

# First results of Uranium-236 in the South Atlantic Ocean

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## Abstract

In the last years,  $^{236}\text{U}$  ( $T_{1/2}=2.34\cdot 10^7$  y), which is essentially an anthropogenic radionuclide, has been established as a new oceanographic tracer thanks to its conservative behaviour in seawater. At the Centro Nacional de Aceleradores (CNA), in Seville (Spain), it has been recently demonstrated that  $^{236}\text{U}$  can be determined at environmental levels on the 1 MV Accelerator Mass Spectrometry (AMS) system. In the framework of the existing collaboration between the National Marine Information and Research Centre (NatMIRC), in Namibia, and the IAEA Environment Laboratories in Monaco,  $^{236}\text{U}$  has been analysed by AMS at CNA in a set of seawater samples collected in the northern Benguela upwelling system, on the Namibian coast. In surface samples,  $^{236}\text{U}/^{238}\text{U}$  atom ratios at the level of  $10^{-10}$  have been obtained, in agreement with the expected values for the so-called global fallout. The  $^{236}\text{U}$  inventories reported up to now in seawater are discussed.

## Introduction

The naturally occurring isotopes of uranium are the alpha emitters  $^{238}\text{U}$  ( $T_{1/2}=4.51\cdot 10^9$  y),  $^{235}\text{U}$  ( $T_{1/2}=7.038\cdot 10^8$  y) and  $^{234}\text{U}$  ( $T_{1/2}=2.455\cdot 10^5$  y), and their atomic abundances are, respectively, 99.2742%, 0.7204% and 0.0054%. These isotopes have been extensively measured in seawater for oceanographic studies because of their conservative behaviour in the seawater. Typical  $^{238}\text{U}$  concentrations in the oceans are approximately 3.5 ppb (3.5  $\mu\text{g/L}$  or 3.5  $\text{mBq/L}$ ), so they can be easily assessed by conventional radiometric and/or mass spectrometry (MS) techniques (Boulyga and Heumann 2006). In contrast,  $^{236}\text{U}$  ( $T_{1/2}=2.342\cdot 10^7$  y) has mainly an anthropogenic origin. It has been estimated that about  $10^6$  kg of  $^{236}\text{U}$  has been produced by men since the onset of the nuclear age in the 1940's (i.e. in atomic bombs mainly via the reaction  $^{238}\text{U}(n,3n)^{236}\text{U}$  induced by fast neutrons; in nuclear reactors via the reaction  $^{235}\text{U}(n,\gamma)^{236}\text{U}$  induced by thermal neutrons; and in minor amounts from the alpha-decay of its parent,  $^{240}\text{Pu}$  ( $T_{1/2}=6524$  y)). On the other hand, it has been estimated that about 35 kg of  $^{236}\text{U}$  have a natural origin (i.e. produced by the neutron activation of the natural  $^{235}\text{U}$  by cosmogenic and nucleogenic neutrons) (Christl et al. 2012). In recent years, several studies have been focused on the determination of  $^{236}\text{U}$  in seawater (Casacuberta et al. 2014, Casacuberta et al. 2016, Chamizo et al. 2015b, Christl et al. 2012, Christl et al. 2013, Sakaguchi et al. 2012).  $^{236}\text{U}$  is a powerful oceanographic tracer because of its long half-life and the fact that, unlike natural uranium isotopes, it has not reached steady-state in the oceans because it has been released to the general environment in the last 70 years (Sakaguchi et al. 2012, Christl et al. 2012). However,  $^{236}\text{U}$  concentrations in the environment, and specifically in the oceans, are extremely small. For instance, surface waters from the Northern Hemisphere are strongly influenced by the so-called *fallout* (i.e. atmospheric debris produced in the nuclear explosions). However, their activity concentrations, which are at the  $\text{fg/L}$  or  $\text{nBq/L}$  level, cannot be assessed by radiometric counting techniques (Casacuberta et al. 2014, Sakaguchi et al. 2012). The

