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Review

The isotopic and elemental patterns of uranium ore as tools for its provenance determination: A systematic review

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ABSTRACT

This systematic review explores the potential of isotopic and elemental analysis of uranium ore as tools for provenance determination. This capability is vital for effective criminal investigations involving illicit trafficking, environmental contamination, terrorism and nuclear proliferation. A comprehensive literature review was conducted across multiple databases, focusing on studies that utilized isotopic and elemental analysis techniques for uranium origin assessment. The included studies were critically evaluated for methodological rigor. The review found that both isotopic and elemental analysis provides valuable insights into the provenance of uranium ore. Isotopic analysis, in particular, has been established as a powerful tool for tracking the origin of uranium ore. It has potential applications in forensic investigations, nuclear security and safeguards. However, the full potential of elemental patterns remains underutilized. Integrating both techniques could enhance the discriminatory power of provenance assessments. Key challenges, such as limitations in existing databases, analytical techniques and sample contaminations, hinder the definitive determination of uranium origin. To address these challenges, the review emphasizes the need for collaboration between researchers and law enforcement agencies. This collaboration should focus on expanding geochemical databases, developing standardized methodologies, and fostering interdisciplinary cooperation. By overcoming these limitations, the forensic community can significantly improve its ability to investigate uranium-related crimes, contributing to national security, safeguards and environmental protection.

1. Introduction

1.1. Background

Uranium, a radioactive element vital for nuclear energy, medicine, industry, and research, presents a significant global security and health risk. It's potential for misuse in illicit activities, including theft, smuggling, and nuclear proliferation, combined with its association with environmental contamination, poses a serious threat [1–6]. Accurately determining the origin of uranium is essential for tracing its path from mine to market, aiding in criminal investigations and preventing nuclear proliferation [7].

The isotopic and elemental patterns within uranium ore offer a unique fingerprint that can link a specific uranium sample to its geographic origin [8–10]. By understanding these patterns, law enforcement agencies can develop robust forensic tools to trace the movement of illicit uranium [11]. This systematic review aims to comprehensively examine the current state of knowledge regarding the application of isotopic and elemental patterns in uranium provenance determination for the purpose of criminal investigation and prosecution.

The review synthesizes existing research, knowledge gaps, and proposes future research directions to enhance the use of uranium isotopic and elemental signatures as forensic evidence [12,13]. By understanding the potential of these techniques, law enforcement agencies can strengthen their ability to dismantle criminal networks involved in uranium trafficking, prevent nuclear proliferation, and ensure the effective prosecution of related offenses.

Forensic investigations into uranium-related crimes necessitate

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robust methodologies for tracing the element to its origin [2,9,14]. Isotopic and elemental signatures within uranium ore offer potential as powerful tools for provenance determination. Nevertheless, a comprehensive evaluation of current knowledge, identification of research gaps, and assessment of the practical application of these techniques in forensic contexts are crucial for developing effective countermeasures to uranium-related criminal activities.

Ultimately, this review aims to contribute to the development of a robust forensic framework for combating uranium-related criminal activities, safeguarding national and international security, and protecting public safety. The insights derived from this study will be invaluable for law enforcement agencies, policymakers, and researchers seeking to prevent the illicit use of uranium.

1.2. Statement of the Problem and purpose

Illicit uranium trafficking, nuclear proliferation, and environmental contamination present significant global challenges. Accurate determination of uranium origin is crucial for effective law enforcement, counter-proliferation, and environmental remediation. While isotopic analysis is widely used for uranium provenance, the potential of elemental patterns as a complementary tool remains under-explored. This review aims to address the gap in knowledge regarding the comprehensive application of both isotopic and elemental patterns. By synthesizing existing research, it will assess the current state of knowledge, identify limitations, and explore the potential of these techniques in forensic investigations. Ultimately, this study seeks to contribute to the development of robust forensic methodologies for combating uranium-related criminal activities and safeguarding national security.

2. Methodology

This systematic review employed a rigorous methodology to identify, assess, and synthesize relevant research. A comprehensive literature search was conducted across four major academic databases: Web of Science, Google Scholar, Scopus and PubMed. The search utilized a combination of keywords (uranium, provenance determination, isotopic analysis, elemental patterns, criminal investigation, and forensic science) and Medical Subject Headings (MeSH) terms to maximize the retrieval of relevant studies. Studies were included based on specific criteria, excluding those that did not directly address the use of isotopic and elemental analysis for uranium ore provenance determination. The quality of included studies was assessed using the Critical Appraisal Skills Programme (CASP) checklist. Data extraction involved collecting information on author details, publication year, study design, analytical techniques, key findings, and limitations.

