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Natural and artificial radionuclides in a marine core. First results of ^{236}U in North Atlantic Ocean sediments

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ABSTRACT

There are very few data available of ^{236}U in marine sediment cores. In this study we present the results from the first oceanic depth profile of ^{236}U in a sediment core sampled in the North Atlantic Ocean, at the PAP site (4500 m depth, Porcupine Abyssal Plain (PAP) site, 49°0′ N, 16°30′ W). Additionally, the sediment core was radiologically characterized through the measurement of anthropogenic ^{137}Cs, ^{239}Pu, ^{240}Pu, ^{129}I and ^{14}C and natural ^{210}Pb, ^{40}K and ^{226}Ra.

The measured ^{236}U concentrations decrease from about $90{\cdot}10^{6}$ at g^{-1} at the seafloor down to $0.5{\cdot}10^{6}$ at g^{-1} at 6 cm depth. They are several orders of magnitude lower than the reported values for soils from the Northern Hemisphere solely influenced by global fallout (i.e. from $2700{\cdot}10^{6}$ to $7500{\cdot}10^{6}$ at g^{-1}). ^{236}U/^{238}U atom ratios measured are at least three orders of magnitude above the estimated level for the naturally occurring dissolved uranium. The obtained inventories are $1{\cdot}10^{12}$ at m^{-2} for ^{236}U, 80 Bq m^{-2} for ^{137}Cs, 45 Bq m^{-2} for $^{239+240}$Pu and $2.6{\cdot}10^{12}$ at m^{-2} for ^{129}I. Atomic ratios for ^{236}U/^{239}Pu, ^{137}Cs/^{236}U and ^{129}I/^{236}U, obtained from the inventories are 0.036, 0.11 and 2.5 respectively. Concentration profiles show mobilization probably due to bioturbation from the abundant detritivore holothurian species living at the PAP site sea-floor. The range of ^{236}U, ^{137}Cs, $^{239+240}$Pu and ^{129}I values, inventories and ratios of these anthropogenic radionuclides are more similar to the values due to fall-out than values from a contribution from the Nuclear Fuel Reprocessing Plants dispersed to the south-west of the North Atlantic Ocean. However, signs of an additional source are detected and might be associated to the nuclear wastes dumped on the Eastern North Atlantic Ocean.

1. Introduction

Uranium-236 is a long-lived radioisotope ($T_{1/2}$ = $2.35{\cdot}10^{7}$ yr) (Flynn et al., 1972) that decays by alpha emission to ^{232}Th and is an almost entirely anthropogenic isotope. It is produced in nuclear reactors through the capture of a thermal neutron by ^{235}U or ^{239}Pu (followed by alpha decay of ^{240}Pu), or in nuclear weapon tests from the ^{238}U(n, 3n)^{236}U reaction induced by fast neutrons. It is also naturally produced in inferior amounts by galactic cosmic ray induced secondary neutrons and by neutrons originating from (α, n) reactions in the natural terrestrial environment. These processes lead to natural ^{236}U/^{238}U atom ratios that range from 10^{-14} (i.e. estimated for typical rocks) to 10^{-9} (i.e. maximum value measured in uranium ores) (Steier et al., 2008). However, the amount of ^{236}U in the environment mainly has an anthropogenic origin, leading to isotopic ratios between 10^{-9} and 10^{-4} in the environment (Steier et al., 2008; Wendel et al., 2013; Winkler et al., 2012 and references therein).

A total amount of 35 kg of mobile ^{236}U coming from natural sources has been estimated (Christl et al., 2012; Steier et al., 2008 and references therein). On the other hand, (i) between 900 and 1400 kg of ^{236}U were released globally during the period of atmospheric nuclear weapons testing (Sakaguchi et al., 2009; Winkler et al., 2012), (ii) inventories of 95 ± 32 kg have been recently assessed for the liquid effluents coming from the nuclear fuel reprocessing plants (NFRP) in Britain (i.e. Sellafield and Springfields), and in France (i.e. La Hague) (Christl et al., 2015), and (iii) about 100 kg of ^{236}U were dispersed over eastern Europe as a consequence of the Chernobyl accident (Castrillejo et al., 2017). The Sellafield and La Hague effluents dominate the concentrations of ^{236}U in the North Atlantic and Arctic Oceans (Casacuberta et al., 2014, 2016).