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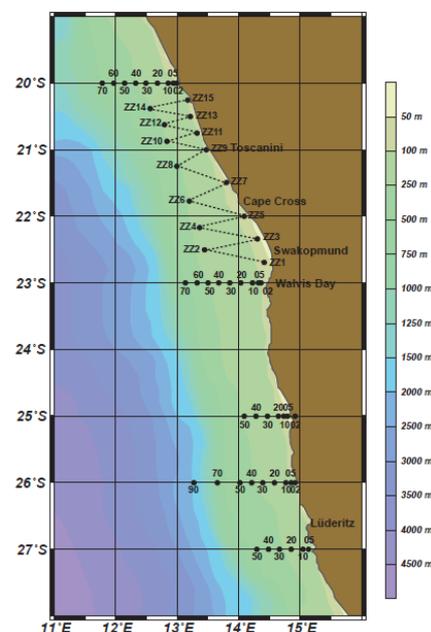
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study of  $^{236}\text{U}$  in environmental samples has recently become possible as a result of the high sensitivity reached by modern MS systems. When  $^{236}\text{U}$  enters the environment, it is mixed with the existing  $^{238}\text{U}$  and a wide range of  $^{236}\text{U}/^{238}\text{U}$  atomic ratios arises. For instance, in surface seawater from the Northern Hemisphere only influenced by the so-called *global fallout* (i.e. particles injected into the stratosphere during the high-yield nuclear atmospheric tests), atomic ratios at the  $10^{-9}$  level have been determined (Casacuberta et al. 2014, Sakaguchi et al. 2012), and in the Equatorial Pacific Ocean, ratios at the  $10^{-10}$  level have been determined in surface seawater and at  $10^{-12}$  level in deep waters (Chamizo et al. 2015b). The main limitation for the determination of  $^{236}\text{U}$  by MS techniques is the presence of scattering events and/or molecular isobars produced by the abundant isotopes,  $^{235}\text{U}$  and  $^{238}\text{U}$ , which cannot be chemically suppressed. MS techniques such as TIMS (*Thermal Ionization Mass Spectrometry*) and HR-ICP-MS (*High Resolution Inductively Couple Plasma Mass Spectrometry*) can only measure ratios down to the  $10^{-10}$  level (Lee et al. 2015, Roos 2008). Accelerator Mass Spectrometry (AMS) features with the lowest  $^{236}\text{U}/^{238}\text{U}$  background atomic ratios mainly thanks to the destruction of the molecular isobars in the so-called stripping process, which occurs in the terminal of an electrostatic tandem accelerator. In the last years, compact AMS systems, working at accelerator terminal voltages of 1 MV and below, have demonstrated their potential to measure  $^{236}\text{U}$  at environmental levels (Christl et al. 2015, Chamizo et al. 2015). However, because of the recent development of these high sensitive AMS techniques, there is still very few information about  $^{236}\text{U}$  in the environment and, furthermore, these data are especially rare in the South Hemisphere. In this context, the first  $^{236}\text{U}$  results in the South Atlantic Ocean have been obtained on the 1MV AMS system at the Centro Nacional de Aceleradores (CNA), in Seville, Spain. The studied area was the coast of Namibia, in the south eastern Atlantic Ocean, where the marine environment is strongly influenced by the northwards flowing, cold Benguela upwelling current (Sakko 1998).