A qualitative thematic analysis was used to synthesize the extracted data, identifying recurring themes and patterns. As appropriate, a quantitative *meta*-analysis was conducted to summarize the data on the accuracy and precision of different techniques. The results of the review are presented in a manuscript following Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) guidelines [43],

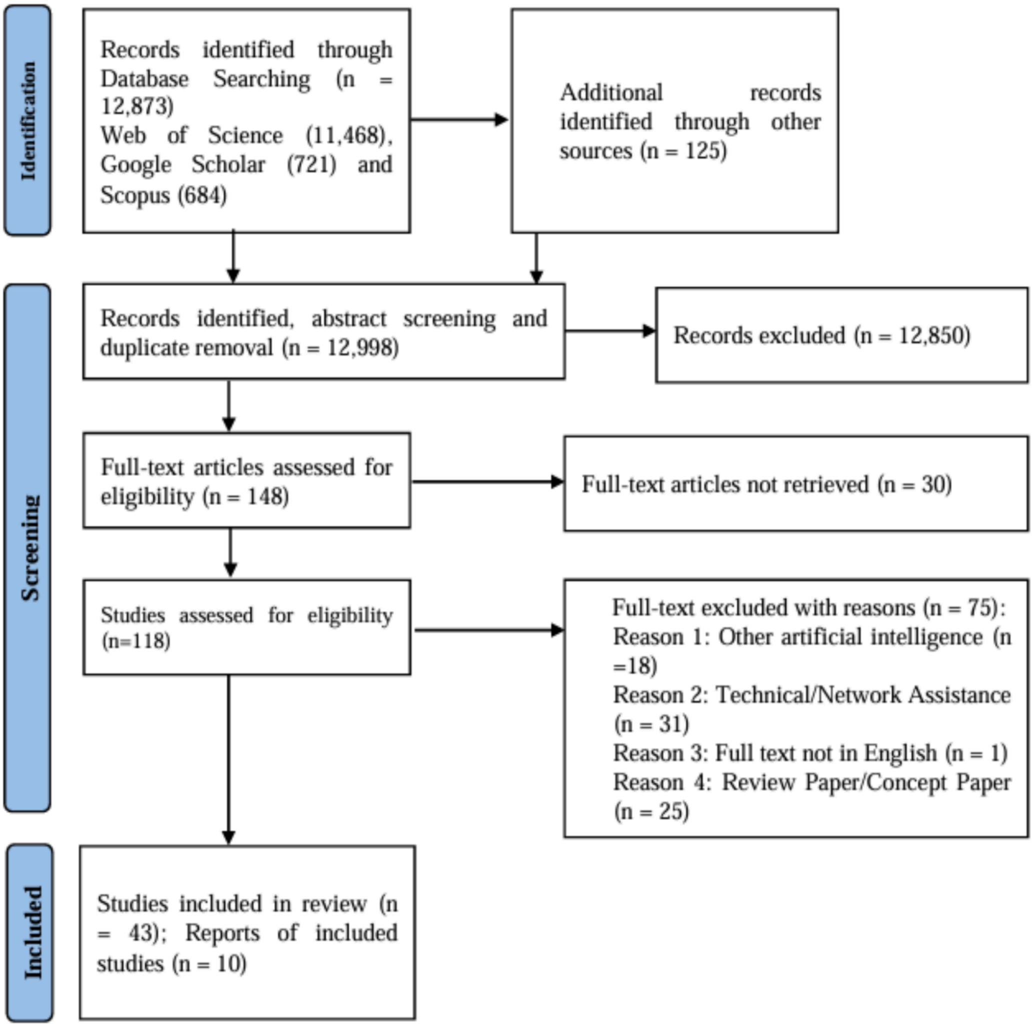

Fig. 1. PRISMA 2020 Flowchart of the study collection [43].

ensuring transparency and clarity in the reporting of the search strategy, study selection, data analysis, and findings Fig. 1. The impact of excluding potentially relevant studies was assessed, and the limitations of the review, such as reliance on published literature, are acknowledged.

3. Literature review

3.1. Theoretical framework on isotopic, chemical and elemental characteristics of the uranium ore for identifying the origin of uranium

3.1.1. Analysis of the ratios of isotopes of uranium and thorium (^{234}U/^{238}U; ^{235}U/^{238}U, ^{236}U/^{238}U and ^{232}Th/^{238}U) in uranium ore

A theoretical review of the literature explores the use of uranium and thorium isotope ratios for determining the origin of uranium ore [15]. Studies from various countries have demonstrated variations in these isotope ratios, particularly in the ^{235}U/^{238}U ratio [16,17]. Factors such as the type of uranium deposit (e.g., sandstone-hosted versus magmatic) and geological environment influence these variations. A number of researches suggest that analyzing isotope ratios can effectively differentiate between uranium ores from diverse origins (Table 1). This method has potential to trace the source of uranium, a critical component in numerous nuclear applications [18,19].

