

Supernova Produced and Anthropogenic ^{244}Pu in Deep Sea Manganese Encrustations

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Abstract

After the detection of ^{60}Fe from a nearby supernova in a terrestrial sample the search for SN produced radionuclides has been extended to the r-process radionuclide ^{244}Pu . For this purpose an efficient chemical separation method has been developed to extract traces of plutonium from samples in the kilogram range. The AMS (accelerator mass spectrometry) method for actinides was optimized for a nearly background free detection of plutonium isotopes with a high overall efficiency. Measurements of anthropogenic (i.e. a-bomb produced) plutonium isotopes in deep sea manganese encrustations have been carried out to examine the probability for plutonium to be incorporated in deep sea manganese encrustations and its mobility therein. In a first measurement of a pre-bomb layer of a ferromanganese crust a weak ^{244}Pu signal has been detected, consistent with the SN signal suggested by the ^{60}Fe data.

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1. Introduction

The detection of apparently interstellar ^{60}Fe ($T_{1/2} = 1.5$ Myr) activity in a deep sea ferromanganese crust by Knie et al. (1999) motivated a search for other long-lived radionuclides produced in recent ($< 10^7$ yr), nearby ($< 10^2$ pc) supernova (SN) explosions. One very promising isotope for this purpose is ^{244}Pu ($T_{1/2} = 82.6$ Myr), which can be directly deposited on Earth together with the SN debris. Furthermore, ^{244}Pu , which was produced by former SNe, should be present in the interstellar medium (ISM), thus a comparable signal can be expected from ISM swept up by the explosion front (Ellis et al., 1996).

Contrary to ^{60}Fe , which is produced inside the solar system only in negligible amounts, plutonium originating from the atmospheric thermonuclear bomb tests during the 50's and 60's (Balagna et al., 1965; Taube, 1974) is a severe background. In order to achieve a higher explo-

sive force, the fusion device of the bomb is surrounded by a shell of ^{238}U , where fission reactions are induced by fast neutrons generated in the bomb's inner part. Due to multiple neutron capture reactions some percent of these ^{238}U atoms form uranium isotopes up to $A = 255$ (Diamond et al., 1960; Balagna et al., 1965; Dorn and Hoff, 1965; Taube, 1974). Succeeding β^- decays of these short-lived heavy uranium isotopes yield the long-lived plutonium isotopes beyond mass 238. The resulting isotope ratios are characteristic for bomb plutonium.

If the isotopic ratio $^{239}\text{Pu}/^{244}\text{Pu}$ from plutonium of anthropogenic origin is well known, it would be possible to identify excess ^{244}Pu of interstellar origin. It is advantageous, that this ratio should be in the order of 10^3 for bomb plutonium, which is substantially higher than the ratio expected from stellar nucleosynthesis. However, because of the extremely low magnitude of the expected interstellar signal, the mobility of plutonium in the samples has to be examined. This is to avoid too high contamina-

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tions of the pre-bomb material with bomb plutonium.

Additionally, it has to be known, which fraction from plutonium was finally incorporated into the samples. A measurement of the amount of bomb test plutonium in the samples' layers of appropriate age yields the incorporation efficiency ε_i .

2. Samples

As SN produced ^{60}Fe was already successfully measured in a deep sea ferromanganese crust by Knie et al. (1999), we decided to investigate, whether deep sea manganese encrustations (i.e. ferromanganese crusts and manganese nodules) are suitable samples for the detection of SN produced ^{244}Pu . Besides of their high age, the main advantage of encrustations is their extremely low growth rate of only few mm Myr^{-1} , thus plutonium needs to be extracted from a relative low amount of matrix material. Two samples from the equatorial Pacific were at our disposal. The distance between the samples was only a few kilometers, their depths were nearly identical. It is therefore assumed, that both samples had the same chemical conditions and the same plutonium concentration in their environment.

