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Natural and anthropogenic ^{236}U in environmental samples

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Abstract

The interaction of thermal neutrons with ^{235}U results in fission with a probability of ~85% and in the formation of ^{236}U ($t_{1/2} = 2.3 \times 10^7$ yr) with a probability of ~15%. While anthropogenic ^{236}U is, therefore, present in spent nuclear fuel at levels of ^{236}U/U up to 10^{-2}, the expected natural ratios in the pre-anthropogenic environment range from 10^{-14} to 10^{-10}. At VERA, systematic investigations suggest a detection limit below $^{236}\text{U/U} = 5 \times 10^{-12}$ for samples of 0.5 mg U, while chemistry blanks of $\sim 2 \times 10^7$ atoms ^{236}U per sample limit the sensitivity for smaller samples. We have found natural isotopic ratios in uranium reagents separated before the onset of human nuclear activities, in uranium ores from various origins and in water from a subsurface well in Bad Gastein, Austria. Anthropogenic contamination was clearly visible in soil and rivulet samples from Salzburg, Austria, whereas river sediments from Garigliano river (Southern Italy) were close to the detection limit. Finally, our natural in-house standard Vienna-KkU was calibrated against a certified reference material (IRMM REIMEP-18 A).

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1. The natural uranium inventory

'Natural uranium', as it can be found in the Earth's crust, consists of two primordial isotopes (^{238}U, ~99.3%; ^{235}U, ~0.72%) and the radiogenic isotope ^{234}U (equilibrium abundance of ~0.005%). ^{236}U with a half-life of $2.342(3) \times 10^7$ years [1] is continuously produced by neutron-capture on ^{235}U.

The thermal neutron capture cross-section ($\sigma[^{235}\text{U}(n,\gamma)] =$ 98 barn) is about 1/6 of the fission cross-section ($\sigma[^{235}\text{U}(n,f)] = 583$ barn). At underground depth, below the influence of cosmogenic particles (~30 m), the dominant sources of neutrons are from (α,n) reactions on light nuclei, where the α-particles are emitted in the U and Th α-decay series [2] and from spontaneous fission of ^{238}U [3]. In some high-grade uranium ores, induced fission of ^{235}U makes a minor contribution. The difficulty of modeling is discussed in [4], for the similar case of natural ^{239}Pu (and ^{36}Cl) production. The thermal neutron flux will

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strongly depend on the concentration of light targets elements for (α,n), on water and uranium content for efficient thermalization and on neutron poisons such as boron, gadolinium and samarium. [4] suggests that at least an uncertainty of 30% of a modeled ^{236}U/U ratio has to be expected. For some applications, shorter-lived radionuclides (^{36}Cl, ^{41}Ca) may provide experimental neutron flux data.

^{236}U/U ratios have been measured previously in several uranium ores [5–8]. Typical ratios obtained range from 1×10^{-11} to 1×10^{-9}. The total amount of uranium ore on Earth is difficult to assess, but the "ultimately recoverable global natural uranium resource base" is presently estimated to 2.9×10^{10} kg U [9]. Assuming an isotopic ratio of $^{236}\mathrm{U/U} = 5 \times 10^{-11}$, this corresponds to an inventory of 1.5 kg ^{236}U.

Different from ores, typical rocks have a uranium and thorium content of only several mg/kg, thus a much lower ^{236}U/U ratio is expected. For rock types investigated in [2] and at depths where cosmogenic production is negligible, values between 1×10^{-14} and 5×10^{-14} can be calculated. On average, these rock types have a density of 2.6 g cm^{-3} and a uranium content of 1.8 mg/kg. Assuming $^{236}\mathrm{U/U} = 3 \times 10^{-14}$ results in an additional inventory of 22 kg ^{236}U in the uppermost 1000 m of land surface.