Unfortunately, due to analytical limitations, most previous studies have concluded the potential origin of uranium based on a limited number of characteristics [9,20]. While these limited characteristics can be informative, definitive conclusions often require complementary data [12,13]. This review aims to examine the potential of isotopic and elemental patterns in uranium ore as tools for provenance determination.

3.1.2. Analysis of the ratios of stable isotopes of lead (^{204}Pb/^{206}Pb, ^{207}Pb/^{206}Pb and ^{208}Pb/^{206}Pb) in uranium ore

The determination of stable lead isotope ratios in uranium ore has been employed as a characterization tool in various countries [21]. Among other findings, significant variations in lead isotopic ratios (^{204}Pb/^{206}Pb, ^{207}Pb/^{206}Pb, and ^{208}Pb/^{206}Pb) for uranium ore from Rössing, Namibia (Table 2) were identified. Similarly, variations were observed in lead isotope ratios from Phalabora, South Africa (Table 3). The ability to distinguish uranium-bearing minerals from three Australian mines was demonstrated based on lead isotopic compositions using multivariate statistical analysis [21,31]

The long-lived uranium isotopes, ^{235}U and ^{238}U, decay to ^{207}Pb and ^{206}Pb, respectively. The ratio of these radiogenic lead isotopes is dependent on the age of the rock, making it a valuable tool for estimating uranium deposit age. In a closed system with no uranium or lead loss or gain, the ^{207}Pb/^{206}Pb ratio increases steadily over time since the ore body's formation. Consequently, variations in the ^{207}Pb/^{206}Pb isotope ratio can be correlated with the age of the uranium deposit [14].

Table 1

Isotope ratios of n(^{234}U/^{238}U), n(^{235}U/^{238}U) and n(^{236}U/^{238}U) measured for six uranium ore samples [15].

Country of Origin	Milling Facility	n(^{234}U/n(^{238}U)	n(^{235}U/n(^{238}U)	n(^{236}U/n(^{238}U)
Gabon	Comuf Mounana	5.434(42) x 10^{-5}	0.007 235 7(36)	9.32(69) x 10^{-9}
Czech Republic	DIAMO, Straz Pod Ralskem	8.355(49) x 10^{-5}	0.007 256 0(36)	2.540(92) x 10^{-8}
Canada	CAMECO Rabbit Lake Op.	5.444(48) x 10^{-5}	0.007 256 8(36)	2.8(1.7) x 10^{-10}
Namibia	Röessing Uranium Mine	5.460(41) x 10^{-5}	0.007 256 8(36)	2.0(1.8) x 10^{-10}
France	Cogema Lodere	5.154(28) x 10^{-5}	0.007 257 2(36)	1.9(1.7) x 10^{-10}
France	CETAMA Amethyste	5.340(33) x 10^{-5}	0.007 235 2(36)	2.0(1.4) x 10^{-10}

Table 2

Results of Lead (Pb) isotope ratio measurements of Namibia mine tailing samples [22].

Sample ID	$\frac{^{204}Pb}{^{206}Pb}$	$\frac{^{207}Pb}{^{206}Pb}$	$\frac{^{207}Pb}{^{206}Pb}$
TSF2-1	0.0589	0.9672	2.3113
TSF2-2	0.0612	1.0153	2.4330
TSF2-3	0.0584	0.9658	2.3160
TSF2-4	0.0614	1.0148	2.4278
TSF2-5	0.0610	1.0111	2.4209
TSF2-6	0.0606	0.9993	2.4004
TSF2-7	0.0605	1.0059	2.4007
TSF2-8	0.0605	1.0045	2.4075
TSF2-9	0.0606	1.0065	2.4037
TSF2-10	0.0606	1.0020	2.3969
Mean	0.064 ± 0.00091	0.9992 ± 0.01708	2.3918 ± 0.04078
Measured NIST SRM 981	0.05888 ± 0.0001	0.91441 ± 0.0036	2.17061 ± 0.0013
Certified NIST SRM 981 [33]	0.059042 ± 0.000037	0.91464 ± 0.0033	2.1681 ± 0.0008

Table 3

Results of Lead (Pb) isotope ratio measurements of South African mine tailing samples [22].