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Up to now, due to the difficulties in the measurement of low $^{236}U/^{238}U$ ratios, the availability of ^{236}U measurements in the environment was limited. After the improvement of the analytical detection techniques, AMS (Marsden et al., 2001), ICP-MS (Ketterer et al., 2003) or TIMS (Richter et al., 2010), ^{236}U concentrations data in several environmental compartments have become available in recent years. Furthermore, recent studies demonstrate the potential use of ^{236}U as an oceanic circulation (Christl et al., 2013; Sakaguchi et al., 2012), coral (Nomura et al., 2017; Winkler et al., 2012) and ice core (Wendel et al., 2013) tracer.

However, the biogeochemical characteristics of anthropogenic ^{236}U in seawater and particles are still not well constrained (Sakaguchi et al., 2012), e.g. its main oxidation state in the ocean is undetermined and also how this affects effectively to its scavenging by particles. Furthermore, preliminary results point out to a possible speciation between the dissolved and the particulate phases in seawater, which favours the ^{236}U concentration in the sediment (Eigl et al., 2017; Sakaguchi et al., 2012). Finally, it is necessary to better constrain the anthropogenic ^{236}U released, especially from the NFRP, and more particularly of Sellafield, and its pathways through the Atlantic and Arctic oceans. Recently, it has been modelled ^{236}U inputs from the NFRP, but further studies will be needed in order to increase the accuracy of the estimations (Christl et al., 2015). This is of primary importance in order to use $^{236}U/^{238}U$, and also $^{129}I/^{236}U$, as a sensitive tool for the calculation of tracer ages and ventilation rates in the North Atlantic domain (Casacuberta et al., 2016).

Pu as opposed to U is considered almost exclusively anthropogenic. Plutonium has a very different geochemical behaviour than uranium, it is complex because different valences plutonium isotopes coexist and constitute inorganic and organic ligands. In seawater it is a particle-reactive element that is exported in the water column with the sinking particles, and is re-mineralized in deep waters or stored in the sea-bed sediment (Chamizo et al., 2015). ^{210}Pb is a particle reactive tracer of natural origin widely used for core dating purposes (San Miguel et al., 2003) and evaluation of particle export fluxes (Le Moigne et al., 2013). ^{137}Cs and ^{129}I behave in seawater as a conservative tracer, with its distribution governed by physical transport ocean currents processes (Periáñez et al., 2016; Villa-Alfageme et al., 2015). Natural uranium is conservative in water (Owens et al., 2011) and the same behaviour has been predicted for ^{236}U (Casacuberta et al., 2014). However, ^{236}U adsorption properties in marine sinking particles and its mobility in the sediments is not well constrained. The four anthropogenic radionuclides have a contribution in seawater from fallout and they have been regularly released from NFRPs such as Sellafield and la Hague (Chamizo et al., 2015; Periáñez et al., 2016; Villa-Alfageme et al., 2015).

Therefore, there are several challenges that still must be solved when using anthropogenic ^{236}U as a tracer: i) to define its biogeochemical behaviour, ii) furthermore, it is necessary to better constrain the anthropogenic ^{236}U sources and specially quantify the NFRP annual releases and its pathways through the Atlantic and Arctic oceans. We investigated these topics, through the analysis of a marine sediment core collected in the North Atlantic at the Porcupine Abyssal Plain (PAP) site.

The analysis of a combined set of radionuclides with different sources (i.e. anthropogenic and naturally occurring) and with different behaviours in seawater (i.e.particle reactive, and conservative) is crucial to investigate on the biochemistry of anthropopgenic ^{236}U.