## Materials and Methods

### Samples

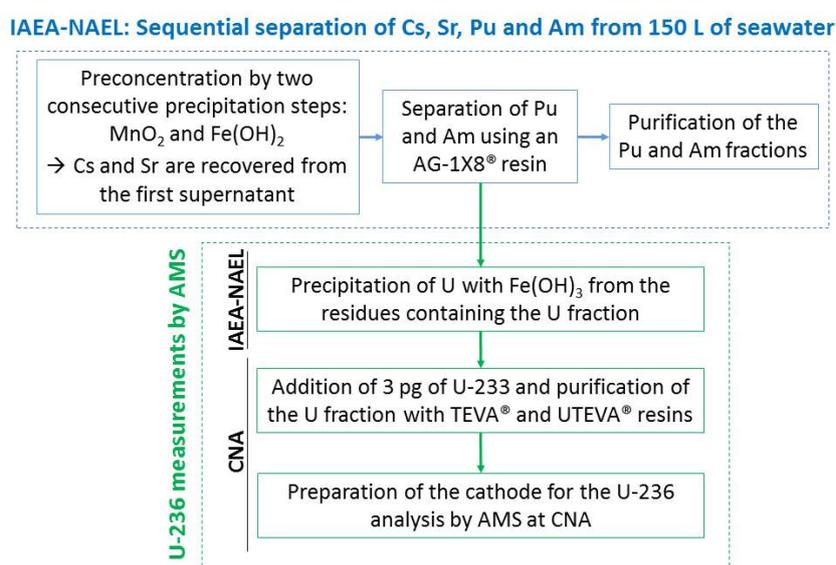
Sampling was done along the Namibian coast, in the framework of the existing collaboration between the National Marine Information and Research Centre (NatMIRC), in Namibia, and the IAEA Environment Laboratories (IAEA-NAEL) in Monaco. The aim of the project was to establish a baseline study of marine radioactivity and offshore trace elements levels in this area (Fig. 1). Surface seawater samples (150 L) were collected for the determination of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$  and Pu isotopes and processed in the IAEA-NAEL following a sequential method for the simultaneous extraction of these elements (Lee et al. 2003, La Rosa et al. 2001, Lee et al. 2001, Povinec et al. 2001). In order to analyse  $^{236}\text{U}$  in these samples, the residues produced throughout that procedure containing most of the uranium were further processed at CNA, following a similar strategy than the one described in (Chamizo et al. 2016).



**Figure 1.** Map of the sampling location of the seawater samples collected by the IAEA-NAEL and the NatMIRC in the Namibia's coast, South Atlantic Ocean.

## Radiochemical procedure

The radiochemical method performed in a first instance at IAEA-NAEL is mainly constituted in three steps (Fig. 2). First, actinides are preconcentrated by two sequential  $\text{MnO}_2$  and  $\text{Fe}(\text{OH})_2$  precipitations and separated from the Sr and Cs which are recovered from the first supernatant. Using an AG1-X8<sup>®</sup> resin, a first purification is performed and Pu and Am are separated. Finally, Pu and Am fractions are additionally purified. Following this method, U is not retained in the AG1-X8<sup>®</sup> resin, so the first eluates and the corresponding washings were combined and further processed. U was preconcentrated from these fractions by  $\text{Fe}(\text{OH})_3$ -co-precipitation, which, once dried, it was brought to the CNA for its final purification. The samples were dissolved in 8M  $\text{HNO}_3$ , spiked with 3 pg of  $^{233}\text{U}$  and loaded into TEVA resins, in order to remove the Pu traces. Finally, the U fractions were purified using UTEVA<sup>®</sup> resins. AMS cathodes were prepared by adding 1 mg of Fe(III) to the uranium solutions for their co-precipitation. Precipitates were transferred to quartz crucibles, dried and oxidized at 650°C. Finally samples were mixed with 3 mg of Nb powder and pressed into appropriated aluminum cathodes (Chamizo et al. 2015).



**Figure 2.** Radiochemical method followed in this work for the recovery of the uranium fraction from the residues obtained from the initial processing at IAEA-NAEL of 150L seawater samples. The first steps of the procedure were carried out at the IAEA-NAEL and the final uranium purification at the CNA.

## Reagents and spikes

$^{238}\text{U}$  is an abundant natural radionuclide so it is important to minimize its presence in the samples.  $^{238}\text{U}$  traces in reagents and laboratory equipment would underestimate the measured  $^{236}\text{U}/^{238}\text{U}$  atom ratios, which is the final result of an AMS measurement. To minimize its contamination, glassware was avoided when possible and, in the different  $\text{Fe}(\text{OH})_3$  co-precipitation steps, an  $\text{Fe}^{3+}$  solution provided by *High Purity Standards* (HPS, England), whose certified  $^{238}\text{U}$  concentration is at the <0.5 ppb level, was used. Furthermore, in the purification of the uranium fraction at CNA, *Suprapur* grade reagents were used.