Sample ID	$\frac{^{204}Pb}{^{206}Pb}$	$\frac{^{207}Pb}{^{206}Pb}$	$\frac{^{208}Pb}{^{206}Pb}$
C5W-1	0.0482	0.8159	2.0108
C5W-2	0.0484	0.8115	1.9457
C5W-3	0.0498	0.8221	2.0326
C5W-4	0.0507	0.8269	2.0349
C5W-5	0.0488	0.8144	2.0434
C5W-6	0.0488	0.8221	2.0217
C5W-7	0.0496	0.8193	1.9589
C5W-8	0.0495	0.8192	2.0331
C5W-9	0.0510	0.8304	2.0427
C5W-10	0.0487	0.8187	2.0481
Mean	0.04935 ± 0.0017	0.82005 ± 0.0194	2.01719 ± 0.0606
Measured NIST SRM 981	0.0588 ± 0.0001	0.91441 ± 0.0036	2.17061 ± 0.0013
Certified NIST SRM 981 [33]	0.059042 ± 0.000037	0.91464 ± 0.00033	2.1681 ± 0.0008

Despite comprehensive analysis, technical limitations necessitated the use of limited number of characteristics for preliminary assessments of uranium's origin. This is one of the reasons why the majority of previous research had not yet fully explored the complex interplay between different isotopic characteristics [1,12]. While a limited number of characteristics may provide some insights, a more comprehensive analysis is necessary for definitive conclusions regarding uranium provenance [1]. This review aims to scrutinize the power of integrated nuclear forensic characteristics for uranium origin assessment.

3.1.3. Analysis of the rare earth elements concentration, patterns, trace elements and mineralogy in uranium ore

Several trace elements are essential for geological studies, but rare earth element (REE) distribution patterns are the most commonly reported characteristics used for uranium ore origin assessment [14]. The REE patterns in uranium-bearing minerals, such as uranium ore concentrates (UOC), can be linked to specific geological environments [21]. Due to their low solubility and similar chemical properties, REE abundances generally persist through uranium chemical processing. In addition to UOC, uranium soil samples, such as those from the Serule uranium deposits in Botswana, have been analyzed using ICP-MS to reveal distinct REE signatures.

For example, analysis of the chondrite normalized rare earth elements patterns for uranium soil samples from Serule deposits, revealed the trend of the concentration showing that Lanthanum (La) > Cerium

(Ce) > Neodymium (Nd) > Samarium (Sm) > Europeum (Eu) > Gadolinium (Gd). Also, Terbium (Tb) ≈ Dysprosium (Dy) ≈ Holmium (Ho). However, this was not the case as in some samples it was observed that Ce > La. The typical REE pattern for many geological samples, including those from the Serule deposits, shows a characteristic trend: La > Ce > Nd > Sm > Eu > Gd and Tb ≈ Dy ≈ Ho. This pattern reflects the increasing ionic radius of the lanthanide series, which influences their geochemical behavior. Larger ions like La are more easily incorporated into minerals, leading to higher concentrations [5].

In addition, researchers have successfully matched REE patterns in intercepted uranium samples to their known origins [4,23,24]. REE patterns are crucial for identifying uranium ore types due to their consistency through most hydrometallurgical processes. For example, unconformity-related deposits often exhibit bell-shaped REE patterns centered on Tb or Dy [21,25]. The LREE/HREE ratio of uranium can also be used to differentiate between deposits from various origins. For example, uranium from the Rössing deposit in Namibia has a LREE/HREE ratio of 0.41, while uranium from the Pine Creek deposit in Australia has a ratio of 0.19.

While REE patterns have been used to infer uranium origin, their limitations without considering other factors have been recognized [21,22,26]. Nuclear forensics has advanced, but a single definitive parameter for uranium identification remains elusive. Combined isotopic characteristics and multivariate statistics are increasingly employed to address this challenge [21,27,28]. This review aims to explore the potential of a comprehensive model for determining uranium provenance by examining and integrating various characteristics.

3.1.4. *Analysis of the ratios of stable and unstable (long-lived) isotopes of neodymium (^{143}Nd/^{144}Nd) in uranium ore*

The ^{143}Nd/^{144}Nd isotope ratio has been widely used to assess uranium origin in locations such as Arlit, Niger; Ranger, Australia; Neclebras, Brazil; Faraday, Canada; and Phalabora, South Africa; (Table 4). Neodymium, present in trace amounts in uranium-based nuclear materials, is unaffected by radioactive decay, making it a reliable reference isotope. The ^{143}Nd/^{144}Nd ratio exhibits significant variation between uranium minerals, offering a valuable tool for distinguishing uranium from different sources. Furthermore, the ^{143}Nd/^{144}Nd ratio demonstrates greater consistency within uranium mineral samples and concentrates compared to other isotopes like lead (Pb) and strontium (Sr). This consistency highlights its importance as a key characteristic for uranium origin determination [1,9].

The stability of ^{144}Nd against radioactive decay makes neodymium isotope composition a potential indicator of uranium deposit type and age. Beyond its established use in geological chronometry and provenance studies, the ^{143}Nd/^{144}Nd isotope ratio has emerged as a valuable nuclear characteristic, reflecting the age and mineral composition of uranium deposits [14,29,30].

