-based traceability does stop working. Impurities (trace elements levels) are expected to decrease throughout the processing chain and no longer be present in battery-grade graphite concentrate.

Samples must undergo sample preparation, including dissolution, pressing into pellets and treatment of liquids with pipettes and filters. Additionally, the weighing process can introduce uncertainties and is highly operator-dependent.

The application of analytical and statistical methods at the very end of the APO brings certain uncertainties and data quality still needs to be improved by enhanced method development. Machine operators may cause differences in results due to different machine handling and tuning. The precision and accuracy of the produced data play a crucial role, as well as the availability of suitable certified calibration and reference materials. A reference database needs to be available to compare unknown samples and needs to be constantly updated.

The three tested and applied parameters to identify the origin of graphite deposits in this study provide information at various levels. Trace element analysis provided the most detailed information, allowing differentiation down to the level of the mine. However, this parameter is highly dependent on the processing and further investigation is needed to understand how the parameter behaves throughout the processing chain of graphite. The information provided by carbon isotope ratio parameter also provided information, to some extent, but is more related to the carbon source material. Therefore, Ukraine could be distinguished from the rest of the world and samples from Sri Lanka due to the non-organic (hydrothermal origin) of the graphite precursor material. The remaining samples could not be further differentiated from other deposits based solely on this parameter and were classified as RoW (rest of the world).

Using Raman spectroscopy, amorphous graphite is distinguished from other deposits based on the different crystallinity. Amorphous graphite in this study is limited to the deposits in Kaisersberg (Austria) and Hunan Lutang (China). Thus, these two deposits could be distinguished from all others.

A multi-parameter modeling approach at the end of the Results chapter, considering all three parameters, significantly increased the differentiation power of the three methods.

Overall, establishing a proof of origin method for mineral raw materials is an important part of supply chain management and can help ensure compliance with regulations/certifications and ethical sourcing practices.

8.4. Further research implications and aspects

1. Optimization and Automation:

How can the developed method be further optimized for enhanced efficiency?

The sample preparation process is the most error-prone. This is where the majority of errors occur, as impurities may be introduced and the material can become contaminated very easily. Portable analytical methods enable greater efficiency in analytics. Analysis can take place directly on-site without the need for standardized laboratory facilities and personnel can be trained for these applications. With the elimination of complicated sample preparation, the accuracy of results also increases simultaneously. Sample preparation processes can introduce impurities and overall biases through digestion methods. However, portable methods require method development and usually show lower limits of detection in comparison to stationary laboratory methods and can currently not be used for determining the origin of graphite. Portable LIBS and XRF methods had been tested, but need to undergo special method development for the use in APO techniques. Limitations using pLIBS and pXRF devices were inhomogeneous sample material, the analysis of small areas of the material only and the data processing and data reduction. The database of samples and analyzed parameters constantly needs to be enlarged and updated, as well as data quality must be improved.

Are there opportunities for automation of the analytical and statistical process?

The data evaluation process can be optimized. Machine-learning applications can assign unknown samples to known ones in the database. However, the entire process of analytical work always requires trained personnel. Complete automation is not possible, although some steps, such as in the sample preparation process, might be automated. Automated statistical data evaluation can save a significant amount of time.

2. Exploration of New Analytical Techniques

What emerging analytical techniques could be explored to improve the precision and sensitivity of the analytical proof of origin method?

LA-LIBS tandem systems can analyze samples even faster by generating more measurements in a shorter time. Advanced TOF-laser-(ablation) technologies and mass spectrometers enable increasingly higher sensitivities and speed.

Portable analytical applications can be combined or connected with processing facilities. This makes portable methods location-dependent again, but not tied to a certified laboratory. It allows for a rapid analysis without the need for specific sample preparation, potentially also on conveyor belts in the processing of raw materials. Sensitivities of those techniques still need to be improved to determine trace element levels.

If the sample preparation process can be automated or simplified and the application of analytical methods could become more user-friendly and less time-consuming in the future, analysis for industry applications could take place directly on-site. Note: Certified independent laboratory facilities will still be required in case of doubt for independent assessments (customs, certification systems, other).

Are there any portable devices to be used for this application?

Currently available on the market, handheld LIBS (Laser-Induced Breakdown Spectroscopy) devices could be applied after calibration and method development for the required application and raw material. Carbon isotope measurements still require well equipped laboratories. Trace element detection requires high sensitivities for the required limits of detection and need a sample preparation prior to applying the analytical method, which is currently not feasible with any portable device.

3. Extension to Other Materials:

Can the developed method be adapted or extended to analyze the origin of other mineral raw materials? Or other carbon-based materials?

The analytical proof of origin must be independently developed for each raw material. The parameters defining the APO are individual for each raw material and are influenced by geological formation processes, material properties, grain sizes and mineralogy. Trace elements are a parameter that can be used for many raw materials. However, their analysis is complex and material-dependent. Each material requires its own matrix-matched calibration materials, as well as various sample preparation and analytical methods.

The application of this methodology to other carbon-rich materials is very likely possible. Method development for graphite analysis was combined with the analysis of carbon-rich materials from methane pyrolysis at the Chair of General and Analytical Chemistry. Carbonaceous material from methane pyrolysis has a broad range of applications, spanning from various technological to agricultural applications. Also, by-products from the battery industry can be considered as carbon-rich materials due to the high graphite content (both synthetic and natural). Anode materials or black mass, a material

obtained from recycling Li-ion batteries might be a potential material using the developed analytical methodology.

4. Real-world Application and Validation:

Can the method be validated and applied in real-world scenarios? Is there a potential for its application in industry and certification standards?

In the present stage, APO for graphite is cost-intensive. The analytical proof of origin using laboratory methods is not likely to become a standard application due to its high costs, complexity, and time-consumption. APO can be applied in specific cases of doubt or questionable origin. This methodology directly refers to the raw material itself and thus might verify the origin of a raw material based on its inherent material properties. Most existing proof of origin methods refer to (digital) documents only, which can be falsified and are potentially vulnerable to fraud. The potential for industrial applications is more likely in on-site technologies. The approach developed in this study may be relevant for certified laboratory facilities involved in standardized certification systems.

How consistent are the parameters along the value chain?

This was tested and verified using carbon isotope ratios. The ratio remains consistent along the processing steps from flotation feed to purified graphite and only changes due to the natural spatial variability within a deposit. Trace elements in different (intermediate) products along the supply chain still need to be collected and analyzed. After each processing step, the content of impurities decreases and it is to be expected that the content of significant trace elements will therefore also decrease. If remaining elements are directly bound to the graphite structure still needs to be explored. However, effects of processing on fingerprinting results still needs to be understood.

5. Environmental and Ethical Considerations:

Can the developed method be employed to address environmental and ethical concerns related to graphite production and sourcing?

The CO₂ emissions of a raw material can be estimated and calculated by knowing the origin of the material and its trade routes.

Certain ethical problems or human rights violations are more prevalent in some regions than in others. For example, raw materials from Europe may receive less emphasis on ethical considerations, in comparison to raw materials originating from Africa. Chinese material is often criticized due to its strongly negative environmental impacts.

The analytical proof of origin (APO) of the critical raw material graphite marks a significant step towards securing and tracing graphite supply chains. The developed methodology does not only ensure the authenticity and origin verification of graphite but also aligns with the requirements of the EU supply chain law. By implementing traceability measures, industries can achieve greater transparency, reduce the risk of counterfeit materials and enhance the sustainability of their supply chains.

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Appendix

A. Worldwide Database of Graphite Carbon Isotope Values ($\delta^{13}\text{C}$)

sample ID	continent	mine/province	country	type	$\delta^{13}\text{C}$ (‰)	Reference
Cx-07	South America	Peresopolis	Brazil	Amorph	-28.7	(Manoel and Dexheimer Leite 2018)
Cx-12	South America	Peresopolis	Brazil	Amorph	-29.01	(Manoel and Dexheimer Leite 2018)
Cx-15	South America	Peresopolis	Brazil	Amorph	-29.23	(Manoel and Dexheimer Leite 2018)
Cx-19	South America	Peresopolis	Brazil	Amorph	-28.36	(Manoel and Dexheimer Leite 2018)
20TP01	Asia	Tianping	China	Flake	-28.8	(Ma et al. 2021)
20TP02	Asia	Tianping	China	Flake	-26.8	(Ma et al. 2021)
20TP03	Asia	Tianping	China	Flake	-27.2	(Ma et al. 2021)
20TP04	Asia	Tianping	China	Flake	-26.3	(Ma et al. 2021)
20TP05	Asia	Tianping	China	Flake	-27.6	(Ma et al. 2021)
20TP06	Asia	Tianping	China	Flake	-27.7	(Ma et al. 2021)
20TP07	Asia	Tianping	China	Flake	-27.7	(Ma et al. 2021)
20TP08	Asia	Tianping	China	Flake	-27.3	(Ma et al. 2021)
PD-10	Asia	Liugezhuang	China	Flake	-21.3	(Wang et al. 2019)
PD-17	Asia	Liugezhuang	China	Flake	-24.7	(Wang et al. 2019, Meng 2016)
Z25	Asia	Hadamengou	China	Flake	-25.86	(Wang et al. 1988)
Z26	Asia	Hadamengou	China	Flake	-24.55	(Wang et al. 1988)
Z27	Asia	Hadamengou	China	Flake	-16.1	(Wang et al. 1988)
Z28	Asia	Hadamengou	China	Flake	-20.54	(Wang et al. 1988)
Z20	Asia	Miaogou	China	Flake	-28.97	(Wang et al. 1988)
Z21	Asia	Miaogou	China	Flake	-14.6	(Wang et al. 1988)
Z22	Asia	Miaogou	China	Flake	-15.41	(Wang et al. 1988)
Z23	Asia	Miaogou	China	Flake	-13.95	(Wang et al. 1988)
Z16	Asia	Xinghe	China	Flake	-20.49	(Wang et al. 1988)
Z17	Asia	Xinghe	China	Flake	-20.78	(Wang et al. 1988)
Z18	Asia	Xinghe	China	Flake	-20.64	(Wang et al. 1988)
DL-4	Asia	Daliangziku	China	Flake	-17.9	(Wang et al. 2019)
DL-9	Asia	Daliangziku	China	Flake	-18	(Wang et al. 2019)
LG-6	Asia	Liugezhuang	China	Flake	-22.3	(Wang et al. 2019)
B5304-1	Asia	Bairiqili	China	Flake	-21.6	(Yan et al. 2020)
B5304-3	Asia	Bairiqili	China	Flake	-21.6	(Yan et al. 2020)
B5304-2	Asia	Bairiqili	China	Flake	-23	(Yan et al. 2020)
B3458-1	Asia	Banhongshan	China	Flake	-23.1	(Yan et al. 2020)
B3461-1	Asia	Banhongshan	China	Flake	-23.2	(Yan et al. 2020)
B3461-2	Asia	Banhongshan	China	Flake	-23.4	(Yan et al. 2020)

B3763-4	Asia	Banhongshan	China	Flake	-23.4	(Yan et al. 2020)
B3763-5	Asia	Banhongshan	China	Flake	-23.9	(Yan et al. 2020)
B20002-1	Asia	Datonggou	China	Flake	-23.7	(Yan et al. 2020)
B2103-1	Asia	Datonggou	China	Flake	-23.3	(Yan et al. 2020)
B2103-2	Asia	Datonggou	China	Flake	-24	(Yan et al. 2020)
B2103-3	Asia	Datonggou	China	Flake	-23.8	(Yan et al. 2020)
B2011TC1-1	Asia	Datonggou	China	Flake	-23.7	(Yan et al. 2020)
B2011TC1-2	Asia	Datonggou	China	Flake	-23.3	(Yan et al. 2020)
JHS-1	Asia	Jinhongshan	China	Flake	-23.1	(Yan et al. 2020)
JHS-2	Asia	Jinhongshan	China	Flake	-26.3	(Yan et al. 2020)
JHS-3	Asia	Jinhongshan	China	Flake	-22.9	(Yan et al. 2020)
JHS-4	Asia	Jinhongshan	China	Flake	-23.6	(Yan et al. 2020)
JHS-5	Asia	Jinhongshan	China	Flake	-23.1	(Yan et al. 2020)
JHS-6	Asia	Jinhongshan	China	Flake	-24.8	(Yan et al. 2020)
B2003-1	Asia	Koukouertu	China	Flake	-22.5	(Yan et al. 2020)
B2003-2	Asia	Koukouertu	China	Flake	-23.5	(Yan et al. 2020)
BTC2-1-3	Asia	Koukouertu	China	Flake	-22.5	(Yan et al. 2020)
BTC3-1-2	Asia	Koukouertu	China	Flake	-23.2	(Yan et al. 2020)
B6305-1	Asia	Zelikeng	China	Flake	-22.3	(Yan et al. 2020)
B6305-3	Asia	Zelikeng	China	Flake	-22.3	(Yan et al. 2020)
B6305-4	Asia	Zelikeng	China	Flake	-22.8	(Yan et al. 2020)
B6305-5	Asia	Zelikeng	China	Flake	-22.5	(Yan et al. 2020)
B6308-1	Asia	Zelikeng	China	Flake	-23.1	(Yan et al. 2020)
B6308-3	Asia	Zelikeng	China	Flake	-23	(Yan et al. 2020)
OY-XH-6a-1	Asia	Huangtuyao	China	Flake	-25.5	(Yang et al. 2014)
OY-XH-6a-2/1	Asia	Huangtuyao	China	Flake	-25.48	(Yang et al. 2014)
OY-XH-6a-2/2	Asia	Huangtuyao	China	Flake	-25.5	(Yang et al. 2014)
OY-XH-6b-1	Asia	Huangtuyao	China	Flake	-15.77	(Yang et al. 2014)
OY-XH-6b-2	Asia	Huangtuyao	China	Flake	-16.77	(Yang et al. 2014)
OY-XH-6c-1A2	Asia	Huangtuyao	China	Flake	-19.05	(Yang et al. 2014)
OY-XH-6c-1B1	Asia	Huangtuyao	China	Flake	-20.63	(Yang et al. 2014)
OY-XH-6c-1B2	Asia	Huangtuyao	China	Flake	-20.93	(Yang et al. 2014)
OY-XH-6c-1B3	Asia	Huangtuyao	China	Flake	-20.76	(Yang et al. 2014)
OY-XH-6c-1B4	Asia	Huangtuyao	China	Flake	-20.72	(Yang et al. 2014)
OY-XH-6d1-1	Asia	Huangtuyao	China	Flake	-25.63	(Yang et al. 2014)
OY-XH-6d1-2	Asia	Huangtuyao	China	Flake	-25.66	(Yang et al. 2014)
OY-XH-6d2	Asia	Huangtuyao	China	Flake	-25.5	(Yang et al. 2014)
OY-XH-6d3-1	Asia	Huangtuyao	China	Flake	-25.28	(Yang et al. 2014)
OY-XH-6d3-2	Asia	Huangtuyao	China	Flake	-25.56	(Yang et al. 2014)
OY-XH-6d4-1	Asia	Huangtuyao	China	Flake	-20.6	(Yang et al. 2014)
ZK7-4H25	Asia	Magou	China	Flake	-18.65	(Zhu et al. 2021)
ZK7-4H31	Asia	Magou	China	Flake	-22.88	(Zhu et al. 2021)
ZK7-4H42	Asia	Magou	China	Flake	-25.93	(Zhu et al. 2021)
ZK7-4H55	Asia	Magou	China	Flake	-20.94	(Zhu et al. 2021)
ZK7-4H66	Asia	Magou	China	Flake	-18.63	(Zhu et al. 2021)
ZK7-4H95	Asia	Magou	China	Flake	-17.39	(Zhu et al. 2021)

ZK3-7H4	Asia	Magou	China	Flake	-18.13	(Zhu et al. 2021)
ZK3-7H9	Asia	Magou	China	Flake	-22.47	(Zhu et al. 2021)
ZK3-7H14	Asia	Magou	China	Flake	-25	(Zhu et al. 2021)
ZK3-7H24	Asia	Magou	China	Flake	-25.72	(Zhu et al. 2021)
ZK3-7H32	Asia	Magou	China	Flake	-21.88	(Zhu et al. 2021)
ZK3-7H32-rep	Asia	Magou	China	Flake	-21.78	(Zhu et al. 2021)
ZK3-7H37	Asia	Magou	China	Flake	-18.83	(Zhu et al. 2021)
ZK3-7H101	Asia	Magou	China	Flake	-18.39	(Zhu et al. 2021)
ZK3-7H118	Asia	Magou	China	Flake	-16.49	(Zhu et al. 2021)
WLG-1-01	Asia	Wuligou	China	Flake	-15.14	(Zhu et al. 2021)
WLG-1-05	Asia	Wuligou	China	Flake	-15.71	(Zhu et al. 2021)
WLG-1-18	Asia	Wuligou	China	Flake	-14.95	(Zhu et al. 2021)
WLG-1-19	Asia	Wuligou	China	Flake	-16.22	(Zhu et al. 2021)
WLG-2-01	Asia	Wuligou	China	Flake	-18.34	(Zhu et al. 2021)
WLG-2-02	Asia	Wuligou	China	Flake	-17.82	(Zhu et al. 2021)
WLG-2-03	Asia	Wuligou	China	Flake	-13.93	(Zhu et al. 2021)
WLG-2-04	Asia	Wuligou	China	Flake	-13.71	(Zhu et al. 2021)
WLG-2-04-rep	Asia	Wuligou	China	Flake	-13.69	(Zhu et al. 2021)
15a2	Asia	Itna	India	Flake	-24	(B. Maibam, P. Sanyal, S. Bhattacharya 2015)
15A1	Asia	Itna	India	Flake	-24.6	(Maibam et al. 2015)
15A2	Asia	Itna	India	Flake	-24.6	(Maibam et al. 2015)
15b1	Asia	Itna	India	Flake	-24.9	(Maibam et al. 2015)
15b2	Asia	Itna	India	Flake	-24.8	(Maibam et al. 2015)
15B1	Asia	Itna	India	Flake	-25.2	(Maibam et al. 2015)
14a	Asia	Thumbasog e	India	Flake	-20.6	(Maibam et al. 2015)
14b	Asia	Thumbasog e	India	Flake	-21.3	(Maibam et al. 2015)
14A	Asia	Thumbasog e	India	Flake	-21.5	(Maibam et al. 2015)
14B	Asia	Thumbasog e	India	Flake	-20.5	(Maibam et al. 2015)
14C	Asia	Thumbasog e	India	Flake	-20.6	(Maibam et al. 2015)
GR-KLD-1-3	Asia	Koliakode	India	Vein	-17.5	(Zhang and Santosh 2019)
GR-KLD-2-4	Asia	Koliakode	India	Vein	-12.8	(Zhang and Santosh 2019)
GR-KLD-3-3	Asia	Koliakode	India	Vein	-12.8	(Zhang and Santosh 2019)
GR-KLD-4-2	Asia	Koliakode	India	Vein	-13.8	(Zhang and Santosh 2019)
GR-KLP-3	Asia	Kulappara	India	Vein	-11.7	(Zhang and Santosh 2019)
GR-KLP-4-1	Asia	Kulappara	India	Vein	-12.1	(Zhang and Santosh 2019)
GR-KLP-4-3	Asia	Kulappara	India	Vein	-13.3	(Zhang and Santosh 2019)
114400	Europe	Guadamur	Spain	Flake	-20.59	(Martín-Méndez et al. 2016)
114632	Europe	Guadamur	Spain	Flake	-27.76	(Martin-Mendez et al. 2016)
114633	Europe	Guadamur	Spain	Flake	-20.92	(Martin-Mendez et al. 2016)
114428	Europe	Guadamur mine site	Spain	Flake	-27.63	(Martin-Mendez et al. 2016)
114429	Europe	Guadamur mine site	Spain	Flake	-27.86	(Martin-Mendez et al. 2016)
114430	Europe	Guadamur mine site	Spain	Flake	-27.86	(Martin-Mendez et al. 2016)

114431	Europe	Guadamur mine site (trench)	Spain	Flake	-27.64	(Martin-Mendez et al. 2016)
114432	Europe	Guadamur mine site (waste)	Spain	Flake	-27.78	(Martin-Mendez et al. 2016)
114433	Europe	Guadamur mine site (waste)	Spain	Flake	-27.75	(Martin-Mendez et al. 2016)
114434	Europe	Guadamur mine site (waste)	Spain	Flake	-27.59	(Martin-Mendez et al. 2016)
114568	Europe	P. Montalbán mine	Spain	Flake	-20.91	(Martin-Mendez et al. 2016)
114554	Europe	P. Montalbán mine	Spain	Flake	-23.15	(Martin-Mendez et al. 2016)
114555	Europe	P. Montalbán mine	Spain	Flake	-23.03	(Martin-Mendez et al. 2016)
114556	Europe	P. Montalbán mine	Spain	Flake	-23.13	(Martin-Mendez et al. 2016)
114557	Europe	P. Montalbán mine	Spain	Flake	-23.01	(Martin-Mendez et al. 2016)
114571	Europe	Puebla Montalbán	Spain	Flake	-23.38	(Martin-Mendez et al. 2016)
114566	Europe	Puebla Montalbán	Spain	Flake	-20.58	(Martin-Mendez et al. 2016)
114567	Europe	Puebla Montalbán	Spain	Flake	-20.46	(Martin-Mendez et al. 2016)
U-10	Europe	SCS lower crust	Spain	Flake	-18.28	(Martin-Mendez et al. 2016)
U-10	Europe	SCS lower crust	Spain	Flake	-19.7	(Martin-Mendez et al. 2016)
00SI0115st1-4	Asia	Digana	Sri Lanka	Vein	-7.6	(Touzain et al. 2010)
00SI0115st1-3	Asia	Digana	Sri Lanka	Vein	-8.5	(Touzain et al. 2010)
00SI0115st-1-1	Asia	Digana	Sri Lanka	Vein	-7.5	(Binu-Lal et al. 2003)
00SI0115st-1-3	Asia	Digana	Sri Lanka	Vein	-7.9	(Binu-Lal et al. 2003)
00SI0115st-1-3	Asia	Digana	Sri Lanka	Vein	-7.7	(Binu-Lal et al. 2003)
00SI0115st-1-4	Asia	Digana	Sri Lanka	Vein	-7.8	(Binu-Lal et al. 2003)
00SI0115st-1-6-1	Asia	Digana	Sri Lanka	Vein	-6.3	(Binu-Lal et al. 2003)
00SI0115st-1-6-1	Asia	Digana	Sri Lanka	Vein	-7	(Binu-Lal et al. 2003)
00SI0115st-2-1	Asia	Digana	Sri Lanka	Vein	-7.3	(Binu-Lal et al. 2003)
00SI0115st-2-1	Asia	Digana	Sri Lanka	Vein	-7.2	(Binu-Lal et al. 2003)
00SI0115st-2-2	Asia	Digana	Sri Lanka	Vein	-4.5	(Binu-Lal et al. 2003)
00SI011st2-2	Asia	Digana	Sri Lanka	Vein	-1.6	(Binu-Lal et al. 2003)
00SI011st2-2	Asia	Digana	Sri Lanka	Vein	-1.7	(Binu-Lal et al. 2003)

OOSI011st2-2	Asia	Digana	Sri Lanka	Vein	-2.3	(Binu-Lal et al. 2003)
OOSI0114st-5	Asia	Magaha-Unuwinna	Sri Lanka	Vein	-8.7	(Binu-Lal et al. 2003)
OOSI0115st-1-4 Inta Crystalline	Asia	Digana	Sri Lanka	Vein	-7.64	(Binu-Lal et al. 2003)
OOSI0115st-1-4 Inta Crystalline	Asia	Digana	Sri Lanka	Vein	-7.65	(Binu-Lal et al. 2003)
OOSI0115st-1-4 Inta Crystalline	Asia	Digana	Sri Lanka	Vein	-7.67	(Binu-Lal et al. 2003)
OOSI0115st-1-4 Inta Crystalline	Asia	Digana	Sri Lanka	Vein	-7.61	(Binu-Lal et al. 2003)
OOSI0115st-1-4 Inta Crystalline	Asia	Digana	Sri Lanka	Vein	-7.59	(Binu-Lal et al. 2003)
OOSI0115st-1-4 Inta Crystalline	Asia	Digana	Sri Lanka	Vein	-7.59	(Binu-Lal et al. 2003)
OOSI0115st-1-4 Inta Crystalline	Asia	Digana	Sri Lanka	Vein	-7.59	(Binu-Lal et al. 2003)
OOSI0115st-1-3 Intra Crystalline	Asia	Digana	Sri Lanka	Vein	-8.92	(Binu-Lal et al. 2003)
OOSI0115st-1-3 Intra Crystalline	Asia	Digana	Sri Lanka	Vein	-8.81	(Binu-Lal et al. 2003)
OOSI0115st-1-3 Intra Crystalline	Asia	Digana	Sri Lanka	Vein	-8.2	(Binu-Lal et al. 2003)
OOSI0115st-1-3 Intra Crystalline	Asia	Digana	Sri Lanka	Vein	-9.36	(Binu-Lal et al. 2003)
OOSI0115st-1-3 Intra Crystalline	Asia	Digana	Sri Lanka	Vein	-7.98	(Binu-Lal et al. 2003)
OOSI0115st-1-3 Intra Crystalline	Asia	Digana	Sri Lanka	Vein	-7.82	(Binu-Lal et al. 2003)
OOSI0115st-1-6-1 Intra Crystalline	Asia	Digana	Sri Lanka	Vein	-7.8	(Binu-Lal et al. 2003)
OOSI0115st-1-6-1 Intra Crystalline	Asia	Digana	Sri Lanka	Vein	-6.91	(Binu-Lal et al. 2003)
OOSI0115st-1-6-1 Intra Crystalline	Asia	Digana	Sri Lanka	Vein	-6.65	(Binu-Lal et al. 2003)
OOSI0115st-1-6-1 Intra Crystalline	Asia	Digana	Sri Lanka	Vein	-6.65	(Binu-Lal et al. 2003)
OOSI0115st2-2 Intra Crystalline	Asia	Digana	Sri Lanka	Vein	-0.64	(Binu-Lal et al. 2003)
OOSI0115st2-2 Intra Crystalline	Asia	Digana	Sri Lanka	Vein	-1.37	(Binu-Lal et al. 2003)
OOSI0115st2-2 Intra Crystalline	Asia	Digana	Sri Lanka	Vein	-1.39	(Binu-Lal et al. 2003)
O10W77	Asia	Bogala	Sri Lanka	Vein	-7.76	(Touzain et al. 2010)
BSSF	Asia	Bogala	Sri Lanka	Vein	-10.34	(Touzain et al. 2010)
BNPG	Asia	Bogala	Sri Lanka	Vein	-9.64	(Touzain et al. 2010)
BCSF	Asia	Bogala	Sri Lanka	Vein	-9.45	(Touzain et al. 2010)
BCFR	Asia	Bogala	Sri Lanka	Vein	-9.43	(Touzain et al. 2010)
Close to nuclei	Asia	Bogala	Sri Lanka	Vein	-8.29	(Touzain et al. 2010)
na	Asia	Bogala	Sri Lanka	Vein	-7.79	(Dobner et al. 1978)
na	Asia	Bogala	Sri Lanka	Vein	-8.61	(Dobner et al. 1978)
na	Asia	Bogala	Sri Lanka	Vein	-7.42	(Dobner et al. 1978)

na	Asia	Bogala	Sri Lanka	Vein	-7.3	(Dobner et al. 1978)
na	Asia	Bogala	Sri Lanka	Vein	-7.2	(Dobner et al. 1978)
na	Asia	Bogala	Sri Lanka	Vein	-7.18	(Dobner et al. 1978)
na	Asia	Bogala	Sri Lanka	Vein	-8.11	(Dobner et al. 1978)
na	Asia	Bogala	Sri Lanka	Vein	-8.23	(Dobner et al. 1978))
na	Asia	Bogala	Sri Lanka	Vein	-7.35	(Dobner et al. 1978)
na	Asia	Bogala	Sri Lanka	Vein	-8.66	(Dobner et al. 1978)
na	Asia	Bogala	Sri Lanka	Vein	-7.3	(Dobner et al. 1978)
na	Asia	Bogala	Sri Lanka	Vein	-7.49	(Dobner et al. 1978)
BSL88-30-4a	Asia	Bogala	Sri Lanka	Vein	-8	(Hoernes et al. 1994)
BSL88-30-4b	Asia	Bogala	Sri Lanka	Vein	-7.6	(Hoernes et al. 1994)
BSL88-30-5	Asia	Bogala	Sri Lanka	Vein	-7.9	(Hoernes et al. 1994)
KSSF	Asia	Kahatagaha –Kolongaha	Sri Lanka	Vein	-8.7	(Touzain et al. 2010)
KNPG	Asia	Kahatagaha –Kolongaha	Sri Lanka	Vein	-8.95	(Touzain et al. 2010)
KCSF	Asia	Kahatagaha –Kolongaha	Sri Lanka	Vein	-5.61	(Touzain et al. 2010)
KCFR	Asia	Kahatagaha –Kolongaha	Sri Lanka	Vein	-9.29	(Touzain et al. 2010)
Close to nuclei	Asia	Kahatagaha –Kolongaha	Sri Lanka	Vein	-7.9	(Touzain et al. 2010)
010W76	Asia	Bogala	Sri Lanka	Vein	-8.6	(Weis et al. 1981; Touzain et al. 2010)
009W76	Asia	Kahatagaha –Kolongaha	Sri Lanka	Vein	-8	(Weis et al. 1981; Touzain et al. 2010)
BSL88-31-1	Asia	Kahatagaha –Kolongaha	Sri Lanka	Vein	-6.2	(Hoernes et al. 1994)
BSL88-31-3a	Asia	Kahatagaha –Kolongaha	Sri Lanka	Vein	-6.3	(Hoernes et al. 1994)
BSL88-31-3b	Asia	Kahatagaha –Kolongaha	Sri Lanka	Vein	-6.7	(Hoernes et al. 1994)
BSL88-31-3c	Asia	Kahatagaha –Kolongaha	Sri Lanka	Vein	-7	(Hoernes et al. 1994)
BSL88-31-4	Asia	Kahatagaha –Kolongaha	Sri Lanka	Vein	-6.7	(Hoernes et al. 1994)
Graphite Gneiss	Europe	Zavalje	Ukraine	Flake	-35.08	(Zagnitko et al. 2013)
Garnet-graphite gneiss	Europe	Zavalje	Ukraine	Flake	-30.65	(Zagnitko et al. 2013)
Graphite Garnet biotite gneiss	Europe	Zavalje	Ukraine	Flake	-30.5	(Zagnitko et al. 2013)
Graphite Garnet biotite gneiss	Europe	Zavalje	Ukraine	Flake	-34.5	(Zagnitko et al. 2013)
82-14A-1	North America	Antrim	USA	Vein	-15.8	(Rumble and Hoering 1986)
82-14A-2	North America	Antrim	USA	Vein	-17	(Rumble and Hoering 1986)
82-14A-3	North America	Antrim	USA	Vein	-15.7	(Rumble and Hoering 1986)

82-14A-4	North America	Antrim	USA	Vein	-16	(Rumble and Hoering 1986)
82-14A-5	North America	Antrim	USA	Vein	-15.8	(Rumble and Hoering 1986)
82-14A-6	North America	Antrim	USA	Vein	-16.1	(Rumble and Hoering 1986)
82-14A-7	North America	Antrim	USA	Vein	-17.2	(Rumble and Hoering 1986)
82-14B	North America	Antrim	USA	Vein	-15.9	(Rumble and Hoering 1986)
81-8A	North America	Bristol	USA	Vein	-17	(Rumble and Hoering 1986)
81-8B-1	North America	Bristol	USA	Vein	-16.9	(Rumble and Hoering 1986)
81-8B-2	North America	Bristol	USA	Vein	-16.6	(Rumble and Hoering 1986)
81-8C	North America	Bristol	USA	Vein	-17.2	(Rumble and Hoering 1986)
81-8F	North America	Bristol	USA	Vein	-17.4	(Rumble and Hoering 1986)
81-80	North America	Bristol	USA	Vein	-17.3	(Rumble and Hoering 1986)
81-8P-1	North America	Bristol	USA	Vein	-17	(Rumble and Hoering 1986)
81-8P-2	North America	Bristol	USA	Vein	-17.9	(Rumble and Hoering 1986)
81-8P-3	North America	Bristol	USA	Vein	-16.9	(Rumble and Hoering 1986)
81-8Q-1	North America	Bristol	USA	Vein	-17	(Rumble and Hoering 1986)
81-8Q-2	North America	Bristol	USA	Vein	-17	(Rumble and Hoering 1986)
80-41C-1	North America	Franklin in Pierce	USA	Vein	-9.4	(Rumble and Hoering 1986)
80-41C-2	North America	Franklin in Pierce	USA	Vein	-9.2	(Rumble and Hoering 1986)
BO-41C-3	North America	Franklin in Pierce	USA	Vein	-9.8	(Rumble and Hoering 1986)
80-41D-3	North America	Franklin in Pierce	USA	Vein	-12.4	(Rumble and Hoering 1986)
BO-41D-4	North America	Franklin in Pierce	USA	Vein	-12.9	(Rumble and Hoering 1986)
80-41D-5	North America	Franklin in Pierce	USA	Vein	-12.7	(Rumble and Hoering 1986)
80-41D-6	North America	Franklin in Pierce	USA	Vein	-12.8	(Rumble and Hoering 1986)
80-41D-7	North America	Franklin in Pierce	USA	Vein	-13.3	(Rumble and Hoering 1986)
80-41M-3	North America	Franklin in Pierce	USA	Vein	-11	(Rumble and Hoering 1986)
80-41N-1	North America	Franklin in Pierce	USA	Vein	-12.1	(Rumble and Hoering 1986)
80-41N-2	North America	Franklin in Pierce	USA	Vein	-11.1	(Rumble and Hoering 1986)
80-41N-3	North America	Franklin in Pierce	USA	Vein	-12	(Rumble and Hoering 1986)
80-41AA-4	North America	Franklin in Pierce	USA	Vein	-11.5	(Rumble and Hoering 1986)
80-41AA-5	North America	Franklin in Pierce	USA	Vein	-11.8	(Rumble and Hoering 1986)
80-41AA-6	North America	Franklin in Pierce	USA	Vein	-12.6	(Rumble and Hoering 1986)

80-41AA-7	North America	Franklin in Pierce	USA	Vein	-12.4	(Rumble and Hoering 1986)
80-41AA-8	North America	Franklin in Pierce	USA	Vein	-12.4	(Rumble and Hoering 1986)
80-41AA-9	North America	Franklin in Pierce	USA	Vein	-11.2	(Rumble and Hoering 1986)
80-41AA-10	North America	Franklin in Pierce	USA	Vein	-11.1	(Rumble and Hoering 1986)
80-41AA-11	North America	Franklin in Pierce	USA	Vein	-11.1	(Rumble and Hoering 1986)
80-41AA-12	North America	Franklin in Pierce	USA	Vein	-12.6	(Rumble and Hoering 1986)
80-41AA-13	North America	Franklin in Pierce	USA	Vein	-12.4	(Rumble and Hoering 1986)
80-41AA-14	North America	Franklin in Pierce	USA	Vein	-11.9	(Rumble and Hoering 1986)
80-41AA-15	North America	Franklin in Pierce	USA	Vein	-11.4	(Rumble and Hoering 1986)
80-41BB-3	North America	Franklin in Pierce	USA	Vein	-15.3	(Rumble and Hoering 1986)
80-41BB-4	North America	Franklin in Pierce	USA	Vein	-12.4	(Rumble and Hoering 1986)
80-41BB-5	North America	Franklin in Pierce	USA	Vein	-12.4	(Rumble and Hoering 1986)
80-41BB-6	North America	Franklin in Pierce	USA	Vein	-12.2	(Rumble and Hoering 1986)
80-41BB-7	North America	Franklin in Pierce	USA	Vein	-12.1	(Rumble and Hoering 1986)
80-41BB-8	North America	Franklin in Pierce	USA	Vein	-12.4	(Rumble and Hoering 1986)
80-41BB-9	North America	Franklin in Pierce	USA	Vein	-12	(Rumble and Hoering 1986)
80-41BB-10	North America	Franklin in Pierce	USA	Vein	-12.8	(Rumble and Hoering 1986)
80-41BB-11	North America	Franklin in Pierce	USA	Vein	-15.6	(Rumble and Hoering 1986)
80-41BB-12	North America	Franklin in Pierce	USA	Vein	-15.1	(Rumble and Hoering 1986)
80-41BB-13	North America	Franklin in Pierce	USA	Vein	-12.4	(Rumble and Hoering 1986)
80-41BB-14	North America	Franklin in Pierce	USA	Vein	-16.2	(Rumble and Hoering 1986)
80-41A1-4	North America	Franklin in Pierce	USA	Vein	-11.1	(Rumble and Hoering 1986)
80-41A1-6	North America	Franklin in Pierce	USA	Vein	-11.4	(Rumble and Hoering 1986)
80-41A1-5	North America	Franklin in Pierce	USA	Vein	-12.9	(Rumble and Hoering 1986)
80-41A1-7	North America	Franklin in Pierce	USA	Vein	-13.6	(Rumble and Hoering 1986)
80-41A2-4	North America	Franklin in Pierce	USA	Vein	-12.8	(Rumble and Hoering 1986)
80-41A2-5	North America	Franklin in Pierce	USA	Vein	-13.1	(Rumble and Hoering 1986)
9220G-1	North America	Annandale	USA	Flake	-23.2	(Volkert et al. 2000)
9220F-2	North America	Annandale	USA	Flake	-27	(Volkert et al. 2000)
9220M	North America	Annandale	USA	Flake	-22.8	(Volkert et al. 2000)
9220D-1	North America	Bloomington	USA	Flake	-19.2	(Volkert et al. 2000)

9220K-3	North America	Bloomingdale	USA	Flake	-21.3	(Volkert et al. 2000)
9220J-2	North America	Bloomingdale	USA	Flake	-15	(Volkert et al. 2000)
9220C	North America	Fisher	USA	Flake	-16.4	(Volkert et al. 2000)
9220A	North America	High Bridge	USA	Flake	-25.5	(Volkert et al. 2000)
9220E	North America	Mendham	USA	Flake	-28.1	(Volkert et al. 2000)
9220N	North America	Oak Ridge	USA	Flake	-26.6	(Volkert et al. 2000)
9220B	North America	Sutton	USA	Flake	-28.4	(Volkert et al. 2000)
9220L	North America	Sutton	USA	Flake	-28.1	(Volkert et al. 2000)
9220I	North America	Wanaque	USA	Flake	-24.1	(Volkert et al. 2000)
006S75	North America	Dillon	USA	Vein	-6.1	(Paul L. Weis 1981); Touzain et al. 2010)
001W76	North America	Ticonderoga	USA	Vein	-6	(Weis et al. 1981; Touzain et al. 2010)
AKIT17a	Africa	Aukam	Namibia	Vein	-16.5	(Schiebel 2022)
AKIT17b	Africa	Aukam	Namibia	Vein	-15.78	(Schiebel 2022)
AKIT15	Africa	Aukam	Namibia	Vein	-16.54	(Schiebel 2022)
AKIT14	Africa	Aukam	Namibia	Vein	-16.06	(Schiebel 2022)
AKIT13	Africa	Aukam	Namibia	Vein	-14.68	(Schiebel 2022)
AKIT12	Africa	Aukam	Namibia	Vein	-14.92	(Schiebel 2022)
AKIT11	Africa	Aukam	Namibia	Vein	-14.18	(Schiebel 2022)
AKIT08	Africa	Aukam	Namibia	Vein	-17.31	(Schiebel 2022)

B. Database of Graphite Carbon Isotope Values ($\delta^{13}\text{C}$) from Samples in this Project

sample ID	continent	mine/province	country	type	$\delta^{13}\text{C}$ (‰)						
1A	Europe	Kaisersberg	Austria	amorph	-23.25	-23.27	-23.27				
2A	Europe	Kaisersberg	Austria	amorph	-22.70	-21.98	-21.98	-22.05			
1B	Africa	Balama	Mozambique	flake	-25.27	-25.14	-24.58	-24.58			
2B	Europe	Skaland	Norway	flake	-21.09	-21.09	-20.93				
3B	South America	Minas Gerais	Brazil	flake	-24.84	-24.64	-25.00	-25.68	-25.68	-25.00	
4A	Asia	Hunan Lutang	China	amorph	-21.62	-21.62	-22.34				
4B	Europe	Zavallia	Ukraine	flake	-33.73	-33.25	-34.24				
5B	Asia	Tayginka	Russia	flake	-23.71	-24.16	-24.54	-24.16			
6A	Europe	Kaisersberg	Austria	amorph	-22.45	-21.10	-22.45				
6B	Asia	Inner Mongolia	China	flake	-17.96	-17.95	-19.15				
7B	Asia	Heilongjian	China	flake	-23.21	-23.21	-21.67				
10B	Asia	Ragedara	Sri Lanka	hydro (vein)	-7.25	-13.09	-7.19	-8.08			

11B	Africa	Brickaville	Madagascar	flake	-24.50	-24.96	-21.95	-24.96			
13B	Europe	Kropfmühl	Germany	flake	-23.66	-23.76	-23.66	-24.64			
14B	Europe	Kropfmühl	Germany	flake	-21.90	-21.90	-22.23	-22.43	-22.76		
15B	Europe	Skaland	Norway	flake	-21.27	-22.43	-20.32				
16B	Europe	Skaland	Norway	flake	-20.93	-20.83	-21.36	-21.46			
21A	Asia	Shandong	China	flake	-25.97	-25.97	-25.00				
21B	Africa	Balama	Mozambique	flake	-26.60	-26.51	-26.07	-26.27			
21C	South America	Minas Gerais	Brazil	flake	-25.10	-23.98	-28.29				
21D	Africa	Brickaville	Madagascar	flake	-21.42	-24.96	-25.36				
21E	Asia	Tayginka	Russia	flake	-23.71	-25.28	-24.42				
21F	Asia	Ragedara	Sri Lanka	hydro (vein)	-7.25	-10.37	-13.09				
21G	Europe	Kaisersberg	Austria	amorph	-21.32	-21.32	-21.30	-21.30	-21.84		
21H	Europe	Kaisersberg	Austria	amorph	-22.13	-22.13	-22.00	-22.08	-22.68	-22.67	
21I	Europe	Kaisersberg	Austria	amorph	-22.09	-22.09	-22.09	-23.25	-22.12	-22.12	-22.62
21K	Africa	Balama	Mozambique	flake	-26.29	-26.01	-25.35				
21L	Africa	Balama	Mozambique	flake	-26.47	-26.26	-25.60				
21M	South America	Minas Gerais	Brazil	flake	-24.33	-24.34	-24.50				
21N	South America	Minas Gerais	Brazil	flake	-25.08	-24.58	-24.53				
22A	Asia	Shandong	China	flake	-23.39	-23.73					
22B	Africa	Balama	Mozambique	flake	-25.14	-24.58	-24.58				
22C	South America	Minas Gerais	Brazil	flake	-24.28	-24.18	-24.39	-24.82	-24.72		
22D	Africa	Brickaville	Madagascar	flake	-21.95	-24.96	-23.00				
22E	Asia	Tayginka	Russia	flake	-24.31	-24.08					
22F	Europe	Zavallia	Ukraine	flake	-30.37	-28.98	-29.49				
22G	Europe	Skaland	Norway	flake	-20.83	-19.88	-22.43	-21.46	-21.36		
22H	Africa	Balama	Mozambique	flake	-26.44	-27.18					

