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Nuclear weapons produced ^{236}U, ^{239}Pu and ^{240}Pu archived in a *Porites Lutea* coral from Enewetak Atoll

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ABSTRACT

A slice from a *Porites Lutea* coral core collected inside the Enewetak Atoll lagoon, within 15 km of all major nuclear tests conducted at the atoll, was analysed for ^{236}U, ^{239}Pu and ^{240}Pu over the time interval 1952-1964 using a higher time resolution than previously reported for a parallel slice from the same core. In addition two sediment samples from the Koa and Oak craters were analysed. The strong peaks in the concentrations of ^{236}U and ^{239}Pu in the testing years are confirmed to be considerably wider than the flushing time of the lagoon. This is likely due to the growth mechanism of the coral. Following the last test in 1958 atom concentrations of both ^{236}U and ^{239}Pu decreased from their peak values by more than 95% and showed a seasonal signal thereafter. Between 1959 and 1964 the weighted average of the ^{240}Pu/^{239}Pu atom ratio is 0.124 ± 0.008 which is similar to that in the lagoon sediments (0.129 ± 0.006) but quite distinct from the global fallout value of ~0.18. This, and the high ^{239,240}Pu and ^{236}U concentrations in the sediments, provides clear evidence that the post-testing signal in the coral is dominated by remobilisation of the isotopes from the lagoon sediments rather than from global fallout.

1. Introduction

^{236}U and ^{239}Pu, with half-lives of 2.342 × 10^{7} a (Browne and Tuli, 2006) and 24,110 a (Browne, 2003), respectively, can be used as environmental tracers because their natural abundance is negligible compared with that produced anthropogenically. Furthermore, before the nuclear era no man-made ^{236}U or ^{239}Pu were present on earth (Choppin et al., 2002; Steier et al., 2008). This allows these isotopes to be used as a timing signal in environmental archives like ice cores (Wendel et al., 2013), lake sediments (Hancock et al., 2014) and corals (Lindahl et al., 2011; Sakaguchi et al., 2016; Winkler et al., 2012). Corals, as they grow, incorporate elemental and isotopic information which reflect physical and chemical conditions in the surrounding seawater (Purdy et al., 1989 and references therein), and can therefore provide a record of the ^{236}U and ^{239}Pu levels present at the time of growth. Here we have analysed a *Porites Lutea* coral collected in 1984 within the lagoon (11°36′N, 162°05′W) of the Enewetak Atoll (Fig. 1), which is part of the Marshall Islands, located in the western part of the North Pacific Ocean. It was, together with Bikini Atoll, used by the United States to conduct the bulk of their nuclear weapons testing program between 1949 and 1958 (Rudrud et al., 2007). In spite of its close proximity to the two largest test sites, Hardtack I-Oak (8.9 Mt TNT equivalent; 3 km) and Ivy-Mike (10.4 Mt; 13 km), the coral grew continuously throughout the testing years with an annual growth rate of 7–12 mm/a (Hudson, 1985).

This study is a significant extension of our previous work reported in Froehlich et al. (2016). Here we present a complete record in 2 mm (bi-monthly) increments of ^{236}U and ^{239}Pu concentrations from the same coral core but from a slice cut parallel to it. The cut was along the main coral growth axis and includes the period between 1952 and 1958, covering the four major nuclear testing years at Enewetak Atoll. In our previous work, the ^{236}U and ^{239}Pu concentration peaks were significantly broader in time than the peaks in the ^{14}C concentration, and it was speculated that this may have been due to smearing of the time resolution by the sampling procedure. The new procedure was designed to ascertain whether or not this was the case. The smaller drilling increments improve the timing resolution by a factor of ~2, which allows a more detailed assessment of the historical input of ^{236}U and ^{239}Pu. In addition the sampling width perpendicular to the growth direction was reduced by a factor of ~3, which reduces the possibility of extracting

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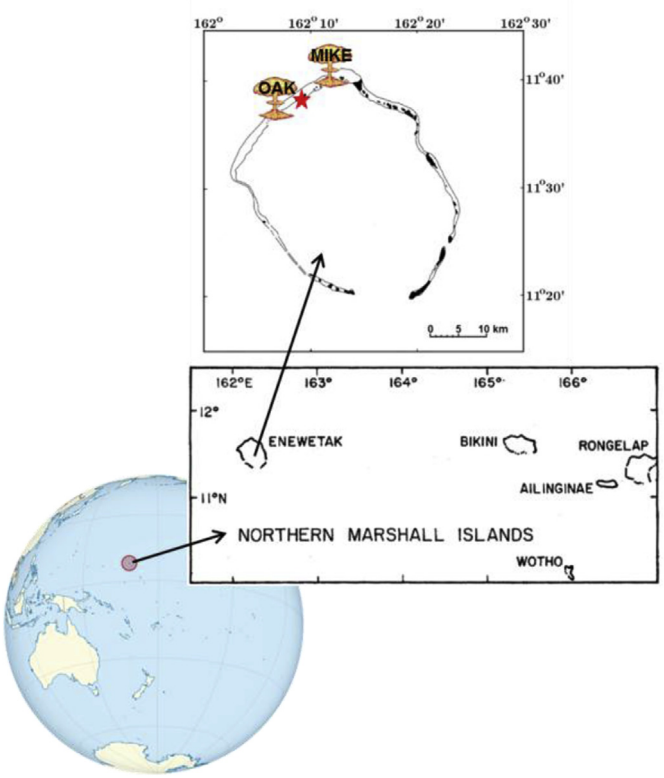

Fig. 1. World map showing the Marshall Islands (red circle) in the western-north Pacific Ocean. The Enewetak and Bikini Atolls are part of the Northern Marshall Islands (figure from Atkinson et al., 1981) and a modified map of the Enewetak Atoll (Henry and Wardlaw, 1990) includes the *Porites Lutea* (core-11 in Hudson, 1985) location as a red star (11°36′N, 162°05′W). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

material from outside the desired time interval.

