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Abstract

236U is mainly derived from anthropogenic nuclear activities such as atmospheric nuclear tests, nuclear accidents, and nuclear fuel reprocessing. Owing to its extremely low natural background and long half-life, it has become an emerging environmental tracer of considerable interest. Accelerator mass spectrometry (AMS) is the most sensitive technique for measuring environmental ultratrace 236U. The theoretical abundance sensitivity of current mainstream AMS systems for the 236U/238U atomic ratio can reach the 10-14 level or lower, providing a reliable means for measuring 236U in media such as soils, ice cores, seawater, and atmospheric aerosols. This paper systematically reviews the principles of AMS measurement of 236U, interference suppression, and advances in instrumentation; summarizes pretreatment procedures including sample digestion, uranium separation and purification, and target preparation; and outlines the current status of tracer applications of 236U in soils, the cryosphere, the ocean, and the atmosphere. In addition, based on our team' s measured results of 236U in forest soils from Changbai Mountain, this paper reports the mechanisms governing the depth-profile distribution of 236U and its correlations with 137Cs and 237Np, and accordingly estimates the total global fallout of 236U from nuclear tests to be approximately 968 ± 290 kg. Finally, existing bottlenecks such as lengthy pretreatment procedures and high costs are analyzed, and future directions are proposed, including compact AMS and rapid uranium isotope detection, automated high-throughput pretreatment, multi-nuclide combined tracing, and expanded tracer applications of natural 236U.

Full Text

Measurement and Applications of Environmental ^{236}U: A Review

Huang Wenjian[1], Liu Zhiyong[2], Wang Huijuan[1], Tu Zhiyi[1], Lai Jianhui[1], Luo Xianghao[1], Wang Hongjie[1], Guan Yongjing[1#]

(1. School of Physical Science and Technology, Guangxi University, Nanning, Guangxi 530004;
2. School of Radiation Medicine and Protection, Soochow University, Suzhou, Jiangsu 215123)

Abstract: ^{236}U mainly originates from anthropogenic nuclear activities such as atmospheric nuclear tests, nuclear accidents, and nuclear-fuel reprocessing. Because of its extremely low natural background and long half-life, it is an emerging environmental tracer that has attracted much attention. Accelerator mass spectrometry (AMS) is the most sensitive technique for measuring ultratrace ^{236}U in the environment. The theoretical abundance sensitivity of current mainstream AMS systems for the ^{236}U/^{238}U atomic ratio can reach the 10^{-14} level or lower, providing a reliable means for measuring ^{236}U in media such as

soil, ice cores, seawater, and atmospheric aerosols. This paper systematically reviews the AMS measurement principles for ^{236}U, interference suppression, and progress in instrumentation research; summarizes pretreatment procedures such as sample digestion, uranium separation and purification, and target-source preparation; and reviews the current status of tracer applications of ^{236}U in media including soil, the cryosphere, the ocean, and the atmosphere. In combination with our team' s measured results for ^{236}U in forest soils from the Changbai Mountains, the depth-profile distribution mechanism of ^{236}U and its correlations with ^{137}Cs and ^{237}Np are reported. On this basis, the total global fallout of ^{236}U from nuclear tests is estimated to be approximately 968 ± 290 kg. Finally, existing bottlenecks such as lengthy pretreatment procedures and high costs are analyzed, and future directions are discussed, including miniaturized AMS and rapid detection of uranium isotopes, automated batch pretreatment, multi-nuclide combined tracing, and expanded tracing with natural ^{236}U.

Keywords: ^{236}U; accelerator mass spectrometry; application

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0 Introduction

Since the first nuclear test in 1945, eight countries worldwide have conducted more than 2,000 nuclear tests in total, releasing large amounts of anthropogenic radioactive nuclides into the environment. Among the many nuclides, ^{137}Cs, $^{239+240}$Pu, and others have been widely used as environmental tracers[1–3], while ^{236}U, owing to its extremely low natural background and high sensitivity to nuclear activities, has in recent years gradually become a new tracer of great interest[4–8]. Environmental ^{236}U mainly originates from atmospheric nuclear tests, nuclear accidents, and nuclear-fuel reprocessing activities. Because ^{236}U has a very long half-life (2.34×10^7 years), once released it can persist in the environment for a long time and continue to migrate, accumulating in soils and sediments through processes such as atmospheric deposition and aquatic transport.

There are significant differences in the ^{236}U/^{238}U atomic ratios of ^{236}U from different sources. As shown in Table 1, ^{236}U in the natural environment is mainly generated from ^{235}U through the neutron-capture reaction (n, γ); the reaction cross section is 111×10^{-26} m^2. Because the neutron flux at the Earth' s surface is extremely low, the production of natural ^{236}U is very small. Even in uranium ore samples containing relatively more ^{236}U, the ^{236}U/^{238}U atomic ratio

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Author biography: Huang Wenjian (2000–), male (Han), from Yulin, Guangxi, master' s student, engaged in nuclear technology research; E-mail:

hwj-x@foxmail.com
Corresponding author: Guan Yongjing, E-mail: yiguan@gxu.edu.cn

usually lower than 5.16×10^{-10} and higher than the average natural level (10^{-14}–10^{-13})[9–12]. The $^{236}U/^{238}U$ atomic ratio in deep rocks is essentially about 3×10^{-14}. Although near-surface rocks are affected by cosmic-ray neutron flux, this ratio can rise to 7×10^{-13}, but it still remains at an extremely low level[13]; the $^{236}U/^{238}U$ atomic ratio from global fallout is about 10^{-9}–10^{-8}[14]. By comparison, in irradiated nuclear fuel or nuclear waste this ratio can be as high as 10^{-4}–10^{-2}, 6–8 orders of magnitude higher than the natural background[15]. Therefore, an anomalous increase in the environmental $^{236}U/^{238}U$ atomic ratio can serve as an effective indicator of nuclear leakage or nuclear discharge.