Close to the surface, cosmic radiation increases the thermal neutron flux by some two orders of magnitude. Calculated depth profiles are given in [2]. Integration yields ^{236}U production rates of typically 0.5 ^{236}U atoms/cm^2 and year (half of this production takes place in the top-most meter). This corresponds to an additional inventory of 10 kg.

The uranium can be released during rock weathering. In contrast to the case of radiogenic ^{234}U, which is leached out of the host mineral more readily than the primordial U isotopes, we expect no fractionation of ^{236}U/U. The recoil of the α decay ($\sim 10^5$ eV) is thought to dislocate the ^{234}U atom and to damage the lattice, which increases its mobility [10]. For the (n,γ) production of ^{236}U, the recoil energy is typically below 1 eV (estimated using prompt-γ data from [11]), which is below the displacement energy of most minerals. A total flux of 0.88×10^{10} g U yr^{-1} is transported by rivers and ground water aquifers to the oceans [12]. The average ^{236}U/U ratio will reflect the ratio in the deep lithosphere, plus additional ^{236}U from cosmogenic production. To get limits for the latter, we consider the extreme cases: all cosmogenic ^{236}U remains on land till it decays or is completely transported to the oceans. The latter case will increase the ^{236}U/U ratio in river runoff by 3.4×10^{-14}. Altogether, the average ratio in river water should lie between 1×10^{-14} and 1×10^{-13}, which directly translates into the expected ^{236}U/U of the pre-anthropogenic oceans. Their total uranium budget of 4.5×10^{15} g has a mean residence time of $(3.2–5.6) \times 10^5$ yr [13] and is well mixed with respect to ^{236}U. The resultant ^{236}U inventory of the pre-anthropogenic oceans is below 0.5 kg.

The amount of anthropogenic ^{236}U produced in nuclear power reactors can be estimated from the total uranium mined, i.e. 2.2×10^9 kg up to 2003 [14]. By assuming that 50% of the ^{235}U were actually used up in reactors, we obtain an anthropogenic inventory at the order of 10^6 kg ^{236}U.

The environmental distribution of anthropogenic ^{236}U is not well assessed yet. ^{236}U/U ratios from 10^{-6} up to 10^{-3} have been measured in soils from the close vicinity of the Chernobyl power plant [15,16]. Up to a distance of 200 km from this site, ratios were found from 2×10^{-7} to 4×10^{-7} [17], while near two uranium processing factories in the USA they range between 10^{-6} and 1.4×10^{-4} [18]. In sediments from the Irish Sea affected by the Sellafield (UK) nuclear fuel reprocessing plant, ratios are 10^{-6} to 10^{-5} [16,18,19]. In commercial reagents and artificial standard materials, ^{236}U/U ratios as high as 10^{-6} have been measured [16,20]. It is difficult to predict how far anthropogenic ^{236}U is spread at the yet unexplored sensitivity level of ^{236}U/U below 10^{-6}. Due to the low specific activity of ^{236}U, significant amounts can be released during fuel reprocessing without violating health protection regulations. Atmospheric bomb tests may have introduced a global surface contamination with ^{236}U similar to the Pu isotopes and releases from nuclear power plants, both during routine operation and due to accidents, are a possible source of regional contamination.

2. Samples

Uranium reagents, prepared from ore in pre-nuclear times, preserve natural isotopic ratios with minimal risk of anthropogenic contamination. A large amount (many kilograms) of uranyl nitrate (sample label "Vienna-KkU" in Table 1) is stored in the basement of the Radiuminstitut in Vienna (merged in the Faculty of Physics in 2007). It was mined and processed before 1918 in Joachimsthal (Czech Republic). The ore grade mined at that time was typically 60% U_3O_8 [21]. ^{236}U/U ratios for this material have been published in [8,22,23].

