

The suitability of ^{236}U as an ocean tracer

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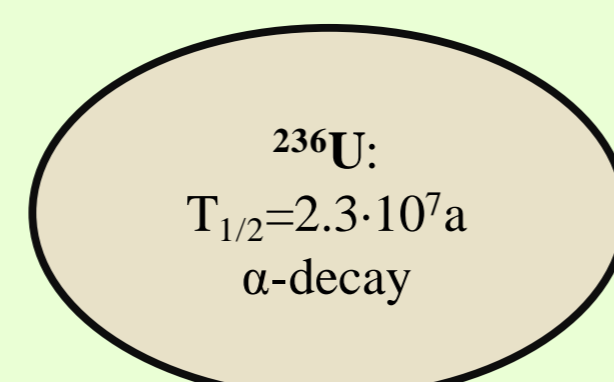
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Introduction

^{236}U is an α -emitter with a half-life of $2.3 \cdot 10^7$ years. It does occur in nature, but only at ultra-trace concentrations. Natural $^{236}\text{U}/^{238}\text{U}$ ratios in the range of 10^{-10} for uranium bearing ores have been reported in literature and ratios of 10^{-14} are expected for typical environmental samples. However, so far no widespread survey has been completed because levels are too low for routine detection by techniques other than AMS [1,2]. If the $^{236}\text{U}/^{238}\text{U}$ ratio is higher than 10^{-9} , it can be assumed that the sample has been exposed to a significant (anthropogenic) neutron flux [2].

We measured sea water samples from two world oceans (Atlantic Ocean, Pacific Ocean), one marginal sea (Irish Sea) and one inland sea (Black Sea).

uranium isotope	natural abundance [3]
^{238}U	99.27%
^{235}U	0.72%
^{234}U	0.005%
^{236}U	$^{236}\text{U}/^{238}\text{U} = 10^{-14} - 10^{-10}$



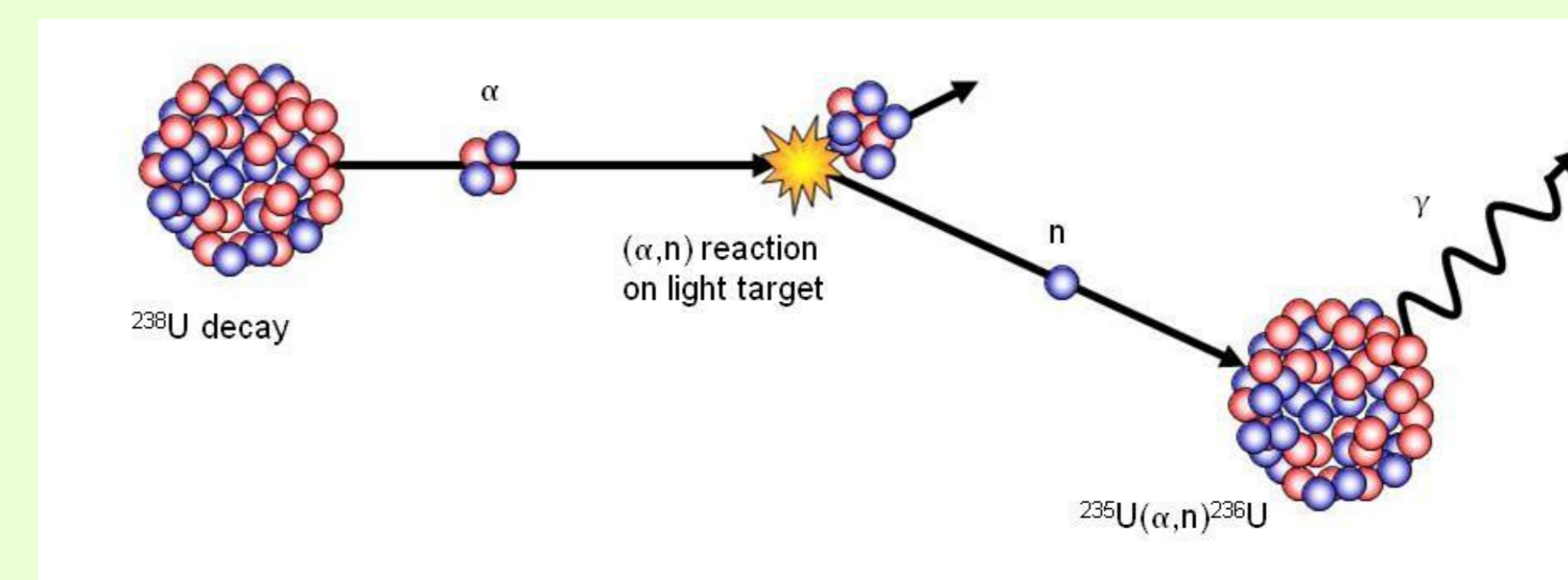
Formation of ^{236}U

- $^{235}\text{U}(n,\gamma)^{236}\text{U}$
- $^{238}\text{U}(n,3n)^{236}\text{U}$
- ^{240}Pu (α -decay)

^{236}U can be formed by $^{235}\text{U}(n,\gamma)^{236}\text{U}$ and $^{238}\text{U}(n,3n)^{236}\text{U}$ reactions or as a result of growth from ^{240}Pu already deposited on land by global fallout [4,5].

