Determination of Plutonium in environmental samples by AMS and Alpha Spectrometry

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Abstract

Environmental samples from nuclear weapons test sites at the atolls of Mururoa and Fangataufa (French Polynesia, south Pacific) have been analyzed for their content of Plutonium isotopes by applying the independent techniques of decay counting (Alpha Spectrometry) and Accelerator Mass Spectrometry (AMS). Here, we propose the combination of both techniques which results in a maximum of information on the isotopic signature of Pu in environmental samples.

Plutonium was chemically separated from the bulk material by anion exchange. ^{242}Pu was used as an internal standard for both AMS and Alpha Spectrometry. The samples for Alpha Spectrometry were prepared by micro-precipitation with NdF_3. After Alpha Spectrometry, the samples were reprocessed for AMS. Pu was co-precipitated with $Fe(OH)_3$ and finally, solid samples were prepared. At the VERA (Vienna Environmental Research Accelerator) facility, the various Pu isotopes were separated by their isotopic masses and quantified by the AMS technique.

A good agreement of the results obtained from the AMS measurements was found with those obtained from Alpha Spectrometry. Overall, the data agree on average within 10 % of each other. Isotope ratios for ^{238}Pu, ^{239}Pu and ^{240}Pu can be extracted from our investigations. Alpha Spectrometry delivers data for the ^{238}Pu and the combination of $^{(239+240)}$Pu concentrations in those samples. In addition, the AMS technique provides information on the individual concentrations of ^{240}Pu and ^{239}Pu.

Introduction

The isotopic signature of the various plutonium isotopes depends on the production process. Measurements of their ratios allow the reconstruction of the history of neutron exposure of a certain region. Generally speaking, longer irradiation and higher fluxes lead to a higher content of the heavier isotopes. Measurements of the isotopic composition of environmental plutonium thus provide information on the origin of the material as the isotope ratios vary with reactor type and nuclear fuel burn-up [Oughton et al. 2001]. For weapons test fallout samples isotope ratios depend on the fissionable material used for weapon construction and the type of weapons test [Irlweck et al., 1999, Yamamoto et al., 1996]. Determination of plutonium isotopes clearly is of interest for nuclear safeguards [Donohue 1998], radiation protection [Priest et al., 1999], and, recently, also for astrophysical applications [Wallner et al. 2000, Winkler et al. 2004a]. For analysis of environmental plutonium, methods rely in general on chemical separation of Pu from the bulk material in combination with alpha spectrometry for determination of ^{238}Pu and $^{239(40)}$Pu. ^{241}Pu can be calculated from the amount of ^{241}Am ingrown since the last separation of the Pu [Irlweck et al., 1998, Mietelski et al., 1999] or can be directly measured by LSC [Hrnecek et al., 2002, Solatie et al., 2002]. For the determination of ^{239}Pu and ^{240}Pu, which can give direct information on the content of fissionable ^{239}Pu, mass spectrometric methods such as ICP-MS and AMS are necessary [Povinec et al., 2001]. ICP-MS has been widely applied for this purpose, although care must be taken to remove interferences like $^{238}UH^+$, which can influence the determination of ^{239}Pu [Hrnecek et al., 2002, Chiappini et al., 1996, Muramatsu et al., 1999, Muramatsu et al. 2001].

AMS (Accelerator Mass Spectrometry) is characterized by low measurement background and high detection efficiency. Most existing facilities can be understood as a combination of two conventional mass separators with an electrostatic tandem accelerator in between. These mass separators have almost 100% transmission but only ‘medium’ resolution (relative mass resolution 10^{-2} to 10^{-3}). Thus, separation of isotopes is possible, but (atomic or molecular) isobars cannot be resolved. The tandem accelerator serves two purposes: The higher particle energy in the second, ‘high-energy’ separator allows for better suppression of the neighbouring masses. For plutonium isotopes, the main advantage of AMS compared to conventional MS

(TIMS, ICP-MS) is the complete destruction of molecular isobars (e.g. ^{238}UH for ^{239}Pu) by stripping to high positive charge states in the terminal of the tandem accelerator [Oughton et al., 2001]. Additionally, this method allows the determination of higher mass plutonium isotopes such as ^{242}Pu and ^{244}Pu in environmental samples [Fifield et al., 1996].