VERA has participated in the blind inter-comparison exercise REIMEP-18 [24] for diluted uranium solutions. Our results submitted for "REIMEP 18 A" is $^{236}\mathrm{U}/^{238}\mathrm{U} = 3.047(76) \times 10^{-8}$. This value was obtained by normalizing to an independently diluted solution "Sol3-1-2 (10^{-8})" [25] and agrees well with the finally certified value $^{236}\mathrm{U}/^{238}\mathrm{U} = 3.0579(83) \times 10^{-8}$ [26]. With "REIMEP 18 A" as reference we were able to calibrate our pre-nuclear uranyl nitrate stock as an in-house standard "Vienna-KkU", $^{236}\mathrm{U}/^{238}\mathrm{U} = (6.98 \pm 0.32) \times 10^{-11}$. The lack of a standard material for ^{236}U/U close to natural isotopic ratio was identified as a main source of uncertainty in [8,22,23].

A single bottle of uranyl acetate ("Kk-Uranylacetat" in Table 1) stored at the Radiuminstitut serves as a further, independent sample. Material "U3O8_MB" was obtained from the Atominstitut of the Technical University, Vienna (result already published in [23]). Sputter targets from two old reagents stored at the ETH Zürich and at the Univer-

Table 1
Samples and results

Sample label	Description	U content (μg/g)	$^{236}U/^{238}U$	^{236}U concentration (atoms/g)*
Vienna-KkU	Uranyl nitrate, K.k. Uranfabrik Joachimsthal, stored at the University of Vienna, (In-house standard)	~500,000**	$(6.98 \pm 0.32) \times 10^{-11}$	$(9.0 \pm 0.4) \times 10^{10}$
U3O8_MB	U_3O_8, K.k. Uranfabrik Joachimsthal, stored at TU Wien	–	$(5.25 \pm 0.9) \times 10^{-11}$	–
Kk-Uranylacetat	Uranyl acetate, old reagent stored at the University of Vienna	–	$(9.5 \pm 0.5) \times 10^{-11}$	–
LW_32_Blank_BE	UO_3, old uranium reagent found at the Universität Bern	–	$(5.9 \pm 0.2) \times 10^{-10}$	–
LW_32_Blank_ZH	U_3O_8, old uranium reagent found at the ETH Zürich	–	$(6.4 \pm 0.2) \times 10^{-10}$	–
ANU-0102	Ore from Jakimov, Czech Republic	147,000	$(4.83 \pm 0.17) \times 10^{-11}$	$(1.8 \pm 0.6) \times 10^{10}$
ANU-099	Ore from Val Redena, Italy	203,000	$(1.23 \pm 0.09) \times 10^{-11}$	$(6.4 \pm 0.5) \times 10^{9}$
ANU-267	Ore from unknown origin	426,000	$(6.04 \pm 0.18) \times 10^{-11}$	$(6.6 \pm 0.2) \times 10^{10}$
LOT2061	Yellowcake	–	$(6.05 \pm 0.44) \times 10^{-12}$	–
LOT2063	Yellowcake	–	$(4.65 \pm 0.16) \times 10^{-11}$	–
Garigliano upstream average	River sediment from Garigliano river, upstream of nuclear power plant	2	$(1.4 \pm 0.2) \times 10^{-9}$	$(2 \pm 1) \times 10^{7}$
T2H	1 g Salzburg soil 0.5 mm depth	<1	$(1.48 \pm 0.01) \times 10^{-6}$	$(5.1 \pm 0.2) \times 10^{8}$
T2G	1 g Salzburg soil, 15 mm depth	<1	$(8.0 \pm 0.4) \times 10^{-7}$	$(5.3 \pm 0.2) \times 10^{8}$
T2F	1 g Salzburg soil, 25 mm depth	2 ± 1	$(3.9 \pm 0.2) \times 10^{-7}$	$(2.09 \pm 0.02) \times 10^{10}$
Forstau_B	1.5 L Salzburg rivulet water	<0.001	$(6.3 \pm 0.4) \times 10^{-8}$	$(1.4 \pm 0.1) \times 10^{5}$
Forstau_F	1.5 L Salzburg rivulet water	<0.0011	$(7.0 \pm 0.4) \times 10^{-8}$	$(2.3 \pm 0.1) \times 10^{5}$
Forstau_G	1.5 L Salzburg well water	<0.001	$(2.1 \pm 0.2) \times 10^{-8}$	$(1.2 \pm 0.1) \times 10^{5}$
Forstau_K	1.5 L Salzburg rivulet water	0.015	$(4.4 \pm 0.1) \times 10^{-8}$	$(11.4 \pm 0.3) \times 10^{5}$
Forstau_L	1.5 L Salzburg rivulet water	<0.001	–	$(0.84 \pm 0.08) \times 10^{5}$
Forstau_P	1.5 L Salzburg rivulet water	<0.001	$(5.8 \pm 0.4) \times 10^{-8}$	$(1.7 \pm 0.1) \times 10^{5}$
Forstau_R	1.5 L Salzburg rivulet water	<0.001	$(11.4 \pm 0.8) \times 10^{-8}$	$(1.4 \pm 0.1) \times 10^{5}$
Forstau_T	1.5 L Salzburg rivulet water	0.003	$(4.0 \pm 0.2) \times 10^{-8}$	$(3.7 \pm 0.2) \times 10^{5}$
Gastein 1	2.5 L subsurface well water, Bad Gastein	~0.1	$(6.5 \pm 2.1) \times 10^{-12}$	–
Gastein 2 A	2.5 L subsurface well water, Bad Gastein	~0.1	$(6.2 \pm 2.3) \times 10^{-12}$	–
Gastein 2B	2.5 L subsurface well water, Bad Gastein	~0.1	$(7.0 \pm 3.1) \times 10^{-12}$	–

