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Status of ^{236}U analyses at ETH Zurich and the distribution of ^{236}U and ^{129}I in the North Sea in 2009

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ABSTRACT

Compact, low energy accelerator mass spectrometry (AMS) has evolved over the past years as one of the most sensitive, selective, and robust techniques for the analysis of heavy and long lived radionuclides. In this study, we will first focus on the analytical capabilities of the compact AMS system TANDY, mainly for ^{236}U analyses, and then present a new dual tracer approach, that combines ^{129}I and ^{236}U. The measured ^{129}I/^{236}U ratios of samples collected in the North Sea in 2009 are in reasonable agreement with the expectations from documented or estimated releases from the two major nuclear reprocessing plants located at Sellafield (GB) and La Hague (F), suggesting that the ^{129}I/^{236}U ratio can be used as a water mass tag in the North Atlantic region. However, our results indicate that, in contrast to ^{129}I, additional contributions of bomb produced ^{236}U cannot be neglected in the North Sea region. This complicates the simple and straight forward use of the ^{129}I/^{236}U ratio as a quantitative tool for the calculation of transport times of North Sea water in the Arctic Ocean.

1. Introduction

^{236}U became accessible as a new anthropogenic tracer for oceanographic studies quite recently due to technical developments in AMS that significantly improved the sensitivity, the background suppression, and the sample throughput for ^{236}U/^{238}U analyses [1–4].

One aim of this study is to document the analytical capabilities of the compact, low energy 0.5 MV AMS system TANDY at ETH Zurich for routine analyses of ^{236}U/^{238}U. The current sensitivity for real measurement conditions and the machine background of the TANDY system will be presented and discussed.

In the second (application related) part of this study, we will present the first ^{129}I/^{236}U ratios measured in North Sea surface waters in 2009. The North Sea is of particular interest for oceanic tracer studies because it represents the source regions for anthropogenic nuclides released by the two major European nuclear reprocessing facilities located in Sellafield (GB) and La Hague (F). Soluble substances released into the ocean by these facilities enter the North Sea where they are mixed before being advected via the Norwegian Coastal Current (NCC) into the Arctic (e.g. [5]).

Previous studies have shown that anthropogenic radionuclides released into the North Sea region can be used as a water mass tag that allows identifying the waters on their way through the Arctic Ocean. For example, ^{129}I has extensively been used to identify Atlantic Ocean waters and to determine their transit times in the Arctic Ocean (e.g. [6,7]). The concentration of a single tracer, however, is progressively diluted while being transported. The ratio of two tracers, in contrast, would be conserved if both are dominated by one unique source. If, in addition, the tracer input functions are well known, this would allow a simple and precise estimation of water mass transit times that is independent of water mass mixing history.

Here, we want to emphasize the potential of ^{129}I/^{236}U as a new tool for the determination of transit times in the North Atlantic and Arctic region because it combines ideal geochemical properties of both constituents (e.g. conservative behavior in sea water) with the excellent analytical capabilities of accelerator mass spectrometry (AMS) to detect ultra-trace amounts of rare nuclides in comparable small volumes of sea water.

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2. AMS of ^{236}U at ETH Zurich

2.1. AMS setup and measurement

All ^{129}I and ^{236}U analyses were performed with the compact ETH Zurich system TANDY [8–10]. Details about the ^{129}I/^{127}I measurements (including AMS setup and performance parameters) are described by C. Vockenhuber et al. (this volume). The measured ^{129}I/^{127}I ratios were normalized to the ETH Zurich in house standard D22 with a nominal value of ^{129}I/^{127}I = (50.35 ± 0.16) × 10^{-12} [10]. The general setup for routine AMS of ^{236}U/^{238}U at ETH Zurich also was documented earlier [11]. The measured isotopic ratios were normalized to the ETH Zurich in house standard ZUTRI with a nominal value of (4055 ± 200) x 10^{-12} [10].

2.2. Sensitivity and background for AMS of ^{236}U at ETH Zurich

The background for AMS of ^{236}U/^{238}U at the ETH Zurich TANDY system has been thoroughly investigated [1,12]. As an add on to these studies, and after the first two years of routine operation with He stripping, the main operational parameters and characteristics of the compact ETH Zurich TANDY system for AMS of ^{236}U/^{238}U are summarized and discussed in the following.

2.2.1. Sensitivity for ^{236}U

For actinide measurements with the ETH Zurich TANDY system a terminal voltage of 300 kV is applied and the ions are detected in the 3+ charge state at energies of about 1.3 MeV [11]. The maximal transmission, i.e. the ratio of the $^{238}U^{3+}$ ion current on the high energy side relative to the injection current of negative $^{238}U^{16}O$ molecules, is close to 40% (red squares in Fig. 1) when using He-stripping [2]. However, due to the existence of triply charged UH-molecules [13] the measurements of ^{236}U/^{238}U have to be performed at a higher stripper gas density than needed for optimal transmission (shaded area in Fig. 1). At He-densities above 0.15 μg/cm^2 the molecular background is sufficiently suppressed while transmission decreases only to about 37%. At lower He-densities UH^{3+} molecules survive the stripping process, which is indicated by an exponential increase of the measured