2.1. The ferromanganese crust VA13-2

The first sample, a flat part of the ferromanganese crust VA13-2, originates from the equatorial Pacific ($9^\circ 18' \text{N}$, $146^\circ 03' \text{W}$, 4830 m depth) and was recovered 1976 during an expedition of the German research vessel *Valdavia*. It is depicted in figure 1. This crust was dated by Segl et al. (1984) with the well established ^{10}Be method. The part used in this work had a thick-

ness between 4 and 5 cm, its lowest layer an age of about 14 Myr. Besides of the measurements for anthropogenic plutonium, a large piece with a surface area of 25 cm^2 and a weight of 120 g was used for the search of SN produced ^{244}Pu on Earth. The top layer from this part was removed in order to avoid a contamination with bomb plutonium.

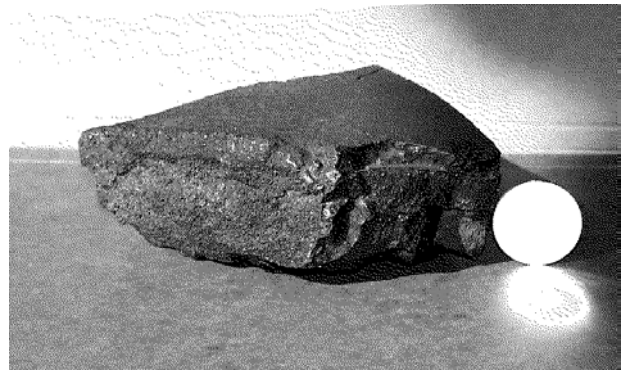


Fig. 1. Part of the ferromanganese crust VA13-2.

2.1. The manganese nodules 251GBH

The second sample (251GBH), a number of manganese nodules, originates from the equatorial Pacific at $9^\circ 19.6' \text{N}$, $146^\circ 05.4' \text{W}$ from a depth of about 5 km. It was collected in 1982 during an expedition of the German research vessel *Sonne*. It is assumed, that the nodules started their growth approximately 15 Myr ago (v. Stackelberg, 2000). They were roughly spherical with a diameter $d = (1.5 \pm 0.5) \text{ cm}$. The weight of the total sample was 857 g, the density of the dried material 1.5 g cm^{-3} , thus the area of ocean floor covered by the nodules was $(5.7 \pm 2.0) \times 10^2 \text{ cm}^2$.

3. Plutonium transport in the Pacific ocean

In the 80's, remaining bomb plutonium was still found in the Pacific ocean, mainly in water samples collected from depths of 450 m and 1000 m, respectively (Bowen et al., 1980; Livingston and Anderson, 1983). A downward flux of plutonium could be measured, mainly caused by sedimentation of biological particles (e.g. fecal pills and phytoplankton) produced in the upper layers of the ocean. Plutonium is adsorbed because of its high particle reactivity. By comparing the amount of plutonium introduced initially onto the water surface with the vertical plutonium flux, a residence time of 420-460 yr in ocean water was estimated (Livingston and Anderson, 1983; Fisher et al., 1988).

In Pacific sediment cores (30°N, 160°W) taken during the years 1974-1978, activities around 130 dpm m⁻² were reported by Livingston and Anderson (1983). Contrary to manganese encrustations, one can assume that nearly all plutonium that has reached the ocean floor incorporated into sediments, i.e. $\epsilon_i \approx 100\%$. However, the activity introduced onto the water surface area at this location was about 6.2×10^3 dpm m⁻² (Bowen et al., 1980). At the time VA13-2 was recovered (1976) about 2.1 % of the bomb plutonium had already reached the ocean floor.

251GBH was recovered six years after VA13-2. It has to be considered, that within the time span, additional plutonium arrived at the ocean floor. Using the mean residence time in the ocean of 440 yr, additional 1.4 % (in total 3.5 %) of the plutonium in the water column must have arrived at the ocean floor during the six years until 1982.