When thermal neutrons interact with ^{235}U , the probability for fission is about 85%, whereas formation of ^{236}U happens with a probability of 15% [3].

A great amount of ^{236}U is produced as a by-product in nuclear reactors, also via the thermal neutron capture on ^{235}U [4].



Estimated global inventory

- natural ^{236}U : ca. 30 kg
- anthropogenic ^{236}U : more than 10^6 kg

The global inventory of natural ^{236}U is assumed to be about 30 kg ^{236}U in the upper layers of land surface and less than 0.5 kg in the oceans.

The anthropogenic inventory of ^{236}U is in the order of 10^6 kg, based on the assumption that the total uranium mined till 2003 adds up to $2.2 \cdot 10^9$ kg and assuming 50% of the ^{235}U were actually used up in reactors [3].

Chemical Procedures

The chemical procedure to the uranium extraction is based on the co-precipitation of uranium with $\text{Fe}(\text{OH})_3$, 20 μL of ^{233}U spike [$(4.76 \pm 0.05) \cdot 10^{10}$ atoms ^{233}U per g, IRMM-058; sample identification 0008] and 20 mL of Fe^{3+} solution (1 mg Fe^{3+}/mL) are added to 2 L of the acidified (pH=2) water samples. After heating the sample solution for 6-8 hours, NH_4OH solution is added to adjust a pH of 8-9, which leads to the forming of a red brown $\text{Fe}(\text{OH})_3$ precipitate. This precipitate is then dissolved in 3M nitric acid and uranium is extracted by a column filled with UTEVA[®] resin.

Targets for AMS are prepared by co-precipitation with $\text{Fe}(\text{OH})_3$, calcination in a furnace and pressing the resulting oxides into aluminium sample holders, which are convenient for the use in the ion source of the AMS facility.

Accelerator Mass Spectrometry – AMS

AMS is well suited for the detection of heavy, long-lived isotopes, because it measures the isotopes directly rather than their infrequent decay and it is robust against interfering molecular isobars [6,7].

The Vienna Environmental Research Accelerator (VERA) was especially adapted for the isotope ^{236}U and its lowest $^{236}\text{U}/^{238}\text{U}$ ratio reported on real samples is $5 \cdot 10^{-12}$ [7]. While previous measurements could identify ^{236}U only in the vicinity of known contaminated sites (Sellafield, Chernobyl, etc.), our measurements in various compartments of the environment are evolving into a consistent picture of the dispersion of anthropogenic ^{236}U .