Ion chromatography extraction was performed in a vacuum box, and TEVA<sup>®</sup> and UTEVA<sup>®</sup> resins (Eichrom Industries, Inc.) in 2 mL cartridges with 100-150  $\mu\text{m}$  particle size material were used.

$^{233}\text{U}$  spike was provided by the Laboratory of Ion Beam Physics, ETH Zürich (IRMM-051).

## AMS measurements

AMS determinations were performed on the 1 MV AMS system at the Centro Nacional de Aceleradores (CNA, Seville, Spain). The current status of the facility is discussed in (Chamizo et al. 2015b) and (Scognamiglio et al. 2016). Briefly, uranium isotopes are extracted from the  $\text{Cs}^+$  sputtering ion-source as negative oxide ions ( $\text{UO}^-$ ). These anions are analysed by a first  $90^\circ$  sector magnet in the low-energy side and, then, they are accelerated along the first section of a tandem accelerator which contains the stripper gas (He) in its terminal. The molecular ions are broken up, stripped to positive charge states and the atomic breakup products undergo a second acceleration along the second acceleration tube. On the high-energy side, ions are selected first by a  $90^\circ$  sector magnet and later by a  $120^\circ$  electrostatic deflector.  $^{238}\text{U}$ , an abundant isotope, is analysed by measuring the  $^{238}\text{U}^{3+}$  beam-current in an offset Faraday cup. The minor isotopes,  $^{236}\text{U}$  and the tracer  $^{233}\text{U}$ , are detected in a gas ionization chamber. (Chamizo et al. 2015a)

The final AMS results are the atom ratios. The main limitation to determinate the  $^{236}\text{U}/^{238}\text{U}$  ratio, which characterizes the sample of interest, is the presence of  $^{235}\text{U}$ . This abundant natural isotope may interfere to  $^{236}\text{U}$  analysis, due to their very similar trajectories in the different cinematic filters and to the formation of molecular isobars. The corresponding background (i.e. spurious  $^{236}\text{U}$  counts coming from the  $^{235}\text{U}$  in the sample) was subtracted from our results using the ratio between the counts on mass 239 and  $^{238}\text{U}$  as a proxy, assuming no real  $^{239}\text{Pu}$  is present in the sample (Chamizo et al., 2015). At CNA, the current  $^{236}\text{U}/^{238}\text{U}$  background ratio is  $9 \cdot 10^{-11}$  (Scognamiglio et al., 2016). The  $^{233}\text{U}/^{238}\text{U}$  and  $^{236}\text{U}/^{233}\text{U}$  ratios can be used to quantify the  $^{236}\text{U}$  and  $^{238}\text{U}$  concentrations by using the so-called isotope-dilution method. However, for the samples studied in this work,  $^{233}\text{U}$  was added in an intermediate step of the procedure as explained before (Fig. 2). In order to obtain the  $^{236}\text{U}$  concentrations in the original sample, the radiochemical yield of the steps carried out at the IAEA-NAEL (i.e. before the addition of the  $^{233}\text{U}$  spike) was estimated from the expected  $^{238}\text{U}$  concentration in the original sample, which can be extrapolated using the empirical formulas based on the salinity data (Owens et al. 2011, Pates and Muir 2007).