In this work samples from a measurement project organized by the IAEA to assess the radiological situation at the atolls of Mururoa and Fangataufa (French Polynesia) [IAEA, 1998, Danesi et al., 2002], which have already been analyzed for their content of ^{241}Am, $^{239(40)}$Pu and ^{238}Pu [Hrnecek et al., 1999, Irlweck et al., 1999] were reanalyzed for Pu and assessed for their ^{240}Pu/^{239}Pu ratio by AMS. One goal of this work was to establish a measurement procedure for Pu isotopes at the VERA AMS facility [Vockenhuber et al., 2003] at the Institut für Isotopenforschung und Kernphysik, University of Vienna.

Experimental

Standards, reagents and samples

For all samples in this work, ^{242}Pu reference material (SRM 4334G) from NIST (Gaithersburg, Maryland, USA) was used as an internal standard for alpha spectrometry and AMS. For separation of Pu, AG 1x8 anion exchange resin, 100 – 200 mesh (Biorad) was used. The resin was preconditioned with 100 ml 8 M HNO_3 before use. All reagents used were of analytical grade.

Samples from Mururoa atoll were top soil samples from the Faucon area at the atoll (9.3.6, 9.3.18 and 9.3.25) and coral bedrock from the Colette area (7.2.4.1 and 7.2.4.2). From Fangataufa, loose coral rock samples (11.4.1, 11.4.3 and 11.4.4) from the Kilo location were analyzed. Sample sizes used for analysis were 2.5 to 6 g.

Chemical separation of Plutonium

After addition of ^{242}Pu, the samples were leached with 8 M HNO_3 for 3 hours and then filtrated. The filtrate was evaporated to dryness and the residue dissolved in 20 ml 1 M HNO_3. 100 mg $(NH_4)_2Fe(SO_4)_2 \cdot 6H_2O$ was added to the solution to reduce Pu to the trivalent state. Then the solution was adjusted to 8 M HNO_3 by addition of 65% HNO_3 and Pu was oxidized to Pu(IV) with 0.5 g $NaNO_2$. For the separation of Pu, a column containing 10 g AG 1X8 resin was used. After loading of the sample solution, the column was washed with 30 ml 8 M HNO_3. To remove Th from the column, 50 ml 32% HCl was used. Finally, Pu(III) was eluted from the column with 50 ml 0.1 M NH_4I – 9 M HCl. This fraction was evaporated to dryness and fumed several times with 5 ml 65% HNO_3 and some drops of H_2O_2. For preparation of the samples for Alpha Spectrometry, the residue was dissolved in 20 ml 1 M HNO_3 and 100 mg $(NH_4)_2Fe(SO_4)_2 \cdot 6H_2O$ was added to reduce Pu to Pu(III). Then Pu(III) was oxidized to the tetravalent state by addition of 1 ml 25% $NaNO_2$. After addition of 50 µg Nd^{3+}, Pu was co-precipitated with NdF_3 by adding 5 ml 40% HF and leaving the solution for 30 min at room temperature. Finally, the NdF_3 solution was filtered through a 0.1 µm membrane filter, dried and mounted for alpha decay counting. To stay flexible for the subsequent reprocessing of the samples for the AMS measurements following the decay counting, cellulose nitrate membrane filters (NC 10, Schleicher & Schuell, Germany) were used for the filtering step [Hrnecek et al., 2002].

Alpha Spectrometry measurements

For the alpha spectrometry measurements, low background ion-implanted silicon detectors (Ultra-AS, Ortec) with an active area of 450 mm^2 were used. All samples were placed at a distance of 3 mm from the detector at $5 \cdot 10^{-3}$ mbar pressure in the measurement chamber. The counting time was set to 40 000 s. For the evaluation of the spectra, Genie 2000 2.1 software (Canberra) was used. For these measurements, the detection limits calculated according to Currie (1968) were 0.4 mBq/sample for $^{239(40)}$Pu and ^{238}Pu.