Bowen et al. (1980) reported for two sites close to the location of our samples somewhat lower plutonium activities which were introduced into the ocean (3.6×10^3 dpm m⁻² and 4.7×10^3 dpm m⁻²). We assume a value of $(4.1 \pm 0.6) \times 10^3$ dpm m⁻² for the samples' location. The activities were determined by means of α spectrometry which does not distinguish between the two dominating isotopes ²³⁹Pu and ²⁴⁰Pu. But using a typical a-bomb ratio of $^{240}\text{Pu}/^{239}\text{Pu} = 0.25 \pm 0.05$ atom/atom, it can be estimated, that $(3.9 \pm 1.0) \times 10^9$ atoms ²³⁹Pu cm⁻² were introduced into the ocean above the samples. At the time of the recovery of VA13-2, $(8.2 \pm 2.1) \times 10^7$ atoms cm⁻² had reached the ocean floor, six years later, when 251GBH was recovered, $(1.4 \pm 0.4) \times 10^8$ atoms cm⁻².

4. Experimental methods

A chemical extraction method for plutonium in deep sea manganese encrustations has been developed based on procedures for actinide extractions from sediments (Yamamoto, 1982; Buesseler and Halverson, 1987):

After disintegrating the sample material in aqua regia, a ²³⁶Pu spike was added to the solution. It served as a reference isotope for AMS as well as a radioactive tracer to determine the yield of the chemical extraction. Plutonium was recovered by a co-precipitation with calcium oxalate and an extraction chromatography with an AG1x8 anion exchange resin, which was repeated three times. These separation steps allowed the reduction of the matrix material from a kilogram range until far below one milligram. The residue was dissolved in hydrofluoric acid after adding 50 μg of iron as a car-

rier. This solution was dropped into small holes drilled in high purity graphite cylinders, dried, and converted to oxide in a muffle furnace. Chemical yields up to 50 % could be achieved. The carbon cylinders were used as cathodes in an sputter ions source, from where plutonium was extracted as PuO^- , separated by mass, and injected into the Munich MP Tandem Accelerator (see figure 2). The negative ions were accelerated to the positive (+12.5 MV) terminal and stripped by a thin carbon foil, resulting in a total molecular disintegration, thus molecular background can be entirely rejected in a succeeding mass separation.

After the accelerator the 11^+ charge state with an energy around 150 MeV was selected. This high energy allows an extremely efficient suppression of any remaining background with nuclear physics detection methods, in this case a time-of-flight path succeeded by a ΔE - E_{rest} telescope.

These are key points for the very high sensitivity achievable with AMS. During a measurement turn it was switched between the isotope of interest, e.g. ^{244}Pu , and the reference isotope ^{236}Pu . The background level is determined in runs of blank samples, which was spiked with ^{236}Pu , too.

An overall efficiency (formation of PuO^- ions, transmission from the source to the detector, detector efficiency) up to 10^{-4} could be achieved. During all ^{244}Pu blank runs no background events were detected. For a more detailed description of the method, see Wallner et al. (2000).

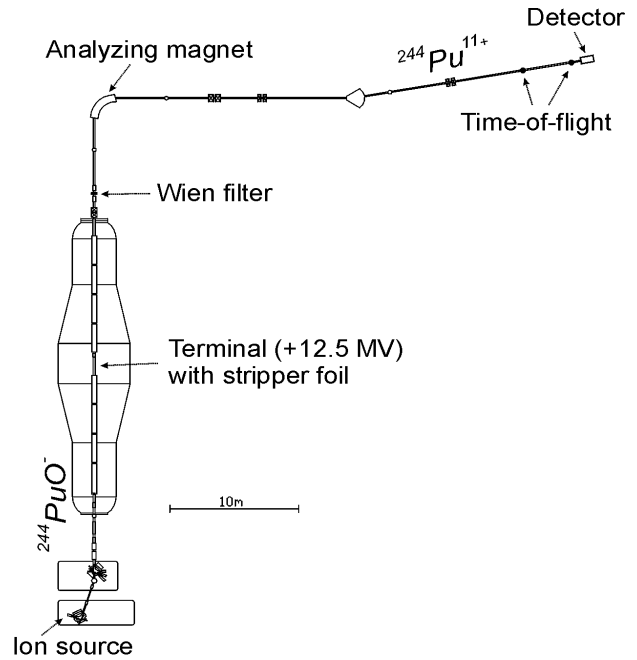


Fig. 2. AMS setup for the detection of actinides.