Table 1. Typical ranges of the $^{236}U/^{238}U$ atomic ratio in different source categories

Source category	Typical range of $^{236}U/^{238}U$ atomic ratio
Deep rocks (no cosmic rays)	3×10^{-14}
Near-surface rocks	7×10^{-13}
Uranium ore	$\leq 5.16\times10^{-10}$
Global fallout	$\sim 10^{-9}$–10^{-8}
Irradiated nuclear fuel or nuclear waste	$\sim 10^{-4}$–10^{-2}

AMS is an ultra-high-sensitivity mass-spectrometric analytical technique developed in the late 1970s. Its core principle is to ionize the atoms to be measured in a sample, accelerate the ions with an accelerator to the mega-electron-volt (MeV) energy range, use a stripper to dissociate molecular-ion interferences, and then directly count specific isotopes with magnetic analyzers, electrostatic analyzers, and particle detectors. Compared with conventional mass spectrometry, AMS can effectively eliminate molecular background and interference from isobars, and its theoretical abundance sensitivity can reach the 10^{-15}–10^{-16} level, 6–8 orders of magnitude higher than conventional mass spectrometry. It is currently the most effective method for measuring ultratrace ^{236}U in environmental samples. In addition to AMS, ICP-MS/MS (inductively coupled plasma tandem quadrupole mass spectrometry) can also effectively measure ^{236}U; by eliminating molecular-ion interferences through reaction gas, its detection limit can satisfy the analysis of soil and seawater samples at global-fallout levels. TIMS (thermal ionization mass spectrometry) provides extremely high isotope-ratio precision in samples with relatively high uranium contents and is commonly used in nuclear forensics and dating studies. MC-ICP-MS (multi-collector inductively coupled plasma mass spectrometry) combines high throughput with the ability to analyze multiple isotopes simultaneously, making it suitable for nuclear emergency response and combined source-tracing scenarios. These methods each have their own strengths and complement AMS.

With technological progress, ^{236}U has shown tracer value in a variety of environmental media. This paper systematically reviews the principles and latest domestic and international progress in accelerator mass-spectrometric measurement of ^{236}U; meanwhile, from the perspectives of multiple environmental media such as soil, permafrost, the ocean, and the atmosphere, it summarizes the current application status of ^{236}U as a novel environmental tracer, in the hope of providing a reference for research in related fields.

1 Principles and Technical Progress of Accelerator Mass-Spectrometric Measurement of ^{236}U

1.1 Basic Principles of AMS Measurement of ^{236}U

AMS measurement of ^{236}U usually uses a cesium-sputter negative ion source: the sample is mixed with a conductive matrix (such as Nb powder) and pressed into a target, and negative ions of UO^- (or UF_5^-) are produced by bombardment with a Cs^+ ion beam. Some of the cesium ions, after losing energy, are adsorbed on the surface of the target cone, forming a cesium-covered layer, thereby reducing the work function of the target element and obtaining a larger negative-ion current. After preliminary mass selection by injection into a magnetic analyzer, the negative ions enter a tandem accelerator, where charge exchange occurs in the stripping gas at the terminal of the accelerator,

The negative ions are converted into positive ions and further accelerated. For uranium-series elements, helium is currently the optimal stripping gas:

He has a relatively high ionization potential, which facilitates stripping uranium ions to a higher 3+ charge state; at the same time, its small atomic number can significantly reduce ion-scattering losses. After stripping and secondary acceleration, high-energy U^{3+} ions first pass through a high-energy magnetic analyzer, whose main function is to eliminate high-abundance isobaric ions of the nuclide being measured. These ions originate from molecular "fragments" produced during the stripping process, as well as from continuous momentum spectra formed by ion scattering and charge exchange. Subsequently, the accelerated ions pass again through a magnetic analyzer, electrostatic deflector, or velocity selector; by selecting the mass-to-charge ratio of the particles to be analyzed, part of the molecular background and scattering background is further removed. Finally, ion identification and counting are performed by a gas ionization detector or a time-of-flight detector.

1.2 Main interferences in ^{236}U AMS measurements and suppression strategies

The core challenge in ^{236}U AMS measurement is that the ^{238}U content in environmental samples is usually 10^9–10^{11} times higher than that of ^{236}U, and even trace interfering ions can overwhelm the ^{236}U signal. Therefore, effective

recognition and suppression of background interference are the key factors determining measurement sensitivity.

Guan et al. systematically analyzed three major sources of interference and their suppression methods on the CIRCE 3 MV AMS system[1]. For molecular-ion interference, such as $^{238}U^{14}N^{-}$ and $^{236}U^{16}O^{-}$, which have the same mass, nitrogen can be eliminated by preheating the target (800 °C) in the ion source, and residual molecular ions can be completely dissociated during stripping in the tandem accelerator. For ^{238}U tailing interference, the intensity of ^{238}U is 9–11 orders of magnitude higher than that of ^{236}U, and the low-energy tail formed by scattering and energy loss can fall into the ^{236}U mass window; this interference is mainly suppressed by combining a high-resolution injection system with high-energy-end magnetic and electrostatic analyzers. For charge-exchange interference, during acceleration and analysis, ^{238}U ions may undergo charge exchange with residual gas (e.g., $^{238}U^{4+} \rightarrow {}^{238}U^{5+}$) and then enter the $^{236}U^{5+}$ channel; a high-resolution electrostatic analyzer can spatially separate them from $^{236}U^{5+}$.

