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Anthropogenic ^{244}Pu in the environment

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Abstract

The presence of ^{244}Pu ($t_{1/2}$ = 81 Ma) in the early solar system is long established [Nature 187 (1960) 36; Science 172 (1971) 837]. ^{244}Pu is also one of a number of isotopes which may be detectable as live radioactivity on earth, originating from the Interstellar Medium (ISM). There has been a recent attempt to measure a ISM isotope signature of ^{244}Pu [ApJ 558 (2001) L133]. This rises the question of the presence of an anthropogenic contribution in the environment. In order to better assess the ^{244}Pu fall-out on earth, we have started a program at VERA to measure ^{244}Pu in surface sediments of Lake Michigan, routinely sampled for ^{239,240}Pu measurements. Considerable amount of information on other isotopes exist for Lake Michigan. We have reproduced the expected ^{241}Pu/^{239}Pu and ^{240}Pu/^{239}Pu ratios for the global fallout. The measured ^{244}Pu/^{239}Pu was considerably higher than expected and was therefore most likely the result of contamination. Measurement of samples specifically prepared for the measurement of ^{244}Pu will be tried in the future.

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1. Introduction

Some of the heaviest nuclides are produced in the astrophysical r-process. The production site mainly considered for the r-process are supernovae. If the supernova is close enough a deposition of nuclides on earth from the supernova ejecta and swept-up ISM can be expected (Ellis et al., 1996). In search of isotope signatures from such events only radionuclides are to be considered. The radionuclide has to be short-lived enough so that there is no primordial background from the formation of the solar system. On the other hand a long-lived radionuclide can accumulate in the ISM and make a higher contributions by means of

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swept-up matter by the supernova blast. The r-process yield of the nuclide is also of importance.

There are also several long-lived radionuclides which should be produced or ejected by supernovae, but are not produced by the r-process. Among them is ^{60}Fe. This nuclide is expected to be produced in considerable amounts by supernovae (Clayton, 1971; Woosley and Weaver, 1995).

Accelerator mass spectrometry (AMS) is a technique suited for measuring isotope signatures at the lowest levels. The detection of a ^{60}Fe (Knie et al., 1999) signature in a ferro-manganese crust using AMS has spawned search for signatures in geological records from other radionuclides. Likely candidates are ^{244}Pu (Wallner et al., 2000), ^{182}Hf (Vockenhuber et al., 2002, these proceedings) and ^{247}Cm. A detection of ^{244}Pu together with a signal of ^{60}Fe would be an experimental indication that the r-process goes on in supernovae.

Apart from supernova-induced deposition, there has been an attempt to measure the steady-state flux of ^{244}Pu from the ISM (Paul et al., 2001) in a deep-sea surface sediment (from 5800 m water depth at 9°30′ N, 174°18′ W). Since the surface layer of the sediment has not been removed before the chemical processing of the sediment, plutonium isotopes from atmospheric bomb-testing were present in the sample material. In the AMS measurement 1 count of ^{244}Pu was found. After comparison with the other plutonium isotopes it was concluded that this is compatible with fall-out from bomb tests.

2. Anthropogenic level of ^{244}Pu

The neutron flux in a detonation of a nuclear bomb is able to produce considerable amounts of ^{244}Pu by multiple neutron capture on plutonium and uranium isotopes. While there are measurements of ^{244}Pu near test sites (Diamond et al., 1960), the ambient level of ^{244}Pu from the stratospheric fall-out of the big atmospheric tests is still not known. The AMS measurement mentioned above (Paul et al., 2001) indicates a ^{244}Pu/^{239}Pu-ratio of $\sim 10^{-4}$. However, a measurement of ^{244}Pu manganese nodules (from 5000 m water depth at 9°18′ N, 146°03′ W) (Wallner, 2000) yielded a ratio of about $\sim 10^{-3}$.

3. Measurement of sediment samples from Lake Michigan

In order to solve this puzzle we dedicated a beam-time (19–24 May 2003) at the Vienna Environmental Research Accelerator (VERA) to the measurement of plutonium isotopes from Lake Michigan Sediments. In our heavy ion AMS measurements we use a ToF spectrometer in combination with an ionisation chamber for energy detection (Vockenhuber et al., 2003). The reported pile-up problem (Winkler et al., 2002) has been solved by measuring the pulse-width of the detector pulses as well (see Fig. 1). The electroplated samples we used, were originally prepared for measurement of ^{239,240}Pu via decay counting. The sample material had already been spiked with ^{242}Pu, therefore the fall-out contribution of this isotope could not be measured. Further chemical processing was necessary for obtaining sputter targets suitable for AMS. As there was worry about getting enough counts of ^{244}Pu, eight electroplated samples were processed for a single sputter target.