5. Results

5.1. Anthropogenic plutonium

^{239}Pu was measured in the top layer (0-2 mm, corresponding to 0-0.8 Myr) of the crust VA13-2, resulting in a surface density of $(1.15 \pm 0.55) \times 10^7$ atoms cm^{-2} . To obtain the incorporation efficiency ϵ_i , this number has to be compared with the amount of anthropogenic ^{239}Pu that has reached the ocean floor at the time of the sample's recovery (1976), which has been estimated in section 3 to $(8.2 \pm 2.1) \times 10^7$ atoms cm^{-2} . Hence, $\epsilon_i = (14 \pm 8) \%$ for the crust VA13-2.

Additionally, the entire nodule sample 251GBH was chemically prepared and the concentrations of the plutonium isotopes ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu and ^{244}Pu were determined

with AMS (see also Korschinek et al., 2001). The results are presented in figure 3. It should be noted that ^{244}Pu was measured with a sensitivity of about four orders of magnitude higher than achievable with α spectrometry. The total amount of ^{239}Pu in this sample was $(1.25 \pm 0.12) \times 10^{11}$ atoms. On the ocean floor, the nodules have covered a surface of $(5.7 \pm 2.0) \times 10^2 \text{ cm}^2$, thus the ^{239}Pu surface density had been $(2.2 \pm 0.8) \times 10^8 \text{ atoms cm}^{-2}$. Comparing this number with the amount of ^{239}Pu that had reached the ocean floor $((1.4 \pm 0.4) \times 10^8 \text{ atoms cm}^{-2}$, see section 3) yields an incorporation efficiency ε_i for the sample 251GBH of $(160 \pm 70) \%$, which is about one order of magnitude above the value for VA13-2. This difference can be explained by two reasons: Assuming a spherical shape, the surface area of a nodule is four times the area of the ocean bed that was covered by it. Compared to crusts nodules can be more effectively flushed by water streams near the ocean floor. Hence, they can more effectively adsorb plutonium from the ocean water. Also nodules do not totally cover the sediment. It is possible that they remobilize plutonium from their surroundings.

In order to determine the mobility of plutonium in ferromanganese crusts, a layer of 2-4 mm depth (0.8-1.6 Myr) of VA13-2 was examined for anthropogenic plutonium. No ^{239}Pu was detected. An upper limit of $1.5 \times 10^5 \text{ atoms cm}^{-2}$ can be deduced from this result, i.e. less than 1% of the bomb plutonium was transported from the crust's surface into the 2-4 mm layer. Using the $^{244}\text{Pu}/^{239}\text{Pu}$ ratio of anthropogenic plutonium measured in 251GBH, an upper limit of $200 \text{ atoms cm}^{-2}$ can be given for anthropogenic ^{244}Pu in layers below 2 mm.

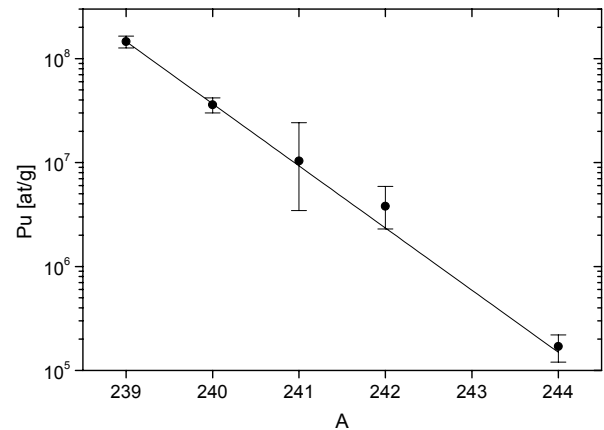


Fig. 3. Measured plutonium concentrations in the manganese nodules 251GBH. The data are decay corrected for the time of the atmospheric bomb tests (1960).