1.3 Research progress in accelerator mass spectrometric measurement technology for ^{236}U

Internationally, the 3 MV AMS system at the VERA laboratory of the University of Vienna, Austria, is one of the representative large-scale facilities for ^{236}U measurement. Winkler et al. systematically investigated the charge-state distribution of uranium and thorium under He stripping and found that, within a terminal-voltage range of 1.0–1.7 MV, the transmission efficiency of the U^{3+} charge state (after the analyzing magnet) can reach 20%–25%, far higher than that of the conventional 5+ charge-state configuration. This study shows that, on a 3 MV accelerator, using a relatively low terminal voltage together with He stripping to the 3+ charge state can significantly improve measurement efficiency while maintaining relatively high ion energy[2]. In China, the GANA 3 MV AMS system built at Guangxi Normal University has measured the $^{236}U/^{238}U$ atomic ratios in coastal seawater, freshwater from the Guilin region, and soil samples in China, with good reproducibility. This system can efficiently enrich and accurately quantify ^{236}U in only 1 L of seawater or freshwater, providing important technical support for nuclear-environment monitoring and studies of ^{236}U geochemical behavior[3]. In recent years, the development of compact AMS systems has provided a new technical route for ^{236}U measurement. The compact multi-nuclide AMS system independently developed by the China Institute of Atomic Energy (CIAE) uses a low terminal voltage of about 230 kV and He stripping to the 3+ charge state, achieving a transmission efficiency of 35.2% and a $^{236}U/^{238}U$ atomic-ratio abundance sensitivity of about 10^{-15}. This system employs an independently developed Bragg-type gas ionization detector together with a 30 nm thick Si_3N_4 entrance window, enabling effective heavy-ion

[1] 16
[2] 17
[3] 18

detection at a low ion energy of about 0.9 MeV[4].

This study indicates that the low-energy compact AMS system combines the advantages of high sensitivity, operational safety, and low cost in the measurement of ^{236}U, and can achieve rapid, low-background analysis of trace uranium isotopes such as ^{236}U and ^{233}U with minimal sample consumption, providing important technical support for nuclear emergency response, nuclear safeguards, and environmental monitoring.

2 Environmental Sample Pretreatment and AMS Target Preparation

2.1 Sample Digestion and Extraction of Uranium

For solid samples such as soils and sediments, uranium must first be quantitatively converted from the solid matrix into solution before further concentration, separation, and purification can be performed. The general procedure is as follows: after the sample is dried, ground, and homogenized, high-temperature ashing is carried out as needed to remove organic matter; subsequently, digestion is performed using a mixed acid system of HNO_3–HF (with $HClO_4$ added when necessary), or by an alkali fusion method (such as borate fusion) to break down the silicate crystal lattice and bring all uranium into solution. Whether digestion is complete directly affects the representativeness of ^{236}U measurement. For refractory samples, fusion is recommended to ensure quantitative recovery. For samples measured by the known isotope dilution method, a known amount of ^{233}U may be added before digestion to correct the recovery of the entire process.

Water samples (seawater, river water, melted ice-core water, etc.) have low uranium concentrations and large volumes, so pretreatment focuses on enrichment: seawater samples are usually first acidified to pH 2, then a Fe^{3+} carrier is added and ammonia is used to adjust the pH to alkaline conditions. Uranium is quantitatively enriched from several liters to several tens of liters of seawater through $Fe(OH)_3$ coprecipitation; ice-core samples can be treated by a similar process after controlled melting under clean conditions. The precipitate obtained by coprecipitation is collected by centrifugation and dissolved, and then purified and separated using extraction-resin chromatographic columns such as DGA and UTEVA to remove interfering substances including thorium, plutonium, and matrix elements. Finally, measurement is carried out using mass spectrometers such as ICP-MS/MS, MC-ICP-MS, or AMS.

2.2 AMS Target Preparation

Among the many mass spectrometric techniques, for AMS measurement of ^{236}U, the $Fe(OH)_3$ coprecipitation method is currently the mainstream method for sample pretreatment and preparation. This method has strong adaptability and

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high stability, and is especially suitable for pretreatment scenarios involving various environmental samples; it is the most widely applied technical approach at the present stage. In this study, sample preparation was carried out based on the classic ferric hydroxide coprecipitation method. The simple operating procedure is as follows: concentrated ammonia is added dropwise to a solution containing uranium and Fe^{3+}, and the system pH is adjusted to 8–9 so that Fe^{3+} is fully precipitated as $Fe(OH)_3$; the coprecipitated product is collected by centrifugation; the resulting precipitate is dried at 80 °C and then transferred to a muffle furnace, where it is calcined at a high temperature of 850 °C so that $Fe(OH)_3$ is fully converted into Fe_2O_3, ultimately yielding a reddish-brown solid. This solid is thoroughly and uniformly mixed with high-purity Nb powder or PbF_2 powder at a certain mass ratio. High-purity Nb powder has good electrical and thermal conductivity, improving the stability of the UO^- beam; PbF_2 promotes efficient extraction of uranium in the form of UF_5^-, after which it is pressed into an AMS-specific target holder. In subsequent AMS measurements, the ion source extracts negative ions in the form of UO^- or UF_5^-; under these conditions, a stable $^{238}U^{3+}$ beam current of approximately 10 nA can be obtained in front of the detector.

In the preliminary stage, this research group used the $Al(OH)_3$ co-precipitation method to prepare samples; that is, after preparing samples by $Al(OH)_3$ co-precipitation, oxidation treatment was completed at 850 °C, and aluminum powder was then mixed in to prepare the target material. Comparison of the experimental performance of the two preparation methods showed that the core advantage of the $Al(OH)_3$ co-precipitation method is its relatively high recovery rate during the co-precipitation-product collection stage, but it has obvious shortcomings. After aluminum powder is added, the conductivity of the target material cannot meet experimental expectations, resulting in a low ion-source extracted beam current. Actual measurement results showed that the $^{238}U^{3+}$ beam current at the tuning stage with this method was only 5 nA, while in actual environmental-sample testing the typical beam current was as low as 50 pA. Although this beam intensity can meet the requirements of basic measurements, an extremely low beam current greatly increases the sample measurement time, and the experimental efficiency is low. By contrast, with the $Fe(OH)_3$ co-precipitation method, relying on the UO^- or UF_5^- ion-extraction mode, for environmental samples the typical $^{238}U^{3+}$ beam current in front of the detector can reach 1 nA. The beam intensity is significantly improved, which can effectively shorten AMS measurement time and greatly improve the measurement efficiency and experimental stability of ^{236}U samples.