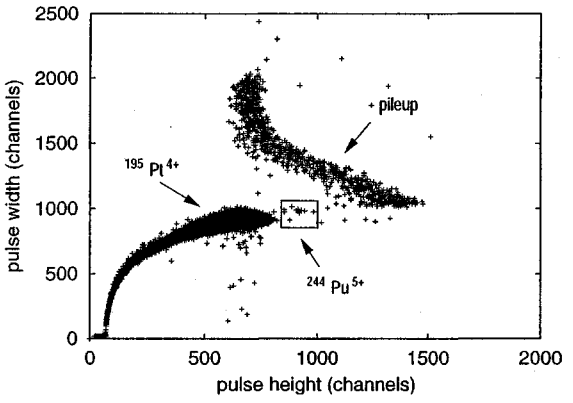

Fig. 1. The pileup of pulses from two $^{195}Pt^{4+}$ ions can be identified by higher pulse width unless the two particles reach the detector at the same time. However, in this case they will sum up to the double height and do not interfere with $^{244}Pu^{5+}$. In the 2D-spectrum of pulse width versus pulse height $^{244}Pu^{5+}$ is separated from the background.

Table 1
The result of the measurement of isotope ratios in sediment samples from Lake Michigan

	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$	$^{244}Pu/^{239}Pu$	$^{239}Pu/^{242}Pu$
Plates 1–8	$(1.73 \pm 0.03) \times 10^{-1}$	$(1.70 \pm 0.10) \times 10^{-3}$	$(2.67 \pm 0.09) \times 10^{-3}$	$(3.98 \pm 0.02) \times 10^{-2}$
Plates 9–16	$(1.36 \pm 0.02) \times 10^{-1}$	$(1.71 \pm 0.09) \times 10^{-3}$	$(1.05 \pm 0.01) \times 10^{-1}$	$(3.71 \pm 0.02) \times 10^{-2}$

The ^{239}Pu count rates in the final detector were between 10 and 20 counts/s. Only statistical uncertainties are given.

We also measured two targets made from plates containing only ^{242}Pu spike material. We found $(1.3 \pm 0.4) \times 10^{-6}$ for the ratio of $^{244}Pu/^{242}Pu$. As the sediment samples were prepared for α-counting they contain about the same activity of ^{242}Pu and $^{239,240}Pu$. That means the spike material would in principle allow for measuring $^{244}Pu/^{239}Pu$ ratios down to 10^{-4}.

Together with the sediment samples a target of our in-house reference material – prepared from different amounts of ^{242}Pu and ^{244}Pu standard solutions – was measured. The $^{244}Pu/^{242}Pu$ ratio of former measurements of this material was reproduced within statistics. From the yield of this target we obtain a detection efficiency of at least (the target was not used up) 6×10^{-5}.

The measured $^{244}Pu/^{239}Pu$ of both sediment sample targets is surprisingly high (see Table 1). One target (plates 9–16) was obviously contaminated with ^{244}Pu. It is not possible to tell where the contamination comes from. However, the amount of ^{244}Pu in this target material points to lab contamination.

The $^{240}Pu/^{239}Pu$ and $^{241}Pu/^{239}Pu$ ratios of the sample made from plates 1–8 reproduce the typical values for the northern hemispheric stratospheric fall-out in this area ($(1.73 \pm 0.02) \times 10^{-1}$ and $(1.65 \pm 0.03) \times 10^{-3}$ for $^{240}Pu/^{239}Pu$ and $^{241}Pu/^{239}Pu$ [1], respectively Krey et al., 1976; Cooper et al., 2000). Taking into consideration the $^{242}Pu/^{239}Pu$ ratio from fall-out plutonium ($(3.6 \pm 0.4) \times 10^{-3}$ Krey et al., 1976), the measured $^{244}Pu/^{239}Pu$ ratio of 2.67×10^{-3} would be too high. It is also a factor of 2 higher than the local fallout value of $(1.18 \pm 0.07) \times 10^{-3}$ for the mike test (Diamond et al., 1960). Therefore, it is most likely also the result of contamination.

[1] Corrected for decay from January 1971 to May 2003.

The use of material prepared for other kinds of measurement is clearly unsatisfying, as there is usually no specification for ^{244}Pu. The sample preparation has to be done specifically for the needs of the measurement of this rare isotope. A measurement of ^{244}Pu from bomb-testing is best carried out by not using a spike at all as there is ^{239}Pu as reference isotope. For the measurement of supernova-produced plutonium a spike of ^{236}Pu with high isotopic purity seems to be the best option to trace the chemical yield.

4. Outlook

Our AMS system is now in a condition that makes it capable of measuring ^{244}Pu at natural levels. Our next step is to measure a representative set of samples for the assessment of the global fall-out of bomb-produced ^{244}Pu. This will also allow to further improve the AMS setup of VERA (e.g. efficiency) and to develop chemical sample preparation, which is necessary in order to investigate possible supernova-produced ^{244}Pu in sediment cores.

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