Uncertainties (1σ) quoted are the statistical uncertainties only. From previous measurements [8], an additional systematic uncertainty of several percent can be deduced.

* The concentration is given for the respective environmental material, i.e. per 1 g of ore, soil or water.

** The uranium concentration refers to typical Joachimsthal ore from that time.

sität Bern (Bern, Switzerland) were provided by Wacker ("LW_32_Blank_ZH" and "LW_32_Blank_BE"). "LOT-2061" and "LOT-2063" (no certified reference materials) were provided by Richter from the IRMM, who verified the absence of anthropogenic ^{236}U down to the detection limit of $\sim 10^{-10}$ with TIMS.

Uranium ores from different origin were processed at ANU and measured for $^{236}U/U$ at VERA. Three ores (labels "ANU-", published in [22]) are shown in Table 1 (a publication of the complete data set, measured both at VERA and at the ANU 14-UD tandem accelerator, is in preparation).

To assess the potential influence of anthropogenic contamination, surface soil samples from the Nassfeld near Böckstein (Salzburg province, Austria) were investigated. The alpine regions of Austria were one of the most affected areas in Western Europe after the Chernobyl accident on April 26, 1986. The samples "T2F-H" are the topmost part of a profile investigated in a separate project [27] for other isotopes.

Waters from several rivulets and one well near Forstau, Salzburger Land, Austria ("Forstau_B" to "Forstau T") are influenced by surface runoff water and thus likely affected by any global or regional contamination. This would not be expected for a cased well within the "Gasteiner Heilstollen", Bad Gastein, Austria (a tunnel drilled for therapeutic radon application), which shows exceptionally high uranium content ("Gastein 1", "Gastein 2A", "Gastein 2B").

Samples from a region less influenced by Chernobyl were obtained from a project connected with the ongoing decommissioning of the Garigliano nuclear power plant, southern Italy [28]. River sediment cores were drilled from the Garigliano river, which provided and drained the reactor cooling water. Samples collected upstream of the power plant ("Garigliano upstream average") serve not only as blanks for the Garigliano project, but can be understood as environmental samples exposed to possible global and regional anthropogenic contamination in that region.