3 Application

3.1 Identification by the $^{236}U/^{238}U$ Atom Ratio and Multi-Nuclide Combined Analysis

The order-of-magnitude scale of the $^{236}U/^{238}U$ atom ratio provides multiple discriminating dimensions for nuclear safeguards verification. In sample analysis, by measuring the $^{236}U/^{238}U$ atom ratio in unknown samples, it is possible to make a preliminary judgment as to whether there are sources other than global fallout; an anomalous increase in the ratio can serve as a sensitive indicator of releases from nuclear facilities or leakage from nuclear accidents. Existing studies mainly involve the establishment of background-value databases for contaminated areas, impact assessment of nuclear accidents, and environmental tracing of nuclear tests. For example, Sakaguchi et al. measured a ^{236}U concentration of approximately 3×10^9 atoms/g in soil affected only by global fallout in Ishikawa Prefecture, Japan, with $^{236}U/^{238}U$ atom ratios between 10^{-8} and 10^{-7}, reflecting the characteristics of global fallout[20]. Regarding the Chernobyl accident, Boulyga and Becker measured $^{236}U/^{238}U$ atom ratios as high as 1×10^{-3} in soil near the nuclear power plant, indicating a strong local release signal[21]. After the Fukushima accident, Sakaguchi et al. measured ratios of 10^{-7}–10^{-6} in contaminated materials[22], whereas Yang et al.' s analytical results for Fukushima soil fell within the range of global fallout, indicating that the Fukushima release was limited[23]. In addition, Quinto and De Cesare, in investigations of the environment around decommissioned nuclear power plants in Italy, also did not find large releases of ^{236}U[24,25], indicating that nuclear power plants under normal operating conditions are not a major source of ^{236}U.

It is worth noting that, in some scenarios, a single $^{236}U/^{238}U$ atom ratio makes it difficult to distinguish contributions from different sources; introducing multi-nuclide combined discrimination can effectively improve the identification capability of nuclear forensics. This study analyzed the correlations between ^{236}U in the surface forest soils of Changbai Mountain and the activities/concentrations of other key nuclides. The results showed that ^{236}U was strongly positively correlated with both ^{137}Cs and ^{237}Np (Pearson correlation coefficients r were 0.78 and 0.85, respectively, $p < 0.001$), indicating that their distributions in soil have a high degree of common origin. Since existing studies have confirmed that the main sources of ^{137}Cs and ^{237}Np in this region are global atmospheric nuclear-test fallout, the high common origin of ^{236}U with the two supports the conclusion that ^{236}U also mainly originates from global fallout, and indicates that they have similar migration and retention characteristics in environmental media. Multi-nuclide correlation analysis also demonstrates important value in identifying nuclear events. Yang et al. found a significant linear correlation between ^{236}U and Pu isotopes in contaminated samples from Fukushima, confirming that these nuclides shared a common source.

in the Fukushima Daiichi Nuclear Power Plant accident[^26]. In contrast, Child and Hotchkis found in soils at a former nuclear-weapons test site in

Australia that the atomic ratio of $^{236}U/^{239}Pu$ was significantly lower than the level of global radioactive fallout, indicating the distinctiveness of local release sources[^27]. The above cases show that joint multi-nuclide discrimination can effectively distinguish contributions from different sources and is a broadly applicable analytical approach in the field of nuclear forensics. Specifically, this method does not rely on a single ratio threshold; rather, it identifies the characteristics of local source terms through the degree and direction of deviation of characteristic ratios relative to global background values, as well as correlations among nuclides.

3.2 Behavior and Tracing of ^{236}U in Soil Profiles

Soil is an important receptor of global ^{236}U deposition. Soil-core (profile) analysis is an important means of assessing the vertical migration behavior of ^{236}U in terrestrial environments. Soil-profile studies of ^{236}U conducted worldwide indicate that its distribution characteristics are jointly controlled by multiple factors, including soil type, environmental stability, hydrodynamic conditions, and vegetation cover. As shown in Table 2, in the Changbai Mountain soil cores of this study, and in other forest soils, the relatively hydrodynamically undisturbed environment favors long-term fixation: the ^{236}U peak is concentrated near the surface and migrates downward slowly. By contrast, in environments with stronger hydrodynamic disturbance, such as peatlands, the profile distribution of ^{236}U is more dispersed or migrates to deeper layers, and its retention capacity is weaker. This comparison highlights the important controlling role of the physical stability of environmental media in the retention of ^{236}U.

In addition, small peaks corresponding to the pre-nuclear-test global fallout were found in the Changbai Mountain forest core in China at 25–30 cm, in the Black Forest peat core in Germany at 41 cm[^28], and in the forest soil core in Queensland, Australia, at 13–16 cm[^29]. This phenomenon has important geochemical implications. Taking the Changbai Mountain core as an example, the sharp increase in organic matter content at the 25–30 cm layer may cause significant changes in soil physicochemical properties. On the one hand, ^{236}U can directly complex with organic matter; on the other hand, decomposition of organic matter consumes oxygen and lowers the redox potential[^30,^31], thereby promoting the accumulation of ^{236}U at this horizon.

