Anthropogenic ^{244}Pu in the Environment

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Abstract

In 1960 Kuroda [1] suggested that ^{244}Pu ($t_{1/2} = 81$ Ma) was present in the early-solar system producing isotopic anomalies of xenon. The final proof for this hypothesis came from a measurement of xenon from spontaneous fission of ^{244}Pu in the laboratory [2]. Although the detection of live ^{244}Pu in nature has been reported in the same year [3], this finding is not unequivocally accepted. In particular, the question of possible contamination with man-made plutonium was raised. Due to the recent interest in detecting live ^{244}Pu from the Interstellar Medium (ISM) [4], the question of the presence of anthropogenic ^{244}Pu in the environment became of interest again. So far, two different ^{244}Pu/^{239}Pu isotope ratios have been reported from locations at low northern latitudes in the Central Pacific [4,5]. A ratio of 10^{-4} was measured in a deep-sea surface sediment from 5800 m water depth at 9°30' N, 174°18'W [4], and a ratio of 10^{-3} was found in manganese nodules from 5000 m water depth at 9°18', 146°03'W [5]. 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. First results on the methodology of ^{244}Pu detection were reported at the AMS-9 conference in Nagoya, September 2002 [6]. In the current contribution we report on measurements of ^{244}Pu in Lake Michigan samples, with the goal to establish the ^{244}Pu fall-out at a location where considerable information on fall-out of plutonium and other radionuclides exists.

Key words: ^{244}Pu, AMS.

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

Accelerator Mass Spectrometry is a technique suited for measuring isotope signatures at the lowest levels. The detection of a ^{60}Fe [8] signature in a ferromanganese crust using AMS has spawned search for signatures in geological records from other radionuclides. Likely Candidates are ^{244}Pu [9], ^{182}Hf [10] 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 [4] in a deep-sea sediment. Since the surface layer of the sediment has not been removed before the chemical processing of the sediment, plutonium isotopes from athmospheric bomb-testing were present in the sample material. In the AMS measurement 1 count of ^{244}Pu was found. After comparision 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 [11], 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 [4] indicates a ^{244}Pu/^{239}Pu-ratio of $\sim 10^{-4}$. However, a measurement of ^{244}Pu manganese knodules [5] yielded a ratio of about $\sim 10^{-3}$.

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

Table 1
The result of the measurement of sediment samples from Lake Michigan. Only statistical uncertainties are given.

3 Measurement of sediment samples from Lake Michigan

In order to solve this puzzle we have 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 [12]. The reported pile-up problem [6] has been solved by measuring the pulse-width of the detector pulses as well. The sample material has been spiked with ^{242}Pu, therefore the fall-out contribution of this isotope could not be measured. As there was worry about getting enough counts of ^{244}Pu eight electroplated samples were processed for a single sputter target.

We also measured two targets made from plates containing only ^{242}Pu spike material. We found $(1.3 \pm 0.4) \cdot 10^{-6}$ 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 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) $6 \cdot 10^{-5}$.

Looking at the results of the sediment samples (see Tab. 1), one target (plates 9–16) was obviously contaminated by an electroplated sample containing ^{244}Pu as spike material. It demonstrates the problem of obtaining samples suitable for measuring ^{244}Pu, which is the result of the fact that up to now it was rarely looked for in nature.

The sample from plates 1–8 shows a $^{244}Pu/^{239}Pu$ ratio which is higher than expected for the global fall-out. The $^{240}Pu/^{239}Pu$ and $^{241}Pu/^{239}Pu$ ratios of this sample reproduce the typical values for the northern hemispheric stratospheric fall-out in this area ($(1.73 \pm 0.02) \cdot 10^{-1}$ and $(1.65 \pm 0.03) \cdot 10^{-3}$ for $^{240}Pu/^{239}Pu$ and $^{241}Pu/^{239}Pu$ [1] [13]). Taking into consideration the $^{242}Pu/^{239}Pu$ ratio from

[1] corrected for decay to May 2003

fall-out plutonium ($(3.6 \pm 0.4) \cdot 10^{-3}$ [13]), the measured ^{244}Pu/^{239}Pu ratio of $2.67 \cdot 10^{-3}$ would be too high. This is most likely a result of contamination too.

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 mass 236 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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