3. Measurement method

The efficiency of the measurement methods at VERA [29] was studied with gravimetrically prepared U_3O_8 sputter targets (~1 mg U_3O_8 each). From the ion currents measured, an ionization yield of 6×10^{-4} and a stripping yield (including ion optical losses) of 5% was determined.

In contrast to reagents and ore samples, where several mg of uranium are readily available, only μg amounts

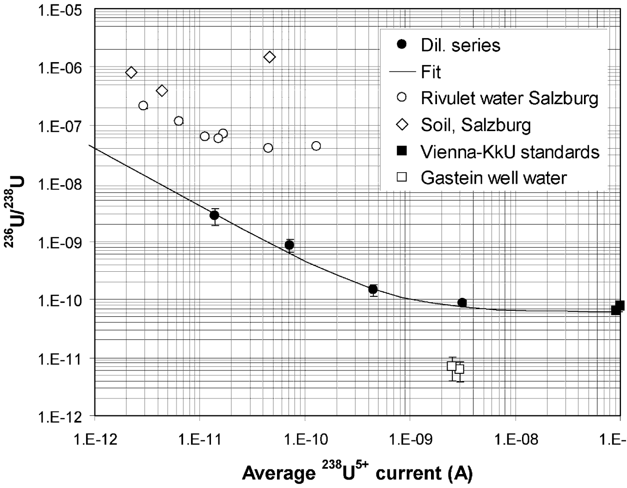

Fig. 1. Samples containing different amounts of uranium and their measured isotopic ratio.

can be extracted from a few grams of typical environmental samples. The performance of our chemical procedures (described in [28]) was studied with artificial samples. Quartz glass was ground in a mortar and different amounts of uranyl nitrate solution were added, to produce samples of several g total mass containing 0, 4, 40 and 400 µg of Vienna-KkU.

A close correspondence exists between the amount of uranium added and the $^{238}U^{5+}$ beam current (Fig. 1), probably because all samples shown were prepared with exactly the same procedure and measured in the same sample wheel. However, even for the sample without any U added, a current corresponding to ~1 µg U is observed, suggesting a laboratory background at this level. Whereas, the $^{236}U/U$ measured for the 400 µg sample agrees with the value determined from mg sized samples, smaller samples show increased ratios. The result is compatible with a chemistry blank of $\sim 2 \times 10^7$ atoms ^{236}U. Similar dilution series have been prepared for two other measurement series (see [28]). Their variation suggests that an uncertainty of ±100% must be assumed for the laboratory background.

The soil and river sediment samples were not completely digested, but only leached, since we were interested only in the anthropogenic influence. The total number of ^{236}U atoms per gram of dry sample mass is most suitable to quantify the anthropogenic contamination. A ^{232}U spike was added to trace the chemical yield by α spectrometry, but these results are not yet available. Fortunately, the good correlation found for the dilution series allows the U concentration and the ^{236}U content to be estimated from the measured $^{238}U^{5+}$ current and $^{236}U^{5+}$ count rate, respectively. We think that this estimate is correct within a factor of two.

4. Results and discussion

Our results for $^{236}U/U$ of ores and pre-nuclear reagents are in the range observed in previous measurements [5–7]. We think the measurements show the natural ^{236}U level.

For two of the three Salzburg soil samples (“T2G” and “T2H”) no $^{238}U^{5+}$ above the laboratory background is found. Both show a similar contamination with anthropogenic ^{236}U ($\sim 5 \times 10^8$ atoms/g soil, blank corrected). The third sample “T2F” shows a significantly higher $^{238}U^{5+}$ current, corresponding to ~2 µg U (blank corrected). Additionally, the sample exhibits a high isotopic ratio of $\sim 1.5 \times 10^{-6}$, corresponding to a ^{236}U concentration of about 2×10^{10} atoms/g soil. Perhaps this exceptionally high ^{236}U concentration is due to a fuel particle from the Chernobyl accident [30].