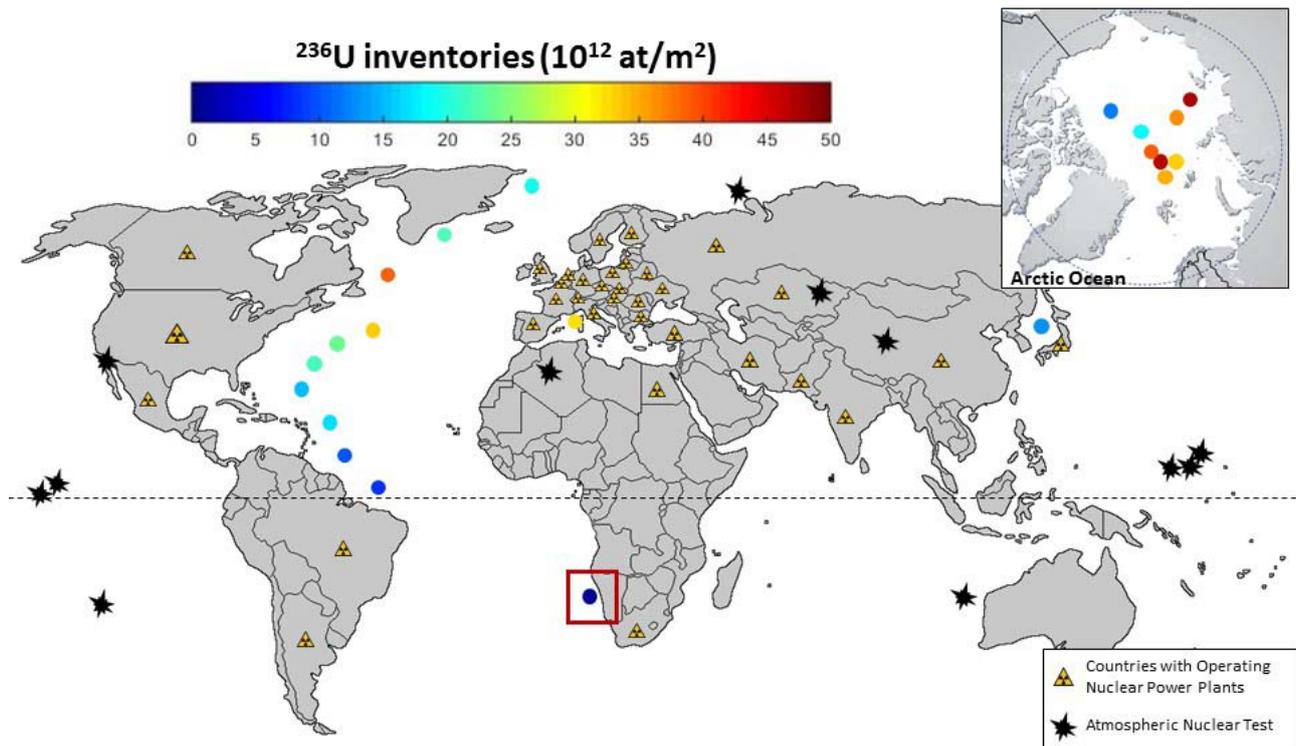
## Results and Discussion

The measured  $^{236}\text{U}/^{238}\text{U}$  atomic ratios are at the level of  $10^{-10}$  in all the surface samples. Regarding to the  $^{236}\text{U}$  concentrations, the results are at the level of the nBq/L or  $10^6$  atoms/L. The average  $^{236}\text{U}/^{238}\text{U}$  atomic ratio and  $^{236}\text{U}$  concentration obtained from the samples located in the  $20\text{-}24^\circ\text{S}$  latitudinal band was  $6.8 \cdot 10^{-10}$ , and 5.3 nBq/L, respectively, with a standard deviation of 30%. From samples located in the  $24\text{-}28^\circ\text{S}$  band, the average  $^{236}\text{U}/^{238}\text{U}$  ratio was  $6.1 \cdot 10^{-10}$  and the average  $^{236}\text{U}$  concentration 4.7 nBq/L, with 12% standard deviation. Therefore, the average results are very similar in both cases but the obtained results are more homogenous in the band  $24\text{-}28^\circ\text{S}$  than in the  $20\text{-}24^\circ\text{S}$ . These values are considerably lower than the reported ones in surface samples from the North Atlantic Ocean, the Mediterranean Sea and the Japan Sea, which are at the level of  $10^{-9}$  and 10 nBq/L for  $^{236}\text{U}/^{238}\text{U}$  atomic ratios and  $^{236}\text{U}$  concentrations, respectively (Casacuberta et al. 2014, Chamizo et al. 2016, Sakaguchi et al. 2012).