While studies indicate that no single isotopic parameter can definitively determine the origin of uranium [10,27], integrated isotopic approaches combined with multivariate statistical analysis can constrain provenance and processing history. This review aims to investigate the potential of neodymium isotope ratios, among the key parameters in uranium origin determination.

3.1.5. *Analysis of the ratios of stable and unstable isotopes of strontium (^{87}Sr/^{86}Sr) in uranium ore*

Strontium (Sr) isotope ratios are among the crucial tools for determining the provenance of uranium ore [3,61]. The study by Elboni et al., investigated the use of elemental and isotopic signatures, focusing on Sr isotopes, in uraninite samples from diverse U.S. deposits. Their research highlighted the significance of Sr isotope ratios, measured in bulk uraninite, as a key fingerprint for provenance determination, revealing distinct signatures correlated with the ore's formation environment. Specifically, they found that low-temperature uranium ores exhibit high radiogenic ^{87}Sr/^{86}Sr ratios (>0.7200), alongside other characteristic elemental signatures. This underscores the importance of characterizing raw uranium ore, including Sr isotope ratios, for accurate attribution in nuclear forensic investigations. Furthermore, the study showed that even after processing, the Sr isotopic signature remains consistent, allowing for the linking of processed uranium ore concentrate (UOC) back to its original geological source [61].

Complementing this, a new, rapid (4–5 h) column chemistry method using nano-NaBiO3 was developed to efficiently isolate U, Sr, Pb, Nd, and Hf, including crucial access to Sr isotope ratios, from uranium ore samples for isotopic analysis [62]. This method's ability to differentiate uranium ores from different regions based on their isotopic ratios, particularly ^{87}Sr/^{86}Sr, reinforces its potential for nuclear forensic science. Because Sr isotopes are incorporated into uranium-bearing minerals during formation and reflect the geological environment, their ratios serve as a powerful fingerprint linking uranium ore to its geographic origin. This rapid and efficient isolation of Sr, combined with precise isotopic analysis, enables comparison of unknown samples to known deposits, greatly facilitating the tracing of illicit nuclear materials and verification of declared origins, thus significantly enhancing nuclear safeguards and forensic capabilities [3,62].

3.1.6. *Analysis of provenance determination of uranium as a critical element in contemporary criminal investigations*

Determining the precise origin of uranium is paramount in contemporary criminal investigations and subsequent legal proceedings [13,32]. The specter of nuclear proliferation and terrorism has elevated uranium provenance to a critical security concern [21]. Given its central role in both nuclear weapons and power generation, uranium is a highly

Table 4

Nd and Sm concentrations and measured ^{143}Nd/^{144}Nd isotope ratios in the investigated uranium ore and ore concentrate samples. Uncertainties (U), given in parentheses with a coverage factor of 2, apply to the last significant digits of the value [9].

Mine/Mill	Country	Nd (ppm)	Sm (ppm)	Sm/Nd	Nd_{Total} (μg)	^{143}Nd/^{144}Nd(U)	ε
Arlit	Niger	2.89	0.8	0.28	0.03	0.51190(21)	−14.4
Rum Jungle	Australia	2.94	1.50	0.51	0.03	0.51106(06)	−30.8
Ranger	Australia	13.4	26.2	1.96	0.16	0.51442(41)	34.8
Olympic Dam 1	Australia	2.34	66.8	0.28	2.74	0.51096(05)	–32.8
Olympic Dam 2	Australia	201	61.1	0.30	2.35	0.51098(04)	–32.3
USA ESI	USA	1.53	0.47	0.31	0.02	0.51291(68)	5.31
Rayrock	Canada	26.4	6.77	0.26	0.31	0.511320(61)	−25.7
Madawasca	Canada	24.8	5.70	0.23	0.29	0.51243(15)	−4.06
Faraday	Canada	21.8	3.63	0.17	0.26	0.51216(13)	−9.32
Phalabora	South Africa	2.13	0.47	0.22	0.02	0.51178(26)	−16.7
Neclebras	Brazil	39.75	13.25	0.33	0.47	0.51255(15)	−1.72
Denison	Canada	115	72.8	0.63	1.34	0.512978(83)	6.63
Stanrock	Canada	9.39	5.19	0.55	0.11	0.51215(10)	−9.43
Stanleigh	Canada	13,1	6.13	0.47	0.15	0.51253(12)	−2.11
Rio Algom	Canada	54,4	25.8	0.48	0.64	0.511951(36)	−13.4

coveted commodity in illicit markets. Tracing uranium to its source is essential to disrupt smuggling networks and prevent nuclear terrorism. Nuclear forensics is instrumental in linking illicit uranium to specific countries, facilities, or individuals involved in its production or theft [26].