Preparation of AMS samples

After completion of the alpha spectrometry measurements, the filter containing the Pu sample was reprocessed for AMS. We used either combustion of the cellulose nitrate filters and dissolving the residue in HCl, or alternatively dissolution of the filters in HNO_3 and wet oxidizing with H_2O_2. Plutonium was precipitated together with ~10 mg iron carrier as hydroxide, and combusted at 800°C in porcelain cups. The resultant oxides were pressed into aluminum sample holders suitable for the ion source of VERA. For all samples the chemically processed material was split into two sample holders to allow reproducibility measurements. In some cases, we also added silver powder as a binder and for increasing the electrical and thermal conductivity. However, we found no additional improvement of their performance as sputter targets in the ion source compared to the pure Fe- and Pu-oxide samples.

AMS measurements

The AMS measurement procedure used at VERA for actinides is described in detail in [Winkler et al. 2004b]. For the sputtering process of plutonium oxide with cesium, PuO^- is the most prolific negative ion, with an expected negative ion yield of 0.3% [Fifield et al. 1996]. These ions are pre-accelerated and pass the first ‘low-energy’ mass spectrometer. No significant beam currents were observed near the respective plutonium masses for those samples. After acceleration and stripping with the 3MV tandem accelerator (Model 9SDH2, NEC), the minimum charge state, which our high-energy analyzing magnet is capable to bend for such heavy ions, is 5^+. A yield of 6% was measured for stripping $^{238}U^{16}O^-$ to $^{238}U^{5+}$, and a very similar value can be assumed for Pu. Particles with different ratios of mass over charge are widely suppressed with the subsequent ‘high-energy’ mass spectrometer. Residual background ions are identified by a time-of-flight (TOF) assembly followed by an ionization chamber to measure the particle energy.

In accelerator mass spectrometry some background ion species may exist which pass through the mass spectrometers exactly like the isotopes of interest (see [Kilius et al. 1997] for a detailed discussion) and thus reach the TOF/Energy system at full intensity. In our case, ^{144}Nd, injected as $^{144}Nd^{112}X^-$ and stripped to $^{144}Nd^{3+}$, exactly matches $^{240}Pu^{16}O^-$ stripped to $^{240}Pu^{5+}$. The intensity of these ions is determined by the ^{144}Nd content of the sputter target and by the forming probability of $^{144}Nd^{112}X^-$, with ^{112}X any compound of mass 112. Usually, the TOF/Energy system can easily identify such background ions (see Figure 1), but excessive count rates would impair the identification. Fortunately, the rate of $^{144}Nd^{3+}$ observed when measuring $^{240}Pu^{5+}$ did not exceed 55 counts/s, despite Nd being used as a carrier in the micro-precipitation procedure.

The efficiency of the time-of-flight/energy detection system is 20-30%. Combining the numbers for the ionization yield, the stripping yield to the 5^+ charge state, and the detector efficiency, we can expect an overall detection efficiency of $\sim 4\times 10^{-5}$ Pu counts in the detector per Pu atom in the sputter target.

Uranium pilot beams from uranium oxide sputter targets were used to set up the accelerator. After tuning the AMS system with the ^{235}U and ^{238}U beams, the electrical fields of the mass selectors were scaled according to the various plutonium masses. At VERA, as at most other AMS facilities, simultaneous measurements of several isotopes are possible in the 'low energy' mass separator. Only one single mass can be injected into the accelerator, other masses—if sufficiently abundant—can be measured as beam currents in Faraday cups, at both low-energy and high-energy side of the AMS system. Usually such an intense isotope beam is used to trace fluctuations of the ion source output and to normalize the detector count rates of the rare species. For our plutonium samples, no isotope is sufficiently abundant for a current measurement. Thus, repeated and fast switching between measurements of the different Pu isotopes in the detector is crucial for a precise determination of the isotopic ratios. The switching time of VERA was about 15 seconds, thus a count time of about 20 to 100 seconds for each isotope yielded a reasonable duty cycle.

The typical measurement procedure was the following: The ^{242}Pu count-rate (spike material) was used to monitor the source output as well as possible machine drifts. The count rates obtained from ^{239}Pu and ^{240}Pu were sandwiched by measurements of the reference count rate of ^{242}Pu. The whole sequence was repeated several times. The first few seconds of sputtering on each target have been rejected in order to allow the source output to stabilize which obviously improved the reproducibility of the data. As mentioned above, ^{238}U has been used for tuning the AMS machine. Because of interferences of this isobar to ^{238}Pu, their concentrations have not been analyzed in this work.

