Antropogenic and Supernova Produced Plutonium Isotopes in Deep-sea Manganese Encrustations ◇

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We have measured in a ferromanganese crust ^{60}Fe with a strong indication of supernova origin [1]. In order to confirm these findings we extended our search for ^{244}Pu. ^{244}Pu ($T_{1/2} = 80\,$My) is formed in core collapse supernovae orders of magnitude less than ^{60}Fe. Nevertheless, as we expect no natural production of ^{244}Pu beside the origin mentioned above, a ^{244}Pu detection should be a strong indication for recently supernova processed material on Earth. Manmade ^{244}Pu, however, was released in the environment by nuclear weapon tests. Therefore, the discovery of ^{244}Pu in natural samples has to be discussed under these consideration. On the other hand, the measurement of this antropogenic $^{239-244}$Pu enables us to determine the adsorption yield of plutonium onto ferromanganese crusts, an indespenable number for quantification of the plutonium flux on Earth. Additionally, ^{244}Pu offers favourable conditions for an AMS measurement:

i) Actinides can be highly enriched, because there is no stable isotope which dilutes the isotope to be measured and forms a macroscopic matrix material.

ii) There is usually only a small amount of possibly interfering neighbouring isotopes which have to be separated by chemistry and AMS.

iii) Because of the lack of stable and hence large amounts of isobars, there is no need of an elaborated isobar separation method.

The sample was spiked with radioactive ^{236}Pu, which serves as chemical tracer during the chemical preparation and as a reference isotope during the AMS measurement. For the measurement of actinides a highly sensitive AMS method was developed. A detection limit of down to 10,000 atoms in the sample could be achieved [2]. The limiting factor is the total efficiency for plutonium detection, i.e. the yield for negative ion formation and beam transmission from the ion source to the detector.

For the extraction of actinides from ferromanganese encrustations a chemical extraction method has been developed, which consitsts of a calcium oxalate precipitation followed by threefold chemical extraction chromatography with an anion exchange resin. This method was applied successfully to a 120 g ferromanganese crust and to 850 g of manganese nodules with extraction yields of 50% and 25%, respectively.

Although the oceanic transport of plutonium is understood partly [3], there is still no information about the adsorption yield of plutonium onto ferromanganese encrustations. Therefore, we measuremed bomb produced plutonium isotopes, which were adsorped onto the surface of ferromanganese nodules from the south pacific at a depth of about 5000 m (see table1).

From our measurements, we can estimate a surface densitiy of about 3×10^8 ^{239}Pu cm^{-2}, which was adsorbed onto the surface of the nodules. In pacific water samples, ^{239}Pu surface densities auround 2 mCi $^{239+240}$Pu km^{-2} were measured near the sample site [5]. Using the measured ^{239}Pu/^{240}Pu ratio of four, one obtains a ^{239}Pu surface density of about 5×10^9 atoms cm^{-2}. Using this value, one achieves an uptake factor (i.e. the ratio of the surface densities in the nodules and in the ocean) of about 5%.

Although this is already a very high value compared to those of other elements, it is more likely much higher, since the residence time τ_r of plutonium in the North Pacific has been measured to 400-500 y [6]. The nodules were collected in 1982, i.e. auround $t = 27\,$y after the large nuclear weapon tests in the Pacific. At this time the fraction F_w of plutonium in water was still: $F_w = \exp(-t/\tau_r) = 94\%$. Therefore, one can assume an uptake factor of 100%.

A first measurement was performed with an 120 g sample of the ferromanganese crust VA 13-2 [7] from the Central North Pacific in order to search for supernova produced ^{244}Pu. The age of the sample ranged from 1My to 14 My, corresponding to a depth of 2-43 mm. To avoid interferences from antropogenic plutonium isotopes, 2 mm of the surface were discarded. The sample surface was 25 cm^2. One event of ^{244}Pu was observed, corresponding to a surface density of 4,000 ^{244}Pu cm^{-2} or a time averaged flux around 300 ^{244}Pu cm^{-2} My^{-1} (for an uptake factor of 100%).

In [1] a total ^{60}Fe flux of 4×10^8 ^{60}Fe cm^{-2} My^{-1} (time averaged over the last 6 My) has been measured. Although the uncertainties are still very large, our first measurement of the ratio ^{244}Pu/^{60}Fe $\approx 8 \times 10^{-7}$ might supply information to check theoretical predictions for r-process calculations.

^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{244}Pu
146 ± 14	36 ± 6	1.5 ± 1.1	3.8 ± 1.5	0.17 ± 0.05

Table 1: Plutonium concentrations in the 850 g nodule sample in 10^6 atoms/gram. The measured isotopic pattern is typical for plutonium originating from nuclear weapon tests.

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