C. Finnish Graphite Samples (Supply Chain Coverage)

Company	Origin	Sample type	Sample name	Sample ID	$\delta^{13}\text{C}$ (‰)
Beowulf Mining	Aitalompi, Finland	Ore	Typical ROM - Crushed core	202100749	-29.09
Beowulf Mining	Aitalompi, Finland	Ore	Typical ROM - Crushed core	202100749	-29.69
Beowulf Mining	Aitalompi, Finland	Ore	Typical ROM - Crushed core	202100749	-27.03
Beowulf Mining	Aitalompi, Finland	Concentrate	Flotation concentrate - Fine Fraction	202100752	-27.70
Beowulf Mining	Aitalompi, Finland	Concentrate	Flotation concentrate - Fine Fraction	202100752	-27.68
Beowulf Mining	Aitalompi, Finland	Concentrate	Flotation concentrate - Fine Fraction	202100752	-27.72
Beowulf Mining	Aitalompi, Finland	Concentrate	Flotation concentrate - Mid Fraction	202100751	-27.95
Beowulf Mining	Aitalompi, Finland	Concentrate	Flotation concentrate - Mid Fraction	202100751	-27.39
Beowulf Mining	Aitalompi, Finland	Concentrate	Flotation concentrate - Mid Fraction	202100751	-27.74
Beowulf Mining	Aitalompi, Finland	Concentrate	Flotation concentrate - Coarse fraction	202100750	-27.15
Beowulf Mining	Aitalompi, Finland	Concentrate	Flotation concentrate - Coarse fraction	202100750	-26.08
Beowulf Mining	Aitalompi, Finland	Concentrate	Flotation concentrate - Coarse fraction	202100750	-27.63
Beowulf Mining	Aitalompi, Finland	Product	Purified spherical graphite (SPG) 2A	202100754	-27.63
Beowulf Mining	Aitalompi, Finland	Product	Purified spherical graphite (SPG) 2A	202100754	-27.39
Beowulf Mining	Aitalompi, Finland	Product	Purified spherical graphite (SPG) 2A	202100754	-27.83
Beowulf Mining	Aitalompi, Finland	Product	Alkaline purified graphite	202100755	-27.45
Beowulf Mining	Aitalompi, Finland	Product	Alkaline purified graphite	202100755	-27.49
Beowulf Mining	Aitalompi, Finland	Product	Alkaline purified graphite	202100755	-29.05
Beowulf Mining	Aitalompi, Finland	Product	Micronized graphite	202100756	-27.26
Beowulf Mining	Aitalompi, Finland	Product	Micronized graphite	202100756	-27.47
Beowulf Mining	Aitalompi, Finland	Product	Micronized graphite	202100756	-29.76
Beowulf Mining	Aitalompi, Finland	Residue	By-product	202100757	-27.47
Beowulf Mining	Aitalompi, Finland	Residue	By-product	202100757	-27.36
Beowulf Mining	Aitalompi, Finland	Residue	By-product	202100757	-28.08
Beowulf Mining	Aitalompi, Finland	Core	Core Samples (Easter Zone)	AITDD17007	-30.58
Beowulf Mining	Aitalompi, Finland	Core	Core Samples (Easter Zone)	AITDD17007	-25.75
Beowulf Mining	Aitalompi, Finland	Core	Core Samples (Easter Zone)	AITDD17008	-27.77
Beowulf Mining	Aitalompi, Finland	Core	Core Samples (Easter Zone)	AITDD17008	-30.93
Beowulf Mining	Aitalompi, Finland	Core	Core Samples (Easter Zone)	AITDD18012	-27.23
Beowulf Mining	Aitalompi, Finland	Core	Core Samples (Easter Zone)	AITDD18012	-26.77
Beowulf Mining	Aitalompi, Finland	Core	Core Samples (Western Zone)	AITDD18018	-26.83
Beowulf Mining	Aitalompi, Finland	Core	Core Samples (Western Zone)	AITDD18018	-30.20

Beowulf Mining	Aitalompi, Finland	Core	Core Samples (Western Zone)	AITDD18016A	-30.44
Beowulf Mining	Aitalompi, Finland	Core	Core Samples (Western Zone)	AITDD18016A	-28.07
Beowulf Mining	Aitalompi, Finland	Core	Core Samples (Western Zone)	AITDD18016B	-28.07
Beowulf Mining	Aitalompi, Finland	Core	Core Samples (Western Zone)	AITDD18016B	-30.34
Beowulf Mining	Aitalompi, Finland	Core	Core Samples (Western Zone)	AITDD18015	-29.51
Beowulf Mining	Aitalompi, Finland	Core	Core Samples (Western Zone)	AITDD18015	-28.48814286
GTK	Emas, Finland	Ore	Flotation Feed	202301520	-27.77
GTK	Emas, Finland	Ore	Flotation Feed	202301520	-28.51
GTK	Emas, Finland	Ore	Flotation Feed	202301520	-29.21
GTK	Emas, Finland	Ore	Flotation Feed	202301520	-27.48
GTK	Emas, Finland	Concentrate	Flotation Rougher Concentrate	202301521	-29.15
GTK	Emas, Finland	Concentrate	Flotation Rougher Concentrate	202301521	-26.74
GTK	Emas, Finland	Concentrate	Flotation Rougher Concentrate	202301521	-28.41
GTK	Emas, Finland	Concentrate	Flotation Rougher Concentrate	202301521	-26.59
GTK	Emas, Finland	Concentrate	Flotation Cleaner Concentrate	202301522	-27.05
GTK	Emas, Finland	Concentrate	Flotation Cleaner Concentrate	202301522	-26.71
GTK	Emas, Finland	Concentrate	Flotation Cleaner Concentrate	202301522	-27.25
GTK	Emas, Finland	Concentrate	Flotation Cleaner Concentrate	202301522	-25.82
GTK	Emas, Finland	Product	Purified graphite	202301523	-26.35
GTK	Emas, Finland	Product	Purified graphite	202301523	-27.00
GTK	Emas, Finland	Product	Purified graphite	202301523	-26.65
GTK	Emas, Finland	Product	Purified graphite	202301523	-25.98
GTK	Raisjoki, Finland	Ore	Flotation Feed	202301524	-27.71
GTK	Raisjoki, Finland	Ore	Flotation Feed	202301524	-27.90
GTK	Raisjoki, Finland	Ore	Flotation Feed	202301524	-28.13
GTK	Raisjoki, Finland	Ore	Flotation Feed	202301524	-26.86
GTK	Raisjoki, Finland	Concentrate	Flotation Rougher Concentrate	202301525 Rough	-28.25
GTK	Raisjoki, Finland	Concentrate	Flotation Rougher Concentrate	202301525 Rough	-26.74
GTK	Raisjoki, Finland	Concentrate	Flotation Rougher Concentrate	202301525 Rough	-26.75
GTK	Raisjoki, Finland	Concentrate	Flotation Cleaner Concentrate	202301525 Clean	-27.42
GTK	Raisjoki, Finland	Concentrate	Flotation Cleaner Concentrate	202301525 Clean	-27.50
GTK	Raisjoki, Finland	Concentrate	Flotation Cleaner Concentrate	202301525 Clean	-27.45
GTK	Raisjoki, Finland	Product	Purified graphite	202301527	-27.56
GTK	Raisjoki, Finland	Product	Purified graphite	202301527	-27.85
GTK	Raisjoki, Finland	Product	Purified graphite	202301527	-27.54
GTK	Raisjoki, Finland	Product	Purified graphite	202301527	-27.42
GTK	Koivuniemi, Finland	Ore	Flotation Feed	202301528	-27.48
GTK	Koivuniemi, Finland	Ore	Flotation Feed	202301528	-26.65
GTK	Koivuniemi, Finland	Ore	Flotation Feed	202301528	-31.44
GTK	Koivuniemi, Finland	Ore	Flotation Feed	202301528	-29.76
GTK	Koivuniemi, Finland	Concentrate	Flotation Rougher Concentrate	202301529	-31.15
GTK	Koivuniemi, Finland	Concentrate	Flotation Rougher Concentrate	202301529	-31.49

GTK	Koivuniemi, Finland	Concentrate	Flotation Rougher Concentrate	202301529	-30.87
GTK	Koivuniemi, Finland	Concentrate	Flotation Rougher Concentrate	202301529	-28.55
GTK	Koivuniemi, Finland	Concentrate	Flotation Cleaner Concentrate	202301530	-31.60
GTK	Koivuniemi, Finland	Concentrate	Flotation Cleaner Concentrate	202301530	-31.52
GTK	Koivuniemi, Finland	Concentrate	Flotation Cleaner Concentrate	202301530	-31.86
GTK	Koivuniemi, Finland	Product	Purified graphite	202301531	-31.56
GTK	Koivuniemi, Finland	Product	Purified graphite	202301531	-31.86
GTK	Koivuniemi, Finland	Product	Purified graphite	202301531	-28.58

C. Raman Spectroscopy Data Table

analysis ID	sample ID		continent	mine/province	country	type	D_STA (mean)	G_STA (mean)	G_shape_factor (mean)	Dmax_pos (mean)	Gmax_pos (mean)	Dmax/Gmax -ratio (mean)	D_shape_factor
fit_1A_01	1A		Europe	Kaisersberg	Austria	amorph	93.47	34.94	6.14	1348.13	1576.82	0.37	38.72
fit_1A_02	1A		Europe	Kaisersberg	Austria	amorph	77.80	41.41	5.33	1349.40	1579.37	0.54	38.54
fit_1A_03	1A		Europe	Kaisersberg	Austria	amorph	74.65	51.70	4.39	1345.59	1579.37	0.68	37.74
fit_1A_04	1A		Europe	Kaisersberg	Austria	amorph	305.25	19.25	14.63	1357.02	1580.64	0.06	46.60
fit_1A_05	1A		Europe	Kaisersberg	Austria	amorph	218.05	16.84	14.61	1346.86	1578.09	0.08	38.48
fit_1A_06	1A		Europe	Kaisersberg	Austria	amorph	56.32	51.95	4.68	1346.86	1580.64	0.92	28.23
fit_1A_07	1A		Europe	Kaisersberg	Austria	amorph	103.52	55.31	3.24	1343.05	1574.28	0.54	41.22
fit_1A_08	1A		Europe	Kaisersberg	Austria	amorph	85.22	43.50	5.02	1350.69	1579.39	0.52	43.28
fit_1A_09	1A		Europe	Kaisersberg	Austria	amorph	127.46	33.58	6.60	1343.07	1567.96	0.27	37.83
fit_1A_10	1A		Europe	Kaisersberg	Austria	amorph	68.05	55.84	3.94	1343.07	1575.58	0.86	29.75
fit_1A_11	1A		Europe	Kaisersberg	Austria	amorph	85.00	49.53	4.11	1354.50	1581.93	0.56	50.71
fit_1A_12	1A		Europe	Kaisersberg	Austria	amorph	74.53	47.49	4.80	1341.80	1574.31	0.62	31.62
fit_1A_13	1A		Europe	Kaisersberg	Austria	amorph	114.79	42.80	7.82	1344.34	1575.58	0.38	59.71
fit_1A_14	1A		Europe	Kaisersberg	Austria	amorph	73.80	56.02	3.46	1341.80	1571.77	0.76	29.96
fit_1A_15	1A		Europe	Kaisersberg	Austria	amorph	80.98	43.15	4.75	1343.07	1569.23	0.52	27.06
fit_1A_16	1A		Europe	Kaisersberg	Austria	amorph	173.98	22.50	11.38	1345.61	1571.77	0.13	33.85
fit_1A_17	1A		Europe	Kaisersberg	Austria	amorph	110.79	35.20	5.36	1337.99	1570.50	0.30	36.17
fit_1A_18	1A		Europe	Kaisersberg	Austria	amorph	122.10	30.00	6.19	1346.88	1571.77	0.25	35.97

fit_1A_19	1A		Europe	Kaisersberg	Austria	amorph	118.81	26.63	7.13	1348.15	1571.77	0.22	29.96
fit_1A_20	1A		Europe	Kaisersberg	Austria	amorph	65.39	40.82	5.53	1351.96	1580.66	0.63	33.91
fit_1B_01	1B		Africa	Balama	Mozambique	flake	5118.34	12.42	21.54	1350.69	1576.83	0.00	59.76
fit_1B_02	1B		Africa	Balama	Mozambique	flake	5121.25	14.01	19.89	1346.88	1581.91	0.00	64.74
fit_1B_03	1B		Africa	Balama	Mozambique	flake	5477.28	12.94	22.33	1353.23	1580.64	0.00	64.24
fit_1B_04	1B		Africa	Balama	Mozambique	flake	5714.83	12.80	22.85	1349.42	1581.91	0.00	64.43
fit_1B_05	1B		Africa	Balama	Mozambique	flake	969.10	12.83	22.58	1343.07	1578.10	0.01	56.98
fit_1B_06	1B		Africa	Balama	Mozambique	flake	595.23	12.95	21.46	1344.34	1580.64	0.02	42.16
fit_1B_07	1B		Africa	Balama	Mozambique	flake	527.01	13.25	22.34	1360.85	1578.10	0.02	52.55
fit_1B_08	1B		Africa	Balama	Mozambique	flake	642.23	13.39	21.60	1357.04	1579.37	0.02	45.36
fit_1B_09	1B		Africa	Balama	Mozambique	flake	812.72	13.59	21.38	1348.15	1579.37	0.02	55.01
fit_1B_10	1B		Africa	Balama	Mozambique	flake	598.82	14.41	20.32	1349.42	1581.91	0.02	48.76
fit_1B_11	1B		Africa	Balama	Mozambique	flake	5496.99	13.27	22.22	1353.23	1581.91	0.00	64.52
fit_1B_12	1B		Africa	Balama	Mozambique	flake	5544.18	13.18	21.54	1337.98	1580.64	0.00	63.84
fit_1B_13	1B		Africa	Balama	Mozambique	flake	402.82	15.10	19.37	1349.42	1578.10	0.04	41.14
fit_1B_14	1B		Africa	Balama	Mozambique	flake	4904.18	13.13	20.02	1344.34	1575.56	0.00	60.13
fit_1B_15	1B		Africa	Balama	Mozambique	flake	148.08	20.23	11.21	1345.61	1580.64	0.13	41.07
fit_1B_16	1B		Africa	Balama	Mozambique	flake	336.45	15.21	19.70	1353.23	1581.91	0.05	44.76
fit_1B_17	1B		Africa	Balama	Mozambique	flake	469.46	14.87	19.41	1343.07	1580.64	0.03	44.77
fit_1B_18	1B		Africa	Balama	Mozambique	flake	5630.97	12.91	22.59	1354.50	1580.64	0.00	64.23
fit_1B_19	1B		Africa	Balama	Mozambique	flake	579.08	13.82	21.15	1353.23	1580.64	0.02	48.82
fit_1B_20	1B		Africa	Balama	Mozambique	flake	5680.58	12.42	24.39	1362.12	1579.37	0.00	63.60

fit_2A_01	2A		Europe	Kaisersberg	Austria	amorph	134.20	39.53	5.26	1345.61	1571.74	0.29	45.40
fit_2A_02	2A		Europe	Kaisersberg	Austria	amorph	86.02	45.14	4.89	1343.07	1569.20	0.54	32.24
fit_2A_03	2A		Europe	Kaisersberg	Austria	amorph	113.47	55.05	4.43	1343.07	1570.47	0.49	47.12
fit_2A_04	2A		Europe	Kaisersberg	Austria	amorph	78.34	49.79	4.43	1346.88	1580.63	0.63	42.66
fit_2A_05	2A		Europe	Kaisersberg	Austria	amorph	94.60	41.67	5.50	1346.88	1578.09	0.44	42.13
fit_2A_06	2A		Europe	Kaisersberg	Austria	amorph	126.46	45.66	5.02	1348.15	1573.01	0.35	48.67
fit_2A_07	2A		Europe	Kaisersberg	Austria	amorph	80.85	59.11	3.65	1351.96	1581.91	0.72	47.30
fit_2A_08	2A		Europe	Kaisersberg	Austria	amorph	115.94	47.01	4.62	1348.15	1576.82	0.40	48.93
fit_2A_09	2A		Europe	Kaisersberg	Austria	amorph	91.86	51.83	3.70	1351.96	1585.72	0.57	52.41
fit_2A_10	2A		Europe	Kaisersberg	Austria	amorph	76.94	50.83	4.33	1346.88	1581.91	0.67	41.43
fit_2A_11	2A		Europe	Kaisersberg	Austria	amorph	99.76	50.94	4.07	1349.42	1581.91	0.51	49.14
fit_2A_12	2A		Europe	Kaisersberg	Austria	amorph	93.10	51.82	4.04	1345.61	1583.18	0.56	48.11
fit_2A_13	2A		Europe	Kaisersberg	Austria	amorph	95.03	45.76	4.35	1351.96	1584.45	0.49	53.86
fit_2A_14	2A		Europe	Kaisersberg	Austria	amorph	101.21	35.65	5.62	1345.61	1573.01	0.36	36.17
fit_2A_15	2A		Europe	Kaisersberg	Austria	amorph	68.14	79.20	2.68	1351.96	1581.91	1.15	44.27
fit_2A_16	2A		Europe	Kaisersberg	Austria	amorph	65.44	76.69	2.44	1346.88	1581.91	1.17	42.81
fit_2A_17	2A		Europe	Kaisersberg	Austria	amorph	90.16	63.01	3.56	1344.34	1583.18	0.70	51.53
fit_2A_18	2A		Europe	Kaisersberg	Austria	amorph	71.12	51.42	3.93	1354.50	1584.45	0.71	42.94
fit_2A_19	2A		Europe	Kaisersberg	Austria	amorph	68.41	50.09	4.49	1346.88	1578.09	0.73	33.87
fit_2A_20	2A		Europe	Kaisersberg	Austria	amorph	94.26	49.14	4.14	1345.61	1576.82	0.52	46.26
fit_2B_01	2B		Europe	Skaland	Norway	flake	265.95	15.54	14.84	1350.56	1579.26	0.06	38.21
fit_2B_02	2B		Europe	Skaland	Norway	flake	207.30	24.19	9.98	1342.93	1576.72	0.12	40.83

fit_2B_03	2B		Europe	Skaland	Norway	flake	246.80	17.32	13.05	1355.64	1579.26	0.07	50.17
fit_2B_04	2B		Europe	Skaland	Norway	flake	198.17	21.03	11.39	1353.10	1580.54	0.11	50.53
fit_2B_05	2B		Europe	Skaland	Norway	flake	216.83	17.33	13.48	1349.29	1580.54	0.08	46.48
fit_2B_06	2B		Europe	Skaland	Norway	flake	5319.78	12.94	21.94	1346.75	1580.54	0.00	63.86
fit_2B_07	2B		Europe	Skaland	Norway	flake	165.89	22.04	10.14	1350.56	1580.54	0.13	46.36
fit_2B_08	2B		Europe	Skaland	Norway	flake	406.92	16.26	17.79	1342.93	1579.26	0.04	42.27
fit_2B_09	2B		Europe	Skaland	Norway	flake	250.01	16.93	13.80	1355.64	1581.81	0.06	46.47
fit_2B_10	2B		Europe	Skaland	Norway	flake	562.42	14.36	19.44	1349.29	1581.81	0.03	45.13
fit_2B_11	2B		Europe	Skaland	Norway	flake	705.42	13.74	21.52	1350.56	1579.26	0.02	46.90
fit_2B_12	2B		Europe	Skaland	Norway	flake	711.23	15.17	18.10	1350.56	1581.81	0.02	53.02
fit_2B_13	2B		Europe	Skaland	Norway	flake	188.22	19.43	11.49	1346.75	1583.08	0.10	46.92
fit_2B_14	2B		Europe	Skaland	Norway	flake	503.12	14.67	19.85	1349.29	1580.54	0.03	46.46
fit_2B_15	2B		Europe	Skaland	Norway	flake	409.07	13.99	16.84	1341.66	1574.18	0.04	35.00
fit_2B_16	2B		Europe	Skaland	Norway	flake	111.80	28.53	6.28	1348.02	1581.81	0.26	47.20
fit_2B_17	2B		Europe	Skaland	Norway	flake	180.02	20.42	11.01	1349.29	1583.08	0.12	44.04
fit_2B_18	2B		Europe	Skaland	Norway	flake	511.70	14.51	19.65	1353.10	1581.81	0.03	48.20
fit_2B_19	2B		Europe	Skaland	Norway	flake	609.59	13.57	13.49	1346.75	1574.18	0.02	47.06
fit_2B_20	2B		Europe	Skaland	Norway	flake	5769.93	13.07	22.33	1345.48	1581.81	0.00	64.49
fit_3B_01	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	647.56	13.37	22.09	1351.84	1581.81	0.02	47.90
fit_3B_02	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5607.04	12.32	23.12	1353.11	1581.81	0.00	61.95
fit_3B_03	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5203.10	12.74	22.49	1326.42	1581.81	0.00	53.00
fit_3B_04	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5343.88	12.50	22.80	1345.48	1581.81	0.00	63.75

fit_3B_05	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5500.60	12.38	22.52	1355.65	1581.81	0.00	58.37
fit_3B_06	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	737.24	13.33	21.90	1342.94	1580.54	0.02	43.99
fit_3B_07	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5552.74	12.86	23.11	1351.84	1580.54	0.00	63.98
fit_3B_08	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5556.50	12.69	23.43	1354.38	1581.81	0.00	63.68
fit_3B_09	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	402.43	13.60	22.51	1346.75	1580.54	0.03	34.48
fit_3B_10	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	1321.39	12.79	22.63	1353.11	1581.81	0.01	56.65
fit_3B_11	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5542.16	12.72	22.91	1436.97	1580.54	0.00	64.03
fit_3B_12	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	331.55	14.15	21.39	1345.48	1581.81	0.04	39.50
fit_3B_13	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	628.53	13.41	21.69	1348.02	1581.81	0.02	48.78
fit_3B_14	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	206.72	16.42	14.76	1353.11	1581.81	0.08	37.56
fit_3B_15	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	599.11	14.38	19.92	1346.75	1581.81	0.02	45.23
fit_3B_16	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	848.57	14.08	20.23	1350.56	1580.54	0.02	58.28
fit_3B_17	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	297.62	15.19	15.90	1344.21	1580.54	0.05	42.48
fit_3B_18	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	153.40	17.32	13.13	1351.84	1581.81	0.11	39.29
fit_3B_19	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	213.35	16.31	15.28	1350.56	1581.81	0.08	39.31
fit_3B_20	3B		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5302.04	12.90	22.42	1339.13	1581.81	0.00	63.75
fit_4A_01	4A		Asia	Hunan Lutang	China	flake	175.35	29.43	9.85	1344.34	1578.11	0.17	58.34
fit_4A_02	4A		Asia	Hunan Lutang	China	flake	174.30	33.36	6.59	1351.96	1579.38	0.20	46.16
fit_4A_03	4A		Asia	Hunan Lutang	China	flake	99.70	168.38	1.65	1345.61	1590.81	1.69	79.12
fit_4A_04	4A		Asia	Hunan Lutang	China	flake	118.54	124.65	2.05	1344.34	1573.03	1.05	87.57
fit_4A_05	4A		Asia	Hunan Lutang	China	flake	79.85	55.19	3.50	1348.15	1581.92	0.69	46.90
fit_4A_06	4A		Asia	Hunan Lutang	China	flake	116.73	32.80	6.58	1349.42	1576.84	0.28	54.56

fit_4A_07	4A		Asia	Hunan Lutang	China	flake	234.60	19.69	14.64	1344.34	1579.38	0.08	47.62
fit_4A_08	4A		Asia	Hunan Lutang	China	flake	101.24	36.40	5.60	1353.24	1579.38	0.36	46.88
fit_4A_09	4A		Asia	Hunan Lutang	China	flake	96.81	38.99	5.08	1350.69	1580.65	0.41	44.39
fit_4A_10	4A		Asia	Hunan Lutang	China	flake	120.25	26.70	7.98	1351.96	1578.11	0.23	44.23
fit_4A_11	4A		Asia	Hunan Lutang	China	flake	66.56	168.67	1.78	1345.61	1578.11	2.57	51.05
fit_4A_12	4A		Asia	Hunan Lutang	China	flake	95.25	42.04	4.99	1349.42	1579.38	0.45	49.22
fit_4A_13	4A		Asia	Hunan Lutang	China	flake	69.21	80.47	2.91	1345.61	1579.38	1.16	46.90
fit_4A_14	4A		Asia	Hunan Lutang	China	flake	312.62	19.88	8.70	1345.61	1570.48	0.07	40.19
fit_4A_15	4A		Asia	Hunan Lutang	China	flake	106.04	164.59	1.56	1348.15	1588.27	1.55	87.41
fit_4A_16	4A		Asia	Hunan Lutang	China	flake	267.31	18.72	15.73	1351.96	1575.57	0.07	44.65
fit_4A_17	4A		Asia	Hunan Lutang	China	flake	92.80	40.94	5.43	1348.15	1579.38	0.44	43.94
fit_4A_18	4A		Asia	Hunan Lutang	China	flake	485.75	16.63	7.76	1348.15	1567.94	0.03	42.43
fit_4A_19	4A		Asia	Hunan Lutang	China	flake	66.20	151.97	1.50	1353.24	1584.46	2.35	63.56
fit_4A_20	4A		Asia	Hunan Lutang	China	flake	68.76	175.66	1.39	1354.51	1588.27	2.53	65.16
fit_4B_01	4B		Europe	Zavalje	Ukraine	flake	5203.91	13.29	21.71	1355.76	1579.36	0.00	63.74
fit_4B_02	4B		Europe	Zavalje	Ukraine	flake	5836.08	12.71	22.70	1351.95	1581.90	0.00	64.56
fit_4B_03	4B		Europe	Zavalje	Ukraine	flake	4904.44	14.47	19.50	1350.68	1580.63	0.00	64.13
fit_4B_04	4B		Europe	Zavalje	Ukraine	flake	527.61	14.06	20.51	1344.32	1581.90	0.03	40.09
fit_4B_05	4B		Europe	Zavalje	Ukraine	flake	5902.30	13.99	23.70	1329.92	1581.90	0.00	65.10
fit_4B_06	4B		Europe	Zavalje	Ukraine	flake	5562.82	12.85	22.78	1348.13	1580.63	0.00	64.30
fit_4B_07	4B		Europe	Zavalje	Ukraine	flake	791.80	13.36	21.60	1349.40	1580.63	0.02	49.02
fit_4B_08	4B		Europe	Zavalje	Ukraine	flake	917.68	13.65	20.61	1353.22	1581.90	0.02	55.41

fit_4B_09	4B		Europe	Zavalje	Ukraine	flake	782.76	13.60	20.72	1348.13	1581.90	0.02	63.45
fit_4B_10	4B		Europe	Zavalje	Ukraine	flake	5640.67	12.82	22.56	1339.24	1581.90	0.00	64.84
fit_4B_11	4B		Europe	Zavalje	Ukraine	flake	5518.19	13.08	21.63	1354.49	1580.63	0.00	64.22
fit_4B_12	4B		Europe	Zavalje	Ukraine	flake	731.75	14.07	19.91	1354.49	1580.63	0.02	63.24
fit_4B_13	4B		Europe	Zavalje	Ukraine	flake	5748.87	13.19	21.76	1339.24	1580.63	0.00	64.09
fit_4B_14	4B		Europe	Zavalje	Ukraine	flake	2432.34	13.47	21.79	1343.05	1580.63	0.01	61.20
fit_4B_15	4B		Europe	Zavalje	Ukraine	flake	5587.14	13.03	22.34	1355.76	1581.90	0.00	64.76
fit_4B_16	4B		Europe	Zavalje	Ukraine	flake	5355.74	13.77	20.85	1392.60	1583.17	0.00	64.77
fit_4B_17	4B		Europe	Zavalje	Ukraine	flake	429.99	14.42	19.96	1353.22	1581.90	0.03	44.05
fit_4B_18	4B		Europe	Zavalje	Ukraine	flake	5559.42	13.77	20.65	1346.86	1581.90	0.00	65.37
fit_4B_19	4B		Europe	Zavalje	Ukraine	flake	5831.73	12.90	22.31	1411.66	1580.63	0.00	61.75
fit_5B_01	5B		Europe	Ural, Tayginka	Russia	flake	6472.21	14.02	26.25	1355.75	1581.91	0.00	65.19
fit_5B_02	5B		Europe	Ural, Tayginka	Russia	flake	5885.91	14.37	23.89	1353.21	1581.91	0.00	64.78
fit_5B_03	5B		Europe	Ural, Tayginka	Russia	flake	5061.00	13.52	22.74	1442.15	1580.64	0.00	62.61
fit_5B_04	5B		Europe	Ural, Tayginka	Russia	flake	4439.57	14.65	24.35	1368.46	1581.91	0.01	63.00
fit_5B_05	5B		Europe	Ural, Tayginka	Russia	flake	5943.28	13.86	24.39	1418.01	1581.91	0.00	64.71
fit_5B_06	5B		Europe	Ural, Tayginka	Russia	flake	5261.25	12.84	22.85	1367.19	1580.64	0.00	63.40
fit_5B_07	5B		Europe	Ural, Tayginka	Russia	flake	7189.48	14.26	30.29	1351.94	1580.64	0.00	64.69
fit_5B_08	5B		Europe	Ural, Tayginka	Russia	flake	6329.63	14.25	26.82	1328.65	1581.91	0.00	64.14
fit_5B_09	5B		Europe	Ural, Tayginka	Russia	flake	5115.91	12.98	22.01	1387.94	1580.64	0.00	63.61
fit_5B_10	5B		Europe	Ural, Tayginka	Russia	flake	304.32	14.38	21.37	1367.19	1580.64	0.04	54.94
fit_5B_11	5B		Europe	Ural, Tayginka	Russia	flake	225.13	16.27	19.46	1362.11	1580.64	0.07	60.96

fit_5B_1 2	5B		Europe	Ural, Tayginka	Russia	flake	210.08	15.51	20.45	1362.11	1580.64	0.07	55.98
fit_5B_1 3	5B		Europe	Ural, Tayginka	Russia	flake	214.23	16.11	19.22	1364.65	1581.91	0.07	57.38
fit_5B_1 4	5B		Europe	Ural, Tayginka	Russia	flake	6019.95	14.37	24.89	1346.86	1580.64	0.00	64.47
fit_5B_1 5	5B		Europe	Ural, Tayginka	Russia	flake	5669.80	15.46	23.05	1346.86	1581.91	0.00	64.35
fit_5B_1 6	5B		Europe	Ural, Tayginka	Russia	flake	6559.55	15.72	26.88	1350.67	1580.64	0.00	64.29
fit_5B_1 7	5B		Europe	Ural, Tayginka	Russia	flake	5721.52	14.46	23.18	1397.68	1581.91	0.00	64.71
fit_5B_1 8	5B		Europe	Ural, Tayginka	Russia	flake	5750.80	16.30	22.10	1353.21	1581.91	0.00	65.04
fit_5B_1 9	5B		Europe	Ural, Tayginka	Russia	flake	5771.94	14.32	23.57	1351.94	1580.64	0.00	64.59
fit_5B_2 0	5B		Europe	Ural, Tayginka	Russia	flake	6505.53	13.95	27.08	1343.05	1581.91	0.00	64.79
fit_6A_0 1	6A		Europe	Kaisersberg	Austria	amorph	179.43	23.02	9.42	1355.77	1575.56	0.12	49.43
fit_6A_0 2	6A		Europe	Kaisersberg	Austria	amorph	161.49	30.97	5.95	1351.96	1585.72	0.19	57.53
fit_6A_0 3	6A		Europe	Kaisersberg	Austria	amorph	248.28	22.20	9.40	1348.15	1583.18	0.08	66.69
fit_6A_0 4	6A		Europe	Kaisersberg	Austria	amorph	166.14	31.95	6.09	1358.31	1584.45	0.19	61.56
fit_6A_0 5	6A		Europe	Kaisersberg	Austria	amorph	151.75	33.18	6.48	1349.42	1581.91	0.22	52.94
fit_6A_0 6	6A		Europe	Kaisersberg	Austria	amorph	85.77	70.01	2.59	1354.50	1585.72	0.82	64.14
fit_6A_0 7	6A		Europe	Kaisersberg	Austria	amorph	122.69	31.21	7.01	1353.23	1583.18	0.26	54.66
fit_6A_0 8	6A		Europe	Kaisersberg	Austria	amorph	116.34	34.98	5.73	1348.15	1585.72	0.29	57.88
fit_6A_0 9	6A		Europe	Kaisersberg	Austria	amorph	151.43	27.37	7.66	1353.23	1583.18	0.18	56.84
fit_6A_1 0	6A		Europe	Kaisersberg	Austria	amorph	159.22	30.22	7.74	1350.69	1579.37	0.19	55.78
fit_6A_1 1	6A		Europe	Kaisersberg	Austria	amorph	180.75	33.59	6.55	1357.04	1585.72	0.19	71.52
fit_6A_1 2	6A		Europe	Kaisersberg	Austria	amorph	172.39	34.11	6.46	1350.69	1585.72	0.19	68.57
fit_6A_1 3	6A		Europe	Kaisersberg	Austria	amorph	134.20	23.47	8.76	1351.96	1581.91	0.18	47.43

fit_6A_14	6A		Europe	Kaisersberg	Austria	amorph	104.72	44.09	4.22	1349.42	1586.99	0.41	62.08
fit_6A_15	6A		Europe	Kaisersberg	Austria	amorph	117.08	45.06	3.63	1358.31	1584.45	0.39	72.24
fit_6B_01	6B		Asia	Inner Mongolia	China	flake	785.62	13.24	21.10	1342.94	1581.81	0.02	47.24
fit_6B_02	6B		Asia	Inner Mongolia	China	flake	306.04	15.09	16.05	1351.83	1581.81	0.05	41.01
fit_6B_03	6B		Asia	Inner Mongolia	China	flake	5359.13	12.63	22.24	1334.47	1581.81	0.00	64.56
fit_6B_04	6B		Asia	Inner Mongolia	China	flake	313.15	15.35	15.51	1342.94	1580.54	0.05	45.47
fit_6B_05	6B		Asia	Inner Mongolia	China	flake	1019.44	12.84	22.27	1355.64	1581.81	0.01	52.82
fit_6B_06	6B		Asia	Inner Mongolia	China	flake	5368.85	13.23	20.30	1391.22	1581.81	0.00	65.08
fit_6B_07	6B		Asia	Inner Mongolia	China	flake	721.21	13.53	21.25	1350.56	1581.81	0.02	49.18
fit_6B_08	6B		Asia	Inner Mongolia	China	flake	845.05	13.39	21.28	1351.83	1580.54	0.02	49.89
fit_6B_09	6B		Asia	Inner Mongolia	China	flake	431.44	16.77	16.52	1351.83	1581.81	0.04	46.97
fit_6B_10	6B		Asia	Inner Mongolia	China	flake	764.33	13.42	21.30	1348.02	1581.81	0.02	52.60
fit_6B_11	6B		Asia	Inner Mongolia	China	flake	1149.79	13.26	23.32	1348.02	1580.54	0.01	58.70
fit_6B_12	6B		Asia	Inner Mongolia	China	flake	1182.81	13.27	21.19	1348.02	1581.81	0.01	57.30
fit_7B_01	7B		Asia	Heilongjian	China	flake	5475.33	13.58	20.71	1355.35	1581.92	0.00	65.48
fit_7B_02	7B		Asia	Heilongjian	China	flake	261.55	15.90	18.10	1348.15	1581.92	0.06	42.59
fit_7B_03	7B		Asia	Heilongjian	China	flake	5307.25	12.86	23.33	1367.21	1579.38	0.00	62.55
fit_7B_04	7B		Asia	Heilongjian	China	flake	5344.68	14.42	20.27	1362.13	1583.19	0.00	65.43
fit_7B_05	7B		Asia	Heilongjian	China	flake	5559.66	14.65	21.13	1335.45	1581.92	0.00	65.56
fit_7B_06	7B		Asia	Heilongjian	China	flake	3991.69	16.36	19.74	1348.15	1584.46	0.02	60.05
fit_7B_07	7B		Asia	Heilongjian	China	flake	214.55	17.54	16.10	1353.24	1583.19	0.08	43.17
fit_7B_08	7B		Asia	Heilongjian	China	flake	6417.07	14.25	23.27	1367.21	1583.19	0.00	66.67

fit_7B_09	7B		Asia	Heilongjian	China	flake	4640.72	15.75	16.44	1349.42	1583.19	0.00	65.67
fit_7B_10	7B		Asia	Heilongjian	China	flake	5907.10	13.93	21.12	1339.26	1584.46	0.00	66.55
fit_7B_11	7B		Asia	Heilongjian	China	flake	2302.64	14.05	20.74	1357.05	1583.19	0.02	58.36
fit_7B_12	7B		Asia	Heilongjian	China	flake	5603.36	16.72	20.64	1344.34	1581.92	0.00	65.62
fit_7B_13	7B		Asia	Heilongjian	China	flake	4093.29	14.61	21.27	1360.86	1583.19	0.01	57.98
fit_7B_14	7B		Asia	Heilongjian	China	flake	7435.08	14.22	29.79	1351.96	1581.92	0.00	65.60
fit_7B_15	7B		Asia	Heilongjian	China	flake	6327.30	14.08	25.00	1348.15	1583.19	0.00	65.47
fit_7B_16	7B		Asia	Heilongjian	China	flake	5804.86	14.02	22.74	1350.69	1581.92	0.00	65.37
fit_7B_17	7B		Asia	Heilongjian	China	flake	5834.85	14.28	22.55	1302.42	1581.92	0.00	65.56
fit_7B_18	7B		Asia	Heilongjian	China	flake	5690.03	14.23	21.52	1368.48	1583.19	0.00	65.74
fit_7B_19	7B		Asia	Heilongjian	China	flake	5659.73	14.02	23.06	1337.99	1580.65	0.00	64.58
fit_7B_20	7B		Asia	Heilongjian	China	flake	6106.25	13.33	25.81	1327.83	1580.65	0.00	64.38
fit_8B_01	8B		Asia	Hubei	China	flake	5886.23	13.82	24.08	1331.64	1580.64	0.00	64.69
fit_8B_02	8B		Asia	Hubei	China	flake	5912.76	12.92	25.11	1355.77	1579.37	0.00	63.74
fit_8B_03	8B		Asia	Hubei	China	flake	6307.85	14.88	25.30	1349.42	1581.91	0.00	65.09
fit_8B_04	8B		Asia	Hubei	China	flake	5658.51	13.42	23.07	1355.77	1581.91	0.00	64.29
fit_8B_05	8B		Asia	Hubei	China	flake	6017.05	12.92	24.54	1340.53	1581.91	0.00	64.89
fit_8B_06	8B		Asia	Hubei	China	flake	423.74	15.12	17.02	1346.88	1584.45	0.03	40.64
fit_8B_07	8B		Asia	Hubei	China	flake	291.66	14.68	16.42	1362.13	1584.45	0.05	56.23
fit_8B_08	8B		Asia	Hubei	China	flake	552.78	17.25	12.64	1353.23	1585.72	0.03	59.00
fit_8B_09	8B		Asia	Hubei	China	flake	233.11	14.68	18.82	1359.59	1584.45	0.06	51.01
fit_8B_10	8B		Asia	Hubei	China	flake	255.39	16.67	16.73	1358.31	1584.45	0.07	50.29

fit_8B_1_1	8B		Asia	Hubei	China	flake	268.81	15.14	17.64	1355.51	1584.22	0.05	39.81
fit_8B_1_2	8B		Asia	Hubei	China	flake	5278.67	14.65	18.41	1342.32	1584.22	0.00	66.95
fit_8B_1_3	8B		Asia	Hubei	China	flake	713.33	13.65	20.40	1354.50	1583.18	0.02	58.48
fit_8B_1_4	8B		Asia	Hubei	China	flake	210.63	17.77	15.54	1353.23	1583.18	0.08	45.38
fit_8B_1_5	8B		Asia	Hubei	China	flake	5990.91	12.41	21.35	1350.69	1584.45	0.00	66.48
fit_8B_1_6	8B		Asia	Hubei	China	flake	401.77	13.69	20.03	1346.88	1583.18	0.03	42.41
fit_8B_1_7	8B		Asia	Hubei	China	flake	5621.42	12.72	19.85	1336.72	1584.45	0.00	66.69
fit_8B_1_8	8B		Asia	Hubei	China	flake	5941.03	13.73	21.61	1401.51	1583.18	0.00	66.04
fit_8B_1_9	8B		Asia	Hubei	China	flake	232.04	14.96	18.89	1351.96	1584.45	0.07	37.92
fit_8B_2_0	8B		Asia	Hubei	China	flake	111.10	22.75	8.36	1367.21	1584.45	0.17	57.13
fit_10B_01	10B		Asia	Ragedara	Sri_Lanka	hydro	5412.39	13.15	21.75	1348.15	1581.91	0.00	64.36
fit_10B_02	10B		Asia	Ragedara	Sri_Lanka	hydro	5359.51	13.40	20.98	1345.61	1581.91	0.00	64.95
fit_10B_03	10B		Asia	Ragedara	Sri_Lanka	hydro	5529.59	12.48	23.35	1391.34	1580.64	0.00	63.93
fit_10B_04	10B		Asia	Ragedara	Sri_Lanka	hydro	5357.90	13.11	21.92	1387.53	1581.91	0.00	64.08
fit_10B_05	10B		Asia	Ragedara	Sri_Lanka	hydro	5512.28	12.97	23.01	1369.75	1581.91	0.00	63.96
fit_10B_06	10B		Asia	Ragedara	Sri_Lanka	hydro	5526.65	12.96	22.33	1407.86	1581.91	0.00	64.56
fit_10B_07	10B		Asia	Ragedara	Sri_Lanka	hydro	4650.12	16.56	16.67	1412.94	1581.91	0.00	65.35
fit_10B_08	10B		Asia	Ragedara	Sri_Lanka	hydro	6050.34	13.74	24.75	1348.15	1580.64	0.00	64.75
fit_10B_09	10B		Asia	Ragedara	Sri_Lanka	hydro	5129.71	14.33	19.93	1330.36	1580.64	0.00	64.62
fit_10B_10	10B		Asia	Ragedara	Sri_Lanka	hydro	347.05	16.09	17.16	1351.96	1581.91	0.05	53.53
fit_10B_11	10B		Asia	Ragedara	Sri_Lanka	hydro	147.69	21.22	9.26	1346.88	1581.91	0.14	49.68
fit_10B_12	10B		Asia	Ragedara	Sri_Lanka	hydro	5246.28	13.72	20.38	1329.09	1581.91	0.00	64.52