In summary, the fixation of ^{236}U in soils is a process controlled by multiple factors. These mechanisms not only affect the distribution characteristics of ^{236}U under different environmental conditions, but also provide an important geochemical theoretical basis for risk assessment and remediation of uranium-contaminated soils.

Table 2. Comparison of peak depths of ^{236}U deposition at different sites worldwide

Study area	Sample type	Depth of 1963 deposition peak/cm	Depth of pre-nuclear small peak/cm	Reference
Changbai Mountain, China	Forest soil core	5–10	25–30	This study
Washington State, USA	Forest soil core	0–5, 5–10	-	[32]
Black Forest, Germany	Peat core	22	41	[28]
Queensland, Australia	Forest soil core	2.05–2.65	13–16	[29]
Madagascar	Peat core	-	-	[33]

Note: "pre-nuclear small peak" refers to the ^{236}U accumulation layer in strata deposited before the start of nuclear testing.

3.3 Application of ^{236}U in Ice-Cap (Ice-Core) Records

Glacier ice cores are ideal natural archives for preserving historical information on atmospheric radionuclide deposition. Compared with other environmental media such as soils, precipitation efficiently captures gaseous and aerosol-borne radionuclides from the atmosphere and seals them in ice layers, forming deposition records with annual resolution.

can be used to evaluate the relative contributions of different nuclear events. The advantage of ^{236}U in ice cores is that almost no decay loss occurs during the post-depositional process; its chemical properties are stable, and it is not readily affected by post-depositional migration in ice cores.

In 2013, Wendel et al. systematically measured, for the first time, the time series of $^{239+240}$Pu and ^{236}U in an ice core from Svalbard in the Arctic. The ice core covered the 1950s to the 1990s, and the ^{236}U/^{239}Pu atomic ratio during the nuclear-test period was 0.18–0.33. The authors pointed out that the significant increase in the ^{236}U/^{239}Pu atomic ratio in the 1975–1976 ice layer may have resulted from inputs from nuclear tests and spent-fuel reprocessing plants. This study established the feasibility of ice cores as an archive of atmospheric deposition of ^{236}U, laying the foundation for subsequent applications of ^{236}U in tracing within the cryosphere[^34]. Wang et al. simultaneously measured $^{239+240}$Pu and ^{236}U in an ice core from the eastern Tianshan Mountains in China. The mean ^{240}Pu/^{239}Pu atomic ratio of this ice core was 0.18 ± 0.02, and the mean ^{236}U/^{239}Pu ratio was 0.27 ± 0.09, consistent with the global fallout range[^35], indicating that the impact of the Lop Nor nuclear tests on glaciers in the eastern Tianshan Mountains was limited.

In summary, ice-core ^{236}U records can reconstruct the history of atmospheric nuclear-test fallout at annual resolution and distinguish global background from local contributions. On this basis, there remains broad room for expanding the application of ^{236}U in the cryosphere. For example, Antarctic ice-core ^{236}U records can be used to evaluate the contribution of Southern Hemisphere nuclear tests (Australia and Pacific test sites) to Antarctica. In addition, time-series studies of ^{236}U in alpine glaciers (such as the Andes and the Himalayas) can provide unique perspectives for understanding atmospheric circulation patterns and transport pathways of radionuclides in tropical and subtropical regions.

3.4 Tracing of ^{236}U in the Marine Environment

3.4.1 Application of ^{236}U in Seawater and Sediments The marine environment is an important receptor of global ^{236}U fallout. Because ^{236}U has relatively high solubility, a single source, and a comparatively uniform distribution relative to other uranium isotopes, it has in recent years become a new tracer for studying ocean currents and water-mass mixing processes. At present, relevant studies have been carried out in typical sea areas such as the South China Sea, the Baltic Sea, the Sea of Japan, and the Mediterranean Sea. Our analysis of surface sediment samples from Beibu Gulf, Guangxi, shows that the ^{236}U/^{238}U atomic ratio is 3.30×10^{-10}–3.62×10^{-8}, and the spatial distribution of ^{236}U shows an evident trend of being higher in the north and lower in the south: concentrations are higher in areas near river mouths and lower in the southern open sea. This indicates that ^{236}U in sediments of the northern gulf is mainly controlled jointly by the long-term accumulation of global atmospheric nuclear-test fallout and local terrestrial inputs[^36]. Generally, the atomic concentration of ^{236}U in open-ocean surface seawater is 10^6–10^7 atoms/L[^37,^38]. However, under the influence of discharges from spent-fuel reprocessing plants (such as Sellafield in the United Kingdom), concentrations in adjacent sea areas (such as the Irish Sea) can rise anomalously to 10^{10} atoms/L[^39,^40]. Differences in ^{236}U concentrations and ratios among different sea areas and depths reflect the transport and mixing processes of water masses from different sources[^41].

By analyzing the concentrations and atomic ratios of ^{236}U and other artificial radionuclides (such as ^{129}I, ^{137}Cs, and Pu isotopes) in different water bodies, the transport pathways and mixing mechanisms of seawater can be more deeply understood. Sakaguchi et al. comparatively studied ^{236}U and ^{137}Cs in seawater of the Sea of Japan and found that the two showed consistent behavioral trends in the Sea of Japan[^37]. Casacuberta et al. used combined tracing with ^{236}U and ^{129}I to successfully distinguish three different Atlantic water currents entering the Arctic Ocean[^42]. In combination with clearly defined sources and detailed release histories, ^{236}U can serve as an effective tool for identifying different water masses and their transit times[^41].