Beyond nuclear threats, uranium is implicated in a range of criminal activities [4]. Illegal uranium mining and smuggling often contribute to environmental degradation and human rights abuses. Identifying the origin of uranium in these cases facilitates the prosecution of perpetrators. Moreover, improper disposal of uranium waste poses severe environmental risks, and tracing its origin helps hold responsible parties accountable. In addition, uranium's unique properties make it susceptible to exploitation in various crimes. Its high density facilitates its use in counterfeiting, while its value has attracted the attention of money launderers. Determining uranium origin is essential to combat these criminal activities.

Developing comprehensive databases of uranium signatures is crucial for accurate origin determination [6,11]. While complex, establishing uranium provenance is increasingly vital for safeguarding national security, protecting the environment, and upholding the rule of law [14]. As technology advances and global threats evolve, the capacity to trace uranium will become even more critical.

3.2. *Current state of knowledge*

3.2.1. *The challenge of uranium provenance determination*

Despite the critical importance of determining uranium provenance in criminal investigations, significant challenges persist. Among other challenges include complexity of uranium signatures [24]. Uranium isotopic and elemental signatures can be influenced by a multitude of factors, including geological formation, weathering, enrichment processes, and human activities. This complexity can make it difficult to definitively link a sample to a specific origin [28]. Another significant drawback in the progress of uranium provenance determination is a database limitation. Although there are growing efforts to establish comprehensive uranium signature databases, many regions still lack sufficient data, hindering comparison and analysis [23].

Similarly, analytical limitations pose a significant challenge to the advancement of uranium provenance determination. While analytical techniques have significantly improved, challenges persist in achieving the necessary precision and accuracy for reliable provenance determination, particularly for small or contaminated samples. In addition, the absence of a robust legal and regulatory framework hinders the progress of uranium provenance determination. The use of isotopic and elemental data in criminal investigations often requires a strong legal and regulatory framework to ensure the admissibility of evidence and protect sensitive information.

3.2.2. *Current state of the Art*

Despite the challenges, significant progress has been made in recent years in the development of uranium provenance determination [29]. Advances in isotopic and elemental analysis have significantly enhanced ability to determine the provenance of uranium ore. High-precision mass spectrometry allows for accurate measurement of uranium isotope ratios, such as $^{235}U/^{238}U$, which can be used to distinguish between different uranium deposits. In addition, techniques like X-ray fluorescence spectroscopy (XRF) and inductively coupled plasma mass spectrometry (ICP-MS) enable detailed analysis of the elemental composition of uranium ores, including elements like thorium, lead, and rare earth elements. By comparing the elemental patterns of different samples, researchers can differentiate between uranium ores from various geological sources. To facilitate these analyses, several countries and international organizations have established uranium signature databases. These databases serve as valuable reference points for comparing samples and identifying potential origins.

3.2.3. *Nuclear forensic case studies for uranium provenance*

The growing number of case studies underscores the practical application of uranium provenance determination in real-world contexts [10,34]. This includes among others, criminal investigations and environmental assessments, highlighting the crucial role of nuclear forensics in investigating illicit uranium trafficking and nuclear proliferation. While these techniques show significant promise, further development and standardization are essential to guarantee their widespread use and reliability. The following case studies illustrate the importance of this subject and the effectiveness of nuclear forensics in tracing uranium provenance.

In 2003, the interception of suspected yellowcake in Italy triggered an investigation that revealed illicit activities. Isotopic and elemental analyses, using techniques like ICP-MS, XRF, and XRD, linked the material to uranium deposits in Niger [35]. This connection was established by comparing the seized material's isotopic composition and elemental patterns to those of known uranium deposits. Another case in early 2009 involved the seizure of a radioactive green powder labeled "Gamma Source" in Australia. Nuclear forensic analysis, focusing on isotopic composition, impurity content, and other characteristics, traced the powder back to the Mary Kathleen uranium mine [4].

In 2010, a significant quantity of uranium was intercepted in South Africa. Isotopic and elemental analysis again proved crucial, pinpointing the uranium's origin to a specific region within the country. This successful tracing contributed to non-proliferation efforts [11]. The 2011 Fukushima nuclear disaster further highlighted the importance of nuclear forensics. Isotopic and elemental analysis of released uranium enabled scientists to determine its origin, track its dispersal, and assess risks to human health and the environment, informing cleanup and risk assessment strategies [36].