fit_10B_13	10B		Asia	Ragedara	Sri_Lanka	hydro	5154.06	13.97	19.89	1348.15	1581.91	0.00	64.92
fit_10B_14	10B		Asia	Ragedara	Sri_Lanka	hydro	604.56	17.03	16.27	1343.07	1579.37	0.03	58.90
fit_10B_15	10B		Asia	Ragedara	Sri_Lanka	hydro	5341.27	13.02	21.02	1383.72	1581.91	0.00	64.91
fit_10B_16	10B		Asia	Ragedara	Sri_Lanka	hydro	5485.09	12.97	22.23	1418.02	1580.64	0.00	64.49
fit_10B_17	10B		Asia	Ragedara	Sri_Lanka	hydro	930.49	13.29	21.90	1360.85	1579.37	0.01	64.78
fit_10B_18	10B		Asia	Ragedara	Sri_Lanka	hydro	5709.22	13.05	21.92	1346.88	1581.91	0.00	64.85
fit_10B_19	10B		Asia	Ragedara	Sri_Lanka	hydro	5623.48	12.75	22.78	1355.77	1580.64	0.00	64.50
fit_10B_20	10B		Asia	Ragedara	Sri_Lanka	hydro	5389.95	13.06	21.90	1365.93	1581.91	0.00	50.59
fit_11B_01	11B		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	5271.67	13.81	20.63	1367.19	1583.18	0.00	64.64
fit_11B_02	11B		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	5746.81	12.79	23.78	1320.18	1581.91	0.00	64.10
fit_11B_03	11B		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	2483.15	14.54	22.32	1344.32	1581.91	0.02	52.85
fit_11B_04	11B		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	423.88	16.43	17.49	1350.67	1580.64	0.04	54.78
fit_11B_05	11B		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	5750.12	14.46	23.14	1348.13	1581.91	0.00	64.83
fit_11B_06	11B		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	6414.91	13.93	26.49	1338.39	1580.64	0.00	64.86
fit_11B_07	11B		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	5386.15	13.09	22.33	1346.86	1580.64	0.00	63.93
fit_11B_08	11B		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	5569.88	12.87	22.53	1397.68	1581.91	0.00	64.34
fit_11B_09	11B		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	6208.43	13.95	25.35	1355.75	1581.91	0.00	64.94
fit_11B_10	11B		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	5656.90	13.17	21.61	1337.96	1581.91	0.00	64.58
fit_11B_11	11B		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	5787.78	13.94	23.47	1353.21	1581.91	0.00	64.81
fit_11B_12	11B		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	411.66	14.15	20.88	1351.94	1581.91	0.03	38.85
fit_11B_13	11B		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	5878.82	13.92	24.14	1355.75	1580.64	0.00	64.62
fit_11B_14	11B		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	400.74	14.58	20.05	1344.32	1581.91	0.04	40.90

fit_11B_15	11B		Africa	Vatomina/Sahamamy Sahasoa	Madagas car	flake	6052.74	14.03	24.96	1341.78	1581.91	0.00	64.64
fit_11B_16	11B		Africa	Vatomina/Sahamamy Sahasoa	Madagas car	flake	5784.49	13.65	24.24	1343.05	1579.37	0.00	63.95
fit_11B_17	11B		Africa	Vatomina/Sahamamy Sahasoa	Madagas car	flake	6094.31	14.44	25.13	1343.05	1580.64	0.00	64.57
fit_11B_18	11B		Africa	Vatomina/Sahamamy Sahasoa	Madagas car	flake	6500.81	13.79	27.54	1353.21	1580.64	0.00	64.45
fit_11B_19	11B		Africa	Vatomina/Sahamamy Sahasoa	Madagas car	flake	5796.48	13.87	23.09	1341.78	1581.91	0.00	65.08
fit_11B_20	11B		Africa	Vatomina/Sahamamy Sahasoa	Madagas car	flake	267.57	15.81	19.53	1344.32	1580.64	0.06	43.31
fit_13B_01	13B		Europe	Passau	Germany	flake	618.58	14.52	20.33	1355.63	1580.51	0.02	61.54
fit_13B_02	13B		Europe	Passau	Germany	flake	953.07	13.94	21.32	1350.54	1580.51	0.01	62.54
fit_13B_03	13B		Europe	Passau	Germany	flake	720.81	14.20	21.00	1350.54	1580.51	0.02	53.44
fit_13B_04	13B		Europe	Passau	Germany	flake	808.83	14.33	21.17	1354.35	1579.24	0.02	64.20
fit_13B_05	13B		Europe	Passau	Germany	flake	697.16	13.48	23.33	1354.35	1580.51	0.02	55.23
fit_13B_06	13B		Europe	Passau	Germany	flake	5719.31	12.96	23.17	1344.19	1580.51	0.00	63.49
fit_13B_07	13B		Europe	Passau	Germany	flake	1269.19	13.19	24.32	1351.81	1580.51	0.01	69.99
fit_13B_08	13B		Europe	Passau	Germany	flake	5478.12	12.71	22.50	1344.19	1580.51	0.00	64.11
fit_13B_09	13B		Europe	Passau	Germany	flake	5656.29	13.09	23.22	1342.92	1577.97	0.00	63.22
fit_13B_10	13B		Europe	Passau	Germany	flake	5360.16	13.39	21.92	1355.63	1580.51	0.00	64.07
fit_13B_11	13B		Europe	Passau	Germany	flake	5501.26	12.65	24.16	1306.07	1579.24	0.00	62.86
fit_13B_12	13B		Europe	Passau	Germany	flake	714.46	13.71	22.27	1349.27	1580.51	0.02	57.61
fit_13B_13	13B		Europe	Passau	Germany	flake	656.73	13.68	21.57	1353.08	1580.51	0.02	55.10
fit_13B_14	13B		Europe	Passau	Germany	flake	1171.13	13.59	22.33	1349.27	1580.51	0.01	67.31
fit_13B_15	13B		Europe	Passau	Germany	flake	2550.21	13.10	22.61	1355.63	1580.51	0.01	63.58
fit_13B_16	13B		Europe	Passau	Germany	flake	514.52	13.72	22.10	1354.35	1581.79	0.03	48.64

fit_13B_17	13B		Europe	Passau	Germany	flake	5562.31	12.75	23.67	1355.63	1580.51	0.00	63.47
fit_13B_18	13B		Europe	Passau	Germany	flake	5460.77	12.77	23.01	1345.46	1580.51	0.00	63.38
fit_13B_19	13B		Europe	Passau	Germany	flake	5694.01	12.66	23.92	1340.38	1580.51	0.00	63.77
fit_13B_20	13B		Europe	Passau	Germany	flake	953.52	13.59	23.66	1355.63	1580.51	0.01	59.67
fit_14B_01	14B		Europe	Passau	Germany	flake	5781.37	15.49	22.17	1355.78	1581.93	0.00	65.40
fit_14B_02	14B		Europe	Passau	Germany	flake	216.17	15.61	13.85	1359.59	1581.93	0.07	45.11
fit_14B_03	14B		Europe	Passau	Germany	flake	6220.87	16.62	23.25	1343.07	1581.93	0.00	65.56
fit_14B_04	14B		Europe	Passau	Germany	flake	195.65	18.86	15.70	1348.15	1581.93	0.09	46.52
fit_14B_05	14B		Europe	Passau	Germany	flake	135.27	18.55	11.67	1355.78	1581.93	0.14	45.83
fit_14B_06	14B		Europe	Passau	Germany	flake	5662.37	15.63	21.21	1433.28	1581.93	0.00	65.68
fit_14B_07	14B		Europe	Passau	Germany	flake	6112.49	13.97	25.26	1346.88	1581.93	0.00	64.68
fit_14B_08	14B		Europe	Passau	Germany	flake	5495.49	13.49	20.63	1357.05	1583.20	0.00	65.62
fit_14B_09	14B		Europe	Passau	Germany	flake	6807.43	13.98	27.48	1343.92	1583.20	0.00	65.40
fit_14B_10	14B		Europe	Passau	Germany	flake	367.58	19.03	14.07	1344.34	1580.66	0.05	53.91
fit_14B_11	14B		Europe	Passau	Germany	flake	350.60	15.79	18.30	1355.78	1580.66	0.04	41.80
fit_14B_12	14B		Europe	Passau	Germany	flake	7215.66	15.27	29.24	1351.96	1581.93	0.00	65.02
fit_14B_13	14B		Europe	Passau	Germany	flake	4949.73	15.00	19.08	1334.18	1581.93	0.00	48.52
fit_14B_14	14B		Europe	Passau	Germany	flake	243.80	15.85	19.12	1344.34	1581.93	0.06	40.90
fit_14B_15	14B		Europe	Passau	Germany	flake	6536.51	14.17	26.17	1340.53	1580.66	0.00	65.36
fit_14B_16	14B		Europe	Passau	Germany	flake	6852.26	14.02	29.16	1365.94	1580.66	0.00	64.51
fit_14B_17	14B		Europe	Passau	Germany	flake	6893.79	14.61	28.29	1339.26	1580.66	0.00	64.95
fit_14B_18	14B		Europe	Passau	Germany	flake	7055.48	14.21	29.79	1380.34	1581.93	0.00	64.69

fit_14B_19	14B		Europe	Passau	Germany	flake	6422.57	14.14	26.44	1348.15	1581.93	0.00	64.86
fit_14B_20	14B		Europe	Passau	Germany	flake	4737.21	13.91	21.15	1415.49	1578.12	0.00	61.97
fit_15B_01	15B		Europe	Skaland	Norway	flake	2124.65	15.59	18.49	1350.69	1579.36	0.01	59.80
fit_15B_02	15B		Europe	Skaland	Norway	flake	297.12	16.85	17.43	1351.96	1581.91	0.06	50.78
fit_15B_03	15B		Europe	Skaland	Norway	flake	823.43	16.68	15.92	1340.53	1576.82	0.02	63.03
fit_15B_04	15B		Europe	Skaland	Norway	flake	269.44	17.55	16.70	1348.15	1580.63	0.07	49.87
fit_15B_05	15B		Europe	Skaland	Norway	flake	209.30	16.77	15.02	1343.07	1580.63	0.07	44.45
fit_15B_06	15B		Europe	Skaland	Norway	flake	260.17	18.63	12.80	1350.69	1581.91	0.07	52.59
fit_15B_07	15B		Europe	Skaland	Norway	flake	486.50	13.61	22.21	1344.34	1579.36	0.03	46.92
fit_15B_08	15B		Europe	Skaland	Norway	flake	5197.45	14.06	20.81	1357.04	1580.63	0.00	64.01
fit_15B_09	15B		Europe	Skaland	Norway	flake	5563.67	12.87	23.29	1343.07	1580.63	0.00	63.35
fit_15B_10	15B		Europe	Skaland	Norway	flake	5552.93	12.89	22.61	1351.96	1580.63	0.00	64.40
fit_15B_11	15B		Europe	Skaland	Norway	flake	5216.54	12.99	22.32	1346.88	1578.09	0.00	61.60
fit_15B_12	15B		Europe	Skaland	Norway	flake	422.28	15.07	19.44	1349.42	1580.63	0.04	48.50
fit_15B_13	15B		Europe	Skaland	Norway	flake	315.47	15.48	16.36	1349.42	1580.63	0.05	48.94
fit_15B_14	15B		Europe	Skaland	Norway	flake	5635.38	12.62	23.70	1368.48	1579.36	0.00	62.71
fit_15B_15	15B		Europe	Skaland	Norway	flake	5569.80	13.14	22.73	1359.58	1579.36	0.00	62.17
fit_15B_16	15B		Europe	Skaland	Norway	flake	346.37	18.59	15.07	1351.96	1579.36	0.06	54.33
fit_15B_17	15B		Europe	Skaland	Norway	flake	287.29	18.66	15.66	1346.88	1579.36	0.07	50.72
fit_15B_18	15B		Europe	Skaland	Norway	flake	808.75	13.34	21.77	1350.69	1579.36	0.02	44.64
fit_15B_19	15B		Europe	Skaland	Norway	flake	820.53	13.51	18.43	1348.15	1574.28	0.02	50.61
fit_15B_20	15B		Europe	Skaland	Norway	flake	5424.00	13.52	21.85	1344.34	1579.36	0.00	63.66

fit_16B_01	16B		Europe	Skaland	Norway	flake	5493.52	15.56	20.58	1379.93	1580.67	0.00	65.63
fit_16B_02	16B		Europe	Skaland	Norway	flake	5037.10	15.51	21.13	1348.17	1578.13	0.00	61.39
fit_16B_03	16B		Europe	Skaland	Norway	flake	5185.24	17.44	19.91	1362.14	1579.40	0.00	64.49
fit_16B_04	16B		Europe	Skaland	Norway	flake	5635.51	16.22	22.24	1345.63	1581.94	0.00	64.77
fit_16B_05	16B		Europe	Skaland	Norway	flake	217.78	20.18	14.46	1351.98	1580.67	0.09	54.01
fit_16B_06	16B		Europe	Skaland	Norway	flake	4760.99	13.90	20.33	1390.09	1575.59	0.00	60.48
fit_16B_07	16B		Europe	Skaland	Norway	flake	5522.36	17.38	20.35	1346.90	1581.94	0.00	65.32
fit_16B_08	16B		Europe	Skaland	Norway	flake	4705.34	18.20	17.00	1346.90	1583.21	0.00	65.12
fit_16B_09	16B		Europe	Skaland	Norway	flake	6206.85	16.44	23.74	1344.36	1583.21	0.00	65.28
fit_16B_10	16B		Europe	Skaland	Norway	flake	172.70	19.59	15.79	1350.71	1580.67	0.11	50.29
fit_16B_11	16B		Europe	Skaland	Norway	flake	250.36	19.94	13.98	1355.79	1583.21	0.08	58.78
fit_16B_12	16B		Europe	Skaland	Norway	flake	4758.13	17.87	17.11	1354.52	1583.21	0.00	65.27
fit_16B_13	16B		Europe	Skaland	Norway	flake	198.11	16.73	19.70	1351.98	1580.67	0.08	50.00
fit_16B_14	16B		Europe	Skaland	Norway	flake	5912.51	16.09	22.83	1351.98	1581.94	0.00	65.18
fit_16B_15	16B		Europe	Skaland	Norway	flake	441.34	15.32	18.12	1357.06	1581.94	0.03	58.42
fit_16B_16	16B		Europe	Skaland	Norway	flake	5107.03	13.76	19.66	1391.36	1580.67	0.00	64.95
fit_16B_17	16B		Europe	Skaland	Norway	flake	5934.74	14.59	23.53	1387.55	1581.94	0.00	65.16
fit_16B_18	16B		Europe	Skaland	Norway	flake	6192.21	14.38	24.55	1350.71	1581.94	0.00	65.30
fit_16B_19	16B		Europe	Skaland	Norway	flake	291.10	16.15	17.85	1351.98	1581.94	0.06	42.25
fit_16B_20	16B		Europe	Skaland	Norway	flake	306.62	15.25	19.30	1344.36	1581.94	0.04	40.91
fit_21A_01	21A		Asia	Shandong	China	flake	5363.33	12.82	22.75	1348.16	1580.65	0.00	63.34
fit_21A_02	21A		Asia	Shandong	China	flake	335.45	14.63	20.58	1351.97	1580.65	0.04	44.27

fit_21A_03	21A		Asia	Shandong	China	flake	401.68	15.26	17.76	1348.16	1580.65	0.04	47.11
fit_21A_04	21A		Asia	Shandong	China	flake	307.45	16.13	15.58	1349.43	1579.38	0.05	46.19
fit_21A_05	21A		Asia	Shandong	China	flake	1213.65	12.95	21.59	1348.16	1581.92	0.01	58.24
fit_21A_06	21A		Asia	Shandong	China	flake	395.47	16.96	17.15	1349.43	1581.92	0.04	54.37
fit_21A_07	21A		Asia	Shandong	China	flake	485.52	13.49	22.26	1349.43	1581.92	0.03	42.59
fit_21A_08	21A		Asia	Shandong	China	flake	5196.58	13.94	20.15	1329.11	1579.38	0.00	64.08
fit_21A_09	21A		Asia	Shandong	China	flake	354.05	15.72	18.62	1353.24	1580.65	0.04	49.46
fit_21A_10	21A		Asia	Shandong	China	flake	360.58	15.61	18.77	1348.16	1581.92	0.04	43.96
fit_21A_11	21A		Asia	Shandong	China	flake	5597.29	12.76	22.97	1411.68	1581.92	0.00	64.00
fit_21A_12	21A		Asia	Shandong	China	flake	444.78	13.93	22.80	1340.54	1580.65	0.03	42.60
fit_21A_13	21A		Asia	Shandong	China	flake	5441.70	12.77	22.79	1340.54	1580.65	0.00	63.68
fit_21A_14	21A		Asia	Shandong	China	flake	493.96	13.93	20.80	1351.97	1580.65	0.03	46.23
fit_21A_15	21A		Asia	Shandong	China	flake	362.23	16.12	18.00	1350.70	1580.65	0.04	48.02
fit_21A_16	21A		Asia	Shandong	China	flake	5615.81	13.50	20.90	1339.27	1583.19	0.00	65.18
fit_21A_17	21A		Asia	Shandong	China	flake	421.39	15.35	18.94	1345.62	1581.92	0.04	46.45
fit_21A_18	21A		Asia	Shandong	China	flake	298.40	15.21	16.24	1351.97	1581.92	0.05	45.66
fit_21A_19	21A		Asia	Shandong	China	flake	5151.30	12.79	22.62	1414.22	1580.65	0.00	62.86
fit_21A_20	21A		Asia	Shandong	China	flake	461.64	14.84	19.11	1349.43	1580.65	0.03	52.95
fit_21B_01	21B		Africa	Balama	Mozambique	flake	460.14	14.69	19.93	1354.49	1581.91	0.03	49.33
fit_21B_02	21B		Africa	Balama	Mozambique	flake	288.03	17.72	16.31	1353.22	1579.37	0.06	47.61
fit_21B_03	21B		Africa	Balama	Mozambique	flake	464.93	14.76	19.75	1346.87	1581.91	0.03	45.85
fit_21B_04	21B		Africa	Balama	Mozambique	flake	5358.70	13.51	22.96	1346.87	1581.91	0.00	63.34

fit_21B_05	21B		Africa	Balama	Mozambique	flake	451.02	14.36	21.36	1343.06	1580.64	0.03	46.36
fit_21B_06	21B		Africa	Balama	Mozambique	flake	351.28	16.77	17.09	1350.68	1581.91	0.05	46.29
fit_21B_07	21B		Africa	Balama	Mozambique	flake	628.07	13.73	21.17	1351.95	1580.64	0.02	50.55
fit_21B_08	21B		Africa	Balama	Mozambique	flake	5678.50	13.34	22.44	1345.60	1581.91	0.00	64.42
fit_21B_09	21B		Africa	Balama	Mozambique	flake	5381.74	13.24	21.95	1357.04	1581.91	0.00	64.12
fit_21B_10	21B		Africa	Balama	Mozambique	flake	316.25	14.55	20.90	1345.60	1581.91	0.05	39.63
fit_21B_11	21B		Africa	Balama	Mozambique	flake	662.35	14.11	20.03	1341.79	1581.91	0.02	43.53
fit_21B_12	21B		Africa	Balama	Mozambique	flake	5457.96	13.40	22.34	1350.68	1581.91	0.00	64.25
fit_21B_13	21B		Africa	Balama	Mozambique	flake	5722.88	13.28	22.72	1327.81	1580.64	0.00	63.96
fit_21B_14	21B		Africa	Balama	Mozambique	flake	5797.94	13.77	24.16	1383.72	1579.37	0.00	64.08
fit_21B_15	21B		Africa	Balama	Mozambique	flake	5582.49	13.60	22.63	1355.77	1581.91	0.00	64.50
fit_21B_16	21B		Africa	Balama	Mozambique	flake	2091.64	15.77	20.11	1348.14	1580.64	0.04	54.57
fit_21B_17	21B		Africa	Balama	Mozambique	flake	357.38	15.25	18.78	1351.95	1581.91	0.04	38.45
fit_21B_18	21B		Africa	Balama	Mozambique	flake	5474.78	12.66	23.43	1350.68	1579.37	0.00	63.07
fit_21B_19	21B		Africa	Balama	Mozambique	flake	277.45	16.82	14.35	1351.10	1582.75	0.06	45.91
fit_21B_20	21B		Africa	Balama	Mozambique	flake	456.70	14.32	20.52	1348.17	1581.28	0.03	51.09
fit_21C_01	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	499.70	15.19	18.17	1340.54	1580.65	0.03	50.50
fit_21C_02	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	468.90	15.76	17.99	1343.08	1581.92	0.03	50.75
fit_21C_03	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	886.08	13.07	22.33	1348.16	1579.38	0.02	59.92
fit_21C_04	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	530.33	13.71	21.98	1354.51	1580.65	0.03	49.54
fit_21C_05	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	1049.02	13.72	21.22	1344.35	1580.65	0.01	65.54
fit_21C_06	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	537.89	13.53	21.75	1345.62	1579.38	0.03	45.09

fit_21C_07	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	523.78	14.47	19.68	1353.24	1580.65	0.03	55.75
fit_21C_08	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	805.26	13.54	20.55	1351.97	1579.38	0.02	55.43
fit_21C_09	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	327.26	16.16	19.02	1346.89	1579.38	0.05	46.74
fit_21C_10	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5369.20	12.94	22.23	1341.81	1580.65	0.00	63.86
fit_21C_11	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5295.61	12.93	22.50	1348.16	1580.65	0.00	63.38
fit_21C_12	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	506.33	14.07	20.87	1343.08	1580.65	0.03	47.73
fit_21C_13	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	306.17	14.76	20.74	1348.16	1580.65	0.05	39.74
fit_21C_14	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	1302.24	12.76	22.73	1349.43	1581.92	0.01	67.62
fit_21C_15	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5434.33	13.23	21.93	1338.00	1581.92	0.00	64.28
fit_21C_16	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	609.18	13.40	22.17	1344.35	1580.65	0.02	46.43
fit_21C_17	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	912.25	13.45	21.82	1348.16	1580.65	0.02	60.15
fit_21C_18	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5709.08	12.54	23.29	1344.35	1580.65	0.00	64.05
fit_21C_19	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5849.67	12.31	23.83	1351.97	1581.92	0.00	64.24
fit_21C_20	21C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	1041.54	13.50	22.01	1355.78	1581.92	0.01	61.48
fit_21D_01	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagascar	flake	782.44	13.87	21.31	1349.41	1580.64	0.02	54.13
fit_21D_02	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagascar	flake	419.35	14.35	20.39	1344.33	1580.64	0.03	39.29
fit_21D_03	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagascar	flake	206.83	17.86	13.85	1349.41	1578.10	0.08	38.15
fit_21D_04	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagascar	flake	5548.37	13.00	23.27	1339.25	1580.64	0.00	63.49
fit_21D_05	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagascar	flake	547.43	14.56	20.39	1349.41	1580.64	0.03	50.21
fit_21D_06	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagascar	flake	492.79	15.60	18.07	1340.52	1581.91	0.03	48.49
fit_21D_07	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagascar	flake	471.64	14.94	19.33	1346.87	1580.64	0.03	46.43
fit_21D_08	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagascar	flake	2192.74	13.85	20.74	1348.14	1580.64	0.02	53.92

fit_21D_09	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	5396.64	12.88	23.18	1360.85	1581.91	0.00	63.48
fit_21D_10	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	1223.94	13.36	22.13	1343.06	1581.91	0.01	66.32
fit_21D_11	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	346.62	16.60	17.35	1344.33	1580.64	0.05	46.22
fit_21D_12	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	5510.18	12.98	21.65	1357.04	1580.64	0.00	64.86
fit_21D_13	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	139.83	22.26	9.93	1349.41	1580.64	0.16	37.49
fit_21D_14	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	5492.25	13.25	22.16	1352.57	1579.81	0.00	63.59
fit_21D_15	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	612.08	14.79	19.53	1342.30	1581.28	0.02	43.47
fit_21D_16	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	417.10	14.34	21.08	1348.17	1579.81	0.04	37.75
fit_21D_17	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	6004.25	12.95	23.43	1355.50	1581.28	0.00	64.77
fit_21D_18	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	415.30	14.83	20.31	1346.70	1581.28	0.04	42.17
fit_21D_19	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	5397.12	12.74	22.29	1368.69	1581.28	0.00	64.60
fit_21D_20	21D		Africa	Vatolina/Sahamamy Sahasoa	Madagas car	flake	530.93	14.11	21.38	1342.30	1581.28	0.02	50.31
fit_21E_01	21E		Europe	Ural, Tayginka	Russia	flake	5013.00	13.61	20.55	1345.61	1580.64	0.00	63.90
fit_21E_02	21E		Europe	Ural, Tayginka	Russia	flake	5334.41	13.63	20.50	1358.31	1581.91	0.00	64.92
fit_21E_03	21E		Europe	Ural, Tayginka	Russia	flake	825.75	14.00	20.86	1354.50	1580.64	0.02	56.94
fit_21E_04	21E		Europe	Ural, Tayginka	Russia	flake	5385.96	12.91	23.10	1367.21	1581.91	0.00	63.47
fit_21E_05	21E		Europe	Ural, Tayginka	Russia	flake	323.53	14.71	20.34	1362.12	1581.91	0.04	58.86
fit_21E_06	21E		Europe	Ural, Tayginka	Russia	flake	5593.58	12.64	23.15	1350.69	1581.91	0.00	63.76
fit_21E_07	21E		Europe	Ural, Tayginka	Russia	flake	5302.72	12.89	22.04	1339.26	1580.64	0.00	63.86
fit_21E_08	21E		Europe	Ural, Tayginka	Russia	flake	5499.24	13.04	22.64	1397.27	1581.91	0.00	64.26
fit_21E_09	21E		Europe	Ural, Tayginka	Russia	flake	5296.32	12.74	22.20	1340.53	1581.91	0.00	63.78
fit_21E_10	21E		Europe	Ural, Tayginka	Russia	flake	5473.25	13.10	22.21	1355.77	1581.91	0.00	64.28

fit_21E_11	21E		Europe	Ural, Tayginka	Russia	flake	5459.32	13.04	21.61	1351.96	1581.91	0.00	64.24
fit_21E_12	21E		Europe	Ural, Tayginka	Russia	flake	5324.68	12.95	22.31	1340.53	1581.91	0.00	63.75
fit_21E_13	21E		Europe	Ural, Tayginka	Russia	flake	5275.86	12.82	22.54	1354.50	1581.91	0.00	63.38
fit_21E_14	21E		Europe	Ural, Tayginka	Russia	flake	917.23	14.02	20.10	1362.84	1581.29	0.01	72.63
fit_21E_15	21E		Europe	Ural, Tayginka	Russia	flake	530.78	13.97	20.94	1349.65	1581.29	0.03	44.70
fit_21E_16	21E		Europe	Ural, Tayginka	Russia	flake	230.69	17.02	13.37	1344.34	1581.91	0.07	46.70
fit_21E_17	21E		Europe	Ural, Tayginka	Russia	flake	5560.47	12.96	22.11	1344.34	1580.64	0.00	64.75
fit_21E_18	21E		Europe	Ural, Tayginka	Russia	flake	488.49	17.11	15.81	1346.72	1581.29	0.03	64.87
fit_21E_19	21E		Europe	Ural, Tayginka	Russia	flake	926.04	14.28	19.74	1343.78	1581.29	0.02	70.34
fit_21E_20	21E		Europe	Ural, Tayginka	Russia	flake	5690.64	12.96	22.92	1388.80	1581.91	0.00	64.10
fit_21F_01	21F		Asia	Ragedara	Sri_Lanka	hydro	5006.46	13.82	19.98	1445.84	1581.79	0.00	64.38
fit_21F_02	21F		Asia	Ragedara	Sri_Lanka	hydro	5099.09	13.95	21.34	1350.54	1581.79	0.00	63.44
fit_21F_03	21F		Asia	Ragedara	Sri_Lanka	hydro	4915.41	13.43	21.77	1381.04	1581.79	0.00	63.07
fit_21F_04	21F		Asia	Ragedara	Sri_Lanka	hydro	1186.14	13.72	21.87	1341.65	1580.51	0.01	56.99
fit_21F_05	21F		Asia	Ragedara	Sri_Lanka	hydro	5230.56	13.16	23.58	1445.84	1580.51	0.00	61.95
fit_21F_06	21F		Asia	Ragedara	Sri_Lanka	hydro	5423.15	12.91	22.82	1341.65	1580.51	0.00	63.64
fit_21F_07	21F		Asia	Ragedara	Sri_Lanka	hydro	5267.33	14.05	20.67	1435.25	1581.79	0.00	64.64
fit_21F_08	21F		Asia	Ragedara	Sri_Lanka	hydro	5400.57	13.09	22.67	1448.38	1580.51	0.00	63.77
fit_21F_09	21F		Asia	Ragedara	Sri_Lanka	hydro	5002.66	12.59	23.17	1425.51	1579.24	0.00	62.20
fit_21F_10	21F		Asia	Ragedara	Sri_Lanka	hydro	5510.66	13.40	21.90	1350.54	1580.51	0.00	64.07
fit_21F_11	21F		Asia	Ragedara	Sri_Lanka	hydro	930.62	14.10	20.42	1350.54	1580.51	0.01	60.31
fit_21F_12	21F		Asia	Ragedara	Sri_Lanka	hydro	581.74	13.85	21.49	1342.92	1581.79	0.02	46.01

fit_21F_13	21F		Asia	Ragedara	Sri_Lanka	hydro	5472.02	13.22	22.71	1353.08	1580.51	0.00	63.60
fit_21F_14	21F		Asia	Ragedara	Sri_Lanka	hydro	5408.11	12.66	23.87	1363.25	1580.51	0.00	62.47
fit_21F_15	21F		Asia	Ragedara	Sri_Lanka	hydro	5045.37	13.31	22.31	1412.80	1580.51	0.00	62.73
fit_21F_16	21F		Asia	Ragedara	Sri_Lanka	hydro	4867.50	15.11	19.16	1348.00	1583.06	0.00	63.76
fit_21F_17	21F		Asia	Ragedara	Sri_Lanka	hydro	1008.52	14.30	19.87	1349.27	1581.79	0.01	68.78
fit_21F_18	21F		Asia	Ragedara	Sri_Lanka	hydro	5435.78	14.60	21.90	1354.35	1580.51	0.00	64.53
fit_21F_19	21F		Asia	Ragedara	Sri_Lanka	hydro	5185.81	13.14	22.78	1414.07	1580.51	0.00	62.65
fit_21F_20	21F		Asia	Ragedara	Sri_Lanka	hydro	4695.04	16.10	18.04	1354.35	1579.24	0.00	63.69
fit_21G_01	21G		Europe	Kaisersberg	Austria	amorph	85.80	52.65	3.72	1351.11	1576.89	0.61	45.83
fit_21G_02	21G		Europe	Kaisersberg	Austria	amorph	93.95	68.13	2.45	1344.34	1586.99	0.72	58.88
fit_21G_03	21G		Europe	Kaisersberg	Austria	amorph	111.72	47.43	3.87	1346.88	1575.56	0.43	53.63
fit_21G_04	21G		Europe	Kaisersberg	Austria	amorph	87.22	66.70	2.87	1344.34	1586.99	0.75	55.84
fit_21G_05	21G		Europe	Kaisersberg	Austria	amorph	63.11	52.90	4.12	1351.96	1585.72	0.87	38.56
fit_21G_06	21G		Europe	Kaisersberg	Austria	amorph	75.10	55.62	3.70	1349.42	1579.37	0.75	45.56
fit_21G_07	21G		Europe	Kaisersberg	Austria	amorph	68.38	75.16	2.73	1351.11	1581.29	1.09	44.15
fit_21G_08	21G		Europe	Kaisersberg	Austria	amorph	74.34	50.84	3.96	1351.11	1582.76	0.68	43.12
fit_21G_09	21G		Europe	Kaisersberg	Austria	amorph	97.98	46.09	4.39	1351.96	1583.18	0.47	53.09
fit_21G_10	21G		Europe	Kaisersberg	Austria	amorph	69.38	72.12	2.72	1344.34	1585.72	1.03	43.13
fit_21G_11	21G		Europe	Kaisersberg	Austria	amorph	65.87	73.19	2.66	1351.96	1583.18	1.09	42.10
fit_21G_12	21G		Europe	Kaisersberg	Austria	amorph	84.28	62.65	3.12	1351.11	1587.16	0.76	53.32
fit_21G_13	21G		Europe	Kaisersberg	Austria	amorph	81.90	56.13	3.61	1348.15	1584.45	0.69	48.46
fit_21G_14	21G		Europe	Kaisersberg	Austria	amorph	85.55	59.62	3.18	1351.96	1585.72	0.70	53.09

fit_21G_15	21G		Europe	Kaisersberg	Austria	amorph	83.57	52.61	3.68	1350.69	1585.72	0.65	50.03
fit_21G_16	21G		Europe	Kaisersberg	Austria	amorph	86.59	57.01	2.97	1350.69	1578.10	0.64	50.17
fit_21G_17	21G		Europe	Kaisersberg	Austria	amorph	97.08	58.55	3.77	1351.96	1579.37	0.59	51.14
fit_21G_18	21G		Europe	Kaisersberg	Austria	amorph	415.94	15.98	19.01	1344.34	1579.37	0.04	39.93
fit_21G_19	21G		Europe	Kaisersberg	Austria	amorph	562.62	15.28	19.17	1351.96	1580.64	0.03	49.42
fit_21G_20	21G		Europe	Kaisersberg	Austria	amorph	83.21	64.25	2.58	1351.96	1583.18	0.77	50.92
fit_21I_01	21I		Europe	Kaisersberg	Austria	amorph	97.22	80.49	3.07	1344.32	1571.74	0.83	52.13
fit_21I_02	21I		Europe	Kaisersberg	Austria	amorph	95.22	66.41	2.53	1351.94	1581.91	0.72	63.08
fit_21I_03	21I		Europe	Kaisersberg	Austria	amorph	773.99	13.50	20.62	1351.94	1581.91	0.02	57.56
fit_21I_04	21I		Europe	Kaisersberg	Austria	amorph	86.51	58.06	3.17	1346.88	1579.39	0.67	51.94
fit_21I_05	21I		Europe	Kaisersberg	Austria	amorph	71.09	59.78	2.78	1348.15	1583.20	0.82	48.79
fit_21I_06	21I		Europe	Kaisersberg	Austria	amorph	81.67	87.88	2.11	1346.88	1581.93	1.09	58.85
fit_21I_07	21I		Europe	Kaisersberg	Austria	amorph	184.12	19.12	16.51	1349.42	1579.39	0.10	42.07
fit_21I_08	21I		Europe	Kaisersberg	Austria	amorph	301.38	16.25	18.20	1362.13	1578.12	0.04	35.69
fit_21I_09	21I		Europe	Kaisersberg	Austria	amorph	151.03	20.46	10.71	1353.24	1579.39	0.13	42.43
fit_21I_10	21I		Europe	Kaisersberg	Austria	amorph	4597.21	15.46	16.99	1362.13	1579.39	0.00	65.01
fit_21J_01	21J		Africa	Balama	Mozambique	flake	5408.53	13.30	23.14	1332.89	1581.87	0.00	63.42
fit_21J_02	21J		Africa	Balama	Mozambique	flake	5349.85	14.34	21.33	1335.43	1581.87	0.00	64.58
fit_21J_03	21J		Africa	Balama	Mozambique	flake	658.94	15.61	17.71	1349.40	1580.60	0.02	58.37
fit_21J_04	21J		Africa	Balama	Mozambique	flake	449.71	14.43	20.61	1353.21	1580.60	0.03	48.26
fit_21J_05	21J		Africa	Balama	Mozambique	flake	204.22	16.43	19.09	1354.48	1581.87	0.08	49.19
fit_21J_06	21J		Africa	Balama	Mozambique	flake	231.70	15.79	15.38	1357.02	1581.87	0.07	52.87

fit_21J_07	21J		Africa	Balama	Mozambique	flake	5624.08	13.36	22.21	1344.32	1580.60	0.00	63.52
fit_21J_08	21J		Africa	Balama	Mozambique	flake	2480.59	14.42	21.80	1344.32	1581.87	0.01	67.16
fit_21J_09	21J		Africa	Balama	Mozambique	flake	450.62	14.67	19.75	1351.94	1580.60	0.03	45.16
fit_21J_10	21J		Africa	Balama	Mozambique	flake	583.99	14.41	19.73	1349.40	1581.87	0.03	57.84
fit_21J_11	21J		Africa	Balama	Mozambique	flake	5813.55	13.77	22.90	1360.84	1581.87	0.00	64.54
fit_21J_12	21J		Africa	Balama	Mozambique	flake	5351.43	14.57	21.69	1348.13	1581.87	0.00	64.24
fit_21J_13	21J		Africa	Balama	Mozambique	flake	526.68	14.96	18.77	1345.59	1581.87	0.03	51.14
fit_21J_14	21J		Africa	Balama	Mozambique	flake	600.15	13.85	20.96	1348.13	1581.87	0.02	53.13
fit_21J_15	21J		Africa	Balama	Mozambique	flake	533.86	14.20	20.40	1343.05	1581.87	0.03	45.63
fit_21J_16	21J		Africa	Balama	Mozambique	flake	5617.58	14.62	22.54	1339.24	1580.60	0.00	64.82
fit_21J_17	21J		Africa	Balama	Mozambique	flake	5053.99	13.79	20.03	1327.81	1581.87	0.00	63.92
fit_21J_18	21J		Africa	Balama	Mozambique	flake	457.68	15.20	19.78	1343.05	1579.33	0.03	64.28
fit_21J_19	21J		Africa	Balama	Mozambique	flake	707.61	16.25	15.94	1351.94	1580.60	0.02	57.48
fit_21J_20	21J		Africa	Balama	Mozambique	flake	5384.83	12.61	22.92	1349.40	1579.33	0.00	48.67
fit_21K_01	21K		Africa	Balama	Mozambique	flake	5332.62	12.82	21.80	1339.24	1581.87	0.00	64.02
fit_21K_02	21K		Africa	Balama	Mozambique	flake	671.83	13.44	21.65	1348.13	1580.60	0.02	44.72
fit_21K_03	21K		Africa	Balama	Mozambique	flake	279.54	16.60	15.20	1348.13	1581.87	0.06	50.55
fit_21K_04	21K		Africa	Balama	Mozambique	flake	400.14	15.16	19.85	1351.94	1580.60	0.04	55.55
fit_21K_05	21K		Africa	Balama	Mozambique	flake	6161.46	14.41	25.43	1348.13	1580.60	0.00	64.59
fit_21K_06	21K		Africa	Balama	Mozambique	flake	5225.34	14.10	20.31	1345.59	1580.60	0.00	64.39
fit_21K_07	21K		Africa	Balama	Mozambique	flake	601.11	15.23	18.80	1357.02	1581.87	0.02	52.45
fit_21K_08	21K		Africa	Balama	Mozambique	flake	5469.42	13.99	20.13	1348.13	1581.87	0.00	64.92

fit_21K_09	21K		Africa	Balama	Mozambique	flake	5387.45	12.95	22.80	1346.86	1579.33	0.00	63.56
fit_21K_10	21K		Africa	Balama	Mozambique	flake	5642.12	14.60	22.99	1353.21	1580.60	0.00	64.54
fit_21K_11	21K		Africa	Balama	Mozambique	flake	334.04	15.92	18.91	1348.13	1579.33	0.05	43.97
fit_21K_12	21K		Africa	Balama	Mozambique	flake	693.00	14.10	20.08	1340.51	1581.87	0.02	48.32
fit_21K_13	21K		Africa	Balama	Mozambique	flake	5469.18	13.23	22.02	1353.21	1580.60	0.00	64.27
fit_21K_14	21K		Africa	Balama	Mozambique	flake	5527.29	12.90	22.84	1350.67	1580.60	0.00	63.32
fit_21K_15	21K		Africa	Balama	Mozambique	flake	5386.12	13.13	22.02	1349.40	1579.33	0.00	64.29
fit_21K_16	21K		Africa	Balama	Mozambique	flake	6200.02	14.35	26.32	1348.13	1580.60	0.00	64.13
fit_21K_17	21K		Africa	Balama	Mozambique	flake	630.25	13.29	22.30	1351.94	1580.60	0.02	49.78
fit_21K_18	21K		Africa	Balama	Mozambique	flake	975.67	13.99	19.94	1351.94	1581.87	0.01	58.80
fit_21K_19	21K		Africa	Balama	Mozambique	flake	464.73	14.42	20.43	1346.86	1580.60	0.03	46.33
fit_21K_20	21K		Africa	Balama	Mozambique	flake	357.47	15.58	18.83	1357.02	1580.60	0.04	46.95
fit_21L_01	21L		Africa	Balama	Mozambique	flake	400.21	14.06	21.16	1348.02	1580.54	0.04	41.92
fit_21L_02	21L		Africa	Balama	Mozambique	flake	722.75	13.59	21.25	1355.65	1580.54	0.02	55.34
fit_21L_03	21L		Africa	Balama	Mozambique	flake	5640.49	12.58	23.31	1339.13	1581.81	0.00	39.54
fit_21L_04	21L		Africa	Balama	Mozambique	flake	5554.10	12.95	22.87	1351.84	1581.81	0.00	64.18
fit_21L_05	21L		Africa	Balama	Mozambique	flake	5526.05	12.58	23.08	1358.19	1580.54	0.00	63.96
fit_21L_06	21L		Africa	Balama	Mozambique	flake	5398.11	13.06	22.44	1364.54	1581.81	0.00	50.87
fit_21L_07	21L		Africa	Balama	Mozambique	flake	5619.58	12.57	22.83	1388.68	1581.81	0.00	63.86
fit_21L_08	21L		Africa	Balama	Mozambique	flake	889.37	13.17	21.73	1355.65	1581.81	0.01	54.16
fit_21L_09	21L		Africa	Balama	Mozambique	flake	5587.17	12.52	23.61	1364.54	1581.81	0.00	63.65
fit_21L_10	21L		Africa	Balama	Mozambique	flake	1149.38	12.88	22.36	1354.38	1580.54	0.01	64.50

fit_21L_11	21L		Africa	Balama	Mozambique	flake	5201.72	13.93	20.75	1344.21	1581.81	0.00	51.60
fit_21L_12	21L		Africa	Balama	Mozambique	flake	283.80	15.86	15.45	1345.48	1581.81	0.05	44.55
fit_21L_13	21L		Africa	Balama	Mozambique	flake	5981.90	13.87	24.84	1349.29	1580.54	0.00	64.51
fit_21L_14	21L		Africa	Balama	Mozambique	flake	279.24	15.45	19.68	1351.84	1580.54	0.06	41.13
fit_21L_15	21L		Africa	Balama	Mozambique	flake	400.75	14.26	20.83	1353.11	1580.54	0.04	45.30
fit_21L_16	21L		Africa	Balama	Mozambique	flake	5667.54	12.72	23.44	1353.11	1580.54	0.00	63.88
fit_21L_17	21L		Africa	Balama	Mozambique	flake	836.01	12.70	23.25	1348.02	1581.81	0.02	44.06
fit_21L_18	21L		Africa	Balama	Mozambique	flake	255.56	15.68	17.94	1346.75	1580.54	0.06	43.68
fit_21L_19	21L		Africa	Balama	Mozambique	flake	5377.32	12.86	22.91	1367.08	1581.81	0.00	64.01
fit_21L_20	21L		Africa	Balama	Mozambique	flake	5553.84	12.81	22.83	1445.86	1580.54	0.00	64.31
fit_21M_01	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	691.47	13.39	21.19	1353.24	1580.63	0.02	47.55
fit_21M_02	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	256.70	15.63	15.97	1344.35	1578.09	0.06	40.08
fit_21M_03	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	398.45	13.96	21.91	1345.46	1580.51	0.04	41.41
fit_21M_04	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5676.78	12.72	22.88	1346.73	1580.51	0.00	63.71
fit_21M_05	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	383.59	15.04	19.07	1349.27	1581.78	0.04	47.22
fit_21M_06	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	1021.18	13.10	21.59	1350.54	1581.78	0.01	51.08
fit_21M_07	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	564.02	13.25	22.21	1350.54	1581.78	0.02	43.37
fit_21M_08	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5857.21	12.45	24.63	1345.46	1580.51	0.00	63.76
fit_21M_09	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	933.80	13.27	22.07	1350.54	1581.78	0.01	53.03
fit_21M_10	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	1116.02	13.19	22.54	1350.54	1580.51	0.01	58.15
fit_21M_11	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	958.45	12.96	22.55	1342.92	1579.24	0.01	57.22
fit_21M_12	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5823.29	12.30	24.60	1361.98	1580.51	0.00	63.83