3.4.2 Application of ^{236}U in Marine Biological Samples

At present, ^{236}U has been applied to various marine biological materials, among which corals and shells are the most widely used because of their unique temporal and spatial advantages. Both corals and shells are mainly composed of $CaCO_3$ and enrich uranium from seawater through biomineralization, enabling the reconstruction of the history of nuclear-explosion inputs, changes in water-mass sources, and the time scales of other nuclear events. Because of their annual growth bands and closed fossil skeleton structures, corals have become natural high-resolution archives for recording the history of seawater ^{236}U. Shells, meanwhile, facilitate multi-site spatial sampling and allow the rapid construction of regional-scale maps of the spatial distribution of ^{236}U.

Huang et al. first reported AMS measurement results for ^{236}U, ^{237}Np, and $^{239+240}$Pu in shells (oysters, clams, and scallops) along the coast of southern China. In samples collected from the South China Sea and East China Sea sites, the study found that ^{236}U concentrations ranged from $(0.358–3.968) \times 10^6$ atoms/g, and the $^{236}U/^{238}U$ atomic ratios were $(0.046–0.524) \times 10^{-8}$, consistent with expected levels in surface seawater affected by global fallout, indicating that ^{236}U in shells from Chinese seas mainly originates from global fallout from atmospheric nuclear tests[43].

Sakaguchi et al. used coral cores from Iki Island, Japan, and measured $^{236}U/^{238}U$ atomic ratios from 1935 to 2010 by AMS, reconstructing the input history of ^{236}U in Japanese surface waters. The results showed three significant peaks in 1955, 1959, and 1963, corresponding respectively to local inputs from hydrogen-bomb tests at the Pacific Proving Grounds and to global fallout; among them, the 1959 peak reached 6.15×10^{-9}[44]. Froehlich et al. combined ^{236}U and Pu isotopic characteristics to study the chronological information contained in coral samples from the Marshall Islands, observing that these isotope ratios can provide information related to the nature of each nuclear-weapon test[45].

3.5 Application of ^{236}U in Atmospheric Deposition Records and Aerosol Source Apportionment

The main pathway by which ^{236}U enters the atmosphere is atmospheric nuclear-test explosions: nuclear-fuel debris is directly injected into the stratosphere and is subsequently transported and deposited on a global scale through atmospheric circulation. Compared with environmental media such as soil and water bodies, the ^{236}U signal in atmospheric aerosols is closer to the original deposition events and may be affected only by small amounts of resuspended surface material. This characteristic makes aerosols ideal "snapshot" samples for studying the atmospheric deposition history of ^{236}U. On the basis of this characteristic, existing studies have demonstrated their application value from three perspectives: reconstruction of deposition history, tracing of long-distance transport, and identification of local anomalies.

Wallner et al. analyzed air filters from Austria for 1962–1966 and showed that

the $^{236}U/^{238}U$ atomic ratio exhibited a seasonal feature with spring maxima; the $^{233}U/^{236}U$ atomic ratios, $(0.15–0.49) \times 10^{-2}$, were far lower than the global-fallout integral value, confirming that ^{233}U mainly came from earlier nuclear tests (around 1955) rather than those of the 1960s[46]. Shinonaga et al., in aerosol samples collected approximately 120 km from the Fukushima Daiichi Nuclear Power Plant, used time-series data for Pu, anthropogenic U, and Cs, combined with wind-trajectory analysis, and for the first time verified with combined Pu and ^{236}U data that fuel material from the Fukushima nuclear accident could be transported over long distances in the atmosphere[47]. Ohno et al. analyzed the $^{236}U/^{238}U$ and $^{235}U/^{238}U$ atomic ratios in atmospheric deposition in Tokyo and Akita during 1963–1979. The $^{236}U/^{238}U$ atomic ratio in the Akita samples showed a global-fallout peak in 1963, while the $^{235}U/^{238}U$ atomic ratio remained close to the natural value throughout. In the Tokyo samples, the $^{236}U/^{238}U$ atomic ratio increased again in the 1970s, while the $^{235}U/^{238}U$ atomic ratio simultaneously decreased to a minimum, confirming local depleted-uranium (DU) release[48].

4 Global ^{236}U Deposition Inventories and Flux Estimation

4.1 Spatial Distribution Pattern of Inventories

The deposition inventories of ^{236}U in soils vary significantly among different regions of the world. They exhibit a pattern of high values at mid-latitudes in the Northern Hemisphere and a bidirectional decrease toward the south and north (Fig. 1). This distributional feature is first manifested as hemispheric asymmetry: inventories in the Northern Hemisphere are generally higher than those in the Southern Hemisphere, with the high-value center located in the mid- to high-latitude belt of 40–50°N. This is closely related to the fact that most historical atmospheric nuclear tests were located in this belt and to the control exerted by mid-latitude atmospheric circulation[^49]. In addition, although annual precipitation plays an important regulating role in regional flux, it is not the dominant controlling factor; the effects of latitude and source strength take precedence over precipitation.

Tianshan, China (43°N), lies deep in the interior of the Eurasian continent, at high elevation, with an annual mean precipitation of only 229 mm and relatively low wet-deposition efficiency, yet its inventory still reaches 3.5×10^{12} atoms/m^2[^35]. By comparison, the Svalbard Archipelago in the Arctic (79.83°N) has annual mean precipitation of 370–450 mm, slightly higher than Tianshan; however, because the overall atmospheric deposition flux at high latitudes is lower, its inventory is instead lower, at only 1.63×10^{12} atoms/m^2[^34]. Although Queensland, Australia, in the Southern Hemisphere is extremely humid in terms of precipitation (annual mean 2200 mm), because the total amount of global radionuclide deposition in the Southern Hemisphere is intrinsically far lower than that in the Northern Hemisphere, its inventory is only $(8.5 \pm 0.3) \times 10^{11}$ atoms/m^2[^29], less than one quarter of that in Tianshan. In mid-latitude Japan in the Northern Hemisphere, however, the situation is entirely different:

Bar chart comparing ^{236}U inventories among sites: Svalbard Archipelago, Black Forest in Germany, Washington State in the United States, Tianshan in China, Changbai Mountain in China, Ishikawa Prefecture in Japan, the Equator, and Queensland in Australia. The x-axis is labeled "^{236}U inventory / (10^{12} atoms $\cdot$ m^{-2})".