In 2024, Australian authorities intercepted an unknown sample of yellowcake. Nuclear forensic analysis, including isotopic measurements of uranium and lead, and trace element analysis, was employed to determine its origin. Comparison with a database of uranium ore samples from known locations successfully identified the source. This particular case study [37] demonstrated the power of combining multiple analytical techniques for effective uranium provenance determination.

These case studies demonstrate the crucial role of isotopic and elemental patterns in determining uranium provenance. This capability is vital not only for preventing illicit trafficking, proliferation, and misuse, but also for safeguarding the environment. By tracking uranium movement, investigators can identify potential sources of contamination and mitigate the environmental impact of uranium-related activities.

3.3. *Knowledge gaps and future research directions*

Despite substantial advancements in uranium provenance determination, several knowledge gaps persist. As notable areas of uncertainty may include several, some few cases can be pointed out. For example to ensure the reliability and comparability of uranium provenance studies, it is essential to establish standardized analytical methods and data reporting protocols. The standardized analytical methods are essential for ensuring consistency and comparability in uranium provenance studies. These protocols dictate how the analysis is performed, encompassing sample preparation, instrumentation, and data reduction.

For sample preparation, protocols may specify a particular grain size for analysis, such as isolating the 63–125 μm fraction for U-Pb geochronology, to minimize variability and ensure comparable data [38]. Standardized acid digestion procedures are crucial for isotopic analyses, guaranteeing complete sample dissolution without contamination. For instance, samples might be dissolved using a mixture of HF, HNO_3, and HCl following a defined temperature and time schedule. When targeting single mineral grains, protocols detail separation methods like density or magnetic separation, coupled with purity criteria, such as selecting zircon grains exhibiting greater than 95 %

purity.

In addition, standardization extends to instrumentation and calibration. Protocols may specify the type of mass spectrometer, like MC-ICP-MS, to ensure data compatibility [59]. The use of certified reference materials (CRMs) is paramount for accurate measurements; protocols specify which CRMs to use and their frequency of use, for example, bracketing every five unknown samples with CRM IRMM-046 for U isotopic ratio calibration. Analytical conditions, including laser spot size for LA-ICP-MS and ion beam intensities, also require standardization, such as performing U-Pb dating with a 30 μm diameter laser spot at a 5 Hz repetition rate. [39]. Data reduction protocols specify how to correct for background signals through blank measurements and how to correct isotopic ratios for mass bias using standardized procedures, such as an exponential law based on a known standard. For geochronology, protocols detail the decay constants used and the method for calculating ages and uncertainties, often referencing recommendations from the International Union of Geological Sciences. [40].

On the other hand, standardized data reporting protocols are equally critical, dictating what data are reported and their format to ensure transparency and reproducibility [41]. Essential parameters include sample information (location, geological context, sample type, and preparation), analytical details (instrument type, calibration standards, and conditions), raw data (measured isotopic ratios, concentrations, and uncertainties), and calculated values (ages, corrected isotopic ratios, and uncertainties). Data tables should follow standardized formats for easy comparison, such as reporting U-Pb geochronology data with sample ID, grain number, U and Pb isotopic ratios, calculated ages, and associated uncertainties [42]. Consistent units, significant figures, and clear uncertainty reporting (e.g., 2σ standard deviations) are crucial. Metadata, including analytical methods, data processing, and potential issues, should be included.

Data repositories like EarthChem facilitate data sharing, and protocols may require data deposition. Standardized plots, like Tera-Wasserburg concordia diagrams, should be used, and clear citation of standardized methods is essential [44]. A concise reporting guideline might state that U-Pb geochronology will be performed on zircon separates using LA-ICP-MS at a specific laboratory, following a referenced protocol, calibrating with the GJ-1 zircon standard, processing with Iolite software, calculating ages with recommended decay constants, reporting data in a standardized table, and uploading all data to EarthChem. Adhering to these standardized methods and reporting protocols allows the scientific community to build a robust database of uranium provenance information, leading to more accurate and meaningful interpretations [60].

Similarly, expanding uranium signature databases to encompass a wider range of geological environments is crucial for more accurate provenance determinations. A wider range of geological environments in this regard, refers to a diverse array of geological settings. For example within igneous environments, uranium occurs in various rock types such as granites, pegmatites [45,46] and volcanic formations [47] each possessing unique characteristics that influence the isotopic fingerprint of the uranium they contain [48,49].

Likewise, sedimentary environments host uranium in rocks [50], like sandstones [51], shales [52], and limestones [53], where the conditions of their formation, including the presence of organic matter or groundwater flow, play a crucial role in shaping the uranium's isotopic makeup. Even metamorphic environments, where existing rocks are transformed under intense heat and pressure, can contain uranium, with the metamorphic processes themselves capable of altering its isotopic signature [54,55].