fit_21M_13	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	247.48	16.39	14.81	1354.35	1581.78	0.06	42.67
fit_21M_14	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5515.86	12.51	23.70	1341.65	1580.51	0.00	63.56
fit_21M_15	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	1035.45	13.06	22.72	1351.81	1581.78	0.01	65.01
fit_21M_16	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	613.05	13.87	21.32	1349.27	1581.78	0.02	48.55
fit_21M_17	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	751.24	13.36	21.49	1345.46	1581.78	0.02	49.06
fit_21M_18	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5497.39	12.64	23.28	1361.98	1581.78	0.00	63.83
fit_21M_19	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	837.81	12.98	24.21	1344.19	1580.51	0.02	52.00
fit_21M_20	21M		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5630.22	12.53	23.33	1355.62	1580.51	0.00	64.02
fit_21N_01	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5363.76	13.14	22.44	1337.84	1580.51	0.00	63.47
fit_21N_02	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	778.86	13.10	22.20	1351.81	1581.78	0.02	46.47
fit_21N_03	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	284.04	15.38	15.60	1355.62	1581.78	0.05	40.90
fit_21N_04	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	800.11	13.46	21.79	1359.44	1580.51	0.02	55.04
fit_21N_05	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5705.75	12.57	23.70	1348.00	1581.78	0.00	64.03
fit_21N_06	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	1195.22	13.01	23.02	1351.81	1581.78	0.01	63.46
fit_21N_07	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	624.89	13.29	22.28	1354.35	1581.78	0.02	46.43
fit_21N_08	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	492.86	14.29	20.62	1348.00	1581.78	0.03	49.39
fit_21N_09	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	463.33	13.85	21.54	1350.54	1580.51	0.03	40.85
fit_21N_10	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	442.35	13.95	20.27	1354.35	1580.51	0.03	46.61
fit_21N_11	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	767.52	13.72	20.83	1346.73	1581.78	0.02	51.75
fit_21N_12	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	239.18	16.61	13.86	1351.81	1580.51	0.07	44.49
fit_21N_13	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	332.81	14.52	21.34	1354.35	1580.51	0.04	42.31
fit_21N_14	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	677.86	14.92	18.31	1349.27	1583.05	0.02	47.21

fit_21N_15	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5249.22	13.43	20.88	1425.50	1581.78	0.00	64.18
fit_21N_16	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5530.33	12.81	22.65	1341.65	1581.78	0.00	64.26
fit_21N_17	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5178.22	13.47	20.65	1361.98	1581.78	0.00	64.62
fit_21N_18	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	516.69	13.85	18.74	1354.35	1581.78	0.03	51.76
fit_21N_19	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5384.20	13.24	21.76	1342.92	1580.51	0.00	64.28
fit_21N_20	21N		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	769.84	14.53	19.55	1344.19	1580.51	0.02	47.84
fit_22A_01	22A		Asia	Shandong	China	flake	362.34	15.89	17.61	1350.70	1581.93	0.04	48.68
fit_22A_02	22A		Asia	Shandong	China	flake	5732.28	16.08	22.69	1372.29	1579.39	0.00	64.68
fit_22A_03	22A		Asia	Shandong	China	flake	5564.95	15.72	22.14	1340.53	1581.93	0.00	64.63
fit_22A_04	22A		Asia	Shandong	China	flake	7103.17	13.71	29.35	1406.60	1581.93	0.00	65.16
fit_22A_05	22A		Asia	Shandong	China	flake	6051.25	13.57	25.28	1382.46	1580.66	0.00	64.48
fit_22A_06	22A		Asia	Shandong	China	flake	6225.02	14.44	25.28	1374.84	1581.93	0.00	64.96
fit_22A_07	22A		Asia	Shandong	China	flake	5900.32	13.39	23.95	1391.35	1581.93	0.00	64.49
fit_22A_08	22A		Asia	Shandong	China	flake	5525.45	14.72	23.05	1355.78	1580.66	0.00	63.77
fit_22A_09	22A		Asia	Shandong	China	flake	252.79	17.95	16.20	1341.80	1580.66	0.07	51.17
fit_22A_10	22A		Asia	Shandong	China	flake	6076.71	14.46	24.56	1348.15	1579.39	0.00	64.93
fit_22B_01	22B		Africa	Balama	Mozambique	flake	6024.41	15.72	24.73	1341.80	1578.12	0.00	64.25
fit_22B_02	22B		Africa	Balama	Mozambique	flake	241.19	15.14	20.51	1348.15	1580.66	0.06	36.66
fit_22B_03	22B		Africa	Balama	Mozambique	flake	230.58	17.24	15.89	1349.42	1579.39	0.08	46.18
fit_22B_04	22B		Africa	Balama	Mozambique	flake	5121.61	13.34	21.54	1346.88	1579.39	0.00	63.54
fit_22B_05	22B		Africa	Balama	Mozambique	flake	5839.10	14.11	24.64	1349.42	1580.66	0.00	63.96
fit_22B_06	22B		Africa	Balama	Mozambique	flake	5784.97	12.75	23.43	1359.59	1581.93	0.00	64.20

fit_22B_07	22B		Africa	Balama	Mozambique	flake	6310.13	14.13	27.10	1348.15	1580.66	0.00	64.00
fit_22B_08	22B		Africa	Balama	Mozambique	flake	242.44	17.12	17.23	1345.61	1581.93	0.07	40.74
fit_22B_09	22B		Africa	Balama	Mozambique	flake	6361.01	14.27	26.51	1339.26	1580.66	0.00	64.58
fit_22B_10	22B		Africa	Balama	Mozambique	flake	399.28	13.89	21.28	1350.70	1581.93	0.04	37.70
fit_22C_01	22C		S_America	PedraAzul, Itaperica, Salta da Divisa		flake	1000.03	14.56	19.01	1348.17	1578.13	0.01	67.60
fit_22C_02	22C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5122.24	13.93	19.30	1438.37	1581.94	0.00	64.08
fit_22C_03	22C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	1247.75	12.75	22.61	1351.98	1580.67	0.01	63.65
fit_22C_04	22C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5732.92	13.81	24.21	1353.25	1580.67	0.00	63.91
fit_22C_05	22C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	260.06	16.32	18.96	1348.17	1581.94	0.07	34.35
fit_22C_06	22C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5947.68	13.99	24.69	1378.66	1580.67	0.00	64.45
fit_22C_07	22C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5484.33	13.97	23.08	1391.36	1580.67	0.00	63.70
fit_22C_08	22C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	5750.78	14.14	23.14	1345.63	1580.67	0.00	64.86
fit_22C_09	22C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	262.86	17.81	18.61	1360.87	1580.67	0.05	70.07
fit_22C_10	22C		S_America	PedraAzul, Itaperica, Salta da Divisa	Brasil	flake	263.14	16.27	18.43	1348.17	1580.67	0.06	39.32
fit_22D_01	22D		Africa	Vatomina/Sahamamy Sahasoa	Madagascar	flake	122.03	26.42	8.03	1350.69	1579.39	0.22	44.90
fit_22D_02	22D		Africa	Vatomina/Sahamamy Sahasoa	Madagascar	flake	5722.35	14.23	23.30	1343.07	1580.66	0.00	64.60
fit_22E_01	22E		Europe	Ural, Tayginka	Russia	flake	687.47	13.08	22.06	1344.32	1580.64	0.02	47.92
fit_22E_02	22E		Europe	Ural, Tayginka	Russia	flake	587.75	13.42	21.33	1357.02	1580.64	0.02	54.37
fit_22E_03	22E		Europe	Ural, Tayginka	Russia	flake	703.10	13.41	21.01	1358.29	1580.64	0.02	60.13
fit_22E_04	22E		Europe	Ural, Tayginka	Russia	flake	888.30	13.16	21.51	1348.13	1580.64	0.02	54.89
fit_22E_05	22E		Europe	Ural, Tayginka	Russia	flake	1190.16	12.67	23.73	1340.51	1579.37	0.01	60.56
fit_22E_06	22E		Europe	Ural, Tayginka	Russia	flake	5423.09	12.71	21.95	1359.56	1581.91	0.00	64.46

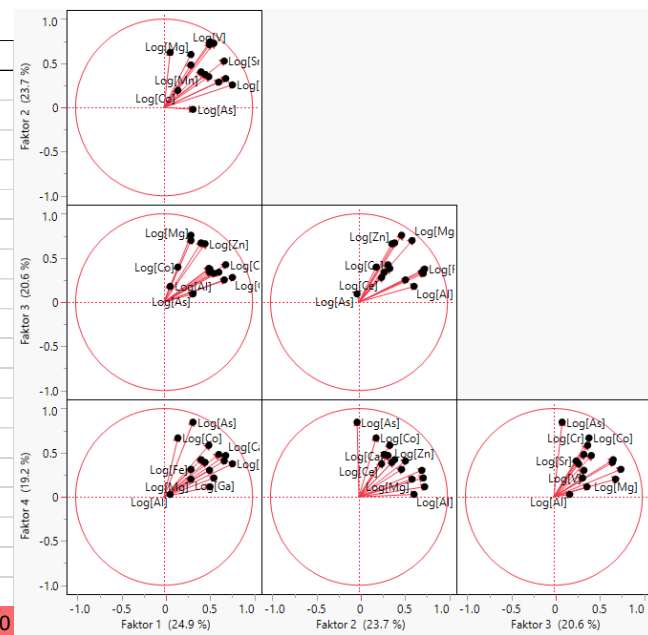
fit_22E_07	22E		Europe	Ural, Tayginka	Russia	flake	780.64	13.33	21.20	1346.86	1579.37	0.02	62.19
fit_22E_08	22E		Europe	Ural, Tayginka	Russia	flake	5447.27	13.15	22.11	1353.21	1581.91	0.00	64.35
fit_22E_09	22E		Europe	Ural, Tayginka	Russia	flake	5349.27	13.12	21.57	1424.36	1581.91	0.00	64.20
fit_22E_10	22E		Europe	Ural, Tayginka	Russia	flake	756.65	13.58	21.01	1358.29	1581.91	0.02	64.55
fit_22E_11	22E		Europe	Ural, Tayginka	Russia	flake	734.96	13.62	19.97	1367.19	1581.91	0.02	72.72
fit_22E_12	22E		Europe	Ural, Tayginka	Russia	flake	312.24	15.50	18.63	1351.94	1581.91	0.05	43.54
fit_22E_13	22E		Europe	Ural, Tayginka	Russia	flake	5307.67	13.18	21.88	1351.94	1580.64	0.00	64.02
fit_22E_14	22E		Europe	Ural, Tayginka	Russia	flake	6106.76	13.85	24.82	1386.25	1581.91	0.00	64.99
fit_22E_15	22E		Europe	Ural, Tayginka	Russia	flake	4940.76	14.34	20.15	1357.02	1580.64	0.00	63.58
fit_22E_16	22E		Europe	Ural, Tayginka	Russia	flake	4722.97	14.85	19.03	1386.25	1580.64	0.00	63.73
fit_22E_17	22E		Europe	Ural, Tayginka	Russia	flake	5536.35	12.77	23.05	1315.10	1580.64	0.00	64.05
fit_22E_18	22E		Europe	Ural, Tayginka	Russia	flake	639.33	14.42	20.08	1348.13	1580.64	0.02	52.20
fit_22E_19	22E		Europe	Ural, Tayginka	Russia	flake	384.44	14.75	19.57	1348.13	1580.64	0.04	42.50
fit_22E_20	22E		Europe	Ural, Tayginka	Russia	flake	5599.68	13.04	22.75	1359.56	1580.64	0.00	64.15
fit_22F_01	22F		Europe	Zavalje	Ukraine	flake	320.04	16.42	17.10	1348.15	1580.66	0.05	44.71
fit_22F_02	22F		Europe	Zavalje	Ukraine	flake	116.03	26.13	7.80	1349.42	1580.66	0.23	44.35
fit_22F_03	22F		Europe	Zavalje	Ukraine	flake	6390.58	13.99	26.29	1353.24	1581.93	0.00	64.91
fit_22F_04	22F		Europe	Zavalje	Ukraine	flake	161.06	16.52	19.90	1360.86	1579.39	0.10	48.16
fit_22F_05	22F		Europe	Zavalje	Ukraine	flake	414.60	16.39	17.04	1348.15	1580.66	0.04	50.13
fit_22F_06	22F		Europe	Zavalje	Ukraine	flake	5923.21	14.45	24.05	1354.51	1580.66	0.00	64.76
fit_22F_07	22F		Europe	Zavalje	Ukraine	flake	321.45	17.01	16.37	1349.42	1581.93	0.05	43.16
fit_22F_08	22F		Europe	Zavalje	Ukraine	flake	145.85	26.82	8.27	1348.15	1579.39	0.19	47.05

fit_22G_01	22G		Europe	Skaland	Norway	flake	157.74	23.54	13.01	1348.17	1579.40	0.15	46.09
fit_22G_02	22G		Europe	Skaland	Norway	flake	5585.12	14.48	22.35	1350.71	1581.94	0.00	64.77
fit_22G_03	22G		Europe	Skaland	Norway	flake	232.31	17.17	18.00	1355.79	1580.67	0.07	46.17
fit_22G_04	22G		Europe	Skaland	Norway	flake	5875.03	14.46	24.73	1362.14	1579.40	0.00	63.98
fit_22G_05	22G		Europe	Skaland	Norway	flake	296.47	16.10	18.76	1346.90	1579.40	0.05	47.33
fit_22G_06	22G		Europe	Skaland	Norway	flake	5535.25	14.33	21.33	1387.55	1579.40	0.00	64.06
fit_22G_07	22G		Europe	Skaland	Norway	flake	6141.09	14.90	25.06	1360.87	1580.67	0.00	64.75
fit_22G_08	22G		Europe	Skaland	Norway	flake	6568.65	14.89	26.83	1343.08	1579.40	0.00	64.91
fit_22G_09	22G		Europe	Skaland	Norway	flake	163.25	16.60	20.22	1350.71	1580.67	0.10	43.61
fit_22G_10	22G		Europe	Skaland	Norway	flake	5785.29	13.76	24.80	1340.54	1579.40	0.00	63.57

D. LA-ICP-MS Trace Element Data, Correlation Matrices and Factor Analysis

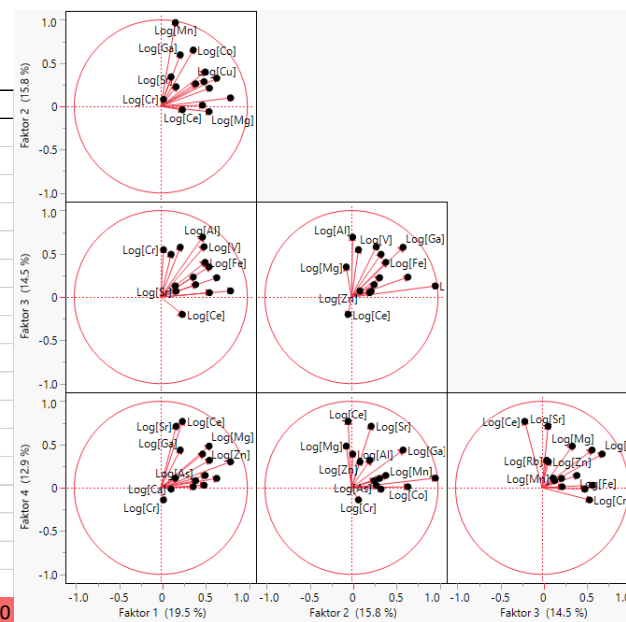
Correlation matrix for LA-ICP-MS trace element data for Kaisersberg (AUT)

Kaisersberg (AUT)	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	Th	U
Mg	1.00																	
Al	0.82	1.00																
Ca	0.74	0.72	1.00															
V	0.83	0.88	0.93	1.00														
Cr	0.75	0.80	0.92	0.95	1.00													
Mn	0.80	0.73	0.86	0.90	0.85	1.00												
Fe	0.91	0.77	0.87	0.91	0.85	0.97	1.00											
Co	0.86	0.69	0.78	0.80	0.72	0.83	0.90	1.00										
Cu	0.47	0.36	0.77	0.62	0.62	0.57	0.58	0.50	1.00									
Zn	0.80	0.72	0.91	0.92	0.90	0.95	0.94	0.81	0.68	1.00								
Ga	0.80	0.91	0.90	0.98	0.94	0.85	0.85	0.73	0.60	0.87	1.00							
As	0.55	0.49	0.75	0.72	0.77	0.72	0.69	0.67	0.58	0.75	0.71	1.00						
Rb	0.80	0.87	0.92	0.99	0.94	0.87	0.88	0.76	0.62	0.90	0.98	0.72	1.00					
Sr	0.77	0.84	0.95	0.95	0.94	0.83	0.84	0.75	0.64	0.87	0.96	0.72	0.95	1.00				
Y	0.15	0.04	0.44	0.23	0.23	0.16	0.19	0.18	0.86	0.29	0.22	0.24	0.23	0.29				
Zr	0.38	0.56	0.46	0.41	0.39	0.28	0.31	0.41	0.21	0.31	0.46	0.24	0.39	0.51	1.00			
Ce	0.31	0.14	0.61	0.42	0.42	0.38	0.40	0.37	0.93	0.51	0.37	0.41	0.41	0.45	0.10	1.00		
Th	0.41	0.50	0.57	0.55	0.59	0.49	0.47	0.38	0.39	0.50	0.58	0.43	0.55	0.60	0.48	0.36	1.00	
U	0.29	0.18	0.59	0.40	0.41	0.34	0.35	0.31	0.95	0.47	0.40	0.40	0.40	0.45	0.18	0.95	0.33	1.00



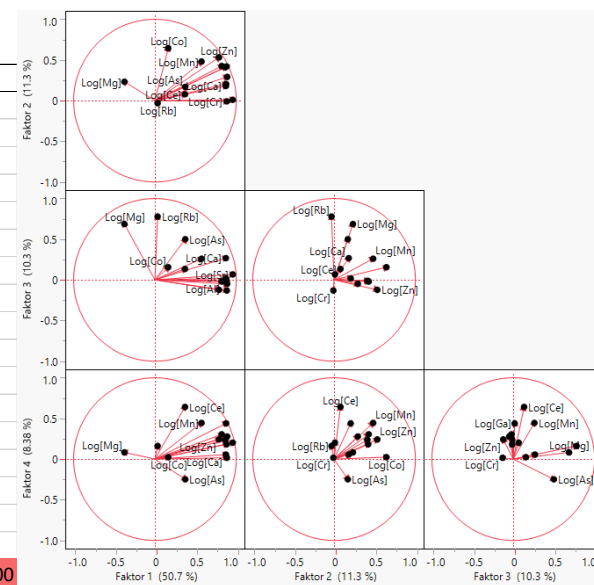
Correlation matrix for LA-ICP-MS trace element data for Minas Gerais* (BRA)

Minas Gerais* (BRA)	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	Th	U	
Mg	1.00																		
Al	0.67	1.00																	
Ca	0.25	0.29	1.00																
V	0.25	0.61	0.38	1.00															
Cr	0.24	0.30	0.55	0.31	1.00														
Mn	0.00	-0.04	0.16	0.16	0.02	1.00													
Fe	0.46	0.50	0.38	0.52	0.21	0.21	1.00												
Co	0.02	0.02	0.15	0.31	0.05	0.52	0.17	1.00											
Cu	0.57	0.48	0.29	0.43	0.11	0.33	0.62	0.44	1.00										
Zn	0.65	0.37	0.26	0.23	0.01	0.07	0.47	0.13	0.76	1.00									
Ga	0.58	0.66	0.33	0.48	0.24	0.52	0.48	0.06	0.41	0.31	1.00								
As	0.33	0.35	0.14	0.37	0.00	0.35	0.28	0.81	0.47	0.17	0.29	1.00							
Rb	0.62	0.31	0.26	0.19	0.02	0.07	0.48	0.06	0.70	0.98	0.30	0.09	1.00						
Sr	0.56	0.38	0.21	0.13	0.12	0.07	0.31	-0.01	0.27	0.27	0.50	0.21	0.26	1.00					
Zr	0.16	0.12	-0.15	-0.09	-0.25	0.10	0.18	-0.09	0.04	0.17	0.17	-0.01	0.17	0.31	1.00				
Ce	0.08	0.05	-0.09	-0.08	-0.21	-0.04	0.04	-0.08	-0.03	0.03	0.02	0.02	0.02	0.38	0.48	1.00			
Th	0.17	0.11	0.16	0.03	0.04	0.09	0.21	0.07	0.19	0.24	0.16	0.15	0.23	0.30	0.36	0.33	1.00		
U	0.03	0.06	0.20	0.13	0.03	0.16	0.18	0.08	0.05	0.03	0.16	0.09	0.04	0.27	0.18	0.79	0.26	1.00	



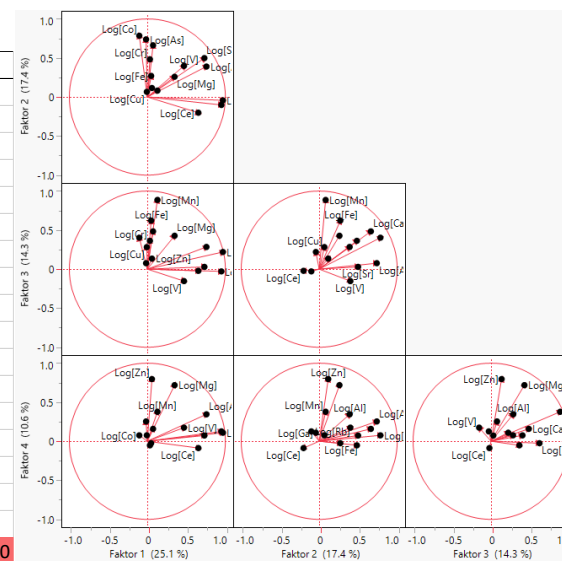
Correlation matrix for LA-ICP-MS trace element data for Shandong* (CHN)

Shandong (CHN)	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	Th	U
Mg	1.00																	
Al	-0.06	1.00																
Ca	0.09	0.66	1.00															
V	-0.09	0.88	0.73	1.00														
Cr	-0.28	0.61	0.73	0.56	1.00													
Mn	0.04	0.68	0.46	0.65	0.33	1.00												
Fe	-0.09	0.86	0.75	0.99	0.53	0.66	1.00											
Co	0.00	0.12	0.07	0.13	-0.08	0.35	0.14	1.00										
Cu	-0.12	0.93	0.78	0.93	0.64	0.68	0.95	0.12	1.00									
Zn	-0.03	0.92	0.53	0.86	0.37	0.63	0.86	0.15	0.89	1.00								
Ga	-0.17	0.70	0.78	0.72	0.77	0.53	0.72	0.04	0.78	0.55	1.00							
As	-0.04	0.16	0.46	0.58	0.21	0.23	0.58	0.04	0.34	0.15	0.34	1.00						
Rb	0.28	-0.04	0.16	0.07	0.00	-0.01	0.08	0.08	0.04	-0.01	0.32	0.18	1.00					
Sr	-0.18	0.27	0.69	0.53	0.63	0.29	0.55	-0.03	0.48	0.11	0.71	0.74	0.17	1.00				
Zr	-0.02	-0.03	-0.01	-0.03	-0.07	0.03	-0.03	-0.04	-0.04	-0.02	-0.05	0.00	0.08	-0.02	1.00			
Ce	-0.05	-0.08	-0.14	-0.05	-0.16	-0.01	-0.05	-0.06	-0.08	-0.04	-0.08	0.02	-0.11	-0.06	-0.01	1.00		
Th	-0.07	0.34	0.65	0.56	0.48	0.40	0.59	0.03	0.52	0.23	0.61	0.68	0.17	0.85	0.41	-0.02	1.00	
U	-0.11	0.76	0.74	0.95	0.51	0.60	0.96	0.10	0.87	0.74	0.70	0.72	0.09	0.65	0.02	0.12	0.69	1.00



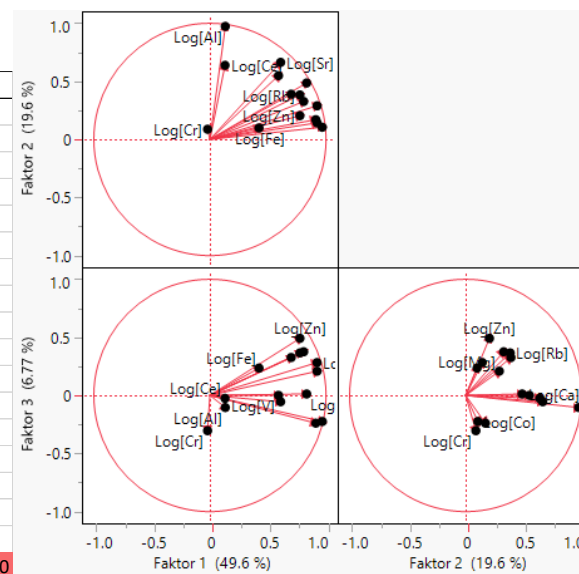
Correlation matrix for LA-ICP-MS trace element data for Hunan Lutang* (CHN)

Hunan Lutang* (CHN)	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	Th	U
Mg	1.00																	
Al	0.72	1.00																
Ca	0.59	0.39	1.00															
V	0.22	0.38	0.21	1.00														
Cr	0.26	0.42	0.41	0.04	1.00													
Mn	0.78	0.61	0.51	0.10	0.37	1.00												
Fe	0.14	0.06	0.28	0.17	0.18	0.34	1.00											
Co	0.34	0.33	0.68	0.22	0.57	0.33	0.23	1.00										
Cu	0.05	0.10	0.20	-0.12	-0.10	0.30	-0.07	0.08	1.00									
Zn	0.72	0.36	0.36	0.25	0.05	0.55	-0.18	0.18	-0.09	1.00								
Ga	0.41	0.79	0.04	0.20	-0.02	0.29	0.01	-0.07	0.02	0.07	1.00							
As	0.30	0.24	0.42	0.59	0.08	0.19	0.20	0.70	0.11	0.16	-0.05	1.00						
Rb	0.32	0.72	-0.06	0.20	-0.14	0.15	-0.09	-0.12	0.05	0.01	0.98	-0.04	1.00					
Sr	0.40	0.72	0.31	0.49	0.03	0.19	0.13	0.29	0.05	0.16	0.72	0.39	0.70	1.00				
Zr	-0.10	-0.08	-0.25	-0.15	-0.17	0.01	-0.24	-0.25	0.57	0.00	-0.06	-0.15	0.04	-0.20	1.00			
Ce	0.26	0.63	-0.05	0.09	-0.17	0.09	-0.09	-0.12	0.01	-0.07	0.93	-0.06	0.94	0.63	-0.09	1.00		
Th	-0.07	0.30	-0.15	-0.05	0.33	-0.21	-0.15	0.23	-0.10	-0.36	0.27	0.03	0.33	0.10	0.11	0.36	1.00	
U	-0.15	-0.04	-0.34	0.05	0.00	-0.15	-0.29	-0.14	0.28	-0.02	-0.14	-0.03	-0.03	-0.20	0.82	-0.13	0.46	1.00



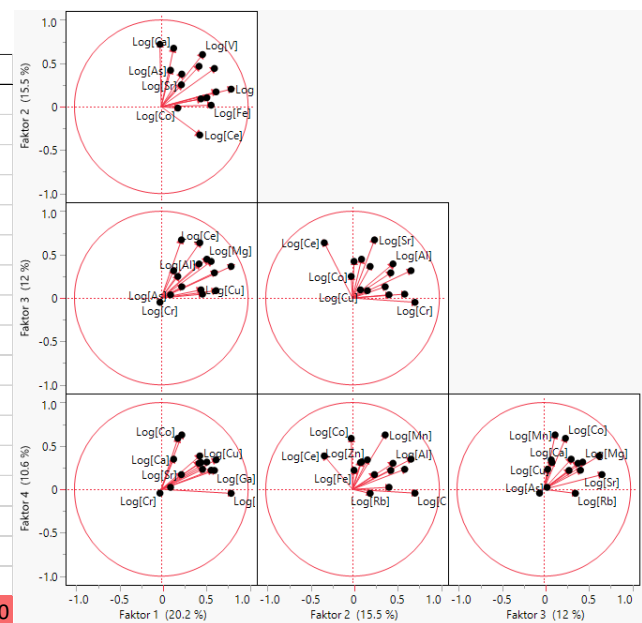
Correlation matrix for LA-ICP-MS trace element data for Inner Mongolia* (CHN)

Inner Mongolia* (CHN)	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	Th	U
Mg	1.00																	
Al	0.04	1.00																
Ca	1.00	0.08	1.00															
V	0.54	0.18	0.56	1.00														
Cr	-0.18	-0.13	-0.15	-0.19	1.00													
Mn	1.00	0.03	1.00	0.58	-0.17	1.00												
Fe	0.16	-0.01	0.18	0.83	-0.01	0.21	1.00											
Co	1.00	0.03	1.00	0.58	-0.17	1.00	0.21	1.00										
Cu	0.99	0.07	0.99	0.55	-0.20	0.99	0.17	0.99	1.00									
Zn	1.00	0.04	0.99	0.53	-0.18	1.00	0.15	1.00	0.99	1.00								
Ga	1.00	0.04	0.99	0.54	-0.18	1.00	0.16	1.00	0.99	1.00	1.00							
As	1.00	0.03	0.99	0.53	-0.18	1.00	0.15	1.00	0.99	1.00	1.00	1.00						
Rb	1.00	0.03	0.99	0.53	-0.18	1.00	0.15	1.00	0.99	1.00	1.00	1.00	1.00					
Sr	0.64	0.32	0.68	0.84	-0.21	0.67	0.71	0.67	0.66	0.63	0.64	0.63	0.63	1.00				
Zr	-0.15	0.37	-0.13	-0.07	0.34	-0.16	0.10	-0.15	-0.11	-0.16	-0.15	-0.16	-0.16	0.03	1.00			
Ce	-0.07	0.37	-0.05	-0.01	0.01	-0.08	0.06	-0.06	-0.04	-0.07	-0.07	-0.07	-0.07	0.10	0.39	1.00		
Th	-0.10	0.28	-0.08	0.64	0.09	-0.08	0.61	-0.08	-0.10	-0.11	-0.10	-0.11	-0.11	0.35	0.22	0.27	1.00	
U	-0.08	0.07	-0.06	0.68	0.12	-0.03	0.95	-0.03	-0.07	-0.09	-0.09	-0.09	-0.10	0.55	0.30	0.15	0.67	1.00



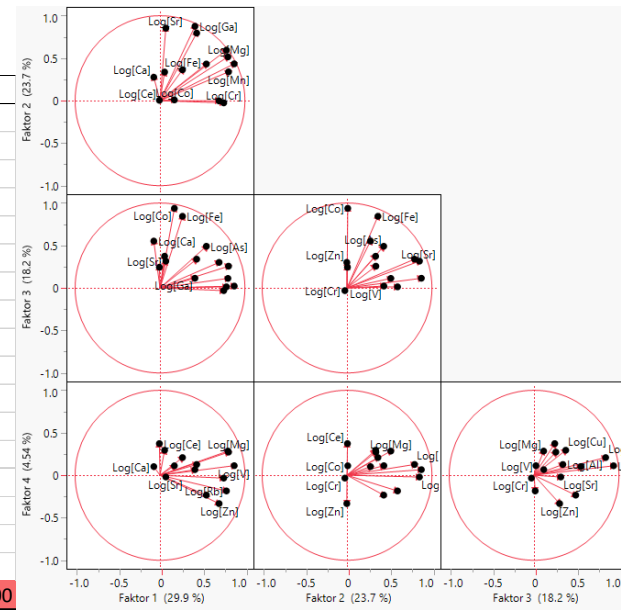
Correlation matrix for LA-ICP-MS trace element data for Heilongjian* (CHN)

Heilongjian* (CHN)	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	Th	U
Mg	1.00																	
Al	0.62	1.00																
Ca	0.56	0.97	1.00															
V	0.50	0.94	0.94	1.00														
Cr	0.18	0.41	0.52	0.56	1.00													
Mn	0.57	0.93	0.96	0.88	0.38	1.00												
Fe	0.62	0.55	0.44	0.44	0.07	0.45	1.00											
Co	0.36	0.45	0.51	0.30	0.09	0.64	0.35	1.00										
Cu	0.50	0.50	0.47	0.49	0.24	0.49	0.63	0.32	1.00									
Zn	0.68	0.80	0.77	0.82	0.30	0.79	0.58	0.38	0.57	1.00								
Ga	0.65	0.98	0.92	0.95	0.43	0.87	0.63	0.34	0.57	0.84	1.00							
As	0.10	0.45	0.50	0.50	0.32	0.44	0.26	0.17	0.39	0.51	0.46	1.00						
Rb	0.67	0.67	0.56	0.66	0.24	0.51	0.58	0.09	0.45	0.69	0.76	0.37	1.00					
Sr	0.54	0.45	0.46	0.32	0.31	0.42	0.62	0.39	0.36	0.34	0.48	0.34	0.35	1.00				
Zr	0.68	0.93	0.89	0.87	0.34	0.90	0.66	0.52	0.55	0.84	0.91	0.33	0.59	0.46	1.00			
Ce	0.70	0.30	0.24	0.11	-0.19	0.36	0.60	0.56	0.36	0.39	0.30	0.00	0.48	0.55	0.44	1.00		
Th	0.53	0.28	0.20	0.12	-0.06	0.20	0.82	0.22	0.37	0.35	0.36	0.21	0.44	0.77	0.34	0.62	1.00	
U	0.46	0.41	0.40	0.30	0.07	0.43	0.54	0.41	0.24	0.43	0.45	0.16	0.28	0.81	0.51	0.58	0.67	1.00



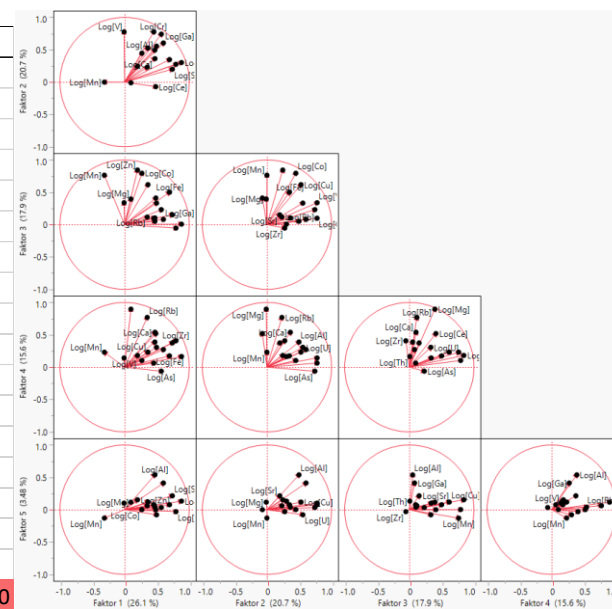
Correlation matrix for LA-ICP-MS trace element data for Kropfmühl (GER)

Kropfmühl (GER)	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	Th	U
Mg	1.00																	
Al	0.73	1.00																
Ca	-0.05	0.19	1.00															
V	0.97	0.69	-0.08	1.00														
Cr	0.87	0.56	-0.15	0.92	1.00													
Mn	0.98	0.71	-0.04	0.94	0.84	1.00												
Fe	0.05	0.66	0.39	-0.02	-0.08	0.04	1.00											
Co	0.04	0.64	0.37	-0.05	-0.10	0.04	0.99	1.00										
Cu	-0.02	0.29	0.01	-0.09	-0.25	-0.01	0.48	0.51	1.00									
Zn	0.01	-0.04	0.01	0.03	0.13	0.12	-0.03	0.05	-0.04	1.00								
Ga	0.78	0.94	0.05	0.76	0.68	0.72	0.52	0.49	0.19	-0.05	1.00							
As	0.26	0.75	0.18	0.23	0.21	0.25	0.83	0.83	0.32	0.19	0.69	1.00						
Rb	0.87	0.66	-0.10	0.90	0.89	0.80	0.03	0.01	-0.15	0.04	0.84	0.33	1.00					
Sr	0.08	0.70	0.20	0.02	-0.03	0.05	0.90	0.89	0.45	-0.05	0.63	0.85	0.15	1.00				
Zr	0.44	0.28	0.02	0.28	0.24	0.43	0.06	0.10	-0.07	-0.08	0.35	0.09	0.41	0.12	1.00			
Ce	0.00	-0.02	-0.04	-0.02	-0.02	0.04	0.03	0.03	0.05	0.10	-0.06	-0.03	-0.11	0.00	-0.04	1.00		
Th	0.14	0.50	0.12	0.10	0.15	0.21	0.65	0.67	0.29	0.30	0.40	0.71	0.08	0.62	-0.01	0.23	1.00	
U	0.35	0.32	0.02	0.20	0.11	0.40	0.28	0.36	0.25	0.28	0.26	0.26	0.15	0.21	0.61	0.27	0.44	1.00



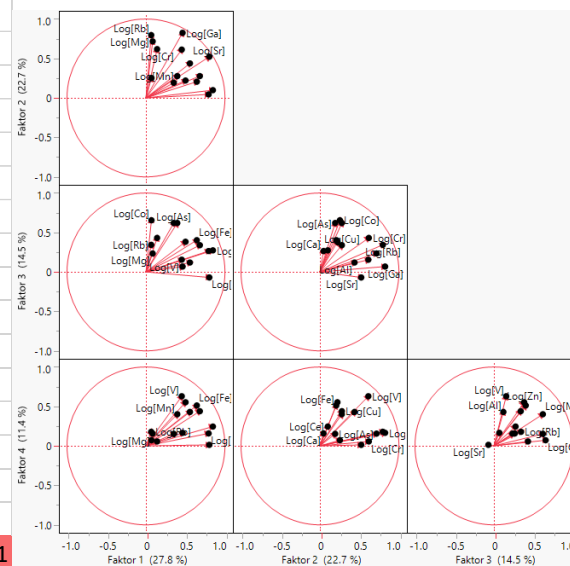
Correlation matrix for LA-ICP-MS trace element data for Brickville* (MAD)

Brickville* (MAD)	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	Th	U
Mg	1.00																	
Al	0.13	1.00																
Ca	0.24	0.81	1.00															
V	0.10	0.73	0.53	1.00														
Cr	0.08	0.81	0.66	0.94	1.00													
Mn	-0.02	0.01	0.02	0.17	0.09	1.00												
Fe	0.09	0.69	0.48	0.92	0.96	0.11	1.00											
Co	0.10	0.69	0.49	0.92	0.96	0.16	1.00	1.00										
Cu	0.10	0.69	0.49	0.92	0.97	0.12	1.00	1.00	1.00									
Zn	0.10	0.69	0.48	0.92	0.96	0.13	1.00	1.00	1.00	1.00								
Ga	0.11	0.84	0.67	0.93	0.99	0.09	0.96	0.96	0.97	0.96	1.00							
As	0.09	0.71	0.52	0.93	0.98	0.11	1.00	1.00	1.00	1.00	0.97	1.00						
Rb	0.77	0.39	0.46	0.29	0.29	-0.04	0.21	0.22	0.22	0.21	0.30	0.23	1.00					
Sr	0.11	0.71	0.49	0.91	0.96	0.10	1.00	1.00	1.00	1.00	0.97	1.00	0.24	1.00				
Zr	0.19	0.53	0.41	0.72	0.75	0.07	0.75	0.75	0.75	0.75	0.74	0.76	0.38	0.76	1.00			
Ce	0.43	0.48	0.38	0.57	0.62	0.05	0.66	0.66	0.66	0.66	0.64	0.66	0.60	0.67	0.52	1.00		
Th	0.08	0.72	0.50	0.92	0.97	0.09	1.00	0.99	0.99	0.99	0.97	1.00	0.23	1.00	0.75	0.67	1.00	
U	0.10	0.84	0.92	0.69	0.78	0.08	0.61	0.62	0.62	0.61	0.77	0.65	0.37	0.61	0.52	0.37	0.63	1.00



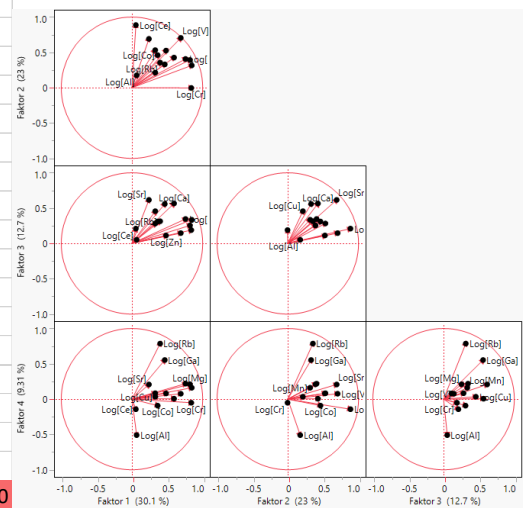
Correlation matrix for LA-ICP-MS trace element data for Balama (MOZ)

Balama (MOZ)	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	Th	U
Mg	1.00																	
Al	0.84	1.00																
Ca	0.18	0.44	1.00															
V	0.92	0.91	0.32	1.00														
Cr	0.94	0.90	0.30	0.96	1.00													
Mn	0.48	0.50	0.33	0.59	0.58	1.00												
Fe	0.05	0.13	0.30	0.14	0.13	0.48	1.00											
Co	0.34	0.26	0.09	0.29	0.34	0.67	0.11	1.00										
Cu	0.24	0.39	0.45	0.41	0.38	0.58	0.69	0.11	1.00									
Zn	0.25	0.29	0.32	0.29	0.30	0.56	0.88	0.18	0.68	1.00								
Ga	0.56	0.69	0.33	0.79	0.70	0.40	0.08	0.12	0.37	0.06	1.00							
As	0.49	0.49	0.34	0.52	0.55	0.52	0.52	0.22	0.72	0.52	0.30	1.00						
Rb	0.98	0.86	0.19	0.95	0.96	0.50	0.06	0.34	0.25	0.22	0.66	0.49	1.00					
Sr	0.27	0.47	0.55	0.50	0.41	0.27	0.12	0.01	0.36	0.06	0.73	0.27	0.35	1.00				
Zr	0.54	0.57	0.35	0.67	0.58	0.43	0.20	0.12	0.35	0.31	0.63	0.36	0.59	0.50	1.00			
Ce	-0.01	0.18	0.64	0.13	0.09	0.24	0.25	-0.01	0.38	0.21	0.14	0.32	0.00	0.60	0.22	1.00		
Th	0.22	0.42	0.70	0.42	0.37	0.58	0.53	0.10	0.67	0.52	0.38	0.54	0.24	0.63	0.49	0.78	1.00	
U	0.14	0.25	0.46	0.28	0.22	0.56	0.74	0.07	0.71	0.76	0.15	0.56	0.13	0.38	0.41	0.60	0.78	1



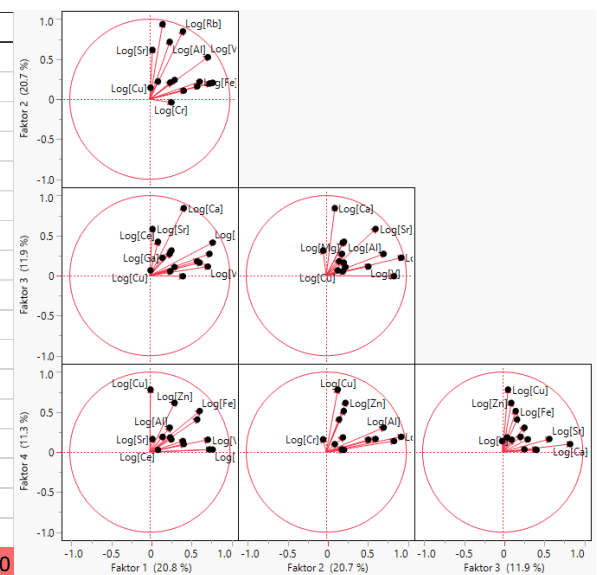
Correlation matrix for LA-ICP-MS trace element data for Skaland (NOR)