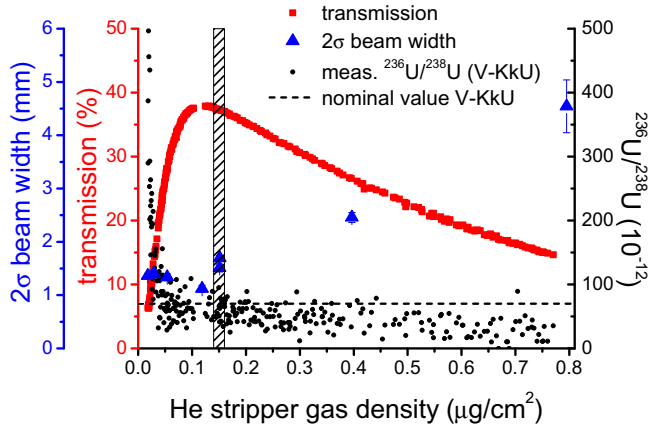

Fig. 1. Characteristic parameters for ^{236}U/^{238}U measurements at the ETH Zurich TANDY system, all plotted as a function of the He gas stripper density: red squares show the transmission through the accelerator and the He gas stripper ($^{238}U^{3+}$/$^{238}U^{16}O^-$), blue triangles represent the 2σ width of the $^{238}U^{3+}$ beam at the high energy side, and black dots represent the measured ^{236}U/^{238}U values of the V-KkU standard, while the black dashed line indicates its nominal value [4]. The shaded area indicates the conditions for routine measurements. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

^{236}U/^{238}U ratio of the Vienna-KkU standard (V-KkU, [4]), significantly above its nominal value (black dots and dashed line in Fig. 1). Scans of the $^{238}U^{3+}$ beam indicate that the 2σ-beam width is less than 2 mm at standard operating conditions (blue triangles in Fig. 1). Consistently, the ^{236}U/^{238}U ratio of the V-KkU is measured at about 90% of its nominal value. Both observations show that beam losses on the high energy side are minimal. Combining the above results, the ETH Zurich TANDY system has an over-all transmission of about 33% for ^{236}U from the low energy side into the detector.

To estimate the total detection efficiency (defined as detected ions vs. atoms in the sample) for ^{236}U, the efficiency of negative ion formation and extraction from the Cs sputter ion source also has to be considered. The negative UO ion yield from our modified NEC ion source [9] is currently at a level of 10^{-3} to 10^{-4} for standard measurement conditions (i.e. measurement times ca. 30–40 min). To reach higher yields the sample material has to be consumed more efficiently during the measurement process, which currently requires long sputtering times of several hours. Obviously, there is considerable potential for improvements (e.g. by optimizing sample size and/or sample matrix). With the current sample preparation setup (i.e. the U-oxide sample is dispersed in a matrix consisting of about 1 mg Fe-oxide and 2–3 mg Nb-powder) the total detection efficiency is at a level of 10^{-4} for standard measurement. During this time the samples are not consumed but are available for a complete remeasurement.

Summarizing the sensitivity of the ETH Zurich TANDY system for AMS of ^{236}U under real measurement conditions, we currently need at least 10^6 atoms of ^{236}U in a sample to determine the ^{236}U/^{238}U ratio with a precision of 10%. This implies that currently a minimum of 10 l of sea water is needed to determine a ^{236}U/^{238}U ratio of 1 × 10^{-11} within 10% (e.g. as found in the deep Equatorial Atlantic Ocean [14,15]). Contemporary waters in the North Sea region all carry ^{236}U/^{238}U ratios significantly larger than 1 × 10^{-9}. Therefore, a sampling volume of 2 l would be sufficient for an AMS measurement, even at the 3% precision level. The sample volumes available for this study were all between 10 and 20 l, significantly larger than the minimal requirements described above.

2.2.2. Background for ^{236}U

The main source of background for AMS of ^{236}U at the ETH Zurich TANDY system was identified by using a time-of-flight detector to originate from ^{235}U and from real mass 236 ions [1] which later were identified as $^{235}UH^{3+}$ molecules [12]. While molecular interferences can be suppressed sufficiently by increasing the stripper gas density (Fig. 1), ^{235}U remains as a severe source of background for ^{236}U that even tends to increase with higher stripper gas pressure because more gas leaking into the accelerator tubes. ^{235}U is injected into the accelerator together with the $^{236}U^{16}O$, either as a molecule of the same mass (e.g. $^{235}U^{17}O$ or $^{235}U^{1}H^{16}O$) or as a tail from the intense $^{235}U^{16}O$ beam present at the neighboring mass. After breakup of the injected molecules in the stripper $^{235}U^{3+}$ has a different energy than $^{236}U^{3+}$ and therefore should be suppressed. However, based on ion optical calculations, several possible scenarios involving charge exchange on residual gas atoms and/or scattering processes have been identified that would allow $^{235}U^{3+}$ to finally reach the gas ionization detector [1].

Since no completely ^{236}U-free U material is available it is not straightforward to simply measure the machine background for ^{236}U/^{238}U by analyzing a blank sample. The Vienna V-KkU-standard with a nominal ^{236}U/^{238}U ratio of (69.8 ± 3.2) × 10^{-12} [4] is among the materials with the lowest ^{236}U/^{238}U ratios available in significant amounts at ETH Zurich. The machine background of the TANDY AMS system, however, is significantly below this level [1].

As an alternative method, it has been suggested to determine the abundance sensitivity with ^{238}U (i.e. measuring the back-