A full analysis of the artificial and natural radionuclides concentrations in the core was carried out additionally to ^{236}U. ^{129}I ($T_{1/2} = 1.57 \times 10^7$ yr) (Raisbeck and Yiou, 1999), ^{239}Pu ($T_{1/2} = 2.41 \times 10^4$ yr) (Prindle et al., 1978), ^{240}Pu ($T_{1/2} = 6.5 \times 10^3$ yr) (Ahmad et al., 2007), ^{14}C ($T_{1/2} = 5.73 \times 10^3$ yr) (Pettitt et al., 2003) were measured through AMS and ^{137}Cs ($T_{1/2} = 30.17$ yr) (Juget et al., 2016), ^{40}K ($T_{1/2} = 1.248 \times 10^9$ yr) (Mougeot, 2017), ^{210}Pb ($T_{1/2} = 22.3$ yr) (Villa et al., 2003)and ^{226}Ra ($T_{1/2} = 1.6 \times 10^3$ yr) (Villa et al., 2005) were measured by gamma spectrometry.

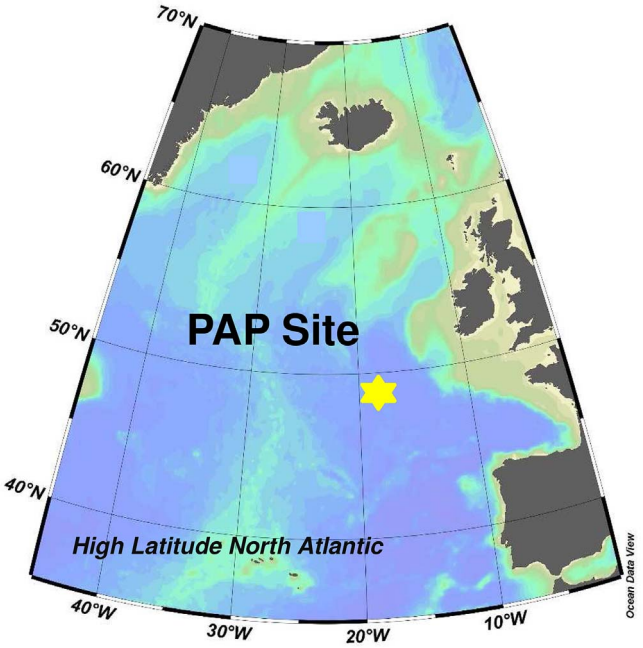

Fig. 1. Map of the sampling site. Porcupine Abyssal plain (PAP) site. Located in the North Atlantic Ocean.

Our results combine a very broad dataset of radionuclides that will be used to i) provide radioecological information of artificial and natural radionuclides in a North Atlantic sediment i) provide a first approach to the transport pathways and biogeochemical behaviour of ^{236}U in the environment and iii) analyse the different sources of ^{236}U to this marine sediment.

2. Sampling and methods

2.1. Sample collection and site description

The sediment core was collected at the PAP site (Fig. 1), at 48° 48.2′N and 16° 27.3′W and 4934 m depth, during the cruise D341 onboard R.R.S *Discovery* in July 2009 (Le Moigne et al., 2013). The samples were obtained using a multicorer and were sectioned into 0.5- or 1-cm intervals. Samples were stored frozen and returned to the shore-based laboratory for subsequent analysis. The Porcupine Abyssal Plain (49° N and 16° 30′ W), PAP site, is located on the boundary between the subpolar and subtropical gyres of the North Atlantic (Villa-Alfageme et al., 2014), adjacent to the Irish continental margin.

Porcupine Abyssal Plain megabenthos includes a large number of marine invertebrates. These animals are increasing in number as more phytodetritus falls to the seabed (Kalogeropoulou et al., 2010). Since 1996, an important change has taken place in the numbers of creatures living over a large area of the North Eastern Atlantic sea floor. Sea anemones, brittle stars, polychaete worms, sea spiders and particularly sea cucumbers on the deep sea floor have all increased significantly in numbers (Billett et al., 2001).

2.2. Actinides measurement

2.2.1. Reagents

For ^{236}U AMS determinations, it is very important to minimize the ^{238}U and ^{236}U contaminations introduced by reagents, spikes and laboratory materials. Therefore, the reagents used during the sample preparation were of the highest purity grade and glassware was avoided when possible. To prepare the AMS cathodes, an ultra-pure iron solution supplied by High Purity Standards (HPS, England) was used.