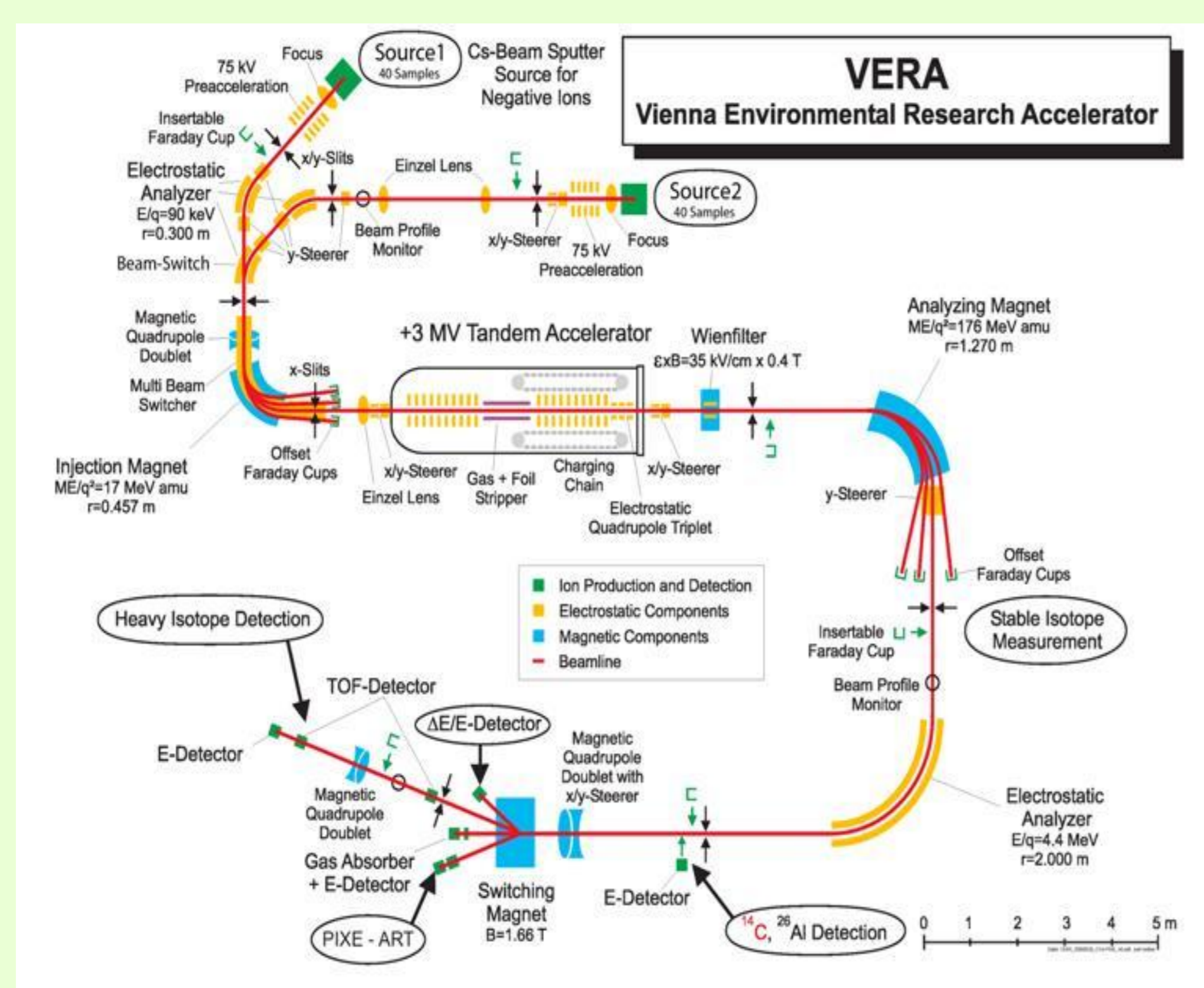


Fig.: Schematic layout of the AMS facility VERA

Sample locations



Results

An isotopic ratio $^{236}\text{U}/^{238}\text{U}$ higher than 10^{-9} is a clear indicator for anthropogenic contamination. Thus, all samples were affected by human nuclear activity and the elevated $^{236}\text{U}/^{238}\text{U}$ levels are likely to result from contamination by global fallout [5].

Atlantic Ocean, Pacific Ocean and Black Sea revealed ^{236}U levels on the same order of magnitude (10^{-9}). For water from the Irish Sea, the detected $^{236}\text{U}/^{238}\text{U}$ isotopic ratio was much higher, showing obviously an influence by the Sellafield reprocessing plant [8,9].

sample	$^{236}\text{U}/^{238}\text{U}$	$\pm 1\sigma$
Atlantic Ocean, La Palma Island, Spain	$1.87 \cdot 10^{-9}$	$5.62 \cdot 10^{-10}$
Black Sea, Romania	$3.63 \cdot 10^{-9}$	$4.89 \cdot 10^{-10}$
Pacific Ocean, Hawaii, USA	$5.74 \cdot 10^{-9}$	$3.07 \cdot 10^{-10}$
Irish Sea	$2.04 \cdot 10^{-6}$	$2.07 \cdot 10^{-8}$

Conclusions

^{236}U can figure as a valuable tracer in hydrology and oceanography and it is a key signature to differentiate uranium sources [10]. ^{236}U has a long residence time of $\sim 500,000$ years in sea water and a sufficiently long half-life ($2.3 \cdot 10^7 \text{ a}$). The natural level is negligible compared to the measured anthropogenic ratios in all investigated seas. ^{236}U is thus suitable as a tracer for the study of ocean dynamics on a global scale.

Compared to ^{137}Cs , where 10 L of sea water are required [11], we obtained our results for ^{236}U on smaller sample sizes ($< 2 \text{ L}$).

^{236}U may also outperform the more established AMS isotope ^{129}I as an ocean tracer, for which the source term is generally restricted to the reprocessing plants and which undergoes more complicated environmental chemistry. Generally, the measurement of several tracer nuclides allows to address dilution and mixing processes.

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