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First ^{236}U data from the Arctic Ocean and use of ^{236}U/^{238}U and ^{129}I/^{236}U as a new dual tracer

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ABSTRACT

The first dataset of ^{236}U/^{238}U in the water column of the Arctic Ocean (AO) is presented and shows the widest range of ratios reported so far in the open ocean, from (5 ± 5) to $(3840 \pm 260) \times 10^{-12}$. Surface samples and depth profiles were collected during two GEOTRACES expeditions in 2011–2012 and analyzed for the concentrations of ^{236}U and ^{129}I, with the aim of investigating whether the combination of ^{236}U/^{238}U and ^{129}I/^{236}U can be used as a new oceanographic tool in the AO. Results show that the distributions of the ^{236}U/^{238}U and ^{129}I/^{236}U atomic ratios are consistent with the different water masses in the AO. High ^{236}U/^{238}U and ^{129}I/^{236}U ratios in the upper water column ($> 2000 \times 10^{-12}$ and >200, respectively) illustrate the penetration of Atlantic waters (AW) into the AO. Lower values were found in Pacific waters (PW) and deep waters of the AO. Rivers seem to represent a temporally and spatially-constrained third anthropogenic source of ^{236}U but more data are needed to confirm this. In a simple mixing model, the combination of ^{236}U/^{238}U and ^{129}I/^{236}U reveals a high contribution ($>99\%$) of natural background waters (pre-nuclear era) in the deep and bottom waters of the Amerasian basin, indicating an apparent water mass renewal time of >1000 years. Despite the relatively high apparent age of the Amerasian Basin deep waters, this work shows the potential of using the dual-tracer approach as a new oceanographic tool in the Arctic Ocean.

1. Introduction

In recent years, an increasing number of studies have measured the anthropogenic occurrence of ^{236}U ($T_{1/2} = 23$ Ma) in the ocean and indicated its potential as an oceanographic tracer (Christl et al., 2012; Eigl et al., 2013; Sakaguchi et al., 2012a; Steier et al., 2008). The general suitability of ^{236}U on a basin-wide scale has been demonstrated recently with the presentation of a first transect through the western North Atlantic Ocean (Casacuberta et al., 2014). The strengths of ^{236}U as an oceanographic tracer are: i) U is soluble (hence conservative) in seawater due to its occurrence as a stable carbonate ion complex, $UO_2(CO_3)_3^{4-}$ and there are currently no indications that ^{236}U and natural U behave differently in the ocean (Sakaguchi et al., 2012a; Rutgers van der Loeff and Geibert, 2008; Langmuir, 1978); ii) contrary to the naturally-occurring U-isotopes, anthropogenic ^{236}U has not yet reached steady state in the oceans, thus providing a highly dynamic and transient signal penetrating the deep ocean (Christl et al., 2012); and iii) due to the incorporation and accumulation of U in aragonite, past ^{236}U levels can be determined at high-resolution in geological archives such as corals (Winkler et al., 2012).

On a global scale, the largest source of ^{236}U is considered to be atmospheric nuclear weapons tests, conducted in the 1950s and 1960s, that released around 1000 kg (Sakaguchi et al., 2009). Smaller but still significant amounts of ^{236}U (95 ± 32 kg) have also been discharged locally into the North Atlantic region by the European nuclear reprocessing facilities of Sellafield, UK and La Hague, France (Christl et al., 2015b). Other potential sources of ^{236}U such

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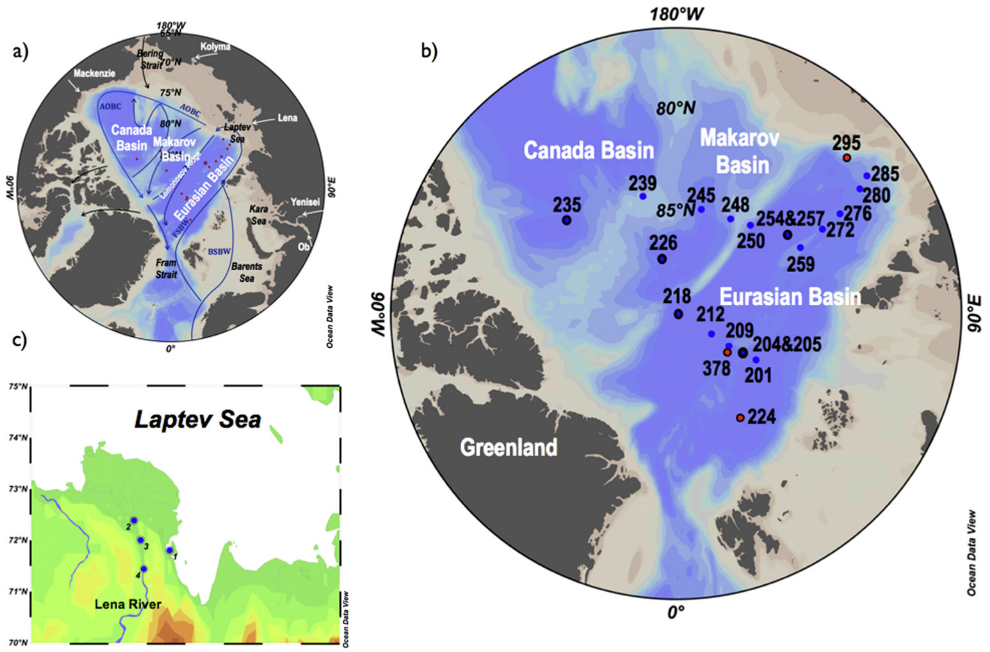