The new information obtained in the present work is that the consecutive record in 2 mm increments has been extended out to late 1964. This period also includes the so-called 'bomb-peak' in global fallout during 1963. In our previous work we only analysed two samples in the 1960s, which were not sufficient to ascertain if the bomb peak was discernible at Enewetak.

In summary, we present a continuous record of ^{236}U and ^{239,240}Pu from a *Porites Lutea* of more than 10 years covering the nuclear and post-nuclear time at Enewetak Atoll with a time resolution of 2–3 months.

2. Materials and methods

A coral slice parallel to the one discussed in Froehlich et al. (2016) was cleaned with an ultrasonic probe and dried overnight at 40 °C in an electric oven. The age model used to guide the sampling process is based on Sr/Ca ratios, as previously described in Froehlich et al. (2016). Consecutive samples were drilled using a Dremel 3000. Sample dimensions were 2 mm along the main growth axis, 20–25 mm wide perpendicular to the growth direction and 2–4 mm deep to be compared with 4 mm × 70 mm x 2–4 mm previously. The growth bands are essentially straight over the narrower width, making them easier to follow than the curvature over the larger distance, thereby reducing the possibility of smearing the time resolution. On average about 130 mg coral material was collected. In addition to the samples from the coral core, two sediment samples (~100 mg) collected in 1985 from the Koa and Oak craters (Wardlaw et al., 1991) were prepared and measured the same way. All acid dilutions and chemical reagents used in the sample preparation were prepared using Milli-Q® (18 MΩ cm) water. The acids were further purified, 65% Suprapur® HNO_3 by sub-boiling distillation, 30% Suprapur® HCl by passing through a column filled with UTEVA® resin in order to remove any uranium that might have been present. The extraction of U and Pu was based on Froehlich et al. (2016) but was streamlined by using a tandem column as described in Srncik et al. (2011). All samples were spiked with ~10^{10} atoms each of ^{242}Pu (SRM 4334H, NIST) and ^{233}U (CRM 111-A, NBL).

2.1. Accelerator mass spectrometry

The AMS measurements for U and Pu were performed using the 14UD heavy ion accelerator at the Australian National University (ANU) (Fifield, 2008; Fifield et al., 2010, 2013). A detailed description of the measurement process is given in Froehlich et al. (2016). The Pu measurement reproducibility reflects the systematic uncertainty of the AMS system and is checked throughout the measurement using a certified reference material (UKAEA Certified reference Material No. UK Pu 5/92,138) and a standard material (Dittmann et al., 2015) with accurately known Pu atom ratios. The reproducibility of both materials during these measurements was 4%, and this has been added in quadrature to the uncertainty due to counting statistics to produce the overall uncertainty on the measurements.

The reproducibility of U was similarly determined to be 7% using a^{233}U/^{236}U in-house material with a known atom ratio of 1.005. In addition, the ^{233}U spike material (CRM 111-A, NBL) contains 0.0166 atom percent of ^{236}U. Since 10^{10} atoms of ^{233}U were added to each sample, ~10^{6} atoms of ^{236}U would also have been added to all coral samples and procedural blanks. This results in an expected ^{236}U contribution between 2 and 12 counts per measurement, depending on the ion source output. Our blanks were consistent with this, and an appropriate correction was applied to each sample. The corrected number of ^{236}U counts in the coral ranged between 22 and 6438.

3. Results

The ^{239}Pu and ^{236}U atom concentrations, the $^{239+240}$Pu activity concentration together with the ^{240}Pu/^{239}Pu and ^{236}U/^{239}Pu atom ratios are given in Table 1 (see supplementary section). Between 1952 and 1958 the ^{239}Pu atom concentration ranges from (25-1189) × 10^8 at/g and the $^{239+240}$Pu activity is between (4-140) × 10^3 mBq/kg, whereas during the post-nuclear time values range between (8-39) × 10^8 at/g and (1.1–4.9) × 10^3 mBq/kg, respectively. The ^{236}U atom concentration shows a similar variation, but the maximum concentration of 174 × 10^8 at/g is almost seven times lower than the maximum ^{239}Pu concentration. A similar factor pertains after the cessation of testing at the Enewetak Atoll, when the ^{236}U concentration ranges from (0.9–8.0) × 10^8 at/g. The ^{240}Pu/^{239}Pu atom ratio varies considerably from 0.584 ± 0.053 in 1952 to 0.069 ± 0.004 in 1958, while the ^{236}U/^{239}Pu atom ratio is lowest in 1952 (0.037 ± 0.005) and highest in 1956 (0.369 ± 0.033). Between 1959 and 1964 the ^{240}Pu/^{239}Pu ratio varies between 0.093 and 0.175 with a weighted mean of 0.124 ± 0.008 and the ^{236}U/^{239}Pu ratio ranges from 0.092 to 0.262 with a weighted mean of 0.144 ± 0.014.

4. Discussion

4.1. ^{236}U and ^{239}Pu atom concentrations

In the previous work, the period December 1952 to May 1953,

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