Detailed results for the Garigliano River sediment samples are shown in [28]. The value given in Table 1 is an average of 12 samples collected upstream of the power plant. The background corrected ^{236}U concentration measured is close to the detection limit.

Fig. 2 shows our soil and sediment results together with literature values of known contaminated sites [17–19]. The high value measured for the Salzburg soil sample “T2F” from 25 mm depth is even higher than that found for a soil 200 km from Chernobyl.

Also all Forstau rivulet water samples show a $^{236}U/U$ well above natural levels. The $^{238}U^{5+}$ currents are comparable with the process blank for five out of eight samples and for one sample (“Forstau L”) even too low for a reliable measurement. One of the other two samples shows a relatively high uranium content of ~10 µg/L. The samples form a consistent dataset with $^{236}U/U$ ratios of about 4×10^{-8} for the samples with the highest uranium content, sloping to higher values for smaller samples (Fig. 1).

A first result on the subsurface Gastein well measured at VERA was already published in [23]: $^{236}U/U = (6.5 \pm 2.1) \times 10^{-12}$ (“Gastein 1” in Table 1). With the new knowledge on laboratory and environmental contamination, this result seems surprisingly low. Thus, we have prepared two new samples from the same water (“Gastein 2A” and “Gastein 2B”). The result agrees well with the already published value. It is an order of magnitude below the typical value for high-grade ore. The two new measurements are included in Fig. 1, despite these measurements

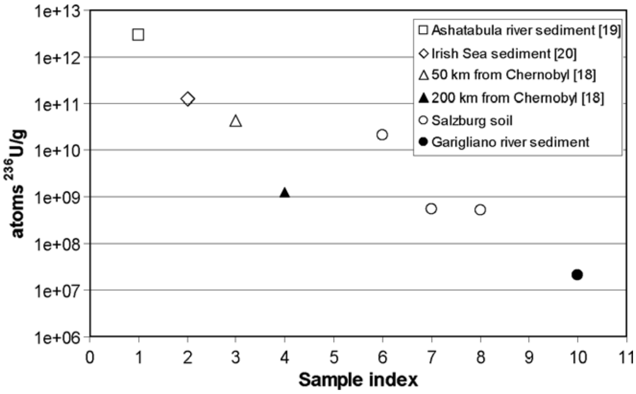

Fig. 2. Soil and sediment samples and comparison with some literature values.

were done in different run series and probably have a different calibration of U content vs. ion current. The observed $^{238}U^{5+}$ current is close to that measured for 400 μg samples from the dilution series.

5. Conclusions

10^6 kg of anthropogenic ^{236}U exists already on Earth. This amount is significantly larger than the relevant natural ^{236}U inventory of ~30 kg ^{236}U in the upper layers of land surface and the <0.5 kg in the oceans. Thus, it is not surprising that anthropogenic ^{236}U is present at levels detectable by AMS also at locations far from known contaminated sites. This ^{236}U contamination can also enter in the laboratory and limits the sensitivity in the present measurement to $\sim 2 \times 10^7$ atoms ^{236}U. River sediment from the Garigliano river, Italy was close to this detection limit. Soils and surface waters from Salzburg, Austria, were clearly affected by anthropogenic ^{236}U. Relatively small samples (several g of soil or 1.5 L of water) were sufficient to obtain significant results.

However, measurements of natural isotopic ratios are still possible. This is clear for reagents separated and stored before the era of nuclear human activities and for ore samples containing large amounts of uranium with relatively high natural isotopic ratios. However, also unaffected water samples exist, as a measurement on a subsurface well in Bad Gastein, Austria, demonstrates.

More work is required to get an overview on the distribution of both anthropogenic and natural ^{236}U in the environment. ^{236}U has potential as a tracer in hydrology and oceanography, it might allow exposure dating of very old surfaces and corresponding erosion rate studies and it might even be useful for uranium prospecting. How well these applications will work depends on whether it is possible to understand the anthropogenic and the natural contribution, which form the background for each other.

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