The  $^{236}\text{U}$  inventory was estimated from a depth profile studied in this area. In Fig. 3 a compilation of the different inventories published up to now in the oceans together with the one obtained from Namibian Coast is displayed. Due to the recent development of the analytical methods, there is only a few available results of the  $^{236}\text{U}$  inventories in the oceans all around the world: Arctic Ocean (Casacuberta et al. 2016), North Atlantic Ocean (Casacuberta et al. 2014), Greenland Sea (López-Lora, 2014), Japan Sea (Sakaguchi et al. 2012) and Mediterranean Sea (Chamizo et al. 2016). Furthermore, the countries with operating Nuclear Power Plants (NPP) and the different places where the Atmospheric Nuclear Test were carried out are marked in the map (Carter & Moghissi, 1977).

The main source of anthropogenic radionuclides in the environment and, most specifically, in the oceans, is the so-called *global fallout*. In the case of  $^{236}\text{U}$ , this source has not been well defined yet because of the few available results in the different environmental compartments. Atmospheric nuclear tests were carried out mainly in the North Hemisphere (Fig. 3), the deposition of radionuclides was higher in that hemisphere, about 3.8 times the one in the South Hemisphere (Hardy et al. 1973). Furthermore, the atmospheric deposition depends strongly on the latitude: if we divide the hemisphere in three bands: 0-30° N (band 1), 30-60° N (band 2) and 60-90° N (band 3), more than 50% of this deposition happened in the band 3 (UNSCEAR, 2000). The expected  $^{236}\text{U}$  inventories from the global fallout in the oceans from the Northern Hemisphere are  $6 \cdot 10^{12}$  atoms/m<sup>2</sup>,  $13 \cdot 10^{12}$  atoms/m<sup>2</sup> and  $5.8 \cdot 10^{12}$  atoms/m<sup>2</sup> in bands 1, 2 and 3, respectively (Casacuberta et al. 2014).

The NPP are potential sources of  $^{236}\text{U}$  into the environment, highlighting, especially, the liquid effluents from the nuclear reprocessing plants (e.g. Sellafield, United Kingdom; La Hague, France) and the accidental releases (e.g. Chernobyl, Fukushima). Unlike the global fallout, these inputs of  $^{236}\text{U}$  are local sources. From the Fig. 3, it is clear that the most of the NPP are situated in the North Hemisphere. Therefore, it is expected that this fact would contribute to increase the differences between Northern and Southern Hemispheres.



**Figure 3.**  $^{236}\text{U}$  inventories reported in different studies: Arctic Ocean (Casacuberta et al. 2016), North Atlantic Ocean (Casacuberta et al. 2014), Greenland Sea (López-Lora, 2014), Japan Sea (Sakaguchi et al. 2012), Mediterranean Sea (Chamizo et al. 2016) and South Atlantic Ocean (this work). Countries with operating Nuclear Power Plants (NPP) and Atmospheric Nuclear Test locations are marked in the map (Carter & Moghissi, 1977).

The  $^{236}\text{U}$  inventories shown in the Fig. 3 are in agreement with the previous discussion of the main sources. The highest values of  $^{236}\text{U}$  were obtained in the North Atlantic Ocean because of the strong influence of the nuclear reprocessing plants of Sellafield and La Hague, among others. The lowest value is the one presented in this work from the South Atlantic Ocean. As it was expected in

advance, the global fallout seems to be the main  $^{236}\text{U}$  source in this region. Furthermore, the plutonium results from these samples also showed the influence of the global fallout. The obtained  $^{240}\text{Pu}/^{239}\text{Pu}$  atomic ratios from the surface samples range from 0.19 to 0.20, which are the typical values of that source. Further studies are necessary to find out more information about other potential regional  $^{236}\text{U}$  sources in the studied area.

## Conclusions

This work extends the current data set of  $^{236}\text{U}$  in seawater, presenting the first results in the South Atlantic Ocean. The results from a set of surface samples and from a depth profile showed the influence of the global fallout in this area. As it was expected because of the heterogeneous distribution of the  $^{236}\text{U}$  released into the environment, the obtained values were lower than the ones reported in the North Hemisphere.

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