Skaland (NOR)	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	Th	U
Mg	1.00																	
Al	0.51	1.00																
Ca	0.79	0.71	1.00															
V	0.91	0.62	0.95	1.00														
Cr	0.70	0.65	0.82	0.87	1.00													
Mn	0.98	0.53	0.87	0.97	0.78	1.00												
Fe	0.99	0.42	0.74	0.88	0.63	0.97	1.00											
Co	0.92	0.49	0.84	0.92	0.71	0.95	0.92	1.00										
Cu	0.69	0.46	0.72	0.74	0.58	0.73	0.69	0.73	1.00									
Zn	0.71	0.52	0.86	0.86	0.72	0.79	0.69	0.78	0.62	1.00								
Ga	0.69	-0.04	0.13	0.36	0.09	0.58	0.75	0.54	0.32	0.17	1.00							
As	0.82	0.13	0.34	0.56	0.29	0.73	0.86	0.71	0.44	0.36	0.94	1.00						
Rb	0.68	-0.06	0.11	0.34	0.08	0.57	0.74	0.53	0.31	0.16	1.00	0.94	1.00					
Sr	0.71	0.16	0.18	0.37	0.11	0.58	0.75	0.55	0.33	0.17	0.97	0.93	0.96	1.00				
Zr	0.36	0.39	0.49	0.49	0.42	0.41	0.34	0.49	0.27	0.46	0.02	0.21	0.01	0.07	1.00			
Ce	0.08	0.09	0.03	0.08	0.05	0.07	0.09	0.12	0.12	0.04	0.09	0.18	0.09	0.11	0.26	1.00		
Th	-0.03	0.03	-0.02	-0.01	0.00	-0.03	-0.03	-0.01	0.12	-0.04	-0.02	-0.02	-0.02	0.00	-0.05	0.80	1.00	
U	-0.03	0.03	-0.02	-0.01	0.00	-0.03	-0.02	-0.01	0.12	-0.04	-0.02	-0.02	-0.02	0.00	-0.05	0.80	1.00	1.00



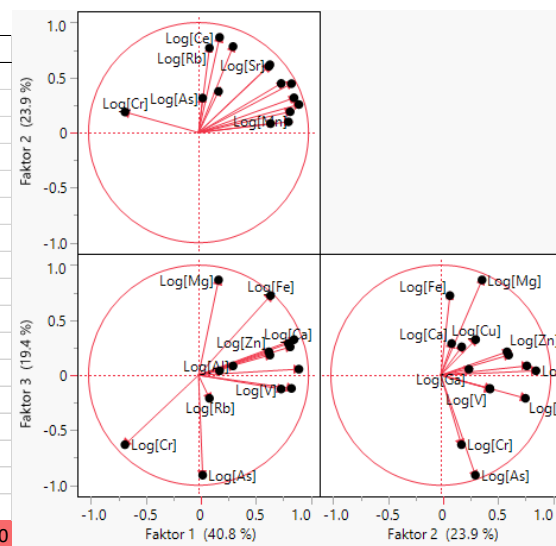
Correlation matrix for LA-ICP-MS trace element data for Tayginka (RUS)

Tayginka (RUS)	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	Th	U
Mg	1.00																	
Al	0.26	1.00																
Ca	0.65	0.24	1.00															
V	0.35	0.86	0.30	1.00														
Cr	0.65	0.11	0.35	0.19	1.00													
Mn	0.49	0.29	0.91	0.38	0.14	1.00												
Fe	0.28	0.56	0.32	0.66	0.20	0.39	1.00											
Co	0.34	0.31	0.30	0.43	0.42	0.33	0.51	1.00										
Cu	-0.05	0.08	-0.05	0.00	-0.02	-0.06	0.16	0.14	1.00									
Zn	0.00	0.06	0.01	0.08	0.23	0.01	0.08	0.71	0.24	1.00								
Ga	0.09	0.65	0.09	0.45	0.05	0.10	0.25	0.24	0.02	0.17	1.00							
As	0.17	0.59	0.15	0.68	-0.02	0.20	0.48	0.19	0.13	-0.04	0.30	1.00						
Rb	0.17	0.76	0.11	0.64	0.04	0.16	0.39	0.28	0.04	0.10	0.91	0.45	1.00					
Sr	0.26	0.84	0.26	0.83	0.12	0.24	0.56	0.30	-0.01	0.04	0.59	0.64	0.65	1.00				
Zr	0.30	0.81	0.22	0.93	0.12	0.27	0.64	0.34	0.02	0.01	0.38	0.73	0.56	0.89	1.00			
Ce	0.18	0.50	0.15	0.45	0.01	0.12	0.27	0.12	-0.05	-0.02	0.28	0.44	0.37	0.40	0.41	1.00		
Th	0.34	0.78	0.24	0.91	0.15	0.27	0.62	0.33	0.00	0.00	0.36	0.73	0.53	0.88	0.98	0.42	1.00	
U	0.30	0.81	0.22	0.93	0.13	0.27	0.64	0.34	0.01	0.01	0.38	0.73	0.56	0.91	1.00	0.41	0.98	1.00



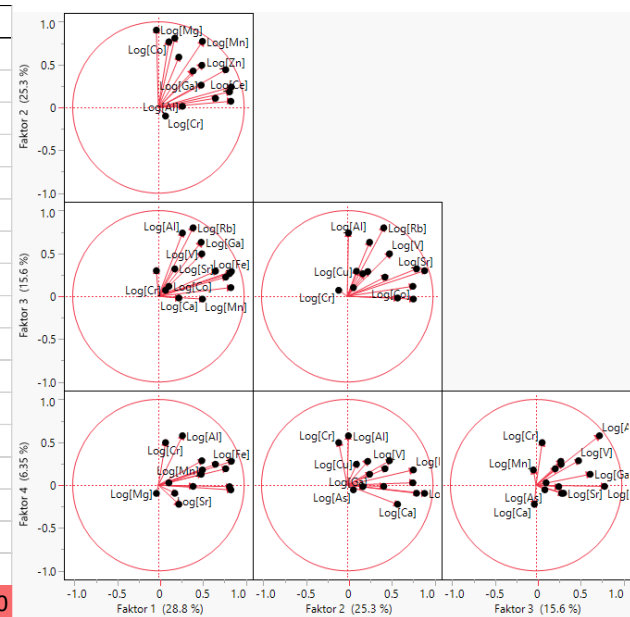
Correlation matrix for LA-ICP-MS trace element data for Ragedara (LKA)

Ragedara (LKA)	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	Th	U
Mg	1.00																	
Al	0.44	1.00																
Ca	0.09	0.15	1.00															
V	-0.11	0.59	0.27	1.00														
Cr	-0.29	-0.02	-0.67	0.02	1.00													
Mn	0.21	0.18	0.89	0.19	-0.75	1.00												
Fe	0.26	0.24	0.82	0.18	-0.86	0.93	1.00											
Co	-0.20	0.47	0.20	0.96	0.04	0.12	0.11	1.00										
Cu	0.15	0.52	0.61	0.24	-0.47	0.58	0.70	0.08	1.00									
Zn	0.43	0.57	0.73	0.49	-0.49	0.73	0.67	0.34	0.55	1.00								
Ga	0.18	0.35	0.82	0.36	-0.73	0.93	0.96	0.27	0.74	0.73	1.00							
As	-0.31	0.39	-0.18	0.85	0.45	-0.30	-0.32	0.89	-0.16	0.02	-0.15	1.00						
Rb	0.79	0.36	-0.27	0.01	0.27	-0.20	-0.22	-0.04	-0.15	0.15	-0.19	0.05	1.00					
Sr	0.62	0.60	0.62	0.42	-0.42	0.63	0.66	0.30	0.62	0.78	0.70	0.01	0.45	1.00				
Zr	0.00	0.60	-0.12	0.83	0.43	-0.25	-0.28	0.80	0.02	0.21	-0.10	0.91	0.35	0.28	1.00			
Ce	0.76	0.55	-0.02	0.21	0.16	-0.02	-0.05	0.11	0.14	0.40	0.00	0.14	0.92	0.67	0.50	1.00		
Th	0.71	0.37	-0.31	0.06	0.36	-0.28	-0.31	0.01	-0.16	0.12	-0.27	0.14	0.99	0.42	0.45	0.94	1.00	
U	-0.01	0.62	0.27	0.99	-0.01	0.22	0.22	0.96	0.23	0.51	0.39	0.83	0.12	0.50	0.83	0.30	0.16	1.00



Correlation matrix for LA-ICP-MS trace element data for Zavallia (UKR)

Zavallia (UKR)	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	Th	U
Mg	1.00																	
Al	0.18	1.00																
Ca	0.49	0.15	1.00															
V	0.26	0.73	0.25	1.00														
Cr	0.04	0.59	0.04	0.56	1.00													
Mn	0.32	0.66	0.56	0.80	0.47	1.00												
Fe	0.07	0.49	0.18	0.68	0.20	0.67	1.00											
Co	0.64	0.57	0.53	0.74	0.47	0.80	0.42	1.00										
Cu	0.15	0.63	0.20	0.82	0.55	0.68	0.49	0.63	1.00									
Zn	0.16	0.74	0.27	0.89	0.50	0.86	0.79	0.67	0.74	1.00								
Ga	0.05	0.28	0.04	0.20	0.06	0.25	0.13	0.19	0.25	0.28	1.00							
As	-0.08	0.41	0.10	0.68	0.20	0.54	0.65	0.40	0.61	0.66	0.18	1.00						
Rb	0.17	0.72	0.18	0.51	0.22	0.47	0.35	0.42	0.44	0.49	0.23	0.33	1.00					
Sr	0.41	0.52	0.40	0.31	0.07	0.38	0.24	0.43	0.22	0.29	0.09	0.12	0.87	1.00				
Zr	0.17	0.69	0.25	0.76	0.39	0.73	0.73	0.59	0.63	0.84	0.16	0.48	0.47	0.34	1.00			
Ce	0.07	0.57	0.35	0.52	0.24	0.47	0.41	0.37	0.51	0.56	0.14	0.39	0.70	0.60	0.46	1.00		
Th	0.21	0.77	0.27	0.81	0.62	0.76	0.44	0.69	0.83	0.77	0.18	0.43	0.62	0.42	0.76	0.62	1.00	
U	0.20	0.44	0.18	0.43	0.19	0.43	0.27	0.42	0.43	0.42	0.15	0.18	0.27	0.19	0.72	0.28	0.66	1.00



LA ICP MS trace element data

LA-ICP-MS	Analyte (µg g ⁻¹)																
sample ID	Mg	Al	Ca	V	Cr	Mn	Fe	Co	Cu	Zn	Ga	As	Rb	Sr	Zr	Ce	U
1A.1	426.71	6961.64	193.59	128.66	94.98	417.94	4410.92	2.66	25.75	49.48	107.21	21.63	126.83	100.15	9.81	34.4	3.66
1A.2	347.58	9029.64	231.29	113.45	90.92	270.05	3493.79	4.21	23.09	39.3	111.23	16.17	110.18	139.3	134.27	44.96	4.41
1A.3	519.91	7312.21	400.04	152.65	108.31	232.7	3817.1	4.14	26.69	59.24	132.78	22.95	174.8	158.67	139.7	36.62	5.32
1A_1	445.27	7091.84	293.33	194.33	108.26	434.23	6332.85	5.77	32.45	69.2	132.79	25.54	207.34	103.28	10.45	65.62	3.86
1A_10	371.14	5715.44	266.58	111.18	71.18	311.52	5107.68	5.95	20.89	27.77	79.84	17.95	112.4	110.87	11.74	57.93	3.45
1A_11	317.63	4597.6	222.18	131.17	78.87	441.39	5472.79	4.57	33.02	46.9	76.67	21.11	136.39	89.4	21.71	89.3	5.56
1A_12	206.67	4563.44	187.9	100.04	66.68	174.33	3165.35	3.06	16.64	26.06	82.92	11.66	83.18	59.32	11.12	27.49	2.21
1A_13	367.95	4540.975	425.28	163.54	91.21	383.86	5891.16	5.35	25.42	41.79	114.34	18.2	171.45	151.49	38.75	89.1	4.2
1A_14	348.76	4518.51	191.27	89.1	61.95	334.81	4237.66	3.8	21.91	29.48	66.05	15.49	100.15	89.3	6.17	41.61	3.27
1A_15	650.5	9189.16	410.94	167.63	80.51	465.45	7523.88	11.13	27.08	61.05	107.17	17.8	164.41	138.97	220.09	46.78	4.09
1A_2	420.93	5357.57	308.35	134.22	79.66	341.95	5061.71	4.33	25.77	62.43	109.33	27.25	182.9	124	12.02	56.63	3.11
1A_3	539.26	5235.46	280.86	145.43	92.42	497.66	7279.63	6.53	25.98	86.52	104.31	20.42	158.4	113.28	11.57	60.8	3.73
1A_4	367.87	3339.72	240.41	108.89	74.24	305.56	4751.46	4.01	27.16	47.38	74.53	18.7	118.88	96.65	22.21	40.11	3.17
1A_5	354.46	5124.24	374.01	141.69	120.61	206.61	4165.33	4.15	29.82	51.18	92.19	19.09	159.64	120.95	8.57	65.43	3.55
1A_6	568.43	4882	395.78	179.31	103.88	517.26	8156.98	10.23	39.81	90.8	98.1	25.94	172.02	150.87	8.63	227.14	5.03
1A_7	561.23	5525.465	478.32	189.32	107.01	957.84	12042.55	9.23	41.55	112.06	108.51	24.11	185.8	122.43	9.11	102.53	4.23
1A_8	456.93	6168.93	261.3	141.19	85.47	581.67	6968.51	6.06	22.29	88	93.3	21.28	141.02	97.55	6.57	47.33	3.34
1A_9	203.54	0.00595	393.57	76.42	60.29	146.39	2795.32	2.95	131.83	52.81	52.1	18.32	82.12	79.73	11.7	534.71	31.27
21G.1	10.85	91.81	7.17	0.635	35.13	2.58	42.41	0.02015	0.078	4.53	0.301	3.1	0.486	1.26	0.0707	0.063	0.00145
21G.2	75.69	328.62	12	1.87	35.14	7.49	280.12	0.204	0.01845	6.48	0.818	3.39	0.713	2.46	0.099	0.105	0.00249
21G.3	44.97	2836.78	16.66	36.13	38.79	3.48	101.94	0.02015	0.203	4.72	26.61	3.37	40.02	14.59	5.5	1.59	0.238
21G.4	8.46	921.78	63.75	5.15	37.2	3.25	33.49	0.051	0.255	4.18	6.44	4.43	3.73	16.06	61.72	84.72	1.53
21G_1	43.47	2956.85	59.69	44.66	39.42	5.93	98.15	0.52	3.74	9.23	34.35	3.78	48.11	55.9	12.78	7	0.8
21G_2	81.25	5592.1	77.64	78.15	44.95	6.3	184.59	0.41	2.64	4.92	62.5	4	89.32	63.39	11.64	6.86	0.55
21G_3	72.12	1479.72	40.9	15.42	33.24	13.38	486.52	0.63	0.82	12.78	10.77	4.72	10.89	12.66	2.85	0.56	0.078
21G_4	16.37	965.73	51.49	11.35	30.02	19.83	39.74	0.02015	2.02	5.11	11.73	3.28	11.34	26.08	8.18	5.39	0.32

21G_5	15.39	1000.97	45.17	10.76	29.11	3.23	36.83	0.166	0.21	3.35	9.85	3.36	11.56	20.63	5.12	11.24	0.221
21G_6	788.06	2956.93	29.98	23.63	31.85	134.59	5683.49	6.77	0.36	20.46	11.34	3.29	8.03	9.68	2.18	1.6	0.082
21G_7	731.56	7163.03	46.09	84.69	38.19	120.8	4428.45	5.26	0.87	15.88	56.19	3.21	89.85	34.75	3.99	0.7	0.102
21H.1	35.26	1854.88	16.99	21.04	38.52	3.87	132.43	0.081	1.08	3.16	11.73	3.26	18.54	14.87	1.93	0.311	0.347
21H.2	4.17	160.32	33.6	10.44	39.24	4	15.1	0.02015	0.72	4.71	1.73	5.23	0.88	20.27	11.56	4.03	1.42
21H.3	8.47	677.92	18.1	9.68	38.3	1.49	13.95	0.02015	1.47	2.24	4.89	3.73	6.06	14.5	1.46	0.358	0.393
21H.4	22.87	1824.35	26.1	17.01	38.97	2.79	44.11	0.096	0.92	3.47	10.56	3.26	17.38	10.25	16.2	2.27	0.53
21H_1	39.37	2480.62	21.51	33.64	27.52	4.6	95.21	0.061	0.332	2.37	21.75	5.38	42.47	12.25	11.22	6.21	2.29
21H_2	48.43	2748.74	27.32	30.16	27.13	4.23	118.8	0.087	0.405	3.85	24.77	4.86	44.09	17.75	5.43	1.2	0.29
21H_3	20.42	1158.56	20.85	16.43	25.63	2.36	44.26	0.061	0.391	3.65	10.66	4.08	16.87	10.21	6.13	1.13	0.477
21H_4	37.35	1777.98	20.02	20.13	25.27	3.01	78.51	0.058	1.17	4.32	20.12	4.43	26.19	19.88	5.45	1.14	0.59
21H_5	20.17	867.06	23.22	8.31	23.78	3.36	58.63	0.225	0.287	4.45	6.83	3.74	10.37	6.91	4.88	6.07	0.211
21H_6	28.31	1818.14	28.87	23.99	29.99	3.54	77.46	0.041	0.62	4.32	17.72	3.6	29.9	20.62	9.47	1.51	0.69
21H_7	14.91	830.46	52.18	9.18	27.33	3.22	32.49	0.145	0.52	4.57	7.17	4.23	9.32	15.83	4.57	3.3	0.233
21I.1	1.85	86.56	15.28	2.09	35.19	0.3	7.22	0.316	1	2.49	1.06	21.15	0.815	7.53	2.92	0.97	0.189
21I.2	1.36	73.65	14.6	6.49	34.99	1.76	4.7	1.19	0.333	2.36	1.42	18.38	0.432	9.93	0.603	0.95	0.365
21I.3	1.53	84.25	17.9	2.05	35.02	0.188	0.4325	2.37	0.86	0.71	1.55	22.94	0.435	7.83	1.67	1.2	0.288
21I.4	2.89	152.82	14.69	5.59	34.97	0.372	9.32	2.11	0.01845	1.05	1.09	18.38	1.03	3.6	0.97	0.501	0.264
21I_1	3.58	226.32	25.21	2.44	23.8	1.51	19.99	0.099	1.59	2.62	1.5	9.8	1.98	5.03	1.34	1.03	0.161
21I_2	2.13	58.4	15.82	0.85	22.89	1.15	15.33	0.26	2.67	2.33	0.39	16.35	0.28	2.91	0.41	0.7	0.132
21I_3	43.01	472.93	17.2	4.22	21.5	7.91	402.21	0.52	1.33	4.37	1.99	4.7	3.09	3.39	1.51	0.57	0.1
21I_4	20.82	539.86	11.21	4.33	20.99	2.55	79.78	1.09	0.2	2.27	2.99	4.71	4.87	3.19	1.03	0.65	0.055
21I_5	3.7	329.65	7.44	2.47	20.92	0.4	11.23	0.81	0.166	1.36	1.53	6.67	2.76	2.26	3.13	0.75	0.168
21I_6	1.5	47.47	18.94	1.57	21.19	6.2	11.53	6.77	0.53	0.75	1.08	12.89	0.187	6.08	0.79	0.98	0.184
21I_7	12.34	536.15	13.89	4.73	20.06	1.71	29.74	1.44	3.3	6.66	3.4	3	5.21	4.02	3.3	3.98	0.088
21C.1	2	133.31	3.62	8.94	41.34	5.25	69.41	0.102	4.86	2.82	0.418	0.078	0.096	0.297	4.15	1.11	0.0372
21C.2	1.54	150.58	3.31	3.78	42.56	6.18	106.67	0.068	5.43	1.78	0.383	0.177	0.0466	0.68	8.01	1.99	0.0556
21C.3	1.52	264.91	3.92	3.52	41.14	6.92	124.78	0.02015	5.74	1.53	0.526	0.102	0.027	0.529	6.13	1.63	0.0448
21C_1	0.74	123.18	2.86	2.83	26.18	209.06	108.66	0.48	5.64	1.19	6.27	0.112	0.057	0.46	4.94	1.94	0.0377
21C_10	0.71	113.15	3.01	1.97	31.03	2.87	73.57	0.043	3.18	0.62	0.3	0.093	0.061	0.11	3.14	0.96	0.0202

21C_11	0.85	298.78	4.59	6.09	26.6	15.46	139.91	0.206	5.3	1.33	0.78	0.147	0.033	0.6	8.45	2.3	0.049
21C_12	0.89	95.43	2.26	2.18	24.81	2.44	67.36	0.171	2.85	1.13	0.17	0.095	0.0218	0.111	3.12	1.01	0.0259
21C_13	1.31	431.44	3.78	6.54	32.57	8.71	294.17	0.5	9.67	2.16	0.68	0.247	0.0253	0.34	7.79	3.69	0.074
21C_14	1.56	947	3.57	10.43	37.65	10.5	83.29	0.108	5.79	1.39	1.03	0.189	0.0135	0.122	3.1	1.05	0.0274
21C_15	4.27	2621.82	6.99	13.61	39.63	9.52	233.08	0.43	15.38	6.35	2.71	0.266	0.064	0.28	8.11	1.88	0.069
21C_2	0.67	105.95	1.51	2.68	24.89	7.54	81.85	0.198	3.43	1.21	0.31	0.093	0.0262	0.29	4.35	1.63	0.024
21C_3	1.51	515.79	3.76	9.17	33.15	9.09	197.69	0.269	6.25	1.43	0.81	0.175	0.045	0.42	9.03	2.61	0.066
21C_4	0.91	436.86	3.75	3.81	31.88	4.55	126.05	0.186	4.85	1.51	0.62	0.204	0.019	0.208	7.68	2.11	0.0348
21C_5	0.91	207.03	3.23	3.44	30.22	4.61	138.46	0.175	4.69	1.05	0.44	0.221	0.0215	0.22	4.74	1.68	0.052
21C_6	0.42	87.76	2.85	1.13	24.92	1.46	49.09	0.02015	1.37	0.43	0.123	0.202	0.0146	0.148	2.22	0.78	0.0152
21C_7	1.03	370.66	2.41	5.86	28.02	5.4	140.78	0.193	7.13	1.63	0.64	0.165	0.025	0.29	6.4	2.09	0.0407
21C_8	3.27	424.05	3.71	5.57	28.44	4.99	116.26	0.113	4.58	1.27	0.67	0.174	0.0238	0.22	7.17	2.07	0.0353
21C_9	1.88	439.23	3.42	3.49	27.75	6.19	135.25	0.195	5.5	2.08	0.59	0.141	0.032	0.27	7.92	2	0.056
21N.1	1.45	741.93	4.47	4.97	58.5	3.78	144.11	1.25	5.14	1.05	0.984	0.0616	0.0391	0.372	6.49	1.59	0.0459
21N.2	0.938	211.02	6.55	2.51	47.62	2.28	67.18	0.28	4.14	1.21	0.517	0.0708	0.018	0.239	5.5	0.881	0.0336
21N.3	1.61	275.69	12.33	2.83	57.59	5.71	147.68	0.186	5.17	1.14	0.545	0.156	0.0241	0.295	6.56	1.52	0.0419
21N_1	0.7	175.25	1.91	2.4	26.88	4.09	102.43	0.05	4.34	1.4	0.36	0.184	0.00605	0.168	4.52	1.63	0.0286
21N_10	0.54	103.84	2.15	2.09	25.89	3.14	79.53	0.235	3.75	0.85	0.27	0.157	0.0224	0.31	3.33	1.22	0.0255
21N_11	0.91	404.42	3.14	5.89	26.76	6.48	334.86	0.7	5.05	1.58	0.53	0.175	0.143	0.17	6.18	1.92	0.049
21N_12	0.92	284.41	3.92	3.05	30.45	5.37	124.71	0.149	4.33	0.8	0.44	0.176	0.0153	0.28	6.35	2.53	0.0352
21N_13	0.87	351.39	3.93	3.58	27.93	4.44	111.29	0.222	4.71	1.37	0.56	0.23	0.029	0.27	10.17	3.08	0.0347
21N_14	1.84	995.73	3.98	11.6	28.79	4.9	123.51	0.178	6.06	2.03	1.51	0.139	0.0214	0.43	10.33	1.98	0.0421
21N_15	1.87	588.79	3.72	6.44	31.45	7.65	187.22	0.259	8	2.17	0.92	0.144	0.048	0.43	8.39	2.72	0.056
21N_2	7.94	538.41	3.39	5.47	29.67	8.21	176.96	0.188	7.28	1.68	1	0.275	0.0127	0.69	8.57	3.5	0.06
21N_3	0.86	142.51	3.32	3.37	26.94	5.41	109.91	0.237	5.15	1.33	0.46	0.169	0.00605	0.54	8.15	2.38	0.0419
21N_4	0.98	435.25	1.9	2.77	24.24	4.02	130.4	0.16	5.36	1.27	0.52	0.304	0.029	0.146	5.92	1.54	0.0395
21N_5	1.86	1947.16	3.76	11.52	35.54	7.46	263.04	0.2	7.41	2.45	2.07	0.21	0.071	0.34	10.32	2.37	0.079
21N_6	0.73	174.28	5.61	14.95	27.39	3.17	152.28	0.16	3.73	0.61	0.3	0.049	0.00605	0.155	4.25	1.43	0.062
21N_7	0.63	149.2	3.24	2.16	25.4	2.42	81.81	0.02015	2.6	0.76	0.28	0.169	0.0165	0.136	2.93	0.99	0.0157
21N_8	2.23	485.84	3.88	8.99	30.35	8.76	163.25	0.373	9.24	6.29	0.84	0.226	0.064	0.31	7.57	2.47	0.053

21N_9	0.78	174.28	1.43	4.04	29.4	5.18	126.12	0.167	4.74	0.99	0.44	0.145	0.00605	0.28	11.58	2.38	0.052
22C.1	1.21	252.55	4.65	2.82	36.26	4.02	60.11	0.38	2.22	1.97	0.507	0.096	0.0336	0.396	12.63	2.8	0.039
22C.2	1.18	282.11	3.54	2.53	35.2	4.92	58.51	0.202	2.11	2.23	0.54	0.113	0.0353	0.82	11.71	2.71	0.0515
22C.3	4.15	871.3	5.34	5.12	38.31	7.5	120.47	0.39	3.19	2.98	1.77	0.243	0.0521	0.86	20.12	4.68	0.072
3B.3	0.857	425.78	6.53	15.28	54.99	21.04	357.39	0.87	5	1.04	1.93	0.157	0.0495	0.689	4.83	1.76	0.185
3B_1	0.647	301.64	8.67	4.81	33.6	16.1	201.61	0.52	2.67	0.91	1.08	0.27	0.0635	0.82	1.69	1.94	0.154
3B_10	0.498	166.96	5.49	3.48	32.11	15.16	139.21	0.47	11.87	0.367	0.94	0.1591	0.0134	0.481	1.94	1.25	0.0685
3B_11	0.89	351.74	6.27	17.85	33.38	191.67	200.21	28.24	18.87	5.82	0.85	1.351	0.0437	0.32	1.2	0.66	0.1093
3B_12	0.678	233.68	6	3.81	32.58	12.43	123.95	0.52	5.85	1.55	0.66	0.176	0.023	0.129	7.25	0.93	0.144
3B_13	0.58	131.56	6.67	8.51	34.9	65.21	159.42	1.9	4.45	1.03	2.74	0.154	0.0192	0.573	1.58	1.19	0.344
3B_14	2.19	833.28	6.72	14.33	39.17	23.43	229.32	0.9	4.91	3.04	1.58	0.227	0.041	0.274	3.76	1.17	0.174
3B_15	0.566	400.21	5.45	6.82	32.29	8.65	89.91	0.295	1.54	0.82	0.91	0.1232	0.00605	0.224	1.078	0.57	0.0348
3B_2	0.521	196.67	8.22	2.6	31.84	10.68	122.35	0.184	2.29	0.673	0.7	0.1343	0.00605	0.228	1.38	0.87	0.0697
3B_3	0.692	553.11	6.84	8.31	33.76	12.95	151.08	0.83	4.87	0.97	1.42	0.1687	0.022	0.229	6.03	0.99	0.145
3B_4	9.57	1318.71	8.22	5.59	71.34	28.69	181.6	1.01	5.54	1.5	3.38	0.218	0.0426	0.621	3.18	1.61	0.1234
3B_5	0.765	378.19	6.51	4.33	36.1	15.62	193.59	0.83	3.33	1.48	0.84	0.19	0.00605	0.142	1.71	0.92	0.1023
3B_6	0.61	175.29	7.57	5.71	37.41	259.19	171.3	2.17	10.37	1.09	3.62	0.1518	0.0536	0.461	2.65	1.53	0.164
3B_7	0.62	309.97	6.12	7.44	35.63	9.42	172.09	0.68	3.46	0.96	0.76	0.1161	0.0412	0.093	1.21	0.5	0.0759
3B_9	1.27	2057.8	8	36.53	47.02	26.94	221.94	1.18	6.59	1.61	4.92	0.329	0.052	0.384	4.38	1.29	0.1144
3B_8	0.82	296.27	8.46	5.75	37.18	12.8	114.19	0.15	2.76	0.516	0.61	0.1491	0.0302	0.117	1.038	0.62	0.0497
21M_11	0.75	160.92	4.73	2.23	29.99	4.04	83.99	0.145	4.45	0.83	0.39	0.097	0.0222	0.38	5.01	1.74	0.032
21M_12	1.71	363.16	5.66	7.31	30.7	20.64	194.79	0.43	5.75	1.4	1.06	0.144	0.065	0.55	11.06	2.64	0.069
21M_13	1.28	262.8	5.81	7.35	28.85	6.3	127.59	0.3	4.44	4.43	0.56	0.122	0.035	0.36	7.24	2.97	0.05
21M_14	1.4	358.96	5.9	5.03	30.34	8.2	158.89	0.28	4.95	2.24	0.92	0.155	0.069	0.6	6.28	2.62	0.041
21M_15	1.13	244.83	4.31	4.23	26.79	10.02	240.86	0.41	6.41	2.57	0.63	0.116	0.05	0.47	7.1	2.65	0.07
21M_2	14.29	2756.34	5.49	12.64	34.96	16.84	258.94	0.78	9.11	3.11	7.14	0.78	0.035	1.58	10.34	2.74	0.096
21M_3	1.08	257.12	3.02	3.68	26.51	6.62	119.8	0.217	6.22	1.95	0.65	0.114	0.0152	0.51	6.32	1.85	0.039
21M_4	1.18	466.34	6.23	3.91	27.83	10.29	165.78	0.181	8.95	1.36	1.51	0.286	0.0253	0.63	6.81	1.98	0.051
21M_5	1.43	343.73	4.4	4.67	29.04	6.91	141.41	0.37	4.88	1.06	0.7	0.22	0.028	0.47	6.54	2.02	0.074
21M_6	1.75	413.15	5.05	5.79	28.84	8.34	162.02	0.39	4.86	1.16	0.88	0.121	0.118	0.82	6.53	2.21	0.06

21M_7	1.29	301.18	4.69	3.92	29.15	7.74	152.73	0.37	5.61	1.69	0.67	0.103	0.031	0.51	9.01	3.1	0.081
21M_8	2	531.71	5.98	9.45	35.25	11.35	205.53	0.47	8.79	3.94	1.23	0.179	0.22	0.45	7.07	2.57	0.104
21M_9	1.87	366.9	6.93	12.02	35.56	11.17	190.64	0.58	8.74	4.02	0.99	0.156	0.075	0.72	8.69	3.01	0.087
21M.1	1.35	573.44	5.74	4.38	43.27	8.95	158.35	0.165	7.04	1.05	1.06	0.115	0.0484	0.89	7.08	1.84	0.0492
21M.2	0.653	37.99	3.74	0.838	40.11	3.65	47.94	0.189	1.7	0.52	0.139	0.135	0.013	0.159	2.43	0.504	0.0153
21M.3	1.22	462.91	5.04	7.71	42.07	5.07	112.93	0.25	4.91	1.5	1.1	0.103	0.0389	0.54	7.38	1.13	0.043
22C_1	1.54	333.02	3.67	4.46	26	142.21	312.35	1.37	6.06	2.02	3.19	0.17	0.131	0.4	50.75	3.08	0.13
22C_10	1.08	238.66	3.7	2.38	25.52	6.48	93.43	0.195	3.25	1.49	0.71	0.127	0.0398	0.52	14.13	3.22	0.0425
22C_11	1.18	413.78	4.67	4.4	25.8	22.08	127	0.56	3.83	1.14	0.98	0.208	0.0498	0.85	19.99	18.4	1.05
22C_12	0.97	246.81	4.27	3.52	28.35	7.18	101.71	0.245	2.74	1.66	0.5	0.306	0.0248	0.39	12.86	2.74	0.071
22C_13	1.4	292	4.18	2.05	24.38	9.54	112.68	0.282	3.25	1.94	0.51	0.185	0.0458	0.32	11.68	2.75	0.0383
22C_14	1.49	408.01	4.26	2.39	27.36	6.76	90.87	0.185	2.38	1.16	0.56	0.19	0.0219	0.36	13.64	2.43	0.0292
22C_15	1.21	298.86	3.96	3.54	26.94	5.05	83.71	0.19	2.22	1.18	1.23	0.135	0.0494	0.68	12.53	2.65	0.056
22C_2	1.38	538.91	6.02	5.29	26.9	20.63	162.27	0.323	4.27	2.21	1.96	0.149	0.0358	0.66	16.54	3.31	0.055
22C_3	1.54	414.92	4.29	5.18	26.15	7.54	109.68	0.5	4.68	3.6	0.78	0.182	0.155	0.46	19.29	3.32	0.121
22C_4	15.71	1815.76	9.16	13.28	33.88	36.78	422.01	2.27	29.51	54.11	4.33	0.318	2.35	0.96	15.67	2.27	0.109
22C_5	1.37	594.41	4.05	4.35	26.53	6.34	108.1	0.293	3.41	2.26	0.87	0.204	0.0485	0.46	14.46	2.93	0.0429
22C_6	1.59	568.15	4.6	4.87	26.86	12.28	121.25	0.412	4.89	4.75	1.25	0.143	0.068	0.52	18.84	3.91	0.0452
22C_7	1.06	361.35	3.78	2.71	25.68	7.67	87.52	0.314	3.06	1.28	0.83	0.063	0.0251	0.48	13.07	2.57	0.033
22C_8	1.36	509.25	3.73	6.14	28.65	12.31	128.93	0.368	6.16	3.73	1.09	0.152	0.067	0.53	15.89	3.5	0.047
22C_9	1.5	489.11	4.33	2.61	25.08	6.3	108.64	0.286	3.36	1.71	0.64	0.456	0.0394	0.33	15.19	4.23	0.064
21A.1	1.41	264.62	8.03	19.58	49.39	1.28	281.55	0.077	9.75	6.7	1.27	0.166	0.105	2.85	3.17	0.84	0.263
21A.2	1.13	297.05	7.61	33.54	49.2	2.42	356.9	0.078	8.32	7.02	1.19	0.245	0.104	1.64	1.24	0.54	0.226
21A.3	1.44	418.2	16.01	40.08	47.88	2.17	759.4	0.262	11.05	11.11	1.45	0.348	0.206	1.84	2.67	0.6	0.347
21A_1	10.75	992.6	13.72	73	47.64	5.22	905.31	0.24	14.13	13.97	2.55	0.468	0.141	3.01	4.42	2.12	0.675
21A_10	0.71	429.09	12.68	35.02	35.29	1.05	458.14	0.163	5.98	11.35	0.67	0.163	0.039	0.88	1.16	0.5	0.279
21A_11	1.66	493.64	23.65	261.82	47.44	5.17	3404.22	0.301	21.78	25.27	3.51	4.59	0.29	8.12	1.69	4.21	2.44
21A_12	1.14	358.49	18.08	34.68	49.18	3.42	606.57	0.02015	15.55	9.93	1.61	0.345	0.108	1.78	2.19	1.08	0.373
21A_13	2.6	1013.38	12.83	75.61	59.18	2.59	325.16	0.122	5.99	7.33	2.6	0.223	0.0594	1.11	1.58	0.72	0.202
21A_14	1.51	396.71	15.19	34.01	41.19	2.41	866.28	0.158	9.2	15.82	1.04	0.293	0.0607	1.37	1.29	0.77	0.41

21A_15	0.86	364.13	11	39.45	38.67	2.35	463.94	0.02015	8.21	7.83	1.17	0.303	0.066	1.36	1.45	0.86	0.25
21A_2	1.07	416.81	9.04	23.99	35.77	0.91	389.19	0.107	7.6	9.86	1.02	0.1267	0.0513	1.25	1.09	0.84	0.229
21A_3	1.5	398.86	14.75	37.59	42.68	3.6	678.07	0.202	14.05	8.59	2.99	0.356	0.224	3.04	2.78	1.74	0.436
21A_4	2.64	341.82	13.79	34	50.49	1.98	299.9	0.02015	6.98	4.64	4.8	0.202	0.72	2.39	1.51	1.14	0.258
21A_5	3.68	3279.83	26.05	413.3	55.73	11.95	5443.82	0.65	61.34	318.86	5.42	0.79	0.178	1.32	2.38	1.04	2.64
21A_6	2.67	498.68	15.81	83.5	42.8	4.72	1542.15	0.47	19.28	17.61	4.28	0.64	0.456	3.44	2.58	1.98	0.791
21A_7	1.11	222.53	12.69	17.91	38.41	2.4	453.91	0.02015	7.61	7.95	3.29	0.241	0.093	3.51	1.02	2.45	0.227
21A_8	0.89	312.58	14.17	32.96	37.65	2.3	405.28	0.172	7.4	11.4	1.21	0.264	0.077	1.54	1.24	0.83	0.375
21A_9	4.14	225.13	13.74	17.73	39.92	1.4	340.71	0.282	8.24	5.59	1.1	0.227	0.067	1.53	2.41	0.82	0.273
22A.1	1.38	2.09	2.51	0.0393	37.44	0.072	6.09	0.08	0.106	0.0445	0.0078	0.18	0.0476	0.0362	0.0115	0.035	0.00073
22A.2	6.08	16.8	7.01	1.32	32.5	0.63	13.73	0.02015	0.41	0.0445	0.117	0.162	0.356	0.363	0.647	0.214	0.0329
22A.3	3.48	6.21	7.06	0.731	33.56	0.69	5.44	0.05	0.119	0.0445	0.0142	0.466	0.135	0.227	0.043	0.0481	0.0065
22A_1	7.95	294.37	3.3	10.63	21.65	13.22	280.99	0.73	3.25	14.7	0.4	0.112	0.072	0.12	0.135	3.31	0.041
22A_10	1.06	2.25	3.29	0.147	22.93	0.205	4.08	0.02015	0.11	0.181	0.017	0.153	0.05	0.074	0.049	0.043	0.00171
22A_11	3.51	39.3	7.06	1.58	24.23	2.27	32.15	0.086	0.85	0.52	0.17	0.35	0.24	0.48	63.09	2.73	0.229
22A_12	34.44	39.53	16.78	0.64	21.75	0.98	78.71	0.071	1.11	0.57	0.107	0.35	0.34	0.31	0.68	0.32	0.0197
22A_13	4.15	25.14	5.78	0.81	22.11	1.26	23.63	0.111	0.72	0.68	0.069	0.3	0.119	0.139	0.66	0.35	0.0124
22A_14	4.82	16.91	3.74	0.228	23.83	0.48	17.77	0.145	0.35	0.4	0.026	0.49	0.38	0.062	0.238	0.13	0.0075
22A_15	7.28	58.24	6.31	0.72	21.95	0.74	14.99	0.15	0.62	0.23	0.106	0.28	0.31	0.19	0.87	0.54	0.0227
22A_2	4.78	66.1	3.2	1.78	21.34	1.22	24.56	0.092	1.39	0.47	0.13	0.25	0.21	0.106	0.36	0.67	0.037
22A_3	6.48	15.49	5.92	0.83	21.35	1.18	30.53	0.081	1.19	0.42	0.064	0.64	0.22	0.126	0.222	0.28	0.255
22A_4	3.53	17.7	7.21	0.67	21.88	3.25	17.82	2.96	0.83	0.49	0.102	0.22	0.25	0.101	1.03	0.26	0.0261
22A_5	1.47	4.75	2.74	0.6	20.89	0.171	5.63	0.065	0.259	0.24	0.031	0.26	0.073	0.091	0.323	0.099	0.0059
22A_6	4.81	17.47	5.19	0.67	24.06	0.57	14.12	0.02015	0.57	0.38	0.089	0.22	0.36	0.119	0.251	0.24	0.0108
22A_7	2.3	7.1	3.93	1.48	21.8	1.42	10.76	0.02015	0.147	0.115	0.2	0.32	0.076	0.22	0.48	85.58	0.62
22A_8	4.24	12.54	4.9	0.243	20.28	0.76	10.49	0.02015	0.48	0.3	0.05	0.27	0.164	0.103	1.24	1.42	0.0186
22A_9	1.73	5.23	2.15	0.19	21.14	0.072	5.84	0.02015	0.164	0.0445	0.0096	0.155	0.05	0.034	0.167	0.17	0.00205
4A.1	52.2	1262.54	75.07	16.89	55.43	49.83	693.45	3.92	6.69	16.11	6.18	52.74	9.12	17.99	13.3	3.83	0.483
4A.2	94.91	1916.52	59.98	24.26	53.01	105.89	1255.65	3.21	12.37	21.62	10.55	45.7	16.37	18.59	24.66	4.88	0.653
4A.3	31.12	1240.54	58.86	13.97	54.41	32.51	459.16	4.54	6.85	13.3	6.26	35.68	13.78	14.73	16.96	4.46	0.666