Fig. 1. a) Map of the Arctic Ocean showing the sampling locations and the general pattern of the surface circulation; black arrows represent Pacific Waters and white arrows are indicative of major rivers discharging into the Arctic Ocean; b) Detailed map of the Arctic Ocean to indicate locations of samples more clearly. Samples taken during *RV Polarstern* expeditions ARK XXVI/3 are indicated in blue and samples from ARK XXVII/3 are indicated in red. Deep profiles are indicated with black circles; c) Location map for the Lena-Delta region showing the sampling sites for the three samples taken at Lena river and one coastal sample. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

as discharges from the Russian or former Soviet Union nuclear facilities to Siberian rivers, or contributions from nuclear accidents such as Chernobyl (Boulyga and Heumann, 2006) or Fukushima (Sakaguchi et al., 2014), are thought to be of minor relevance or are not yet documented. Although input functions are currently not well constrained, major efforts have recently been devoted to constrain the ^{236}U input function from European reprocessing plants, in order to use this radionuclide to track Atlantic waters into the Arctic ocean (Christl et al., 2015b).

In contrast to ^{236}U, the major oceanic source of ^{129}I is not fallout but the liquid discharges from Sellafield and La Hague ($>$6000 kg since 1970s) (He et al., 2013), and only about 50 kg of ^{129}I were released into the atmosphere (and subsequently deposited to the surface of the ocean) as a result of the nuclear weapon tests (Raisbeck and Yiou, 1999). Thus, the combination of ^{236}U with the well-known ^{129}I pointed to the use of ^{129}I/^{236}U as an additional tool in oceanography to constrain water mass sources and transit times. The strengths of combining these two anthropogenic radionuclides are: i) they are both considered to behave conservatively in the open ocean, so variation in their ratios should be water mass dependent (Sakaguchi et al., 2012a; Aldahan et al., 2007) and, ii) their different sources and input functions result in very variable ^{129}I/^{236}U atomic ratios in time and between sources. Small ^{129}I/^{236}U atomic ratio ($<$1 atom atom^{-1}) can be estimated for global fallout and ratios between 250 and 450 would be representative of the European reprocessing plant releases in the 15 years prior to this study (i.e. from 1998 to 2012) (Christl et al., 2015b). Apart from their anthropogenic sources, small amounts of natural ^{236}U and ^{129}I are present on Earth but their inventories are minor compared to the anthropogenic releases and can be considered to be in steady state with respect to exchange between the different compartments of the global environment. The fractions of naturally occurring ^{129}I and ^{236}U in ocean water are referred to as "natural background" in this study, with a corresponding natural ^{129}I/^{236}U ratio that would range between 150 and $\geq$450 (Steier et al., 2008; Raisbeck and Yiou, 1999), but mainly characterized by very low ^{236}U and ^{129}I concentrations (further details in the discussion).

In this study, the first measurements of ^{236}U together with ^{129}I concentrations were made in seawater samples taken in the Arctic Ocean during two GEOTRACES expeditions (ARK XXVI/3 and ARK XXVII/3). The aim is to determine the ^{129}I/^{236}U and ^{236}U/^{238}U atomic ratios of samples representing different water masses in the Arctic Ocean, and to discuss the implications of using the combination of these ratios as a new tool in oceanography.

2. Materials and methods

2.1. Samples

Eight depth profiles and 15 surface seawater samples (Fig. 1, Table S1) were collected in 2011 and 2012 during two *RV Polarstern* expeditions into the Arctic Ocean (ARK XXVI/3 and ARK XXVII/3). Water samples were obtained with 12-L Niskin bottles mounted on a rosette equipped with standard conductivity, temperature and pressure (CTD) sensors (Schauer, 2012; Boetius, 2013). Between 1 and 10 L samples were collected for ^{236}U and ^{129}I-isotope analyses. The unfiltered samples were stored in pre-cleaned (and rinsed with seawater) Nalgene plastic bottles or cubitainers for further processing and analysis.

In August 2014, 4 samples of 1 L each were taken at the mouth of the Lena River, near Tiksi, and upstream of the Stolb Island (Fig. 1c). The salinity of these samples was not determined but was probably very low because samples were collected upstream of the normal tidal range of the river.

Surface waters were also analyzed for δ^{18}O. The combination of salinity with oxygen isotopes allows for the determination of the fractions of meteoric water, sea–ice meltwater or brine influence in addition to marine water (Bauch et al., 2011). In this work, the δ^{18}O data have been used to identify the river fraction in surface waters, indicated in Table S1.

2.2. Chemical separation of uranium and AMS measurement

Between 1 and 10 L were used for the measurement of ^{236}U (see Table S1 for details). Samples were weighed, acidified with