Figure 1: Bar chart comparing ^{236}U inventories among sites: Svalbard Archipelago, Black Forest in Germany, Washington State in the United States, Tianshan in China, Changbai Mountain in China, Ishikawa Prefecture in Japan, the Equator, and Queensland in Australia. The x-axis is labeled "^{236}U inventory / (10^{12} atoms $\cdot$ m^{-2})".

Ishikawa Prefecture likewise has annual mean precipitation of 2200 mm, but its inventory is as high as 1.78×10^{13} atoms/m^2[^50], more than twenty times that of Queensland, fully demonstrating the characteristic that the mid-latitudes of the Northern Hemisphere are the high-value belt of global ^{236}U deposition.

In this study, the soil ^{236}U inventory measured at Changbai Mountain (42°N) was $(9.83 \pm 0.18) \times 10^{13}$ atoms/m^2, one of the highest values among currently reported global deposition sites. This magnitude far exceeds those of stations such as Tianshan and Svalbard. The high inventory at Changbai Mountain is the result of the combined superposition of latitude and monsoon-enhanced ^{236}U deposition, further highlighting the status of the mid-latitude belt of the Northern Hemisphere as the core region of global ^{236}U deposition. Overall, the mid-latitude regions of the Northern Hemisphere, together with abundant precipitation, constitute the most favorable combination for forming high inventories, whereas high latitudes and the Southern Hemisphere, even when precipitation is relatively abundant, are still unable to bridge the fundamental gap brought about by hemispheric and latitudinal belts.

Figure 1. Comparison of ^{236}U inventories in regions at different latitudes (the red dashed line indicates the equator, used to separate Northern Hemisphere and Southern Hemisphere stations)

4.2 Methodological Differences and Uncertainty in Estimating the Global Total

Drawing on methods established in previous studies, this study estimated the total deposition of ^{236}U produced by global nuclear explosions, using forest-soil cores from the Changbai Mountains as the core material. This method follows two validated technical approaches: first, the approach proposed by Quinto et al. based on German Black Forest peat cores, in which the nuclide ratio in complete recovered cores is used to estimate global deposition[^28]; second, the method adopted by Shao et al., in which the ^{236}U/^{137}Cs activity ratio is multiplied by the global total deposition of ^{137}Cs to infer the total amount of ^{236}U[^51]. This study applied the above methods to measured core samples from

the Changbai Mountains. The feasibility of the estimation rests on the following key premises: radioactive nuclides released into the atmosphere (such as ^{137}Cs and ^{236}U) are mainly adsorbed onto aerosol particles and, after long-distance transport, reach the land surface through dry and wet deposition; this process is mainly controlled by meteorological conditions and the physical properties of the particles[^52]. By analyzing the data reported by Wallner et al., we found that the activities of ^{137}Cs and ^{236}U in aerosols are highly correlated ($R^2 = 0.99$, $p < 0.01$)[^53], indicating that the two are attached to similar aerosols and are deposited in similar ways. In addition, both ^{137}Cs and ^{236}U in the Changbai Mountain region of this study originate from global atmospheric nuclear-test fallout, ensuring the uniqueness of the source term for the estimation. Finally, the vertical distribution data of the sediment core samples in this study are complete, with no missing horizons or abnormal disturbance, providing high-quality baseline data for the integration calculation. On the basis of the above evidence, we propose the following two assumptions to complete the estimate of the global total: (1) ^{236}U and ^{137}Cs in the global atmosphere have been fully and uniformly mixed; (2) in the depositional area where the cores in this study were collected, the nuclides deposited mainly underwent downward migration, and fractionation of ^{236}U and ^{137}Cs caused by horizontal migration can be neglected. Based on these premises and assumptions, we estimated the global atmospheric deposition of ^{236}U. First, the total global deposition of ^{137}Cs reported by Shao et al. as of 13 December 2017, 255.5 PBq[^51], was corrected to the sampling date (May 2021), yielding approximately 239.4 PBq. Then, using the ^{236}U/^{137}Cs activity ratio of this core $(9.68 \pm 0.30) \times 10^{-6}$, the total amount of ^{236}U released by global nuclear explosions was inferred to be 968 ± 290 kg (the uncertainty was jointly estimated from the ratio of the atmospheric deposition totals of $^{239+240}$Pu and ^{137}Cs and the $^{239+240}$Pu/^{137}Cs activity ratio in the core). This result is highly consistent with the 900 kg estimated by Sakaguchi et al. based on Japanese soil data[^20]. However, by integrating existing global studies (see Figure 2), estimates of the total release of ^{236}U span a relatively wide range and can be roughly divided into three groups: (1) the approximately 900–1000 kg range, including 968 ± 290 kg from Changbai Mountain and 900 kg from Sakaguchi et al.[^20]; (2) the approximately 1300 kg range, including 1300 ± 448 kg from Shao et al.[^51] and 1060 ± 212 kg from Winkler et al.[^54]; and (3) a higher estimated-value range, such as 1698 ± 850 kg from Quinto et al.[^28] and 2100 kg from Christl et al.[^55]. These differences may arise from differences in sampling locations, sample types, model assumptions, and measurement methods. Although the estimates of ^{236}U differ among studies, overall they remain within the same order of magnitude, with the maximum value (2100 kg) being approximately 2.3 times the minimum value (900 kg). It is worth noting that the estimate of 968 ± 290 kg provided by this study, through coupling with the global total amount of ^{137}Cs and correction using the local activity ratio, provides independent verification from the East Asian continental forest ecosystem for this order-of-magnitude range, and has certain reference value for constraining the uncertainty in the total global release of ^{236}U.