4A_1	43.66	1177.8	65.52	14.24	28.57	66.82	481.71	3.05	45.33	15.59	8.14	40.08	18.63	16.99	89.5	7.56	0.79
4A_10	31.25	831.64	40.62	15.91	26.97	27.64	464.36	2.07	20.77	11.36	5.24	37.04	11.12	15.55	21.27	5.73	0.62
4A_11	40.87	926.7	31.85	15.66	26.48	29.38	431.58	1.76	8.16	15.93	5.97	27.66	16.62	11.35	89.77	7.08	1.18
4A_12	32.56	966.34	36.42	21.25	31.6	40.67	431.96	2.05	5.66	14.4	9.3	29.84	20.25	13.18	43.82	3.4	0.58
4A_13	34.07	938.45	43.97	13.14	25.95	41.23	574.05	2.4	9.97	14.82	5.6	53.33	13.3	14.71	11.87	3.94	0.379
4A_14	27.02	996.68	41.18	18.66	28.75	22.86	340.41	2.55	7.11	16.93	7.03	32.01	15.58	25.46	35.91	3.63	0.63
4A_15	19.45	540.72	40.45	8.47	25.73	41.47	329.77	2.78	9.22	13.7	3.14	31.91	5.94	9.11	21.54	6.93	0.56
4A_2	41.15	935.74	47.29	17.02	27.06	58.14	560.24	2.88	10.83	19.79	6.7	33.13	12.18	15.49	30.33	4.03	0.59
4A_3	91.78	1070.35	81.23	13.96	25.14	62.89	877.56	3.11	6.78	21.63	6.43	37.27	11.34	16.24	11.88	3.29	0.338
4A_4	65.29	1314.83	69.59	31.46	31.89	58.14	1546.52	5.32	14.49	17.5	6.12	156.04	14.12	24.67	23	3.05	0.64
4A_5	26.49	987.97	52.78	28.38	24.61	28.18	386.78	2.18	7.13	16.99	7.4	36.4	16.44	17.22	17.53	5.54	0.65
4A_6	43.46	781.97	35.24	14.6	28.52	39.08	512.23	2.65	6.57	18.24	4.93	28.64	11.48	11.91	22.22	3.85	0.54
4A_7	33.69	966.17	53.03	28.54	26.89	28.06	378.35	2.67	6.9	16.03	5.59	59.14	12.37	17.27	16.02	10.71	0.55
4A_8	71.65	2118.14	50.5	20.46	26.28	55.99	590.2	2.64	9.45	15.69	28.09	39.63	58.39	31.81	14.46	57.26	0.458
4A_9	25.56	809.09	54.02	17.81	33.52	49.47	3411.1	2.65	6.87	11.02	6.98	26.93	11.37	16.58	11.48	4.74	0.402
6B.1	1.7	1.35	15.22	0.0569	46.01	0.71	13.18	0.161	0.071	0.0445	0.011	0.13	0.00605	0.282	1.16	0.291	0.0097
6B.2	1.31	2.42	15.43	0.0469	43.01	0.79	0.88	0.1255	0.0405	0.0445	0.0411	0.136	0.104	0.239	1.007	0.124	0.0181
6B.3	3.06	2.42	16.22	0.137	48	0.287	64.53	0.09	0.09	0.209	0.054	0.259	0.116	0.46	2.99	1.08	0.027
6B_1	0.43	0.172	2.01	0.0161	20.62	0.072	38.87	0.02015	0.051	0.209	0.0085	0.067	0.00605	0.062	0.083	0.103	0.00118
6B_10	2.81	2.26	21.83	0.127	30.66	0.89	35	0.132	0.53	0.26	0.043	0.137	0.126	0.42	3.84	0.72	0.0198
6B_11	2.79	3.42	17.3	0.174	37.16	0.95	43.63	0.156	0.98	0.318	0.079	0.9	0.124	0.49	3.44	1.14	0.027
6B_12	1.69	2.01	12.61	0.093	27.38	0.443	31.73	0.091	0.38	0.248	0.027	0.147	0.043	0.27	1.41	1	0.0144
6B_13	3.5	5.63	20.75	0.248	24.78	0.89	52.58	0.133	0.37	0.88	0.033	0.161	0.046	0.76	1.12	1.03	0.0168
6B_14	2.03	3.18	14.34	0.129	29.14	0.443	27.39	0.126	0.31	0.313	0.033	0.119	0.061	0.38	2.15	3.25	0.0201
6B_15	2.34	3.01	14.48	0.147	32.24	0.83	44.88	0.157	0.296	0.316	0.04	0.151	0.054	0.38	2.36	0.9	0.0152
6B_2	1.8	2.02	11.1	0.167	30.79	0.83	27.77	0.077	0.39	0.182	0.106	0.159	0.082	0.31	1.77	0.29	0.0144
6B_3	3.25	1.62	16.72	0.114	29.27	0.89	29.59	0.139	0.39	0.345	0.023	0.28	0.078	0.51	1.39	0.88	0.0127
6B_4	233.33	3	184.5	1	25	30	179	7	8	92.05	8.75	111.31	20.43	1.31	1.15	0.7	0.0114
6B_5	2.5	3.06	13.18	0.166	13.795	0.56	22.23	0.107	0.36	0.191	0.026	0.21	0.0368	0.66	1.07	0.52	0.0101
6B_6	5.44	2.51	19.77	1.25	32.09	2.59	729.3	0.56	0.51	0.49	0.135	0.79	0.076	1.24	2.78	1.05	0.186

6B_7	1.68	4.32	13.77	0.69	24.35	0.61	55.48	0.049	0.186	0.325	0.066	0.176	0.082	0.4	0.78	0.56	0.0216
6B_8	2.17	3.66	12.5	0.142	35.25	0.58	52.48	0.181	0.211	0.315	0.048	0.198	0.063	0.35	6.12	1.35	0.047
6B_9	2.77	5.15	14.15	0.104	31.45	0.072	43.77	0.02015	0.57	0.85	0.129	0.252	0.167	0.49	4.99	1.29	0.0293
7B.1	17.59	103.45	27.76	13.5	44.25	5.64	148.1	0.49	2.2	7.56	0.83	0.398	0.54	1.01	0.899	0.84	0.448
7B.2	44.39	2318.03	1664.82	410.02	49.32	150.08	734.84	5.08	10.68	42.35	9.3	0.8	5.92	3.96	4.15	2.06	1.27
7B.3	34.37	461.03	424.87	120.4	64.69	23.27	349.98	1.97	7.89	20.7	2.76	0.62	2.4	4.37	1.58	1.39	0.98
7B_1	37.7	244.84	70.73	39.11	30.74	30.39	464.81	2.69	7.49	21.87	1.8	0.268	1.45	2.04	1.99	2.04	0.908
7B_10	31.73	270.34	48.81	22.59	30.84	12.41	383.76	1.93	6.27	26.93	1.59	0.619	1.54	1.6	1.61	1.81	0.766
7B_11	26.98	221.54	88.18	26.99	30.77	12.74	433.95	1.23	3.76	19.24	1.84	0.811	2.34	4.16	1.02	1.61	1.16
7B_12	33.62	272.52	56.04	29.85	33.57	18.49	340.38	2.7	5.87	18.4	2.19	0.359	2.77	2.29	1.19	1.97	0.991
7B_13	37.9	791.53	696.05	57.61	36.8	88.21	404.08	9.63	7.14	21.86	2.68	0.556	1.68	4.55	2.14	3.1	1.28
7B_14	26.35	244.75	53.69	21.98	31.75	12.37	262.94	1.39	4.01	14.76	1.82	0.353	1.37	3.54	1.62	2.04	1.57
7B_15	24.68	216.68	49.05	22.26	33.85	12.97	342.54	5.61	5.5	16.08	1.43	0.486	1.2	1.83	1.34	1.44	0.865
7B_2	19.87	129.33	42.95	17.29	31.49	10.07	188.46	1.07	9.6	11.34	1.19	0.682	1.15	1.42	0.846	1.11	0.451
7B_3	39.9	742.44	378.82	32	33.45	22.26	495.34	1.82	5.55	14.49	3.24	0.403	2.68	3.94	1.74	1.9	1.059
7B_4	33.63	435.4	98.62	34.17	36.05	17.91	779.14	2.81	10.97	22.95	3.37	0.497	3.21	4.27	1.68	2.37	1.33
7B_5	40.68	397.49	63.86	51.83	34.54	12.81	467.37	1.61	6.75	21.85	2.72	0.538	5.9	2.47	1.51	2.88	0.782
7B_6	32.7	268.86	66.8	28.6	34.16	11.92	285.95	1.8	5.34	16.85	1.57	0.334	1.73	2.13	1.32	1.54	0.727
7B_7	36.81	377.89	70.21	24.43	32.28	14.29	414.06	1.9	7.08	15.38	2.22	0.403	1.93	3.41	1.33	2.06	0.881
7B_8	30.45	372.18	73.74	22.07	33.58	14.19	743.93	4.13	7.23	13.91	2	0.45	2.12	3.81	1.92	2.42	0.99
7B_9	23.57	268.77	52.3	43.34	32.05	8.14	246.73	1.51	3.92	15.76	1.79	0.299	2.31	1.72	1.09	1.44	0.596
13B.2	14.18	56.24	15.42	3.15	52.53	12.43	201.68	0.98	11.39	0.566	0.19	0.0676	1.17	0.24	0.1684	0.098	0.0512
13B.2	1140.9	1526.37	39.71	245.65	91.39	423.51	6383.36	19.78	13.33	9.25	6.56	0.346	58.67	2.2	7.82	0.83	0.96
13B.3	6.9	17.56	10.08	1.545	47.75	2.3	32.76	0.08	0.075	0.572	0.0586	0.01915	0.313	0.0715	0.0594	0.341	0.0141
13B.3	43.9	117.01	21.51	10.71	45.67	25.84	401.45	0.307	14.68	1.12	0.62	0.119	1.71	0.62	0.437	14.71	0.348
13B_1	104.92	378.51	565.83	20.29	26.53	40.71	350.8	0.51	9.4	1.41	1.16	0.094	4.51	1.05	1.08	0.73	0.114
13B_10	102.65	420.37	44.68	21.91	26.04	39.98	553.15	0.89	37.04	1.81	1.08	0.145	4.83	1.29	0.44	0.36	0.103
13B_11	2083.59	3756.41	47.67	767.96	206.75	851.84	3408.77	1.45	24.48	17.22	12.34	0.72	89.31	1.26	1.03	2	0.43
13B_12	50.92	977.57	552.45	10.9	28.16	30.15	29176.89	45.85	13.47	1.62	1.12	0.42	1.26	1.04	0.43	0.43	0.16
13B_13	49.55	147.07	13.12	11.13	26.18	21.89	369.98	0.62	70.91	1.26	0.42	0.128	1.81	0.29	0.66	0.55	0.084

13B_14	117.85	262.78	11.42	42.96	37.39	57.96	464.22	0.53	17.66	1.84	0.8	0.152	5.6	0.38	0.48	0.71	0.107
13B_15	57.85	449.08	26.57	13.24	26.7	23.84	328.91	0.41	19.24	1.1	1.54	0.134	3.44	1.26	0.5	0.4	0.097
13B_2	66.14	301.79	18.22	13.48	24.91	23.75	403.32	0.64	52.38	1.9	1.43	0.134	3.19	0.9	0.68	0.57	0.138
13B_3	101.18	308.26	13.64	14.54	25.61	33.68	325.82	0.36	71.62	1.74	0.97	0.112	3.23	0.62	0.41	0.14	0.094
13B_4	52.16	178.64	14.63	16.23	24.23	21.6	766.25	0.97	16.34	2.1	0.54	0.192	3.15	0.4	0.4	1.5	0.5
13B_5	278.48	3865.08	215.95	36.36	32.28	95.99	88943.11	160.77	88.33	7.79	12.4	2.24	19.28	25.67	1.15	1.88	0.55
13B_6	111.55	359.74	25.89	28.27	29.74	58.58	4156.05	16.71	73.7	6.5	1.99	0.2	5.91	1.14	0.5	0.85	0.7
13B_7	151.8	427.62	78.67	60.11	40.54	58.86	515.1	1	5.76	2.16	1.5	0.181	6.88	0.98	3.16	1.56	0.59
13B_8	62.56	238.43	13.36	19.81	25.36	23.85	341.66	0.61	10.7	1.23	0.88	0.138	2.96	0.4	0.8	0.26	0.189
13B_9	212.7	479.08	15.01	34.32	33.33	93.62	1004.02	1.6	34.58	1.95	1.27	0.127	5.06	0.68	0.65	5.62	0.288
14b.1	71.3	144.19	84.28	31.4	63.41	133.67	770.7025	14.72	17.99	231.1	0.167	0.67	10.95	0.1	0.17	2.8375	0.56
14b.2	57.93	116.27	5.5	21.99	63.18	17.4	712.373125	2.51	3	5.84	0.52	0.067	3.59	0.22	0.0166	0.055	0.21
14b.3	51.92	138.66	20.94	23.95	74.2	18.43	697.7907813	3	4.35	4.07	0.0615	0.413	1.86	0.35	0.0354	0.027	0.131
14b.4	254.66	792.16	5.5	104.29	90.58	175.51	694.1451953	1.5	4.865	0.5	5.29	0.52	39.34	3.12	2.33	0.0225	0.119
14B_1	156.07	471.55	24.7	65.15	39.31	34.22	610.58	0.49	5.38	10.49	2.62	0.89	24.34	1.2	0.59	0.386	0.117
14B_2	211.41	917.77	44.2	97.69	56.8	56.8	733.27	0.393	17.57	8.6	3.63	0.617	29.38	5.08	1.36	0.216	0.068
14B_3	268.47	538.99	13.59	116.76	94.35	61.76	734.94	0.276	8.04	7.73	2.8	0.432	29.87	0.48	0.8	0.148	0.078
14B_4	1026.23	1715.59	39.97	427.13	163.67	276.37	2602.74	1	9.08	14.83	11.12	0.488	102.99	3.21	1.08	0.214	0.0449
14B_5	136.8	543.98	16.6	63.3	39.52	35.46	375.48	0.234	7.56	8.12	2.99	0.298	21.02	1.05	0.412	0.098	0.139
14B_6	46.38	1142.59	49.91	16.08	29.09	10.05	277.63	0.197	7.92	4.56	4.21	0.269	20.69	7.11	0.95	0.142	0.0544
14B_7	118.15	231.13	7.32	49.4	33.36	29.12	339.34	0.137	4.25	5.85	1.25	0.332	13.08	0.099	0.063	0.045	0.096
21D.1	2.46	261.68	3.36	9.82	37.59	27.11	124.5	0.58	2.32	2.33	0.424	0.186	0.0481	0.063	0.1258	1.52	0.149
21D.2	4.03	229.26	3.15	13.62	35.41	20.59	193.42	0.395	6.41	8.26	0.547	0.452	0.067	0.101	0.0447	2.75	0.208
21D.3	1.92	124.25	2.3	5.61	37.46	17.97	69.38	0.6	1.41	2.89	0.325	0.392	0.121	0.055	0.0139	2.76	0.0699
21D_1	3.5	452.47	2.65	36.75	23.06	12.06	230.88	0.48	10.46	6.35	0.54	0.42	0.176	0.107	0.0366	2.3	0.253
21D_10	0.71	56.75	2.31	4.92	22.97	4.91	49.11	0.228	1.43	1.32	0.28	0.21	0.0145	0.112	0.017	1.88	0.061
21D_11	3.5	536.74	1.78	4.25	21.26	11.93	104.59	0.68	7.57	16.42	1.3	0.174	0.055	0.049	0.0201	1.37	0.089
21D_12	0.65	43.53	2.56	5.22	21.36	10.67	49.33	0.39	1.18	1.4	0.1	0.32	0.00605	0.019	0.0298	2.82	0.059
21D_13	5.21	1043.44	4.54	16.59	21.78	36.6	251.39	1.53	11.15	26.87	0.95	0.65	0.181	0.142	0.095	3.8	0.18
21D_14	1.16	130.14	2.82	31.64	22.39	280.94	234.47	5.08	16.22	10.55	0.32	0.81	0.062	0.026	0.0289	2.6	0.226

21D_15	1.77	146.38	3.42	60.88	22.21	36.78	258.88	1.45	6.8	6.74	0.36	0.83	0.044	0.044	0.113	3.64	0.188
21D_2	0.6	63.01	2.62	5.53	21.3	4.7	106.66	0.144	1.18	1.57	0.23	0.161	0.00605	0.052	0.078	1.22	0.087
21D_3	5.23	369.88	2.39	20.83	21.72	20.14	247.69	0.77	7.2	8.87	0.58	0.51	0.54	0.047	0.05	4.42	0.154
21D_4	9.75	720.23	6.56	23.62	21.84	20.83	144.69	1.34	7.5	11.6	0.98	0.3	0.47	0.089	0.093	2.14	0.108
21D_5	1.14	160.25	2.57	6.91	22.77	5.5	64.45	0.223	2.39	1.96	0.18	0.31	0.079	0.033	0.033	1.28	0.067
21D_6	6.13	589.9	3.32	128.98	48.59	55.27	187.14	1.86	15.82	8.06	1.26	0.3	0.84	0.1	0.0069	4.43	0.05
21D_7	0.57	44.91	2.63	7.59	23.85	2.15	11.65	0.062	0.93	0.35	0.068	0.186	0.089	0.0097	0.00215	0.27	0.0061
21D_8	1.1	92.77	2.69	12.54	23.07	25.48	107.9	1	2.99	2.5	0.26	0.68	0.136	0.068	0.0258	2.5	0.104
21D_9	2.1	284.56	2.87	14.37	22.26	17.61	169.63	0.61	3.67	3.94	2.3	0.55	0.051	0.97	0.065	10.7	0.278
11B_1	0.465	105.12	2.52	2.37	26.36	0.93	168.15	0.6	3.09	5.97	0.86	1.54	0.166	0.038	0.97	1.09	0.1
11B.1	3.11	1260.59	20.06	10.25	59.93	2.47	274.89	0.657	5.5	2.15	3.13	1.29	0.43	0.704	2.56	2.43	0.174
11B.2	1.67	758.64	10.28	7.93	53.83	1.89	956.91	0.369	3.88	1.25	1.83	0.575	0.239	0.294	2.04	1.96	0.18
11B.3	1.43	1002.79	11.95	8.15	54.24	1.25	93.31	0.175	2.52	1.34	2.25	0.552	0.191	0.389	2.04	2.15	0.166
11B_10	6.6	1903.43	5.56	10.23	36.69	6.1	941.71	0.45	23.81	3.57	4.8	2.4	1.24	0.125	10.07	72.57	0.126
11B_11	1.66	1823.77	5.1	11.37	35.21	4.15	405.37	0.44	5.63	1.52	3.26	1.62	0.453	0.44	2.98	5.21	0.64
11B_12	2.01	799.69	6.2	8.96	34.59	4.24	1525.94	1.33	19.59	2.72	2.24	1.91	0.384	1.01	1.66	3.61	0.268
11B_13	7.51	7476.66	42.34	447.88	760.48	53.44	160912.98	100.8	1428.54	1047.3	99.54	233.01	1.88	35.53	166.31	132.83	15.56
11B_14	1.03	342.78	4.17	6.12	37.9	1.47	331.98	0.51	9.47	2.7	1.18	0.77	0.152	0.3	1.09	1.51	0.09
11B_15	1.32	586.96	3.86	5.67	30.26	2.1	139.1	0.172	1.64	0.82	1.69	0.54	0.193	0.71	3.83	27.14	0.083
11B_2	7.48	2895.34	14.26	27.14	58.68	4.07	648.25	1.37	12.55	6.33	10.73	1.74	0.7	0.85	9.03	6.01	0.229
11B_3	2.3	974.33	4.81	8.53	33.99	2.4	2010.75	0.432	3.53	1.72	2.31	0.8	0.68	0.83	1.61	1.68	0.106
11B_4	7.35	3634.23	6.59	41.02	61.77	8.05	336.96	0.56	7.35	4.65	8.06	1.3	1.96	1.89	6.97	8.5	0.277
11B_5	2.69	1237.14	5.84	58.51	45.54	1.41	194.14	0.46	3.94	2.34	3.01	1.01	0.351	0.58	2.13	2.39	0.136
11B_6	1.12	461.17	3.45	6.46	29.96	1.08	65.35	0.133	1.76	0.86	1.13	0.487	0.297	0.35	1.17	1.71	0.06
11B_7	1.5	845.36	4.46	20.99	49.85	2.9	4836.03	2.2	24.37	25.48	4.55	9.17	0.295	1.02	8.03	5.29	0.58
11B_8	5.46	5877.58	64.05	79.23	203.79	24.18	526.5	2.07	20.56	7.94	25.09	11.45	2.08	0.37	8.56	2.85	19.36
11B_9	6.56	275.3	6.89	117.09	103.66	3.98	571.75	1.02	20.03	2.21	4.16	6.45	2.08	0.41	45.35	4.69	2.86
22D_1	11.89	319.84	5.11	7.98	23.9	14.54	459.16	0.74	26.51	8.82	0.85	0.488	0.91	0.34	1.91	9.75	0.182
22D_10	15.99	701.81	13.81	10.63	24.1	19.4	413.34	0.89	4.6	5.57	1.61	0.416	3.97	0.9	1.7	122.11	0.221
22D_11	5.79	355.75	7.38	7.7	25.82	15.86	331.74	0.71	5.15	4.89	0.66	0.276	0.294	0.32	1.02	16.67	0.19

22D_12	6.23	257.1	8.51	5.46	24.31	13.02	312.39	0.61	10.18	8.88	0.53	0.296	0.51	0.37	0.55	10.07	0.183
22D_13	5.67	276.51	8.64	4.99	24.09	15.56	278.54	0.77	3.66	7.64	0.58	0.283	0.405	0.62	0.52	10.87	0.158
22D_14	3.77	207.4	6.99	4.05	24.41	80.18	258.25	1.4	2.62	6.94	0.53	0.257	0.268	0.39	0.344	7.32	0.136
22D_15	11.63	425.77	10.69	9.18	25.67	20.07	386.72	0.8	10.36	21.05	1.31	0.357	1.4	0.5	0.87	16.28	0.209
22D_2	19.41	449.13	7.36	12.63	24.54	22.71	601.42	1.24	10.86	7.58	1.35	0.485	2.62	0.4	26.21	11.56	0.7
22D_3	4.54	262.53	6.73	5.51	25.24	11.33	240.01	0.42	5.05	3.85	0.62	0.54	0.92	0.43	4.56	8.14	0.146
22D_4	8.43	266.93	10.18	5.69	23.24	9.3	223.34	0.46	5.02	4.74	0.56	0.272	0.35	0.231	0.367	5.92	0.151
22D_5	6.95	335.26	7.15	10.74	26.32	23.4	612.1	0.78	5.41	9.24	0.84	0.68	0.62	0.5	0.56	12.05	0.246
22D_6	8.59	325.75	9.55	8.41	25.68	15.65	428.41	0.82	10.8	5.04	0.83	0.411	0.69	0.49	0.77	19.5	0.242
22D_7	7.06	358.81	8.6	9.94	25.51	21.97	556.43	0.73	7.08	5.52	0.77	0.399	0.68	0.46	0.66	11.67	0.234
22D_8	9.38	401.37	10.07	5.51	26.59	14.51	298.95	0.63	6.51	7.19	1.15	0.42	2.08	0.64	0.61	8.76	0.173
22D_9	11.08	421.36	9.48	9.67	25.41	16.23	502.57	0.97	6.17	13.36	0.92	0.49	0.86	0.48	0.7	23.47	0.321
22D.1	3.47	218.8	7.05	8.09	35.5	10.48	355.05	0.87	5.56	2.39	0.52	0.412	0.59	0.25	0.367	5.26	0.249
22D.2	7.09	434.63	8.6	6.99	36.43	21.42	407.29	1.19	5.88	5.07	1.03	0.37	1.63	0.58	134.43	14.84	1.15
22D.3	6.38	367.06	7.18	9.04	35.03	21.35	435.52	1.31	4.38	5.78	0.77	0.365	1.06	0.385	1.15	22.72	0.377
1B.1	186.26	3870.91	14.16	1346.87	439.75	55.38	816.86	5.94	23.07	78.68	75.57	2.16	56.34	6.6	25.18	0.69	1.15
1B.2	2.14	33.79	13.61	14.69	38.37	6.1	51.21	0.175	5.12	24.68	0.86	0.224	0.218	1.08	11.71	0.65	0.622
1B.3	1.89	22.67	7.15	11.73	39.33	9.66	44.57	0.219	3.47	12.87	0.65	0.251	0.201	0.58	18.3	0.49	1.17
1B_1	2.63	72.83	5.98	26.93	26.19	7.46	56.02	0.187	4.25	12.8	1.24	0.133	0.581	1.24	15.93	0.52	0.54
1B_10	1.34	26.51	5.54	12.16	25.84	2.28	39.46	0.156	2.94	9.59	0.54	0.315	0.145	0.76	6.87	0.32	0.53
1B_11	8.69	276.88	8.12	141.79	43.17	5.3	63.88	0.09	4.94	15.28	11.02	0.23	2.63	2.04	17.52	0.59	0.8
1B_12	2.18	55.87	8.93	22.16	27.49	3.82	46.66	0.138	3.33	13.06	1.5	0.217	0.431	1.88	11.7	0.62	1.4
1B_13	1.64	46.32	5.38	28.99	27.85	3.61	94.35	0.02015	4.52	17.94	1.03	0.313	0.316	1.23	9.42	0.44	0.97
1B_14	1.18	22.06	4.68	9.89	25.44	6.87	41.73	0.136	3.44	9.08	0.63	0.273	0.168	0.55	7.51	0.45	0.453
1B_15	2.73	80.27	5.16	31.86	30.69	30.89	89.11	0.94	6.44	18.2	2.23	0.314	0.63	0.81	8.34	0.28	0.9
1B_2	1.04	22.84	5.13	8.88	25.97	2.77	36.07	0.072	2.36	8.8	0.42	0.179	0.273	0.48	5.41	0.24	0.276
1B_3	1.65	39.3	7.14	18.97	26.71	2.89	69.78	0.138	4.69	19.36	1.59	0.252	0.235	5.86	6.76	1.09	0.72
1B_4	5.02	168.31	7.4	45.9	28.88	3.7	226.11	0.188	16.86	55.37	0.95	0.433	1.75	1.1	10.17	0.78	1.09
1B_5	1.13	26.53	6.6	20.78	26.45	2.47	67.72	0.085	4.31	8.1	0.52	0.232	0.133	2.13	7.28	0.66	0.54
1B_6	1.85	47.86	7.3	27.07	27.94	4.18	80.12	0.215	3.85	14.04	1.77	0.186	0.414	2.49	10.41	1.2	0.87

1B_7	12.21	421.47	7.84	202.55	73.36	4.74	56.96	0.217	4.64	19.77	12.99	0.249	4.84	3	11.87	0.49	0.54
1B_8	2.22	67.89	7.2	29.62	30.57	2.91	60.77	0.125	4.89	11.99	1.46	0.318	0.6	1.1	9.1	0.39	0.72
1B_9	33.67	1212.56	7.06	437.04	73.69	6.5	67.1	0.283	4.04	10.99	43.99	0.201	13.79	6.16	15.91	0.34	0.54
21B.1	0.89	121.82	4.35	8.5	36.78	4.36	75.28	0.27	1.56	4.32	0.61	0.104	0.126	0.73	1.93	0.585	0.221
21B.2	0.76	62.72	6.37	6.8	36.81	2.15	72.64	0.174	1.13	2.96	0.449	0.11	0.129	0.64	2.19	0.967	0.158
21B.3	1.23	217.44	6.91	28.25	41.71	5.49	516.5	0.61	10.23	14.82	0.71	1.24	0.109	0.69	2.54	0.599	1.42
21B_1	1.95	155.6	9.18	14.13	39.43	9.62	360.79	0.42	5.48	14.62	1.4	0.52	0.247	0.96	1.66	1.29	0.307
21B_10	1.37	170.22	8.83	21.3	42.56	7.4	1153.07	0.178	58.92	10.84	1.98	2.14	0.54	0.9	3.58	0.7	0.335
21B_11	1.39	181.94	9.97	13.4	34.22	8.07	199.97	0.127	4.37	9.08	2.03	0.24	0.38	2	3.21	2.45	0.341
21B_12	0.69	131.18	9.51	9.27	31.95	2.53	138.72	0.02015	2.78	5.77	0.47	0.158	0.078	0.47	0.78	0.45	0.226
21B_13	0.68	77.22	11.71	8.34	33.8	3.65	109.12	0.285	2.31	6.18	0.5	0.188	0.073	0.55	2.04	1.39	0.166
21B_14	0.94	171.12	10.45	10.27	32.1	55.09	284.04	13.34	5.29	10.12	1.51	0.21	0.184	1.03	3.25	0.99	0.239
21B_15	0.69	93.52	11.08	8.88	32.31	5.15	175.73	1.36	2.18	4.47	0.56	0.24	0.121	0.45	0.63	0.63	0.152
21B_2	1.45	184.25	6.73	15.69	37.64	6.8	200.58	0.36	5.37	11.05	1.94	0.25	0.42	1.04	1.71	0.85	0.253
21B_3	0.76	98.77	5.81	10.11	34.86	6.76	187.43	0.17	4.74	11.58	0.85	0.23	0.214	0.82	3.51	0.63	0.244
21B_4	0.37	34.35	4.71	3.51	30.77	1.65	56.59	0.108	1.5	2.41	0.2	0.069	0.041	0.175	0.241	0.23	0.066
21B_5	0.91	119.84	9.05	18.14	34.2	5.66	233.66	0.223	3.48	11.47	0.75	0.23	0.26	0.72	1.97	1.11	0.31
21B_6	2.56	352.95	16.64	74.55	44.32	40.23	12971.71	1.21	64.38	208.71	3.23	1.55	0.49	1.78	12.87	2.36	4.42
21B_7	0.64	116.65	9.98	8.98	32.47	28.32	131.97	3.89	3.76	7.54	1.27	0.21	0.08	0.65	0.74	0.7	0.18
21B_8	0.98	205.29	11.51	14.87	33.97	4.75	398.54	0.162	5.41	7.52	1.34	0.32	0.181	2.79	4.81	1.97	0.462
21B_9	1.54	687.02	9.99	234.65	96.48	54.58	3929.05	2.1	47.91	109.84	1.66	0.76	0.121	0.76	7.41	1.41	2.58
21J.1	15.34	423.08	10.86	58.38	53.83	7.42	525.14	0.41	17.06	25.79	2.24	0.31	0.393	2.53	2.46	1.54	1.81
21J.2	0.97	412.25	10.23	36.91	58.04	4.72	202.75	0.255	8.88	14.26	1.27	0.29	0.131	1.86	1.76	1.06	0.468
21J.3	1.19	310.29	11.21	30.37	55.13	5.64	282.6	0.36	9.59	12.34	1.44	0.342	0.134	2.62	2.36	1.58	0.527
21J_1	1.26	535.88	10.92	181.19	47.78	46.82	870.45	0.02015	26.01	20.16	2.58	0.53	0.174	2.17	4.2	2.8	1.88
21J_10	1.91	305.85	11.16	34.77	50.23	4.74	225.75	0.434	9.24	15.03	3.28	0.288	0.46	2.09	1.98	2.03	0.527
21J_11	1.62	368.22	10.94	43.34	54.32	7.01	270.82	0.123	37.2	16.68	3.17	0.46	0.251	2.36	2.64	2.04	0.623
21J_12	0.78	261.43	8.33	21.6	41.54	3.29	138.66	0.21	4.58	7.47	1.38	0.177	0.095	1.86	1.85	1.6	0.275
21J_13	0.76	345.54	10.83	60.73	41.25	7.34	1963.59	0.52	9.88	14.33	1.33	1.64	0.074	1.26	1.31	1.16	0.85
21J_14	1.79	444.88	18.1	58.11	52.41	17.72	1000.4	0.61	15.93	24.98	2.66	0.71	0.278	2.16	4.57	1.73	1.24

21J_15	3.66	444.85	13.25	99.48	54.01	7.37	810.4	0.172	29.84	24.99	10.52	0.46	1.02	2.92	3.9	1.68	0.94
21J_2	2.12	336.19	9.74	26.97	43.14	4.81	198.44	0.231	21.33	11.75	2.27	0.52	0.169	1.85	2.01	1.73	0.401
21J_3	7.5	238.39	8.71	20.69	38.31	3.03	170.94	0.53	10.7	8.33	1.87	0.219	0.118	2.18	2.53	2.05	0.412
21J_4	1.1	338.02	11.96	26.92	42.54	3.66	131.86	0.158	6.37	9.52	2.05	0.37	0.131	4.05	2.37	4.84	0.434
21J_5	1.4	299.33	11.47	26.15	41.49	5.96	185.53	0.67	9.12	12.66	2.74	0.29	0.5	2.48	3.72	2.84	0.463
21J_6	5.49	300.63	9.96	24.5	39.51	2.33	186.37	0.02015	7.24	9.71	1.45	0.204	0.258	0.92	1.06	0.87	0.377
21J_7	1.27	455.24	17.66	39.85	49.02	4.78	346.61	0.02015	7.7	16.23	2.06	0.183	0.252	1.64	2.47	1.71	0.573
21J_8	0.71	170.54	8.47	17.65	39.43	2.86	142.76	0.02015	4.78	6.42	1.04	0.226	0.084	1.08	1.33	1.34	0.256
21J_9	66.82	2793.39	21.5	1079.63	276.98	43.08	1412.34	0.83	49.57	12.2	270.69	1.04	27.4	49.23	35.24	3.13	1.42
21K.1	0.83	252.76	8.59	28.91	36.43	2.57	129.03	0.104	4.44	8.46	1.41	0.192	0.115	2.56	3.74	0.99	0.383
21K.2	1.49	403.82	15.66	63.54	47.84	5.52	441.69	0.3	14.8	17.48	2.81	0.48	0.245	5.43	11.61	2.37	1.17
21K.3	0.5	84.58	5.29	19.92	32.86	1.7	73.23	0.078	2.24	5.15	0.59	0.172	0.053	1.1	1.09	0.438	0.244
21K_1	1.09	400.53	19.73	53.04	32.08	6	216.91	0.168	6.99	11.25	3.38	0.136	0.19	2.71	3.56	1.44	0.392
21K_10	0.86	631.63	15.5	41.93	33.6	1.35	100.4	0.1018	3.66	14.03	22.53	0.118	0.121	22.71	1.73	1.88	0.25
21K_11	2.76	496.71	16.92	57.46	41.3	5.08	274	0.122	8.2	16.54	11.01	0.51	0.232	15.77	21.89	4.4	0.593
21K_12	1.33	470	19.69	173.68	47.73	22.67	2647.34	0.388	34.74	42.63	5.01	1.25	0.165	31.15	8.61	12.46	4.44
21K_13	1.03	77.54	8.62	7.84	26.47	1.47	51.25	0.02015	2.11	3.47	0.81	0.34	0.0362	1.19	1.39	0.517	0.118
21K_14	0.72	256.48	7.55	14.33	26.89	1.89	73.88	0.059	3.47	3.51	0.93	0.335	0.0434	1.45	1.11	0.445	0.159
21K_15	0.97	238.42	10.99	23.73	33.42	3.07	133.53	0.166	5.24	7.49	2.78	0.209	0.209	3.17	4.11	1.75	0.402
21K_2	1.37	392.63	20.12	45.54	40.78	7.43	521.18	0.327	13.15	16.02	17.18	0.55	0.26	27.82	7.41	4.99	1.4
21K_3	1.21	275.98	14.69	40.52	37.94	3.67	242.71	0.154	7.14	8.61	3.74	0.266	0.197	4.98	5.08	2.12	0.476
21K_4	0.94	463.06	12.33	38.28	31.31	4.22	137.51	0.0913	4.16	9.66	3.2	0.164	0.122	5.15	3.39	1.71	0.35
21K_5	2.18	356.65	13.04	59.69	37.47	6.88	216.58	0.17	8.77	12.77	3.47	0.397	0.13	3.27	9.12	2.7	0.631
21K_6	1.33	471.54	13.5	55.25	42.12	5.57	299.9	0.163	7.02	11.41	4.14	0.52	0.214	4.35	6.96	2.16	0.527
21K_7	1.31	361.23	14.31	84.77	42.9	6.46	438.16	0.343	11.2	18.77	3.43	0.325	0.199	4.3	5.15	2.04	0.94
21K_8	1.05	401.43	10.26	55.59	34.09	3.25	162.26	0.152	4.66	9.32	2.69	0.144	0.166	2.57	9.86	1.39	0.417
21K_9	1	520.38	12.41	43.42	42.13	3.83	188.12	0.126	6.41	11.27	4.11	0.242	0.165	3.99	4.78	1.91	0.422
21L.1	2.56	1041.32	12.58	99.11	51.24	3.82	435.82	0.02015	12.63	29.95	2.98	0.408	0.142	4.63	8.96	1.43	1.2
21L.2	1.95	541.81	12.29	36.12	45.38	1.81	146.7	0.02015	9.47	17.18	3.92	0.214	0.235	5.86	2.43	1	0.498
21L.3	3.26	686.22	10.89	99.42	48.76	4.38	439.43	0.168	14.98	61.78	2.3	0.217	0.101	3.2	1.87	0.7	0.99

21L_1	0.84	291.76	8.31	26.55	26.34	1.14	121.14	0.0855	5.19	10.2	1	0.091	0.0508	1.28	0.92	0.35	0.249
21L_10	0.68	120.14	6.83	8.36	24.82	0.345	54.13	0.0502	2.51	4.17	0.5	0.12	0.0196	0.637	0.615	0.146	0.14
21L_11	0.87	168.65	6.95	8.18	28.62	1.08	38.37	0.1046	2.41	6.05	0.6	0.11	0.072	0.736	0.522	0.126	0.126
21L_12	1.26	357.32	9.2	18.85	28.64	1.41	149.09	0.132	6.05	7.21	1.41	0.218	0.113	1.9	1.95	0.41	0.272
21L_13	1.2	302.97	10.39	26.6	31.9	1.12	136.04	0.0798	7.44	7.74	1.34	0.148	0.081	2.01	2.16	0.54	0.348
21L_14	0.75	197.55	7.6	13.42	25.82	0.714	68.23	0.0673	5	5.35	0.76	0.127	0.0229	1.24	0.74	0.254	0.186
21L_15	1.56	293.19	10.19	21.41	29.96	1.42	153.96	0.163	9.12	7.83	1.53	0.185	0.07	2.29	12.68	0.608	0.42
21L_2	2.85	306.86	7.67	24.97	27.94	1.19	123.25	0.109	7.22	9.85	1.94	0.056	0.141	1.99	1.5	0.479	0.29
21L_3	1	338.95	7.7	48.42	27.93	1.3	161.34	0.02015	6.57	13.37	1.2	0.167	0.104	1.45	1.05	0.349	0.454
21L_4	2.13	507.25	9.29	32.84	29.89	0.95	121.46	0.0924	6.66	10.03	1.82	0.176	0.094	1.59	1.75	0.36	0.341
21L_5	18.77	360.55	10.24	41.88	29.33	1.46	170.89	0.0784	5.66	8.92	1.49	0.192	0.075	2.11	7.11	0.39	0.345
21L_6	1.23	348.33	9.77	24.58	28.01	1.05	163.39	0.1079	11.63	9.79	1.86	0.17	0.0451	2.6	1.37	0.51	0.379
21L_7	1.03	325.54	6.66	22.6	25.6	1.34	159.16	0.125	5.8	10.39	10	0.063	0.15	1.73	1.22	0.355	0.286
21L_8	3.37	442.24	9.88	42.45	29.56	3.71	133.58	0.0916	6.81	10.19	1.88	0.179	0.112	2.22	1.68	0.41	0.507
21L_9	1.41	492.31	10.21	49.98	45.37	1.07	485.03	0.293	7.39	22.67	1.61	0.242	0.072	1.4	2.07	0.412	0.5
22B.1	1.05	332.83	10.49	50.02	41.95	4.32	131.93	0.262	5.92	9.99	1.25	0.171	0.198	1.74	3.89	1.47	0.349
22B.2	0.71	213.16	6.69	19.15	37.24	2.37	152.12	0.111	4.47	5.47	0.65	0.193	0.093	0.76	8.59	0.596	0.325
22B.3	1.31	424.04	14.54	60.82	43.68	6.63	220.95	0.149	7.77	14.11	1.61	0.271	0.282	2.16	5.63	1.66	0.62
22B_1	0.96	388.55	14.81	52	39.91	6.95	232.71	0.243	8.65	23.7	1.95	0.32	0.189	1.63	5.06	2.01	0.6
22B_10	0.94	288.95	9.31	28.46	36.97	3.7	197.19	0.147	8.06	16.16	1.4	0.34	0.135	1.17	3.87	1.58	0.404
22B_11	0.57	104.95	8.72	9.2	29.07	2.55	58.32	0.145	1.96	6.77	0.49	0.193	0.05	0.59	1.6	0.75	0.132
22B_12	1.02	490.56	11.73	48.74	36.91	6.24	245.4	0.074	6.73	24.43	1.97	0.244	0.167	2.17	5.89	2.83	0.55
22B_13	9.47	390.3	9.7	48.82	31.77	3.67	234.15	0.076	6.08	16.27	1.1	0.187	0.087	1.13	3.16	1.58	0.49
22B_14	3.69	350.36	10.53	29.75	31.28	3.11	147.55	0.148	10.46	12.9	2.13	0.38	0.16	3.7	6.69	2.32	0.59
22B_15	1.07	1830.48	10.51	54.95	61.8	4.89	270.19	0.216	6.08	11.04	2.26	0.46	0.142	1.16	6.33	1.47	0.447
22B_2	0.91	408.28	12.16	56.1	34.68	5.58	185.5	0.194	6.21	17.1	1.64	0.281	0.134	1.29	4.23	1.61	0.41
22B_3	0.76	302.69	11.61	27.26	30.46	4.93	161.31	0.048	5	13.03	2.43	0.229	0.138	4.47	3.83	1.68	0.378
22B_4	17.82	491.56	9.64	73.44	32	7.91	424.53	0.236	12.69	28.07	1.52	0.38	0.106	1.16	4.15	1.6	0.85
22B_5	0.93	354.21	10.27	43.03	35.84	6.69	178.32	0.23	5.53	14.27	1.42	0.252	0.14	1.28	4.01	2.18	0.419
22B_6	1.07	293.44	13.78	34.33	31.81	4.3	116.82	0.02015	4.44	14.02	1.26	0.286	0.119	1.23	3.84	1.91	0.336

22B_7	1.19	271.03	8.52	91.73	36.77	9.3	637.31	0.332	15.57	41.1	1.45	0.97	0.28	1.14	4.56	1.7	1.06
22B_8	0.55	141.11	6.98	17.82	30.6	2.5	73.69	0.053	2.77	7.94	0.73	0.235	0.086	0.67	1.19	1.02	0.177
22B_9	0.84	271.47	10.53	35.06	46.45	4.85	172.82	0.135	4.99	13.21	1.08	0.31	0.146	0.94	4.09	1.37	0.326
22H.1	0.62	6.47	5.02	0.357	38.83	0.163	9.97	0.05	1.91	0.34	1.66	0.259	0.0165	14.41	0.6	1	0.129
22H.2	0.86	9.8	0.3655	1.57	40.3	0.309	8.72	0.216	0.01845	0.308	0.565	0.113	0.076	0.139	0.139	0.00194	0.00136
22H.3	0.67	6.14	3.69	0.78	40.55	0.71	0.89	0.0445	0.58	0.125	0.244	0.129	0.00605	0.196	1.84	0.0044	0.006
22H.4	0.61	2.48	6.52	0.504	41.29	0.072	2.4	0.243	0.445	0.81	0.171	0.171	0.078	0.042	0.113	0.0177	0.0083
22H_1	0.7	48.57	4.01	3.16	24.27	1.13	4.7	0.02015	0.31	0.0445	1.27	0.04	0.4	0.21	4.38	0.031	0.0037
22H_2	0.41	19.33	6.71	0.051	27.06	0.57	0.4325	0.02015	0.62	1.45	0.015	0.0395	0.00605	0.39	0.125	0.05	0.00058
22H_3	0.37	21.01	8.89	0.045	21.98	0.162	2.24	0.063	0.195	1.94	0.072	0.262	0.00605	0.129	0.132	0.057	0.0045
22H_4	2.45	86.14	0.95	16.14	33.81	0.072	0.88	0.0425	0.49	1.63	5.86	0.0765	1.22	0.99	0.98	0.0174	0.00185
22H_5	1.64	156.74	2.89	4.44	38.7	4.34	3.5	0.84	0.84	8.13	0.42	0.45	0.39	0.13	0.224	0.064	0.00199
22H_6	0.29	9.62	1.36	0.205	20.7	0.149	0.4325	0.02015	0.01845	1.78	0.067	0.067	0.00605	0.034	0.053	0.0074	0.00088
22H_7	0.33	15.95	4.28	0.0131	27.13	0.072	0.4325	0.02015	0.01845	0.13	0.0085	0.23	0.016	0.073	0.098	0.071	0.00167
15B.1	186.7	382.73	17.09	20.9	54.89	49.67	452.95	0.1055	3.13	4.75	2.76	0.01915	14.69	0.408	0.0796	0.148	0.0585
15B.2	13.21	109.94	11.1	1.052	48.41	4	128.81	0.219	0.73	0.652	0.537	0.106	1.68	0.349	0.0565	0.0905	0.0734
15B.3	22.18	217.32	31.49	2.36	49.25	6.98	131.96	1.39	2.71	2.03	0.542	0.01915	0.82	2.31	0.544	0.252	0.123
15B_1	5766.38	0.00595	1160.79	545.61	81.51	2376.6	14737.02	6.09	49.81	28.36	1275.25	9.22	4013.13	219.02	0.378	2.96	0.97
15B_10	12.69	55.47	14.62	1.43	34.66	4.9	111.93	0.48	5.52	1.09	0.7	0.116	0.72	0.41	0.0364	0.47	0.083
15B_11	67.17	355.23	25.53	5.65	36.88	33.23	484.69	0.84	18.57	3.68	3.87	0.073	9.83	1.66	0.069	0.73	0.165
15B_12	36.49	263.36	36.83	6.09	32.18	13.93	376.99	1.03	8.39	3.44	3.88	0.141	3.36	2.36	0.06	0.72	0.151
15B_13	6.83	39.46	12.95	0.67	32	3.86	65.06	0.063	3.93	0.94	0.21	0.11	0.61	0.193	0.0238	0.179	0.0301
15B_14	38.57	151.58	25.82	6.87	34.04	14.08	132.66	0.152	3.25	1.82	0.76	0.129	2.87	1.15	0.057	0.51	0.259
15B_15	15.71	246.45	16.72	2.56	33.81	11.46	239.32	0.58	16.41	1.28	6.78	0.095	18.94	1.59	0.087	0.32	0.076
15B_2	7.58	65.64	7.56	0.9	33.84	7.61	91.31	0.02015	1.13	1.47	0.99	0.111	2.02	0.46	0.72	0.126	0.0216
15B_3	43.84	83.78	16.63	6.93	40.03	11.75	259.46	0.77	4.9	2.24	0.76	0.091	2.89	0.34	0.121	0.38	0.151
15B_4	46.04	170.85	14.41	4.56	36.58	20.4	108.61	0.135	0.88	2.2	4.07	0.141	7.15	0.8	0.0407	0.23	0.0331
15B_5	17.47	107.89	13.76	2.53	35.87	10.37	127.81	0.8	5.15	2	0.9	0.208	0.85	0.6	0.0252	0.34	0.0567
15B_6	16.4	65.71	15.21	2.46	35.61	15.37	283.73	0.178	9.36	2.11	0.44	0.103	0.75	0.31	0.0239	0.26	0.086
15B_7	10.23	40.88	16.62	2.03	34.9	6.28	103.59	0.302	1.61	1.23	0.25	0.097	0.38	0.26	0.0383	0.174	0.0401