The importance of considering this wide range of environments stems from the fact that different geological processes inevitably lead to variations in uranium's isotopic composition. By examining uranium from this broad spectrum of environments, scientists can build a more complete database of these isotopic signatures. This comprehensive database is fundamental to accurate uranium provenance studies, which aim to trace the origin of uranium samples. Consider, for example, unconformity-related uranium deposits, frequently found in regions like Australia and Canada [56].

These deposits, associated with significant geological boundaries, exhibit distinct isotopic signatures. Contrast this with sandstone-hosted uranium deposits, common in the western United States, which form within specific sedimentary environments and consequently develop their own unique isotopic characteristics. Through the analysis of uranium isotopes across these diverse settings, scientists enhance their ability to pinpoint the source of uranium samples, a capability with profound implications for fields ranging from nuclear forensics and environmental studies to resource exploration. This being said, developing robust statistical models that link isotopic and elemental data to specific geographic origins will further enhance the precision of these techniques [57,58].

In addition, integrating uranium provenance data with other forensic evidence can strengthen the overall investigative process. Addressing legal and ethical challenges related to the collection, analysis, and use of uranium data is vital to ensure the responsible application of these techniques. The future of uranium provenance determination lies in addressing knowledge gaps, developing innovative approaches, and overcoming challenges such as sample contamination and data interpretation. By expanding isotopic databases and enhancing analytical techniques, researchers can create more powerful forensic tools to combat uranium-related crime. International cooperation and information sharing among the stakeholders are also essential for addressing the challenges posed by illicit uranium trafficking.

4. Discussions

The isotopic and elemental composition of uranium ore provides a powerful tool for tracing its origin, an essential step in criminal investigations concerning nuclear proliferation, terrorism, and environmental offenses [2–4,12]. While isotopic analysis has been established as a reliable method, the full potential of elemental patterns remains untapped. As evidenced by previous studies [15,19], isotopic analysis is a valuable tool for determining the geographic origin of uranium. However, elemental patterns offer complementary information that can enhance the accuracy of provenance determinations. By integrating these techniques and expanding geochemical databases, researchers can develop more robust models for uranium origin analysis.

This review article aligns with prior research in recognizing the potential of geochemical fingerprinting for uranium provenance determination [1,10]. Both, the review and previous studies [15,21] demonstrate the utility of unique geochemical characteristics as tracers of uranium origin. However, a key distinction lies in the methodological emphasis. While past studies have often relied on a limited set of isotopic or elemental signatures [2,12], this work underscores the efficacy of an integrated approach. By combining isotopic and elemental analysis, it is possible to create a more comprehensive geochemical profile, enhancing the discriminatory power of provenance assessments. This disparity in methodology likely stems from limitations in existing databases and analytical techniques.

As highlighted in previous studies [11,27], current nuclear forensic libraries often lack the comprehensive data necessary for definitive provenance determinations. This limitation has led to conclusions on origin assessment of uranium based on a single type of signature or a limited number of geochemical characteristics. In addition, analytical capabilities may have restricted the range of obtainable signatures. This review advocates for a multi-faceted approach that leverages the strengths of both isotopic and elemental analysis to address these limitations. By integrating these techniques, it is possible to create a more robust foundation for uranium provenance determination, a crucial aspect of investigations into nuclear proliferation, terrorism, and environmental crimes [1,25].

This comparative analysis underscores the growing consensus on the value of geochemical fingerprinting for uranium provenance determination. While this review emphasizes the benefits of an integrated approach, it acknowledges the need to address current limitations in databases and analytical techniques. By overcoming these challenges and fostering collaboration between researchers and law enforcement, the forensic community can significantly contribute to national security and environmental protection [11,12,26]. This collaborative effort will unlock the full potential of isotopic and elemental analysis in combating uranium-related crime.

5. Conclusions

Accurate uranium provenance determination, crucial for investigating nuclear proliferation, terrorism, and environmental crimes, can be achieved through isotopic and elemental analysis of uranium ore. While isotopic analysis is a well-established method, the full potential of elemental patterns remains untapped. To enhance the discriminatory power of provenance analysis, integrating both techniques and expanding geochemical databases is essential [1,10]. However, challenges such as sample contamination and sophisticated criminal methods hinder progress. By addressing these limitations, developing standardized methodologies, and fostering interdisciplinary cooperation, researchers and law enforcement agencies can strengthen the ability to trace illicit uranium and dismantle criminal networks [11]. Ultimately, continued research and development are necessary to fully unlock the potential of isotopic and elemental analysis for uranium provenance determination, contributing to national security and environmental protection.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work, the author(s) utilized AI tools to assist with grammatical error correction and enhance the logical flow of information. The author(s) subsequently reviewed and edited the content as necessary, assuming full responsibility for the accuracy and originality of the publication.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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