Y-axis: Estimated total atmospheric deposition of ^{236}U */ kg*

X-axis categories: Ishikawa Prefecture soil, Japan; Changbai Mountain soil, China; Canada Baffin Bay; surface soil at Zhangjiakou, Beijing; German Black Forest shale; western Pacific seawater.

Figure 2. Total atmospheric deposition of ^{236}U estimated from different samples (error bars are from the original literature)

5 Results and Outlook

As a sensitive indicator of nuclear activity, ^{236}U has demonstrated unique tracer value in various environmental media, including soil, ice sheets, oceans, and the atmosphere, complementing traditional tracers such as ^{129}I, ^{137}Cs, and Pu isotopes. AMS technology has become an important means for ultratrace measurement of ^{236}U, and in recent years important breakthroughs have been achieved in the optimization of stripping devices and the miniaturization of instruments. At the same time, several practical bottlenecks still exist in this field. The pretreatment procedures for ice-core and seawater samples remain lengthy and complex, measurement costs are high, and enrichment by coprecipitation for large-volume samples together with multistep separation and purification causes the full workflow for a single sample to take several days. Coupled with AMS beam-time costs, this seriously constrains the measurement of large batches of environmental samples.

In response to the above bottlenecks, future domestic research may focus on breakthroughs in four specific directions. First, the development of miniaturized AMS systems and rapid detection techniques for uranium isotopes should be pursued, so as to achieve rapid, low-cost, low-sample-volume measurements for nuclear emergency response and nuclear security. Second, automated batch pretreatment workflows should be developed to significantly reduce the difficulty of sample processing. Third, standardized analytical procedures for combined tracing with ^{236}U, ^{129}I, and Pu isotopes should be established, relying on sequential separation of U, Pu, and I fractions from the same digestion solution to realize simultaneous acquisition of multiple nuclides and promote progress in research on multi-nuclide joint analysis. Fourth, the expanded tracer application of natural ^{236}U in low-content geological samples should be developed, serving related studies in uranium-ore exploration, groundwater, and geology. For example, natural ^{236}U accumulates over long periods in uranium deposits through the $^{235}\mathrm{U}(n,\gamma)$ reaction, and its

The $^{236}\mathrm{U}/^{238}\mathrm{U}$ ratio can reflect the history of neutron fluence experienced by uranium ore bodies and is promising for use in reconstructing the metallogenic age of uranium deposits and ancient redox environments.

From the perspective of practical needs, North China, Northwest China, and the southeastern coastal region, as areas where nuclear-energy facilities are con-

centrated, make the establishment of an environmental background baseline for ^{236}U in China of certain practical significance. In addition, after the Fukushima accident, the demand for rapid analysis of ^{236}U in East Asia' s nuclear emergency monitoring system has been increasing. In the field of nuclear forensics, it is also necessary to gradually accumulate a database of ^{236}U isotopic fingerprint data covering uranium mining, concentration, reactor piles, and other stages in China, in order to improve the ability to identify the provenance of nuclear materials. In subsequent work, we will continue to study ^{236}U in sediments from the South China Sea and in terrestrial soils from Northeast and Northwest China, with a focus on the migration and distribution characteristics of this nuclide.

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Measurement and Application of ^{236}U in the Environment: A review

Wenjian Huang[1], Zhiyong Liu[2], Huijuan Wang[1], Zhiyi Tu[1], Jianhui Lai[1], Xianghao Luo[1], Hongjie Wang[1],

Yongjing Guan[1#]

(1. Guangxi Key Laboratory for Relativistic Astrophysics, School of Physical Science and Technology, Guangxi

University, Nanning 530004, Guangxi, China;

2. State Key Laboratory of Radiation Medicine and Protection, School for Radiological and Interdisciplinary

Sciences (RAD-X) and Collaborative Innovation Centre of Radiation Medicine of Jiangsu Higher Education

Institutions, Soochow University, Suzhou 215123, Jiangsu, China)

Abstract: ^{236}U primarily originates from artificial nuclear activities such as atmospheric nuclear tests, nuclear accidents, and nuclear fuel reprocessing. Its extremely low natural background and long half-life make it a highly regarded novel environmental tracer. Accelerator mass spectrometry (AMS) is the most sensitive technique for measuring ultra-trace levels of ^{236}U in the environment. The theoretical abundance sensitivity of current mainstream AMS systems for the $^{236}U/^{238}U$ atomic ratio can reach the order of 10^{-14} or lower, providing a reliable means for measuring ^{236}U in media such as soil, ice cores, seawater, and atmospheric aerosols. This paper systematically reviews the principles of ^{236}U measurement by AMS, interference suppression, and advances in instrumentation. It summarizes pretreatment procedures including sample digestion, uranium separation and purification, and target preparation. The applications

of ^{236}U as a tracer in various environmental media—such as soils, the cryosphere, oceans, and the atmosphere—are reviewed. Based on our team' s measurements of ^{236}U in forest soils from the Changbai Mountains, we report the depth distribution profile of ^{236}U and its correlation with ^{137}Cs and ^{237}Np, thereby estimating the total global fallout of ^{236}U from nuclear tests to be approximately 968 ± 290 kg. Finally, existing bottlenecks such as lengthy pretreatment procedures and high costs are discussed, and future directions are proposed, including miniaturized AMS and rapid detection of uranium isotopes, automated batch pretreatment, multi-nuclide joint tracing, and expanded applications of natural ^{236}U as a tracer.

Key words: ^{236}U; Accelerator Mass Spectrometry; Applications

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 # Corresponding author: Yongjing Guan, E-mail: yjguan@gxu.edu.cn

Note: Figure translations are in progress. See original paper for figures.

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