15B_8	7.53	35.79	14.62	1.07	35.4	25.74	114.01	0.232	1.94	1.62	0.18	0.06	0.256	0.204	0.056	0.167	0.043
15B_9	8.48	53.47	16	1.2	36.5	6.95	157.6	0.44	6.86	1.14	0.37	0.078	0.38	0.71	0.0154	0.27	0.067
2B.1	38.41	234.32	37.19	17.48	40.64	19.68	312.72	1.11	4.5	5.98	1.28	0.449	3.57	4.81	0.519	1.81	0.472
2B.2	40.24	195.53	80.08	18.17	38.94	20.05	293.39	1.19	5.96	6.21	1.12	0.531	2.66	4.9	0.794	2.63	0.443
2B.3	30.68	157.3	54.39	12.31	38.51	16.15	229.07	0.66	4.73	4.86	0.94	0.567	1.9	4.02	0.551	1.31	0.438
2B_1	22.34	164.73	45.78	18.59	28.56	10.89	222.17	0.41	2.64	6.5	0.99	0.01915	1.71	2.19	1.89	2.49	1.28
2B_10	920.84	1334.84	41.53	211.91	303.03	473.65	1138.12	0.9	3.88	7.03	3.3	0.71	1.91	2.93	0.531	2.31	0.458
2B_11	25.25	116.98	20.68	10.15	23.18	11.73	172.25	0.71	5.01	4.19	0.43	0.58	1.09	1.89	0.438	1.59	0.273
2B_12	29.7	228.32	35.67	18.55	25.5	18.5	263.5	0.63	3.75	6.39	1.4	0.86	2.42	3.28	0.528	2.61	0.55
2B_13	31.55	231.56	27.89	16.04	25.41	19.23	396.05	1.53	4.08	6.02	1.25	1.43	2.55	2.67	0.77	2.03	0.64
2B_14	88.31	386.19	40.68	21.8	27.04	28.56	436.9	1.27	6.65	12.36	1.23	0.01915	2.85	3.19	0.73	2.03	0.47
2B_15	28.91	184.37	34.95	17.03	27.52	15.91	330.09	0.61	3.51	7	0.98	0.57	1.92	4.63	0.58	2.2	0.56
2B_2	37.53	234.83	40.34	22.42	26.82	17.41	254.16	1.92	4	5.26	0.99	0.226	1.92	3.38	1.26	3.11	1.13
2B_3	41.86	156.14	33.57	16.78	26.07	14.16	224.64	0.92	3.3	7.8	0.68	0.83	2.22	2.67	0.61	3.46	0.73
2B_4	48.44	274.16	28.88	19.85	25.46	19.15	337.19	0.246	3.41	6.46	1.45	1	3.04	2.85	0.547	8.86	0.407
2B_5	51.43	178.98	30.07	16.11	25.2	11.39	202.18	0.263	1.67	7.08	0.63	0.326	1.17	1.85	0.295	1.04	0.5
2B_6	37.33	306.92	54.15	16.04	28.7	18.21	272.28	0.72	11.75	4.91	1.24	1.19	3.05	4.27	0.69	2.04	0.465
2B_7	40.62	268.51	53.74	13.46	25.31	13.96	225.79	0.85	3.81	4.64	1.1	0.087	2.55	3.15	0.445	2.01	0.448
2B_8	22.57	199.31	40.51	13.02	24.76	12.82	238.14	0.7	3.43	5.75	0.71	0.305	1.51	3.1	0.88	1.76	0.54
2B_9	55.13	181.72	21.18	13.28	24.43	21.19	373.49	0.79	10.85	2.85	0.75	0.62	1.42	1.92	0.471	1.19	0.68
22G_1	32.19	200.57	19.44	3.85	25.4	7.32	181.76	0.61	8.58	1.3	0.69	0.112	1.38	1.59	0.073	0.93	0.169
22G_10	115.26	1250.87	605.71	6.2	23.59	36.32	292.7	0.369	6.77	1.05	5.37	0.149	1.35	5.28	0.204	1.97	0.29
22G_11	1991.25	3106.1	1751.9	168.05	92.97	506.78	2600.85	1.7	13.97	9.6	11.75	0.86	19.32	42.32	0.44	0.92	0.35
22G_12	5745.6	3673.73	8250.13	1373.73	442.14	3293.12	12847.29	8.49	99.28	124.8	20.05	2.5	7.18	8.52	1.83	1.83	0.38
22G_13	115.45	158.9	73.06	18.97	30.45	26.5	260.63	0.49	6.9	1.13	0.66	0.127	1.37	0.67	0.29	0.68	0.197
22G_14	114.34	3197.99	427.42	22.26	34.57	27.24	259.64	0.4	13.35	2.15	37.94	0.43	14.23	49.05	0.55	1.42	1.11
22G_15	32.07	248.64	27.31	3.58	25.43	8.92	168.11	0.43	6.65	1	2.23	0.091	1.43	2.36	0.076	0.6	0.189
22G_2	38.51	73.54	20.82	4.97	24.88	10.48	162.31	0.71	4.4	0.99	0.34	0.096	0.89	0.4	0.055	0.61	0.186
22G_3	13.05	35.12	5.73	1.92	23.53	5.29	55.55	0.73	3.39	0.277	0.21	0.01915	0.6	0.19	0.028	0.19	0.117
22G_4	54.19	167.88	17.98	6.98	26.52	9.94	166.04	0.318	3.61	0.75	0.95	0.069	2.62	0.96	0.115	0.55	0.176

22G_5	27.71	102.73	13.3	3.63	24.64	5.91	158.36	0.65	5.08	66.8	0.47	0.048	1.24	0.72	0.05	0.51	0.141
22G_6	33.54	225.31	25.01	5.21	26.98	11.16	246.8	0.54	84.96	0.89	0.47	0.099	1.26	1.29	0.148	0.72	0.248
22G_7	49.8	99.7	25.28	6.17	24.69	10.12	123.48	0.276	5.18	1.78	0.56	0.104	1.72	0.44	0.043	0.37	0.107
22G_8	21.6	36.11	11.02	2.31	24.07	4.2	53.09	0.158	1.15	0.45	0.2	0.118	0.63	0.136	0.045	0.23	0.051
22G.1	38.09	507.19	57.82	29.39	46.22	14.38	283.43	0.8	25.26	1.01	1.06	0.291	1.57	6.74	0.177	14.87	228.79
22G.2	13.3	45.32	9.27	1.71	33.46	5.63	146.44	1.83	18.17	0.74	0.177	0.092	0.324	0.325	0.0444	0.294	0.165
22G.3	27.07	133.04	14.54	3.95	33.91	11.13	127.53	0.55	9.97	0.92	0.437	0.098	0.97	1.21	0.116	0.767	0.204
22G_9	71.25	137.48	20.65	10.64	37.06	13.35	406.81	0.268	1.9	3.31	0.49	0.25	2.41	0.39	0.048	0.33	0.31
21E.1	12.08	187.13	31.36	3.01	48.67	6.22	138.67	0.374	6.9	2.91	0.611	0.152	0.39	1.38	0.981	0.498	0.144
21E.2	36.16	201.29	36.94	9.81	47.11	21.85	509.55	2.23	9.15	1.5	1.15	0.175	1.6	0.694	0.798	0.352	0.174
21E.3	26.78	382.27	35.82	12.1	49.64	16.01	420.47	3.19	1049.89	744.83	4.62	0.133	4.68	2.65	1.314	0.27	0.167
22E.1	41.69	512.48	63.9	7.41	33.27	25.01	268.48	0.235	2.37	2.23	1.33	0.172	0.94	11.9	1.2	0.367	2.64
22E.2	324.31	122.11	163.6	3.91	89.64	25.09	256.66	0.91	4.91	3.04	0.51	0.118	0.55	1	0.82	0.77	0.084
22E.3	13.59	311	15.57	4.77	34.12	8.72	148.38	0.46	4.87	2.2	6.23	0.173	2.24	6.96	0.275	0.494	0.087
21E_1	20.43	239.06	47.71	3.89	24.6	12.39	133.73	0.251	10.54	2.21	1.56	0.42	1.01	2.48	0.64	3.29	0.169
21E_10	14.43	486.05	21.27	1.63	25.32	7.48	125.77	0.218	3.95	1.81	0.56	0.144	0.49	0.67	0.98	0.11	0.066
21E_11	139.36	2392.03	124.2	108.4	40.9	66.41	2045.27	1.78	196.06	24.7	8.79	1	19.01	36.87	1115.7	7.32	40.93
21E_12	15.5	110.71	109.76	2.84	26.34	19.13	180.38	0.21	17.96	2.21	0.59	0.201	0.6	2.04	3.76	4.23	0.277
21E_13	31.81	233.79	34.28	4.5	26.97	22.16	224.63	0.38	20.83	3.05	1.32	0.124	0.97	1.65	1.54	0.8	0.174
21E_14	24.6	813.97	46.33	3.79	27.77	15.47	245.17	0.291	9.05	3.08	15.3	0.21	22.88	9.66	2.25	0.35	0.088
21E_15	23.98	269.88	27.61	4.58	25.62	21.79	257.19	0.33	9.43	4.96	0.97	0.36	2.09	1.47	0.93	0.31	0.36
21E_2	25.21	220.4	30.97	6.57	24.92	29.87	290.84	0.272	10.8	2.5	1.73	0.154	2.14	1.02	0.51	0.37	0.105
21E_3	20.22	208.54	38.49	4.29	27.33	19.96	239.2	0.38	66.04	3.1	0.68	0.209	0.65	1.59	1.81	0.37	0.184
21E_4	14.83	103.84	23.85	2.9	25.11	10.88	143.6	0.215	9.64	2.22	0.64	0.183	0.59	1.67	0.356	0.44	0.084
21E_5	32.01	545.84	29.67	28.26	25.76	14.37	251.14	0.25	7.96	2.24	5.49	0.211	9.64	1.5	0.38	0.4	0.087
21E_6	53.69	829.23	34.03	16.46	27.35	23.8	360.47	0.42	6.7	7.43	4.19	0.306	7.53	1.75	3.18	14.09	0.208
21E_7	13.79	79.17	17.7	3.26	26.18	11.55	112.18	0.211	5.43	1.92	0.48	0.112	0.51	0.57	0.5	0.7	0.083
21E_8	19.74	387.68	23.57	3.88	27.19	9.6	505.47	0.269	3064.83	3.06	1.66	0.39	3.42	0.83	0.44	0.43	0.121
21E_9	14.49	202.49	24.29	2.04	25.5	11.71	146.2	0.25	10.1	1.92	0.63	0.155	1.1	0.75	0.5	0.21	0.083
22E_1	14.15	67.24	22.41	1.82	25.54	13.25	193.32	0.51	6.28	2.32	0.45	0.352	0.42	0.75	0.98	1.08	0.089

22E_10	9.61	69.2	16.39	2.74	25.67	9.08	137.27	0.16	21.34	1.8	0.34	0.67	0.178	1.07	5.47	0.71	0.112
22E_11	11.24	62.89	15.77	1.24	25.65	7.46	138.66	0.282	4.19	7.09	0.24	0.108	0.212	0.74	0.339	0.43	0.06
22E_12	51.27	70.71	13.79	2.95	28.27	8.62	137.05	0.295	4.91	2.14	0.29	0.146	0.265	0.52	0.61	0.47	0.145
22E_13	41.06	147.68	13.16	7.01	29.28	22.34	225.6	0.33	2.88	3.85	0.93	0.098	1.31	0.5	0.213	0.52	0.072
22E_14	7.43	125	18.69	1.49	27.12	6.42	137.87	0.179	3.01	3.53	0.25	0.132	0.123	1.26	0.342	0.91	0.071
22E_15	12.6	56.21	13.8	1.78	27.28	13.52	160.72	0.298	3.66	1.31	0.41	0.4	0.7	0.39	0.67	0.39	0.055
22E_2	93.89	44.33	12.97	1.46	28.55	9.36	142.84	0.56	11.21	1.76	0.24	0.199	0.303	0.57	0.374	0.62	0.058
22E_3	6.81	54.31	16.3	0.96	24.85	6.37	117.33	0.121	2.33	2.42	0.48	0.132	0.175	0.56	0.6	0.37	0.079
22E_4	10.05	70.94	16.37	1.27	25.73	7.05	113.76	0.172	3.47	1.18	0.31	0.164	0.276	0.58	0.93	0.32	0.055
22E_5	6.78	49.3	13.61	0.6	24.33	10.58	79.33	0.132	1.81	1.1	0.19	0.156	0.086	0.8	0.331	0.39	0.045
22E_6	23.39	59.49	13.69	2.86	24.52	12.28	127.17	0.199	3.37	2.9	0.38	0.122	0.85	0.5	0.372	0.41	0.075
22E_7	26.39	63.31	21.55	4	29.34	20.02	148.79	0.273	3.92	1.98	0.53	0.113	0.71	0.6	0.212	0.43	0.061
22E_8	10.74	437.04	84.86	1.22	25.77	10.62	151.67	0.206	23.81	1.78	2.61	0.101	0.354	10.53	0.52	1.84	0.069
22E_9	7	65.48	15.03	0.83	25.8	9.32	140.05	0.227	12.21	1.53	0.34	0.124	0.177	0.45	0.161	0.44	0.063
5B.1	28.94	712.41	31.2	21.68	56.19	21.2	270.37	0.0875	6.33	5.28	2.06	0.158	1.78	1	2.91	0.126	0.165
5B.2	4.25	39.79	14.14	0.537	52.66	2.75	41.34	0.095	5	1.03	0.133	0.0682	0.082	0.15	0.0903	0.03	0.0293
5B.3	12.23	206.64	49.29	4.77	53.02	17.02	1010.53	0.467	7	12.35	0.918	0.2	0.85	0.89	0.478	0.482	0.134
5B_1	27.85	106.63	8.52	7.15	25.58	10.79	146.93	0.15	2.93	2.27	0.49	0.331	1.17	0.234	2.61	0.053	0.157
5B_10	17.63	712.43	22.35	3.44	24.14	11.01	190.33	0.193	224.18	8.27	0.92	0.123	0.509	0.66	3.52	0.201	0.107
5B_11	14.15	699.7	17.82	2.1	24.42	12.48	135.36	0.222	7.18	2.93	2.51	0.189	4.64	1.06	0.676	0.068	0.061
5B_12	17.99	224.55	18.92	2.32	24.64	12.34	136.19	0.306	4.82	2.76	0.5	0.102	0.394	0.67	0.691	0.35	0.163
5B_13	154.41	327.95	380.31	15.44	35.56	185.56	773.08	1.14	7.59	9.41	0.86	0.201	0.456	1.14	0.81	0.148	0.086
5B_14	10.18	89.61	18.17	1.22	24.64	8.81	104.43	0.155	4.18	1.88	0.243	0.128	0.215	0.48	0.236	0.063	0.051
5B_15	18.09	256.52	17.61	5.65	24.51	11.73	147.19	0.284	10.5	2.32	2.92	0.127	1.22	2.07	0.689	0.116	0.069
5B_2	25.74	273.21	19.18	2.62	25	12.26	112.17	0.17	5.06	3.31	1.08	0.212	0.583	1.04	0.653	0.094	0.093
5B_3	7.26	82.81	8.26	1.37	24.74	8.19	83.82	0.08	3.27	1.8	0.23	0.169	0.22	0.261	0.199	0.044	0.06
5B_4	45.44	299.84	56.58	5.66	23.9	32.96	223.65	0.388	5.88	4.43	0.87	0.161	0.88	1.36	0.61	0.35	0.175
5B_5	11.02	124.6	26.1	2.26	23.82	11.91	124.83	0.295	3.72	2.75	0.304	0.164	0.323	0.55	0.452	0.094	0.073
5B_6	13.15	272.84	12.84	1.15	23.51	8.47	548.83	0.275	2543.98	7.22	0.36	0.177	0.148	0.49	0.613	0.049	0.078
5B_7	22.73	440.04	15	2.42	25.39	10.51	160.91	0.25	13.19	5.02	6.02	0.184	3.29	2.6	1.01	1	0.124

5B_8	40.87	461.69	22.22	20.01	23.65	28.36	196.14	0.84	7.04	4.6	1.67	0.105	5.87	0.67	1.07	0.093	0.085
5B_9	20.93	165.41	12.15	6.19	25.48	14.83	1950.9	1.12	5.87	2.94	0.6	0.156	1.31	0.43	0.354	0.094	0.107
10B_1	10.17	90.53	23.7	2.58	41.96	16.1	527.51	2.68	229.97	19.34	2.65	1.24	0.192	2.18	2.09	1.67	0.24
10B_2	9.23	54.19	20.91	2.33	43.98	14.21	503.71	2.8	105.19	14.27	2.22	1.72	0.094	1.86	0.25	0.49	0.22
10B_3	9.78	68.56	15.48	1.82	43.77	20.92	496.5	2.95	55.83	16.41	2.55	0.8	0.25	1.37	0.08	0.44	0.214
10B_4	7.84	38.92	21.25	1.5	37.71	12.85	390.01	4.14	40.27	12.41	1.81	1.22	0.113	1.34	0.058	0.33	0.164
10B_5	9.13	30.73	31.76	2.01	35.84	23.69	502.6	3.21	35.06	19.13	2.2	0.75	0.092	1.74	0.63	0.95	0.197
10B_6	9.62	43.06	11.44	1.9	33.82	9.92	359.95	2.73	29.82	17.62	1.51	0.66	0.025	1.06	0.072	0.35	0.173
10B_7	8.53	42.02	11.88	1.01	33.13	12.99	536.99	2.85	65.6	9.11	2.06	0.82	0.071	1.5	0.071	0.39	0.169
10BA_1	0.7	80.35	8.03	6.87	67.53	0.8	2.5	13.32	4.07	13.55	0.86	20.23	0.56	1.26	6.66	1.27	0.64
10BA_2	1.1	28.28	9.74	0.00775	67.22	2.26	8.45	0.075	0.93	9.97	0.159	1.34	0.015	0.77	0.34	0.47	0.026
10BA_3	0.88	11.68	3.11	0.25	65.93	1.87	11.88	0.0555	0.6	5.78	0.65	1.54	0.3	0.2	0.052	0.098	0.018
10BA_4	29.25	72.18	5.69	0.93	57.25	3.73	107.3	0.71	8.3	15.2	0.65	0.96	3.68	2.31	2.35	3.91	0.168
10BA_5	11.42	58.1	5.24	0.084	50.6	0.2	59.02	0.092	0.7	8.54	0.108	1.12	0.047	0.2	0.16	0.117	0.0102
10BA_7	3.82	15.92	6.13	0.073	48.9	0.072	13.17	0.045	0.49	5.97	0.069	0.73	0.00605	0.4	0.26	0.29	0.0125
22F.1	94.7	326.28	24.34	21.53	50.78	27.18	1394.87	1.42	9.1	27.98	1.57	1.43	1.63	2.28	4.19	0.211	0.375
22F.2	226.79	300.27	29.38	7.7	52.92	14.92	331.95	0.562	3.66	17.19	1.14	0.87	0.9	1.84	1.77	0.063	0.0415
22F.3	101.72	780.66	29.3	20.3	52.81	17.21	1082.74	0.647	12.33	35.51	1.87	0.718	1.99	2.82	4.09	0.0837	0.094
22F_1	32.95	186.6	10.26	4.61	24.68	6.41	324.41	0.141	13.08	11.69	4.35	0.71	0.92	0.9	0.9	0.119	0.0311
22F_1	189.29	853.94	40.1	33.36	32.51	33.75	1564.51	1.039	29.36	51.48	52.73	0.93	4.38	4.03	18.82	0.663	4
22F_10	102.11	355.38	27.19	17.25	29.48	14.09	649.5	0.59	7.63	40.19	9.69	1.69	1.62	2.01	2.73	0.45	0.133
22F_10	163.51	522.51	31.32	22.67	29.04	20.53	1105.06	0.896	6.2	32.6	118.37	1.08	4.04	3.45	4.2	0.352	0.181
22F_11	63.61	327.54	80.54	9.79	24.93	9.15	818.24	0.344	10.5	26.01	25.25	0.92	1.82	2.11	1.99	2.94	0.238
22F_11	79.05	1000.2	27.43	15.08	27.41	14.23	561.81	0.462	6.48	16.02	27.18	0.81	20.26	24.75	3.24	2.48	0.201
22F_12	49.17	286.67	16.12	49.56	25.46	14.28	2472.77	0.69	5.82	45.75	1.78	2.11	1.16	1.42	6.44	0.32	0.266
22F_12	44.04	148.75	16.09	12.01	25.79	12.45	896.6	0.525	17.25	24.66	1.14	1.68	1.08	1.73	4.3	0.651	0.265
22F_13	40.73	196.03	9.86	9.58	25.78	12.77	1238.72	0.214	4.26	18.1	0.82	0.86	0.66	0.88	1.55	0.2	0.097
22F_13	109.13	328.28	36.76	12.78	27.12	16.02	741.47	0.561	9.27	21.15	10.05	1.73	2.27	2.77	3.51	0.377	0.21
22F_14	46.22	361.4	15.6	44.26	31.44	11.66	1567.03	0.61	58.07	27.87	19.74	2.54	2.61	1.47	3.99	0.43	0.238
22F_14	147.04	532.08	48.46	27.15	25.36	33.69	4998.56	0.651	6.55	79.23	13.34	1.2	5.23	7.27	17.12	0.646	0.152

22F_15	83.92	244.1	12.07	13.78	25.66	24.67	703.31	0.382	5.14	22.21	1.04	0.65	0.72	0.98	2.25	0.16	0.104
22F_15	74.74	359.77	23.5	10.45	27.08	17.81	864.6	0.553	32.01	24.42	1.82	1	2.88	2.87	3.7	0.282	0.145
22F_2	74.98	415.77	18.47	6.83	25.7	7.82	385.22	0.237	7.02	17.52	72.38	1.16	2.4	1.68	1.64	0.53	0.065
22F_2	440.75	1694.19	82.02	113.34	80.14	92.14	4114.18	2.87	93.76	160.66	82.82	2.93	12.14	8.86	18.77	2.6	1.27
22F_3	94.38	328.55	15.79	7.66	25.26	16.8	409.19	0.283	4.53	46.83	0.82	0.54	0.76	1.34	2.49	0.18	0.084
22F_3	180.8	453.33	33.6	24.19	29.18	36.93	1623.31	0.869	9.76	40.04	8.5	1.29	2.14	2.94	3.79	0.285	0.167
22F_4	95.16	409	28.33	21.1	25.16	29.57	5464.64	0.334	13.19	47.29	6.04	2.09	1.52	2.41	4.41	0.74	0.248
22F_4	62.2	216.03	19.76	7.99	26.62	12.19	831.34	0.578	10.42	10.53	23.67	2.01	2.49	1.94	1.53	0.24	0.077
22F_5	86.27	394.61	18.92	17.52	29.87	15.44	1227.68	0.51	5.79	37.79	15.83	1.53	1.84	1.66	3.23	0.28	0.138
22F_5	153.35	170.03	20.54	7.97	25.85	12.52	726	0.401	6.35	11.62	5.23	0.72	1.19	1.73	2.4	0.279	0.363
22F_6	58.11	208.24	25.48	8.83	25.06	10.03	542.92	0.308	7.8	21	0.87	1.94	1.07	1.62	1.8	0.095	0.07
22F_6	112.35	295.27	59.56	12	28.66	13.91	676.53	0.452	6.55	20.26	2.2	0.98	2.29	3.67	2.87	0.95	0.152
22F_7	112.34	502.48	16.5	12.94	29.58	20.38	689.6	0.51	16.83	34.03	398.14	1	2.85	1.54	1.48	0.095	0.173
22F_7	164.06	440.17	26.79	19.62	27.25	24.71	1768.11	0.927	12.75	37.19	52.73	1.32	2.68	2.36	4.59	0.356	0.157
22F_8	100.88	235.79	10.29	29.1	30.25	14.62	555.83	0.195	5.09	18.5	1.82	1.34	3.38	0.89	1.62	0.134	0.094
22F_8	142.05	451.39	21.06	11.76	28.2	14.87	538.14	0.587	5.51	23.09	3.45	1.22	1.24	1.68	2.56	0.177	0.0704
22F_9	56.35	799.61	15.81	30.61	34.1	23.16	1845.37	0.55	4.94	32.62	2.6	1.66	1.41	1.54	4.09	0.16	0.111
22F_9	53.66	255.75	21.67	6.86	25.47	11.65	784.4	0.754	11.8	13.2	27.96	1.86	2.13	2.99	1.75	0.158	0.096
4B.1	13.18	170.26	10.9	4.72	36.79	6.12	215.29	0.257	7.13	9	0.448	0.335	0.396	1	0.9	0.0103	0.0138
4B.2	16.98	498.2	20.05	5.1	39.17	9.6	160.32	0.105	3.28	5.48	0.97	0.687	0.247	0.5	2.12	0.011	0.0172
4B.3	8.66	620.97	6.36	2.83	40.3	2.61	163.72	0.02015	3.92	6.08	1.12	0.19	0.382	0.486	2.47	0.00413	0.00814
4B_1	6.1	193.12	10.14	3.2	33.07	7.29	154.7	0.02015	5.07	4.85	0.35	0.311	0.099	0.44	0.479	0.49	0.0147
4B_10	9.62	225.81	11.37	3.59	31.35	12.27	269.85	0.49	8.55	3.97	0.51	0.73	0.099	0.7	1.58	0.029	0.074
4B_11	3.21	239.18	7.2	6.08	32.67	2.43	59.84	0.067	2.85	1.92	0.53	0.23	0.219	0.34	1.72	0.088	0.0582
4B_12	9.97	175.51	10.51	3.87	33.09	4.35	188.56	0.095	6.26	6.52	0.23	0.3	0.47	0.54	0.82	0.145	0.075
4B_13	5.21	212.13	9.3	1.55	32.39	2.61	128.87	0.413	2.36	2.36	0.28	0.232	0.079	0.33	0.632	0.0076	0.0157
4B_14	13.78	421.14	11.27	7.97	34.08	3.35	145.49	0.08	4.07	5.92	0.8	0.52	0.53	0.53	1.35	0.0151	0.0119
4B_15	15.17	466.8	12.35	8.94	34.73	4.42	171.51	0.55	2.32	10.14	0.63	0.3	0.37	0.69	1.75	0.0051	0.0445
4B_2	12.55	361.11	12.82	6.41	33.9	6.25	265.41	0.421	5.91	6.42	0.82	0.71	0.177	0.87	1.37	0.083	0.0249
4B_3	6.74	238.36	9.61	2.57	32.83	3.97	125.04	0.02015	4.76	7.53	1.46	1.03	0.092	0.55	1.1	0.027	0.0124

4B_4	5.43	106.82	10.51	2	32.12	2.92	121.41	0.186	3.78	3.74	0.21	0.254	0.161	0.48	0.457	0.053	0.0118
4B_5	5.78	152.94	11.01	3.27	31.49	3.24	201.16	0.305	5.32	3.69	0.37	0.49	0.128	0.6	0.577	0.042	0.025
4B_6	3.26	98.11	11	1.45	32.81	2.12	50.97	0.02015	1.26	1.01	0.162	0.184	0.068	0.248	0.403	0.0112	0.0055
4B_7	11.47	225.11	12.44	2.58	31.83	2.86	183.8	0.105	2.94	5.47	0.3	1	0.23	0.56	0.88	0.0146	0.0106
4B_8	8.56	143.84	12.82	3.25	34.78	7.12	166.25	0.215	3.89	8.45	0.34	0.35	0.41	0.58	0.564	0.032	0.0149
4B_9	3.5	135.53	6.89	2.63	31.39	2.57	60.87	0.333	2.84	1.35	0.31	0.22	0.098	0.31	0.83	0.0212	0.0201

E. Solution-based-ICP-MS Trace Element Data and Correlation Matrices

ICP-MS	Analyte ($\mu\text{g g}^{-1}$)																				
sample ID	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
1A	2235	28809	24.70	21.59	196.45	11993	7.80	16.13	31.48	8.45	10.51	1.92	22.37	47.87	3.40	0.13	0.07	77.34	0.18	29.46	3.22
1A	2335	29475	25.21	22.31	207.05	12583	8.21	16.93	33.93	8.70	10.77	2.15	22.93	48.55	3.89	0.15	0.07	77.69	0.18	29.39	3.38
1A	2257	29517	25.52	21.70	201.08	12029	8.27	16.32	33.18	8.61	10.97	1.88	23.23	48.72	3.44	0.14	0.08	80.12	0.18	29.63	3.28
1B	33.96	389	40.81	9.68	13.05	1007	11.19	12.99	30.71	0.38	0.44	1.37	0.62	4.29	10.10	0.55	0.36	11.80	0.05	2.71	2.34
1B	31.85	363	38.82	8.92	11.84	995	9.45	13.32	30.21	0.35	0.39	1.43	0.58	3.50	10.95	0.51	0.33	11.29	0.05	2.39	2.50
1B	30.00	339	37.95	8.78	11.65	958	9.21	12.35	30.40	0.36	0.46	1.39	0.53	3.47	10.28	0.51	0.33	10.33	0.05	2.54	2.37
1B	30.60	345	36.81	8.61	11.88	965	8.99	12.77	31.03	0.34	0.33	1.48	0.52	3.79	10.17	0.48	0.38	10.69	0.05	2.48	2.25
2A	1892	16091	18.95	20.82	128.57	10471	21.13	59.03	40.87	4.88	5.82	2.07	13.67	19.41	1.96	0.21	0.14	56.40	0.10	25.62	2.00
2A	1995	14502	16.65	18.65	122.82	9999	21.19	45.30	47.16	4.34	7.41	1.82	11.74	17.28	1.98	0.16	0.17	51.93	0.10	26.97	1.88
2A	1744	14088	17.19	19.60	113.74	8592	17.63	101.10	63.01	4.34	6.03	1.88	12.35	16.82	1.88	0.24	0.27	50.76	0.09	26.62	1.72
2B	816	2084	37.72	7.71	46.95	3488	19.97	12.68	15.89	0.75	0.94	3.11	5.71	8.96	52.41	0.48	0.19	10.42	0.08	19.49	1.89
2B	774	1871	35.60	7.07	44.51	3396	19.19	12.23	14.69	0.71	0.89	3.10	4.88	8.56	48.84	0.47	0.18	8.03	0.07	19.39	1.75
2B	776	1883	35.44	7.06	43.07	3407	18.70	12.18	14.01	0.72	0.91	3.31	5.02	8.60	51.25	0.47	0.18	7.65	0.07	19.05	1.97
3B	17.56	4839	33.89	17.78	135.85	2897	3.22	13.66	2.80	1.27	0.19	0.65	0.10	1.61	6.01	0.08	0.03	34.71	0.08	0.60	0.55
3B	15.01	3721	33.93	17.10	144.93	3016	3.29	13.85	3.93	1.04	0.19	0.57	0.09	1.50	5.62	0.08	0.03	43.86	0.09	0.64	0.55
3B	16.12	4234	38.28	17.41	124.79	2999	3.45	13.70	3.77	1.15	0.19	0.60	0.10	1.76	5.95	0.08	0.03	33.48	0.08	0.59	0.58

4A	523	7498	20.73	5.26	94.62	4213	13.77	25.54	31.22	2.94	65.06	1.77	17.80	24.78	3.46	0.26	0.29	33.35	0.28	40.80	0.68
4A	533	7885	21.54	5.45	96.02	4281	13.52	26.07	31.79	3.06	65.76	1.65	18.56	25.01	3.29	0.29	0.29	34.26	0.29	38.85	0.68
4A	524	7891	20.42	5.55	92.25	4186	13.02	24.30	33.14	3.06	64.29	1.82	18.59	24.46	3.17	0.25	0.29	33.89	0.28	37.64	0.64
4B	284	3950	16.17	7.86	30.19	3179	4.86	15.05	16.39	1.20	1.08	0.49	1.25	2.42	3.26	1.17	0.06	7.28	0.02	1.08	0.11
4B	275	4204	16.12	8.09	29.48	3041	4.71	14.40	18.37	1.27	0.58	0.51	1.17	2.17	1.57	1.11	0.06	5.65	0.02	1.10	0.10
4B	278	3971	16.02	7.90	30.29	3012	4.37	15.22	16.48	1.22	0.49	0.39	1.17	2.20	1.56	1.10	0.06	6.37	0.02	1.05	0.11
5B	726	3189	14.90	4.91	52.14	2623	6.23	203.54	34.58	1.03	0.34	1.15	2.96	2.25	75.66	0.57	0.48	6.22	0.02	7.08	0.65
5B	717	3331	15.22	5.01	51.53	2703	6.57	233.58	36.19	1.06	0.19	1.02	2.90	2.17	90.81	0.53	0.66	6.06	0.02	6.92	0.62
5B	760	3520	15.72	5.05	52.31	2767	6.71	224.91	38.58	1.11	0.16	1.28	3.08	2.39	76.54	0.57	0.54	6.10	0.02	6.55	0.41
6B	60.22	53.91	0.62	21.11	4.19	891	6.54	1.17	1.57	0.08	0.17	0.05	0.18	0.97	1.76	0.08	0.01	3.41	0.01	0.33	0.06
6B	56.94	49.93	0.50	16.31	4.23	829	5.39	0.98	0.84	0.08	0.10	0.01	0.16	0.91	1.08	0.07	0.01	0.98	0.02	0.31	0.06
6B	57.81	52.38	0.54	16.91	4.20	1256	5.26	0.98	13.17	0.09	0.12	0.00	0.16	0.97	1.13	0.08	0.01	1.01	0.02	0.25	0.06
7B	650	3687	69.78	9.07	38.08	3728	16.78	19.33	51.96	1.31	0.48	4.48	4.11	10.79	28.65	0.23	0.73	18.61	0.12	7.18	2.40
7B	643	3746	69.12	8.97	37.50	3695	16.67	19.04	51.67	1.34	0.49	4.86	4.27	10.80	28.07	0.23	0.74	18.47	0.12	6.71	2.45
7B	627	3595	68.70	8.95	36.68	3597	16.58	18.16	50.25	1.29	0.51	4.59	4.02	10.30	26.45	0.22	0.70	18.50	0.12	6.58	2.65
10B	808	559	0.35	0.42	41.83	1909	1.41	1.63	1.43	0.42	0.08	0.04	0.06	1.97	40.00	0.01	0.00	0.30	0.01	0.59	0.04
10B	162	158	0.13	0.36	64.12	1213	8.78	197.62	5.95	0.12	0.08	0.03	0.14	1.62	42.52	0.05	0.06	0.90	0.02	7.22	0.01
10B	234	508	0.06	0.54	12.53	2416	3.87	33.03	3.81	0.24	0.05	0.05	0.08	0.36	0.21	0.04	0.01	0.39	0.02	2.24	0.02
11B	34.65	21663	54.31	57.51	5.61	24561	7.76	45.87	4.73	11.42	15.00	6.71	1.57	4.18	15.57	0.14	0.03	15.19	0.02	12.81	1.53
11B	35.51	21028	52.01	60.13	5.69	23321	7.70	48.06	6.43	11.53	15.49	4.78	1.59	4.04	15.41	0.12	0.03	15.19	0.02	12.67	1.43
11B	37.44	21774	54.39	58.44	5.45	23398	8.49	52.13	6.10	10.95	15.38	4.85	1.68	5.49	15.32	0.11	0.03	14.91	0.02	13.32	1.53
13B	4145	4331	82.05	23.92	186.48	10738	16.53	293.78	23.07	2.12	0.78	1.49	21.39	4.22	29.97	0.13	0.21	8.83	0.20	1.58	1.62
13B	4271	4413	84.18	24.86	190.89	10841	15.90	305.17	16.81	2.12	0.50	1.14	22.20	3.95	26.11	0.13	0.19	8.93	0.21	1.41	1.41
13B	4217	4331	85.53	25.06	191.27	10823	16.56	307.16	19.05	2.16	1.28	1.24	21.64	3.26	39.28	0.13	0.16	8.64	0.20	1.57	1.70
15B	560	1059	7.25	5.98	24.18	1857	5.70	23.50	5.01	0.56	0.11	0.80	3.68	1.95	4.63	0.08	0.01	6.08	0.02	0.51	0.21
15B	514	962	7.01	5.56	22.78	2062	6.58	19.86	5.42	0.53	0.04	0.95	3.38	1.81	1.27	0.07	0.01	5.27	0.02	0.51	0.22
15B	614	1089	7.89	6.35	25.95	1927	5.56	22.20	5.12	0.59	0.04	0.73	4.16	3.85	10.06	0.06	0.02	7.26	0.02	0.49	0.21
21A	16.52	3299	70.20	27.80	5.34	5827	11.40	24.87	20.39	0.77	0.35	4.39	0.24	5.94	29.61	0.33	0.57	18.44	0.02	0.49	1.54
21A	15.79	3465	70.54	28.26	4.27	6109	11.07	23.52	20.52	0.79	0.37	4.57	0.23	6.27	30.47	0.34	0.83	19.13	0.02	0.47	0.86
21A	17.79	3378	70.41	28.11	4.28	5969	11.25	23.67	21.23	0.78	0.38	4.64	0.24	6.23	28.54	0.33	0.52	18.55	0.01	0.45	0.80

21B	15.13	1980	53.26	16.45	29.83	4339	10.26	17.22	27.98	0.59	0.40	2.21	0.37	4.19	14.71	0.17	0.25	17.62	0.04	0.60	1.13
21B	18.72	2030	54.74	16.91	31.81	4351	11.36	20.84	30.15	0.61	0.46	2.04	0.39	6.35	15.15	0.19	0.20	16.71	0.05	0.62	1.12
21B	18.46	2010	58.82	17.36	29.00	4468	11.28	18.00	29.78	0.63	0.50	1.82	0.39	6.30	14.72	0.15	0.26	16.12	0.05	0.61	1.15
21C	25.78	2560	8.88	13.45	21.64	1482	2.68	14.11	2.90	0.84	0.03	1.08	0.10	1.83	2.55	0.09	0.02	4.96	0.02	0.19	0.12
21C	25.37	2682	9.34	13.94	20.89	1480	2.71	15.03	3.14	0.87	0.07	1.30	0.11	1.88	2.59	0.09	0.02	4.91	0.02	0.18	0.12
21C	25.13	2493	8.92	13.80	20.61	1464	2.61	14.63	2.92	0.82	0.05	1.16	0.10	2.19	2.25	0.10	0.02	4.74	0.02	0.33	0.13
21D	160	3857	129.29	5.17	259.95	3989	60.20	143.38	29.61	1.55	2.48	0.66	2.65	0.71	1.91	0.02	0.11	19.09	0.17	2.42	0.90
21D	140	3730	135.36	5.26	253.62	4130	55.56	34.88	27.89	1.52	2.64	0.50	2.57	0.72	1.78	0.02	0.10	14.02	0.18	2.46	0.89
21D	174	3799	135.04	5.03	281.87	4277	64.68	39.24	31.16	1.55	3.58	0.75	3.34	0.99	2.03	0.02	0.13	12.25	0.20	2.66	0.94
21E	734	2107	14.94	4.82	77.42	3701	6.42	286.52	19.80	0.88	0.30	0.40	4.08	7.32	94.52	0.46	0.17	10.50	0.02	2.80	0.85
21E	747	2194	15.39	5.11	77.13	3738	6.18	291.74	20.50	0.91	0.20	0.34	4.18	6.67	107.23	0.49	0.16	10.37	0.02	3.26	0.79
21E	776	2350	16.28	5.25	81.39	3928	5.96	297.33	26.05	0.98	0.23	0.44	4.38	7.50	100.65	0.47	0.17	11.11	0.02	3.00	0.65
21F	121	102	0.12	0.34	57.29	891	8.39	186.65	7.71	0.09	0.07	0.10	0.16	1.64	56.73	0.05	0.07	0.94	0.02	6.82	0.04
21F	146	42.15	0.05	0.19	226.07	9610	22.53	711.71	2.90	0.70	0.05	0.10	0.02	4.26	5.94	0.11	0.02	0.56	0.53	13.17	0.01
21J	17.64	4169	79.96	37.06	10.61	2643	14.80	18.78	26.73	1.01	0.25	4.23	0.29	6.53	34.09	0.33	0.57	22.08	0.03	0.55	1.08
21J	16.00	3982	77.95	36.28	10.57	2564	14.48	17.82	28.55	0.96	0.31	4.38	0.27	6.23	33.72	0.33	0.62	20.71	0.03	0.49	0.98
21J	15.73	4136	80.32	37.13	10.45	2636	14.76	18.24	26.59	1.00	0.27	4.73	0.28	6.19	32.95	0.31	0.54	20.96	0.03	0.49	1.14
21K	17.56	4371	102.10	30.91	13.75	3269	13.96	19.83	29.28	1.90	0.38	5.59	0.44	12.14	24.11	0.34	0.76	39.80	0.03	1.14	1.25
21K	16.75	4148	99.95	30.45	12.43	3148	13.96	19.32	30.15	1.86	0.36	5.42	0.43	11.78	22.24	0.33	0.70	39.09	0.02	1.10	1.28
21K	15.07	4122	101.20	30.97	12.23	3145	14.27	19.49	29.52	1.85	0.37	5.51	0.42	11.69	22.26	0.34	0.66	38.28	0.02	1.07	1.19
21L	18.77	4806	74.66	18.46	4.39	2319	14.87	20.47	29.44	1.32	0.13	6.11	0.25	6.60	48.41	0.49	1.13	21.18	0.03	0.67	1.00
21L	17.55	4714	73.52	18.26	4.51	2354	14.78	20.49	27.85	1.31	0.16	6.05	0.23	6.91	44.04	0.46	1.16	21.99	0.03	0.69	1.01
21L	18.68	4802	75.45	19.03	5.04	2407	14.68	21.27	30.22	1.34	0.13	6.12	0.24	6.91	50.24	0.51	1.14	22.18	0.03	0.76	1.07
21M	22.71	3435	10.91	9.63	23.07	1797	3.06	12.53	3.75	1.08	0.05	1.76	0.15	1.41	3.61	0.12	0.02	5.54	0.02	0.30	0.13
21M	19.43	3018	9.92	8.83	20.98	1669	2.89	11.72	3.48	0.96	0.06	1.36	0.14	1.20	2.51	0.11	0.02	5.07	0.01	0.26	0.12
21M	18.58	2865	9.48	8.46	21.16	1678	2.65	11.43	3.53	0.92	0.04	1.57	0.13	1.16	3.02	0.11	0.02	5.08	0.01	0.26	0.13
21N	24.10	2534	8.84	13.23	19.20	1409	2.64	13.86	3.24	0.84	0.04	1.28	0.10	1.19	2.28	0.10	0.02	4.60	0.02	0.20	0.12
21N	24.45	2773	9.18	13.59	20.13	1420	2.88	13.93	4.54	0.89	0.03	1.01	0.10	1.12	2.17	0.09	0.02	4.86	0.02	0.21	0.13
21N	22.85	2408	8.58	12.98	18.79	1415	2.53	13.75	4.01	0.80	0.04	1.06	0.10	1.10	2.61	0.09	0.02	4.67	0.02	0.19	0.11
22A	208	564	11.60	0.93	4.73	387	1.43	2.27	1.55	0.17	0.08	0.17	1.07	2.66	6.61	0.12	0.01	3.16	0.05	0.64	0.11