5.2. SN produced ^{244}Pu

In a first measurement of the crust VA13-2 ($A = 25 \text{ cm}^2$, $d = 2-43 \text{ mm}$, $t = 1-14 \text{ Myr}$, $m = 120 \text{ g}$) one single ^{244}Pu event has been detected, which corresponds a ^{244}Pu surface density of about $4000 \text{ atoms cm}^{-2}$ (Wallner et al., 2000). Compared to this, the upper limit for anthropogenic ^{244}Pu is $200 \text{ atoms cm}^{-2}$, as shown above. With $\varepsilon_i = 14 \%$ a total terrestrial ^{244}Pu surface density of $3 \times 10^4 \text{ atoms cm}^{-2}$ can be determined for the period from 1-14 Myr B.P. Besides of the statistical error and the error of ε_i , there is an additional uncertainty due to count rate fluctuations of the reference isotope ^{236}Pu during the AMS, thus this first result is only accurate within a factor of about five.

Because of the lack of ^{239}Pu in this layer, the ^{244}Pu signal cannot be attributed to anthropogenic background. A possible origin is the debris from a nearby SN which had been directly deposited on Earth, as suggested by Ellis et al.

(1996) and experimentally confirmed by Knie et al. (1999). Additionally, ISM swept-up by the SN explosion front comprises steady state ^{244}Pu , which adds to the material produced in the SN. In figure 4 the signals are given for ISM densities of 0.01, 0.1 and 1 cm^{-3} , according to Ellis et al. (1996). Although the uncertainty of this first measurement is still large, the result is in agreement with the expected signal of a SN within a distance of a few 10 pc.

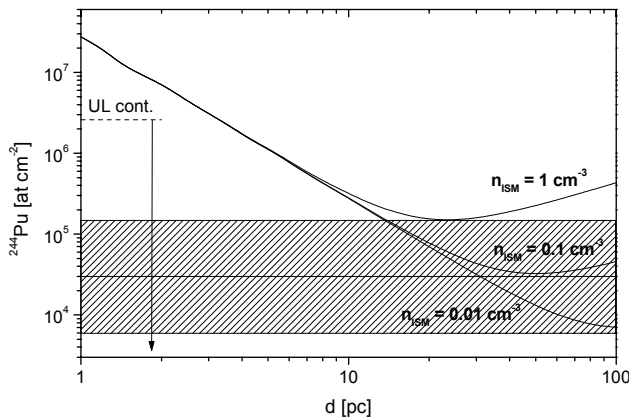


Fig. 4. Expected ^{244}Pu fluence from a SN at the distance d from the Earth, calculated for ISM densities of 1, 0.1, and 0.01 atoms cm^{-3} , respectively. The hatched area indicates the experimental value with its uncertainty. Also shown is the upper limit for a continuous ^{244}Pu flux of 2×10^5 atoms $\text{cm}^{-2} \text{Myr}^{-1}$, given by Paul et al. (2003), multiplied with the time span our sample covers (13 Myr).

6. Conclusion

In a sample of the deep sea ferromanganese crust VA13-2 which covered the period from 1-14 Myr B.P. a ^{244}Pu surface density of about 4000 atoms cm^{-2} was measured. The probability for plutonium to be build in a crust was determined to 14 %, thus this value corresponds to a terrestrial input around 3×10^4 atoms cm^{-2}

during the period from 1-14 Myr. This signal is in agreement with SN production of the detected ^{244}Pu , whereas anthropogenic origin can be excluded because of the missing ^{239}Pu . Measurements of larger samples under improved experimental conditions are in preparation.

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