22A	193	609	7.06	0.99	5.47	503	1.64	2.17	1.51	0.17	0.07	0.20	1.15	2.46	5.30	0.13	0.01	4.31	0.05	0.71	0.10
22B	15.59	4052	102.71	28.76	17.72	2977	15.17	20.10	29.52	1.33	0.33	4.48	0.38	5.49	25.92	0.41	1.51	20.29	0.03	0.88	1.24
22B	13.66	3794	100.02	26.84	16.31	2914	15.34	19.56	28.66	1.26	0.31	4.47	0.34	5.53	24.51	0.38	0.52	22.99	0.03	0.78	1.19
22C	21.64	3254	6.69	6.07	32.58	1311	2.43	8.96	4.01	0.95	0.06	0.30	0.11	1.29	1.80	0.08	0.01	7.31	0.03	0.31	0.11
22C	21.33	3134	6.57	5.97	32.02	1286	2.47	9.02	4.39	0.92	0.04	0.41	0.10	1.35	1.51	0.08	0.01	7.13	0.03	0.30	0.10
22D	128	4813	19.30	3.63	63.15	4883	9.24	21.11	28.76	1.59	0.79	0.43	3.42	1.54	4.39	0.09	0.46	9.07	0.14	6.28	0.50
22D	119	4475	18.48	3.39	59.92	4638	8.58	21.80	28.51	1.48	0.82	0.25	3.18	1.46	4.87	0.09	0.44	8.30	0.13	5.88	0.49
22E	634	1797	13.93	6.21	45.62	2497	4.56	97.55	21.34	0.69	0.11	0.19	2.97	3.47	42.62	0.62	0.21	11.67	0.02	1.13	0.52
22E	632	1769	13.88	5.80	45.77	2480	4.63	105.41	25.89	0.68	0.04	0.18	2.99	3.34	47.17	0.57	0.21	11.51	0.02	1.11	0.41
22F	1656	3076	29.31	9.22	48.36	9516	15.04	43.65	59.45	1.18	1.14	0.71	4.41	6.01	73.52	0.87	1.78	120.12	0.04	2.89	0.26
22F	1510	2826	26.72	8.39	43.95	8805	14.31	39.99	54.63	1.10	1.02	0.71	4.01	7.09	65.84	0.89	1.53	106.39	0.03	2.70	0.28
22G	829	1560	11.02	7.11	23.11	3119	15.46	33.65	3.01	0.75	0.13	1.24	5.01	3.80	16.77	0.05	0.02	9.94	0.02	0.57	0.59
22G	849	1518	11.11	7.29	23.24	3108	14.76	35.05	2.83	0.74	0.10	1.49	5.13	3.63	23.75	0.05	0.02	9.71	0.03	0.58	0.59

Solution-based ICP-MS REE Data

Sample ID	Analyte (ng g ⁻¹)														
	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Y	Ho	Er	Tm	Yb	Lu
1A	25171	52792	6478	25396	5268	971	4318	649	3739	17291	685	1818	242	1396	178
1A	26698	56283	6897	26786	5571	1023	4562	684	3958	18658	726	1938	259	1499	191
1A	26336	55497	6812	26353	5465	1001	4439	671	3904	18578	717	1910	254	1466	187
1B	1326	2383	336	1372	280	59	254	34	199	1282	41	124	19	125	20
1B	1303	2346	336	1384	289	59	257	34	202	1317	41	125	19	123	19
1B	2977	4561	756	3169	618	99	476	51	251	1405	45	126	18	117	18
1B	1401	2448	353	1456	293	58	258	33	192	1257	40	117	17	114	18
2A	23920	49840	6029	24103	4951	787	3532	401	1811	6276	273	636	81	489	68
2A	20206	41758	5121	20187	4239	681	3062	356	1642	5740	252	599	77	468	66
2A	24909	52344	6458	25667	5479	811	3941	452	1992	6694	292	674	85	507	70
2B	1757	4496	448	1806	393	75	390	60	379	2450	80	238	34	214	31
2B	1732	4476	442	1768	377	73	374	58	366	2419	77	228	33	203	30
2B	1763	4382	439	1766	380	75	379	58	372	2446	79	232	33	208	30
3B	462	2902	160	859	352	109	570	101	584	2348	99	233	28	157	21
3B	471	3134	165	860	344	107	551	99	575	2368	99	232	28	155	21

3B	543	3412	190	981	371	110	567	102	584	2373	100	235	28	157	22
4A	10330	17282	2494	12195	1850	359	1670	218	2542	6753	241	663	89	537	77
4A	10004	17311	2590	14126	1952	356	1718	222	2540	6874	248	683	92	553	80
4A	9851	17145	2518	14124	1880	349	1628	216	2894	6815	242	663	90	539	76
4B	407	787	98	385	87	22	99	16	111	696	24	75	12	79	14
4B	325	603	79	327	79	22	99	16	111	692	25	76	12	80	13
4B	568	1037	119	459	96	22	114	18	124	802	27	85	13	87	14
5B	1172	2056	244	903	164	38	141	21	125	747	25	74	11	70	11
5B	1120	1950	230	859	162	47	144	20	128	752	26	77	11	75	12
5B	1024	1778	212	797	153	43	140	20	128	738	26	76	11	73	11
6B	232	9701	76	265	45	6	30	5	25	96	5	13	2	13	2
6B	232	9734	72	255	42	6	28	5	23	80	4	11	2	12	2
6B	244	10450	80	279	47	6	32	6	25	88	4	12	2	12	2
7B	2091	5374	536	2179	462	102	442	65	387	2315	78	217	30	180	27
7B	1920	4850	486	1961	418	98	408	61	374	2273	76	216	30	182	27
7B	1951	4884	489	1961	420	97	410	61	375	2258	76	218	30	184	27
10B	2739	5703	614	2182	400	52	270	32	152	666	25	63	8	54	8
10B	1321	2480	280	1058	242	25	248	38	220	1212	42	113	15	88	13
10B	443	887	101	373	84	10	76	12	69	344	13	36	5	35	5
11B	4136	22413	945	3486	687	121	523	68	337	1042	53	127	16	96	13
11B	3157	22008	719	2671	565	111	436	59	298	902	48	118	15	95	13
11B	5053	21420	1109	4001	756	129	562	73	361	1110	58	135	17	103	14
13B	13683	27922	3467	13877	2816	320	2606	375	2238	11564	434	1183	160	956	130
13B	11617	23579	2918	11699	2415	317	2271	337	2053	10836	406	1129	155	930	129
13B	15076	30551	3781	15128	3039	337	2796	401	2356	12011	453	1224	164	979	133
15B	558	982	114	433	77	19	65	9	55	329	11	32	5	31	5
15B	579	1040	121	467	89	22	80	12	73	440	15	44	6	42	6
15B	561	978	114	434	75	18	64	9	52	308	10	30	4	28	4
21A	1167	1843	293	1213	243	51	178	22	115	605	20	55	8	49	7
21A	1226	1949	310	1292	255	54	191	23	117	605	21	56	8	50	7
21A	1134	1810	287	1192	237	52	176	21	115	610	21	56	8	49	7
21B	1303	3181	368	1593	336	69	279	35	195	1114	38	107	15	95	15
21B	1330	3099	365	1545	316	66	261	33	186	1038	36	101	14	92	14
21B	2121	4111	585	2464	485	87	378	44	227	1197	41	111	15	97	15
21C	545	2618	153	626	188	47	229	37	215	953	38	90	11	58	8
21C	539	2650	150	620	182	48	223	37	214	953	37	89	10	57	8
21C	522	2538	145	602	177	47	223	37	210	942	37	87	10	56	8
21D	9174	36380	2559	10924	2412	491	2668	412	2680	17107	582	1727	246	1566	246
21D	6444	33918	1745	7345	1618	320	1843	287	1902	13220	428	1299	188	1214	195
21D	6956	30965	1977	8537	1906	385	2247	349	2323	15482	522	1570	225	1431	229
21E	2157	3739	434	1506	235	53	166	21	118	640	23	63	9	57	9

21E	1576	2564	298	1037	173	48	128	17	98	557	19	54	8	50	9
21E	2753	4290	474	1585	230	51	162	21	116	639	22	62	9	56	9
21F	1164	2181	243	924	218	22	226	34	201	1105	39	103	13	79	11
21F	947	1673	190	757	206	45	272	44	275	1765	56	153	19	110	16
21J	1641	3345	447	1899	397	84	324	41	231	1407	44	123	17	106	16
21J	1620	3274	435	1864	391	83	316	41	222	1346	42	118	16	101	15
21J	1596	3269	432	1846	391	81	318	40	220	1321	41	116	16	99	15
21K	2098	3989	578	2600	567	116	435	54	294	1711	55	154	22	136	21
21K	2044	3909	572	2550	563	115	437	54	290	1660	55	153	21	136	20
21K	2031	3926	572	2556	560	115	429	52	288	1675	54	152	21	134	20
21L	1016	1531	256	1165	266	60	234	31	177	1202	35	101	14	89	14
21L	1158	1782	296	1331	293	65	248	32	181	1200	36	100	14	88	14
21L	1164	1778	296	1337	298	66	250	32	179	1181	35	100	14	87	13
21M	473	2260	150	712	272	83	478	88	555	2762	103	252	30	160	23
21M	456	2157	143	682	256	78	443	82	514	2586	96	234	28	148	21
21M	442	2110	140	660	251	77	441	81	513	2551	96	235	28	149	21
21N	511	2462	142	587	174	45	218	36	208	915	37	86	10	56	8
21N	526	2596	147	614	184	47	226	37	214	954	37	89	11	58	8
21N	519	2502	145	606	180	47	228	38	216	959	38	90	11	59	9
22A	578	1391	112	398	72	21	88	13	68	505	15	44	5	54	7
22A	693	1634	129	439	73	20	95	14	69	537	15	45	5	50	7
22B	1902	3691	500	2099	432	86	347	47	264	1598	51	146	21	132	21
22B	1777	3524	473	1988	416	83	342	45	258	1594	51	145	20	131	21
22C	559	3977	167	757	297	91	481	84	497	2344	91	223	27	153	23
22C	558	4026	167	750	287	91	483	84	500	2344	91	224	27	153	23
22D	3862	35847	915	3484	700	167	590	90	532	2610	106	305	44	284	42
22D	3689	33995	879	3336	675	163	564	86	510	2457	102	292	42	272	40
22E	1935	3149	373	1340	216	35	166	22	122	651	23	63	9	57	9
22E	1741	2891	343	1229	201	33	154	20	115	623	22	61	9	56	9
22F	919	1701	214	839	183	64	203	35	249	1606	57	180	28	190	31
22F	879	1643	208	821	182	64	197	34	237	1489	54	169	26	180	29
22G	1654	3031	359	1353	242	54	204	29	174	1021	35	100	14	94	14
22G	1611	3009	359	1369	252	56	216	31	187	1096	38	109	16	102	18

Correlation matrix for solution-based-ICP-MS trace element data for Kaisersberg (AUT)

Kaisersberg (AUT)	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
Mg	1.00																			
Al	0.94	1.00																		
V	0.91	0.99	1.00																	
Cr	0.76	0.89	0.93	1.00																
Mn	0.96	1.00	0.99	0.88	1.00															
Fe	0.97	0.93	0.93	0.86	0.95	1.00														
Ni	-0.85	-0.97	-0.95	-0.83	-0.95	-0.81	1.00													
Cu	-0.96	-0.86	-0.83	-0.67	-0.88	-0.95	0.72	1.00												
Zn	-0.89	-0.83	-0.84	-0.77	-0.85	-0.96	0.67	0.95	1.00											
Ga	0.93	1.00	0.99	0.89	1.00	0.93	-0.97	-0.85	-0.83	1.00										
Se	0.31	0.31	0.36	0.64	0.34	0.46	-0.21	-0.23	-0.38	0.32	1.00									
Rb	0.92	1.00	1.00	0.90	0.99	0.92	-0.97	-0.83	-0.82	1.00	0.32	1.00								
Sr	0.93	1.00	0.99	0.87	1.00	0.92	-0.97	-0.85	-0.82	1.00	0.29	1.00	1.00							
Mo	0.95	0.98	0.97	0.87	0.99	0.93	-0.95	-0.84	-0.79	0.98	0.38	0.98	0.98	1.00						
Ag	-0.91	-0.77	-0.71	-0.46	-0.79	-0.83	0.67	0.95	0.82	-0.76	0.01	-0.73	-0.77	-0.77	1.00					
Cd	-0.92	-0.86	-0.86	-0.79	-0.88	-0.98	0.70	0.96	1.00	-0.85	-0.41	-0.84	-0.85	-0.83	0.83	1.00				
Ba	0.93	1.00	1.00	0.89	0.99	0.93	-0.95	-0.86	-0.85	1.00	0.30	1.00	0.99	0.97	-0.76	-0.87	1.00			
Tl	0.94	1.00	0.99	0.87	1.00	0.93	-0.96	-0.87	-0.84	1.00	0.29	1.00	1.00	0.98	-0.78	-0.86	1.00	1.00		
Pb	0.91	0.94	0.90	0.70	0.94	0.82	-0.96	-0.81	-0.69	0.94	0.06	0.93	0.95	0.94	-0.82	-0.72	0.93	0.95	1.00	
U	0.96	1.00	0.99	0.88	1.00	0.95	-0.95	-0.88	-0.85	1.00	0.34	0.99	1.00	0.99	-0.79	-0.88	0.99	1.00	0.94	1.00

Correlation matrix for solution-based-ICP-MS trace element data for Minas Gerais* (BRA)

Minas Gerais* (BRA)	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
Mg	1.00																			
Al	-0.76	1.00																		
V	-0.78	0.84	1.00																	
Cr	-0.23	0.38	0.73	1.00																
Mn	-0.81	0.86	0.97	0.65	1.00															
Fe	-0.82	0.85	0.99	0.69	0.96	1.00														
Ni	-0.66	0.77	0.89	0.68	0.81	0.91	1.00													
Cu	0.22	-0.09	0.30	0.84	0.17	0.28	0.39	1.00												
Zn	-0.16	-0.04	-0.11	-0.37	-0.06	-0.14	-0.07	-0.48	1.00											
Ga	-0.73	0.98	0.80	0.34	0.79	0.82	0.80	-0.07	-0.07	1.00										
Se	0.38	-0.45	-0.40	-0.10	-0.54	-0.31	-0.08	0.35	-0.32	-0.28	1.00									
Rb	0.05	-0.04	-0.29	-0.44	-0.38	-0.17	0.06	-0.19	-0.08	0.14	0.73	1.00								
Sr	0.17	0.11	0.26	0.45	0.22	0.23	0.20	0.47	-0.61	0.08	-0.07	-0.23	1.00							
Mo	-0.76	0.84	0.97	0.72	0.92	0.98	0.91	0.35	-0.20	0.83	-0.21	-0.11	0.23	1.00						
Ag	0.30	-0.36	-0.47	-0.35	-0.57	-0.37	-0.14	0.04	-0.23	-0.19	0.93	0.87	-0.20	-0.29	1.00					
Cd	-0.63	0.82	0.93	0.80	0.90	0.92	0.82	0.42	-0.31	0.79	-0.29	-0.27	0.24	0.94	-0.37	1.00				
Ba	-0.81	0.82	0.96	0.65	1.00	0.96	0.81	0.19	-0.03	0.75	-0.52	-0.38	0.21	0.91	-0.56	0.87	1.00			
Tl	-0.75	0.84	0.97	0.70	0.99	0.94	0.79	0.22	-0.07	0.75	-0.58	-0.46	0.28	0.90	-0.63	0.90	0.99	1.00		
Pb	-0.83	0.88	0.93	0.53	0.96	0.93	0.78	0.06	-0.04	0.82	-0.53	-0.28	0.27	0.88	-0.50	0.83	0.96	0.95	1.00	
U	-0.79	0.85	1.00	0.72	0.99	0.98	0.86	0.27	-0.11	0.79	-0.45	-0.34	0.25	0.95	-0.52	0.93	0.98	0.98	0.95	1.00

Correlation matrix for solution-based-ICP-MS trace element data for Hunan Lutang* (CHN)

Hunan Lutang* (CHN)	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
Mg	1.00																			
Al	0.56	1.00																		
V	0.94	0.23	1.00																	
Cr	0.25	0.94	-0.10	1.00																
Mn	0.73	-0.16	0.92	-0.48	1.00															
Fe	0.93	0.22	1.00	-0.11	0.93	1.00														
Ni	0.11	-0.76	0.45	-0.93	0.76	0.46	1.00													
Cu	0.68	-0.24	0.89	-0.54	1.00	0.89	0.81	1.00												
Zn	-0.15	0.74	-0.48	0.92	-0.78	-0.49	-1.00	-0.83	1.00											
Ga	0.57	1.00	0.25	0.94	-0.15	0.23	-0.76	-0.22	0.73	1.00										
Se	-0.94	-0.23	-1.00	0.10	-0.92	-1.00	-0.45	-0.89	0.48	-0.24	1.00									
Rb	0.54	1.00	0.21	0.95	-0.18	0.20	-0.78	-0.26	0.75	1.00	-0.21	1.00								
Sr	-0.94	-0.81	-0.76	-0.57	-0.45	-0.75	0.24	-0.38	-0.20	-0.82	0.76	-0.80	1.00							
Mo	-0.76	0.12	-0.94	0.44	-1.00	-0.94	-0.73	-0.99	0.76	0.11	0.94	0.15	0.48	1.00						
Ag	0.91	0.15	1.00	-0.18	0.95	1.00	0.52	0.93	-0.56	0.16	-1.00	0.13	-0.70	-0.96	1.00					
Cd	-0.67	-0.99	-0.37	-0.89	0.02	-0.36	0.66	0.09	-0.63	-0.99	0.37	-0.99	0.89	0.02	-0.29	1.00				
Ba	0.85	0.91	0.62	0.72	0.26	0.61	-0.43	0.19	0.39	0.91	-0.62	0.90	-0.98	-0.30	0.55	-0.96	1.00			
Tl	0.99	0.67	0.88	0.39	0.62	0.87	-0.03	0.56	-0.01	0.68	-0.88	0.65	-0.98	-0.65	0.83	-0.77	0.92	1.00		
Pb	-0.21	-0.93	0.14	-1.00	0.51	0.15	0.95	0.58	-0.93	-0.93	-0.14	-0.94	0.54	-0.48	0.22	0.87	-0.69	-0.35	1.00	
U	0.44	-0.51	0.72	-0.76	0.93	0.73	0.94	0.96	-0.96	-0.50	-0.72	-0.53	-0.10	-0.92	0.78	0.38	-0.10	0.30	0.79	1.00

Correlation matrix for solution-based-ICP-MS trace element data for Inner Mongolia (CHN)

Inner Mongolia* (CHN)	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
Mg	1.00																			
Al	0.92	1.00																		
V	1.00	0.94	1.00																	
Cr	0.99	0.86	0.98	1.00																
Mn	-0.81	-0.97	-0.84	-0.72	1.00															
Fe	-0.13	0.27	-0.08	-0.27	-0.47	1.00														
Ni	0.94	0.73	0.92	0.98	-0.56	-0.46	1.00													
Cu	0.97	0.81	0.96	1.00	-0.66	-0.35	0.99	1.00												
Zn	-0.21	0.19	-0.16	-0.35	-0.40	1.00	-0.54	-0.43	1.00											
Ga	-0.19	0.21	-0.14	-0.33	-0.42	1.00	-0.52	-0.41	1.00	1.00										
Se	0.88	0.63	0.86	0.94	-0.44	-0.58	0.99	0.97	-0.64	-0.63	1.00									
Rb	0.98	0.82	0.97	1.00	-0.68	-0.33	0.99	1.00	-0.40	-0.39	0.96	1.00								
Sr	0.75	0.95	0.78	0.65	-1.00	0.56	0.48	0.58	0.49	0.51	0.35	0.60	1.00							
Mo	0.98	0.83	0.97	1.00	-0.69	-0.32	0.99	1.00	-0.39	-0.37	0.96	1.00	0.61	1.00						
Ag	0.85	0.99	0.88	0.77	-1.00	0.41	0.62	0.71	0.33	0.35	0.51	0.73	0.99	0.74	1.00					
Cd	0.98	0.82	0.96	1.00	-0.67	-0.34	0.99	1.00	-0.42	-0.40	0.96	1.00	0.59	1.00	0.72	1.00				
Ba	0.97	0.80	0.96	0.99	-0.64	-0.37	0.99	1.00	-0.44	-0.43	0.97	1.00	0.57	1.00	0.70	1.00	1.00			
Tl	-0.82	-0.98	-0.85	-0.73	1.00	-0.46	-0.57	-0.67	-0.39	-0.41	-0.46	-0.69	-0.99	-0.70	-1.00	-0.68	-0.65	1.00		
Pb	0.49	0.11	0.44	0.61	0.11	-0.93	0.76	0.67	-0.96	-0.95	0.84	0.65	-0.21	0.65	-0.04	0.66	0.69	0.10	1.00	
U	0.99	0.96	1.00	0.96	-0.88	0.00	0.89	0.94	-0.08	-0.06	0.82	0.94	0.83	0.95	0.91	0.94	0.93	-0.89	0.37	1.00

Correlation matrix for solution-based-ICP-MS trace element data for Heilongjian (CHN)

Heilongjian* (CHN)	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
Mg	1.00																			
Al	0.76	1.00																		
V	0.94	0.50	1.00																	
Cr	0.81	0.24	0.96	1.00																
Mn	0.99	0.68	0.97	0.87	1.00															
Fe	1.00	0.80	0.92	0.78	0.99	1.00														
Ni	0.96	0.56	1.00	0.94	0.99	0.95	1.00													
Cu	1.00	0.80	0.92	0.77	0.98	1.00	0.94	1.00												
Zn	0.99	0.85	0.88	0.72	0.97	1.00	0.91	1.00	1.00											
Ga	0.54	0.96	0.23	-0.05	0.45	0.59	0.30	0.60	0.66	1.00										
Se	-0.07	0.60	-0.40	-0.64	-0.18	-0.01	-0.33	0.00	0.08	0.80	1.00									
Rb	0.56	0.96	0.24	-0.03	0.46	0.61	0.31	0.61	0.68	1.00	0.79	1.00								
Sr	0.94	0.93	0.77	0.57	0.90	0.96	0.82	0.96	0.98	0.79	0.27	0.80	1.00							
Mo	1.00	0.79	0.92	0.78	0.99	1.00	0.95	1.00	1.00	0.59	-0.02	0.60	0.96	1.00						
Ag	0.86	0.99	0.64	0.40	0.80	0.89	0.69	0.89	0.93	0.90	0.45	0.90	0.98	0.89	1.00					
Cd	0.89	0.97	0.68	0.46	0.83	0.92	0.73	0.92	0.95	0.87	0.40	0.88	0.99	0.91	1.00	1.00				
Ba	0.60	-0.06	0.84	0.95	0.69	0.55	0.79	0.55	0.48	-0.34	-0.84	-0.33	0.30	0.56	0.11	0.17	1.00			
Tl	-0.87	-0.33	-0.98	-1.00	-0.91	-0.83	-0.97	-0.83	-0.78	-0.05	0.56	-0.07	-0.65	-0.84	-0.49	-0.54	-0.92	1.00		
Pb	0.86	0.33	0.98	1.00	0.91	0.83	0.97	0.83	0.78	0.04	-0.56	0.06	0.64	0.83	0.48	0.54	0.92	-1.00	1.00	
U	-0.99	-0.82	-0.90	-0.75	-0.98	-1.00	-0.93	-1.00	-1.00	-0.63	-0.04	-0.64	-0.97	-1.00	-0.91	-0.93	-0.52	0.81	-0.80	1.00

Correlation matrix for solution-based-ICP-MS trace element data for Kropfmühl (GER)

Kropfmühl (GER)	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
Mg	1.00																			
Al	0.82	1.00																		
V	0.67	0.12	1.00																	
Cr	0.82	0.34	0.97	1.00																
Mn	0.87	0.43	0.95	1.00	1.00															
Fe	0.96	0.64	0.84	0.94	0.97	1.00														
Ni	-0.80	-1.00	-0.09	-0.31	-0.40	-0.61	1.00													
Cu	0.84	0.37	0.97	1.00	1.00	0.95	-0.34	1.00												
Zn	-1.00	-0.77	-0.73	-0.86	-0.91	-0.98	0.75	-0.88	1.00											
Ga	0.20	-0.40	0.86	0.72	0.65	0.45	0.44	0.70	-0.27	1.00										
Se	-0.99	-0.71	-0.79	-0.90	-0.94	-0.99	0.68	-0.92	1.00	-0.36	1.00									
Rb	0.95	0.95	0.41	0.61	0.68	0.84	-0.94	0.63	-0.93	-0.11	-0.89	1.00								
Sr	-0.35	0.25	-0.93	-0.83	-0.77	-0.59	-0.28	-0.81	0.43	-0.99	0.51	-0.05	1.00							
Mo	-0.21	-0.73	0.59	0.39	0.30	0.06	0.75	0.36	0.12	0.92	0.04	-0.49	-0.84	1.00						
Ag	-0.73	-0.21	-1.00	-0.99	-0.97	-0.89	0.18	-0.99	0.79	-0.81	0.84	-0.49	0.90	-0.51	1.00					
Cd	-0.46	0.13	-0.97	-0.89	-0.84	-0.69	-0.16	-0.87	0.54	-0.96	0.61	-0.18	0.99	-0.77	0.94	1.00				
Ba	0.25	0.76	-0.55	-0.35	-0.26	-0.02	-0.78	-0.32	-0.17	-0.90	-0.08	0.53	0.82	-1.00	0.47	0.74	1.00			
Tl	0.83	1.00	0.14	0.36	0.44	0.65	-1.00	0.38	-0.78	-0.39	-0.72	0.96	0.23	-0.72	-0.22	0.11	0.75	1.00		
Pb	-0.85	-1.00	-0.18	-0.39	-0.48	-0.67	1.00	-0.42	0.80	0.35	0.75	-0.97	-0.20	0.69	0.26	-0.08	-0.73	-1.00	1.00	
U	-0.66	-0.97	0.12	-0.11	-0.20	-0.43	0.98	-0.14	0.59	0.61	0.52	-0.85	-0.47	0.87	-0.03	-0.36	-0.89	-0.97	0.96	1.00

Correlation matrix for solution-based-ICP-MS trace element data for Balama (MOZ)

Balama (MOZ)	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
Mg	1.00																			
Al	-0.79	1.00																		
V	-0.78	0.84	1.00																	
Cr	-0.74	0.73	0.81	1.00																
Mn	-0.16	-0.42	-0.21	-0.16	1.00															
Fe	-0.74	0.31	0.36	0.34	0.71	1.00														
Ni	-0.78	0.96	0.87	0.78	-0.42	0.24	1.00													
Cu	-0.84	0.86	0.73	0.53	-0.01	0.63	0.81	1.00												
Zn	0.56	-0.48	-0.31	-0.63	0.10	-0.24	-0.50	-0.30	1.00											
Ga	-0.65	0.84	0.92	0.64	-0.36	0.27	0.79	0.72	-0.15	1.00										
Se	-0.64	0.96	0.80	0.61	-0.59	0.11	0.90	0.76	-0.33	0.89	1.00									
Rb	0.71	-0.80	-0.42	-0.49	0.27	-0.32	-0.76	-0.74	0.61	-0.38	-0.69	1.00								
Sr	-0.50	0.62	0.74	0.57	-0.15	0.38	0.52	0.57	-0.02	0.88	0.66	-0.14	1.00							
Mo	-0.47	0.84	0.44	0.39	-0.63	-0.02	0.79	0.67	-0.40	0.51	0.84	-0.88	0.23	1.00						
Ag	0.53	0.00	-0.11	-0.30	-0.82	-0.89	0.04	-0.27	0.30	0.03	0.22	0.10	-0.20	0.31	1.00					
Cd	-0.34	0.71	0.57	0.27	-0.56	-0.11	0.70	0.56	-0.07	0.61	0.76	-0.52	0.26	0.73	0.47	1.00				
Ba	-0.59	0.68	0.83	0.64	-0.18	0.36	0.60	0.58	-0.13	0.94	0.72	-0.18	0.97	0.26	-0.16	0.32	1.00			
Tl	0.65	-0.91	-0.84	-0.73	0.59	-0.04	-0.91	-0.65	0.42	-0.85	-0.94	0.62	-0.60	-0.74	-0.21	-0.67	-0.71	1.00		
Pb	0.91	-0.75	-0.55	-0.63	-0.18	-0.75	-0.71	-0.84	0.62	-0.43	-0.54	0.85	-0.32	-0.58	0.56	-0.29	-0.35	0.50	1.00	
U	0.93	-0.83	-0.64	-0.64	-0.09	-0.72	-0.77	-0.90	0.53	-0.58	-0.66	0.85	-0.46	-0.64	0.49	-0.39	-0.49	0.61	0.97	1.00

Correlation matrix for solution-based-ICP-MS trace element data for Skaland (NOR)

Skaland (NOR)	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
Mg	1.00																			
Al	0.83	1.00																		
V	0.55	0.91	1.00																	
Cr	0.96	0.91	0.67	1.00																
Mn	0.43	0.85	0.99	0.60	1.00															
Fe	0.91	0.96	0.80	0.90	0.70	1.00														
Ni	0.87	0.98	0.86	0.89	0.76	0.99	1.00													
Cu	0.13	-0.42	-0.75	-0.05	-0.83	-0.23	-0.32	1.00												
Zn	0.30	0.77	0.96	0.47	0.99	0.61	0.69	-0.90	1.00											
Ga	0.99	0.88	0.61	0.97	0.50	0.93	0.90	0.06	0.37	1.00										
Se	0.60	0.92	0.99	0.69	0.95	0.85	0.89	-0.69	0.92	0.65	1.00									
Rb	0.95	0.92	0.69	1.00	0.61	0.91	0.90	-0.08	0.50	0.97	0.71	1.00								
Sr	0.61	0.92	0.98	0.74	0.97	0.81	0.86	-0.69	0.92	0.67	0.96	0.75	1.00							
Mo	0.70	0.96	0.97	0.80	0.94	0.88	0.92	-0.60	0.88	0.75	0.98	0.81	0.98	1.00						
Ag	0.42	0.84	0.99	0.56	0.99	0.71	0.77	-0.84	0.99	0.48	0.97	0.58	0.96	0.93	1.00					
Cd	0.50	0.88	1.00	0.64	0.99	0.76	0.82	-0.79	0.98	0.56	0.98	0.65	0.98	0.96	0.99	1.00				
Ba	0.92	0.73	0.40	0.93	0.32	0.76	0.73	0.24	0.19	0.92	0.41	0.93	0.48	0.55	0.26	0.36	1.00			
Tl	0.57	0.92	1.00	0.70	0.98	0.81	0.86	-0.73	0.95	0.62	0.99	0.71	0.98	0.98	0.98	0.99	0.42	1.00		
Pb	0.45	0.86	0.99	0.59	0.99	0.74	0.80	-0.82	0.98	0.52	0.98	0.61	0.97	0.95	1.00	1.00	0.30	0.99	1.00	
U	0.61	0.93	0.99	0.71	0.96	0.85	0.89	-0.69	0.93	0.67	1.00	0.73	0.98	0.98	0.97	0.98	0.44	0.99	0.98	1.00

Correlation matrix for solution-based-ICP-MS trace element data for Tayginka (RUS)

Tayginka (RUS)	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
Mg	1.00																			
Al	0.55	1.00																		
V	0.96	0.52	1.00																	
Cr	-0.81	-0.63	-0.66	1.00																
Mn	0.70	-0.20	0.65	-0.48	1.00															
Fe	0.69	-0.22	0.65	-0.44	1.00	1.00														
Ni	0.87	0.76	0.76	-0.94	0.42	0.40	1.00													
Cu	0.92	0.29	0.85	-0.81	0.87	0.86	0.81	1.00												
Zn	0.27	0.92	0.31	-0.36	-0.48	-0.49	0.48	-0.03	1.00											
Ga	0.83	0.92	0.80	-0.81	0.21	0.19	0.92	0.64	0.73	1.00										
Se	0.49	0.99	0.44	-0.61	-0.26	-0.29	0.72	0.22	0.92	0.87	1.00									
Rb	0.59	-0.35	0.57	-0.32	0.98	0.99	0.26	0.77	-0.59	0.06	-0.41	1.00								
Sr	0.40	-0.54	0.37	-0.17	0.93	0.94	0.08	0.63	-0.74	-0.16	-0.59	0.97	1.00							
Mo	0.88	0.30	0.82	-0.79	0.83	0.82	0.79	0.98	-0.02	0.64	0.21	0.73	0.58	1.00						
Ag	-0.70	0.07	-0.64	0.64	-0.93	-0.93	-0.54	-0.90	0.32	-0.30	0.14	-0.88	-0.82	-0.88	1.00					
Cd	0.18	0.91	0.19	-0.38	-0.52	-0.53	0.51	-0.03	0.93	0.69	0.90	-0.65	-0.78	0.01	0.33	1.00				
Ba	-0.40	-0.97	-0.34	0.61	0.32	0.35	-0.70	-0.18	-0.90	-0.83	-0.98	0.48	0.64	-0.20	-0.17	-0.94	1.00			
Tl	0.06	-0.48	0.08	0.29	0.46	0.48	-0.20	0.17	-0.66	-0.28	-0.49	0.52	0.59	0.11	-0.20	-0.59	0.56	1.00		
Pb	0.55	0.98	0.49	-0.70	-0.15	-0.18	0.79	0.34	0.86	0.91	0.97	-0.32	-0.50	0.36	0.01	0.88	-0.98	-0.48	1.00	
U	0.46	-0.15	0.30	-0.55	0.78	0.74	0.43	0.72	-0.49	0.15	-0.19	0.70	0.69	0.74	-0.76	-0.33	0.14	0.43	0.00	1.00

Correlation matrix for solution-based-ICP-MS trace element data for Ragedara (LKA)

Ragedara (LKA)	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
Mg	1.00																			
Al	0.76	1.00																		
V	0.93	0.57	1.00																	
Cr	0.33	0.84	0.23	1.00																
Mn	-0.32	-0.68	-0.38	-0.91	1.00															
Fe	-0.21	-0.40	-0.41	-0.68	0.92	1.00														
Ni	-0.58	-0.81	-0.60	-0.88	0.96	0.86	1.00													
Cu	-0.50	-0.77	-0.54	-0.89	0.98	0.89	1.00	1.00												
Zn	-0.71	-0.58	-0.43	-0.08	-0.20	-0.44	0.04	-0.04	1.00											
Ga	0.22	-0.07	-0.02	-0.52	0.78	0.91	0.61	0.67	-0.75	1.00										
Se	-0.48	-0.65	-0.47	-0.63	0.61	0.53	0.67	0.66	0.28	0.32	1.00									
Rb	-0.35	-0.22	-0.04	0.23	-0.57	-0.80	-0.39	-0.46	0.89	-0.95	-0.09	1.00								
Sr	-0.08	-0.59	-0.10	-0.93	0.96	0.84	0.85	0.89	-0.29	0.80	0.56	-0.58	1.00							
Mo	-0.10	-0.37	0.27	-0.16	-0.27	-0.61	-0.21	-0.24	0.72	-0.67	0.06	0.83	-0.15	1.00						
Ag	-0.66	-0.81	-0.70	-0.83	0.92	0.84	0.99	0.98	0.09	0.56	0.68	-0.35	0.78	-0.24	1.00					
Cd	-0.61	-0.66	-0.27	-0.26	-0.09	-0.42	0.09	0.03	0.96	-0.69	0.25	0.87	-0.13	0.85	0.12	1.00				
Ba	-0.69	-0.78	-0.37	-0.39	0.08	-0.25	0.27	0.20	0.92	-0.56	0.29	0.77	0.02	0.77	0.29	0.98	1.00			
Tl	-0.31	-0.56	-0.45	-0.79	0.97	0.98	0.93	0.96	-0.31	0.85	0.61	-0.69	0.90	-0.46	0.91	-0.25	-0.09	1.00		
Pb	-0.68	-0.92	-0.63	-0.89	0.89	0.72	0.97	0.95	0.25	0.42	0.65	-0.17	0.78	-0.01	0.97	0.32	0.48	0.83	1.00	
U	0.53	0.31	0.66	0.21	-0.45	-0.53	-0.56	-0.53	0.10	-0.30	0.12	0.33	-0.24	0.54	-0.60	0.14	-0.03	-0.51	-0.53	1.00

Correlation matrix for solution-based-ICP-MS trace element data for Zavallia (UKR)

Zavallia (UKR)	Mg	Al	V	Cr	Mn	Fe	Ni	Cu	Zn	Ga	Se	Rb	Sr	Mo	Ag	Cd	Ba	Tl	Pb	U
Mg	1.00																			
Al	-0.96	1.00																		
V	1.00	-0.95	1.00																	
Cr	0.87	-0.72	0.90	1.00																
Mn	0.99	-0.94	1.00	0.91	1.00															
Fe	1.00	-0.96	1.00	0.87	0.99	1.00														
Ni	1.00	-0.97	1.00	0.86	0.99	1.00	1.00													
Cu	1.00	-0.96	1.00	0.88	1.00	1.00	1.00	1.00												
Zn	1.00	-0.96	1.00	0.88	0.99	1.00	1.00	1.00	1.00											
Ga	-0.75	0.90	-0.71	-0.34	-0.70	-0.75	-0.76	-0.75	-0.73	1.00										
Se	0.95	-0.90	0.94	0.81	0.92	0.95	0.96	0.94	0.95	-0.68	1.00									
Rb	1.00	-0.96	1.00	0.88	1.00	1.00	1.00	1.00	1.00	-0.74	0.95	1.00								
Sr	0.97	-0.99	0.95	0.74	0.94	0.97	0.98	0.97	0.97	-0.86	0.94	0.97	1.00							
Mo	1.00	-0.96	1.00	0.87	0.99	1.00	1.00	1.00	1.00	-0.75	0.95	1.00	0.97	1.00						
Ag	-0.98	0.94	-0.97	-0.85	-0.97	-0.98	-0.97	-0.98	-0.98	0.71	-0.90	-0.98	-0.95	-0.98	1.00					
Cd	1.00	-0.96	1.00	0.89	1.00	1.00	1.00	1.00	1.00	-0.73	0.94	1.00	0.97	1.00	-0.98	1.00				
Ba	1.00	-0.96	1.00	0.88	1.00	1.00	1.00	1.00	1.00	-0.74	0.94	1.00	0.97	1.00	-0.98	1.00	1.00			
Tl	0.98	-0.93	0.99	0.89	0.99	0.98	0.98	0.98	0.97	-0.71	0.92	0.98	0.93	0.98	-0.93	0.98	0.98	1.00		
Pb	1.00	-0.96	1.00	0.87	0.99	1.00	1.00	1.00	1.00	-0.74	0.95	1.00	0.97	1.00	-0.98	1.00	1.00	0.98	1.00	
U	0.98	-0.99	0.97	0.78	0.96	0.98	0.99	0.98	0.98	-0.84	0.94	0.98	1.00	0.98	-0.97	0.98	0.98	0.95	0.98	1.00

E. Sample List

Sample ID	Mine/Province	Country	Continent	Note
1A	Kaisersberg	Austria	Europe	
2A	Kaisersberg	Austria	Europe	pellet not stable enough for LA-analysis
4A	Hunan Lutang	China	Asia	
1B	Balama	Mozambique	Africa	
2B	Skaland	Norway	Europe	
3B	Minas Gerais	Brazil	South America	
4B	Zavallia	Ukraine	Europe	
5B	Tayginka	Russia	Asia	
6B	Inner Mongolia	China	Asia	
7B	Heilongjian	China	Asia	
10B	Ragedara	Sri Lanka	Asia	
11B	Brickaville	Madagascar	Africa	
13B	Passau	Germany	Europe	
14B	Passau (raw ore)	Germany	Europe	Graphite particles too small for laser-ablation
15B	Skaland	Norway	Europe	
16B	Skaland (raw ore)	Norway	Europe	Graphite particles too small for laser-ablation
21A	Shandong	China	Asia	
21B	Balama	Mozambique	Africa	
21C	Minas Gerais	Brazil	South America	
21D	Brickaville	Madagascar	Africa	
21E	Tayginka	Russia	Asia	
21F	Ragedara	Sri Lanka	Asia	
21G, H, I	Kaisersberg (raw ore)	Austria	Europe	
21J, K, L	Balama	Mozambique	Africa	
21M, N	Minas Gerais	Brazil	South America	
22A	Shandong	China	Asia	
22B	Balama	Mozambique	Africa	
22C	Minas Gerais	Brazil	South America	
22D	Brickaville	Madagascar	Africa	
22E	Tayginka	Russia	Asia	
22F	Zavallia	Ukraine	Europe	
22G	Skaland	Norway	Europe	
22H	Balama (raw ore)	Mozambique	Africa	
22K-S	Zavallia	Ukraine	Europe	Graphite particles too small for laser-ablation

F. AI Declaration

Subject	AI share in %	Tool/Version	Remarks	Prompting
Translation	10	DeepL (Free Version via Browser)	Translation from german to english, DeepL through browser	Online document
Translation	10	ChatGPT-3.5, -4	Translation from german to english	Online document
Finding synonyms	5	ChatGPT-3.5, -4	Improving readability	Online document
Suggestions for the structure of the work (table of content, outlook and recommendations)	5	ChatGPT-3.5, -4,	Improving linguistic readability and understanding for reader	Online document
Summarize own texts and suggest paraphrasing	8	ChatGPT-3.5, -4,	Improving linguistic readability and understanding	Online document