Depending on the goal of the measurement, the number of ions measured and the timing was optimized. The complete measurement was repeated with the same sputter targets in a second beam time to check the reproducibility of the AMS results.

Results and discussion

Figure 1 demonstrates that the high-energy mass separator of VERA can suppress interfering ions from neighboring masses completely. Molecular isobars, which cause background in other mass spectrometric methods like ICP-MS are completely suppressed. The interference of Nd observed for ^{240}Pu could be easily separated in the TOF/Energy detection system.

In Table 1 the results of the AMS measurement are compared with those from Alpha Spectrometry for the soil samples from Mururoa and Fangataufa. In Figure 2, the $^{239(40)}$Pu/^{242}Pu activity ratios from Alpha Spectrometry are compared with the (^{239}Pu+^{240}Pu)/^{242}Pu activity ratios calculated from the plutonium concentrations measured with AMS. Generally, the data of the individual samples agree very well and an average ratio of AMS/Alpha = (1.10 ± 0.04) was obtained for the (^{239}Pu+^{240}Pu)/^{242}Pu isotope ratios. The uncertainties given represent the 1 σ error of the mean value. A possible systematic uncertainty in the AMS data could be attributed to a slightly different ion optical transmission through the AMS system due to the need for scaling of the AMS setup for the different isotopes. For future measurements, such uncertainty contributions may be reduced and quantified by normalization to a reference material of known isotopic composition (standard material). For the AMS measurements two independent run series have been performed. The individual results of those measurements agree quite well with each other; i.e. the deviation of those ratios between the two run series was of the order of a few percent. However, for one sample (11.4.4) the final value obtained from AMS and alpha spectrometry do not agree within their errors. In all single measurements the individual data from that sample gave very similar ratios, which results in small uncertainties. It is that sample which dominates the reduced chi-square value (3.7) in the comparison of AMS and Alpha Spectrometry data. If one rejects that sample then the reduced chi-square has a value of 1.6.

In contrast to alpha spectrometry, AMS can distinguish between the different isotopes ^{239}Pu and ^{240}Pu and provides additional information on the isotopic signature of the various Pu isotopes. ^{242}Pu was added as a spike in this work, therefore no environmental ratio could be determined in this case. Isotopic ratios were measured for both, the Fangataufa and the Mururoa samples. For the Mururoa samples measured in this work, a value for the ^{240}Pu/^{239}Pu ratio of 0.02 was obtained (see Tab. 1). These values agree with those given by R. Chiappini et al. (1999) who found a ratio ^{240}Pu/^{239}Pu lower than 0.03 for areas where safety tests were performed. For the Fangataufa samples (11.4.1, 11.4.3 and 11.4.4) a ratio of 0.05 was obtained, in perfect agreement with a sample from a Fangataufa sediment, where a ratio of 0.05 was also reported by R. Chiappini et al. (1999).

For the ^{238}Pu/$^{239(40)}$Pu ratios determined by Alpha Spectrometry, two distinct ratios of 0.35 ± 0.01 and 0.0051 ± 0.0006 have been found for samples from Fangataufa and Mururoa, respectively. This can be compared with the ratio of ^{238}Pu/$^{239(40)}$Pu of 0.38 found by Mulsow et al. (1999) in water samples in Fangataufa. In addition, the values obtained in this work for Mururoa samples are in good agreement with the ^{238}Pu/$^{239(40)}$Pu ratio of 0.0044 given by Danesi et al. (2002) for samples from the Faucon area.

Conclusions and Outlook:

The comparison of alpha spectrometry and AMS has shown good agreement for “true” environmental samples from two nuclear test sites. However, since the two methods are in some respect complementary, the combination of both methods yields additional information.

The instrumental effort is much higher for AMS than for alpha spectrometry, but the sensitivity is higher and it is possible to distinguish the isotopes ^{239}Pu and ^{240}Pu, which gives more insight into the neutron reassessment. With the same measurement procedure, measurements of the other Pu isotopes, ^{241}Pu, ^{242}Pu and also ^{244}Pu are possible. ^{238}Pu is obscured by interfering ^{238}U in AMS, but can be measured by alpha counting. In this context, it is planned for the future to use ^{236}Pu instead of ^{242}Pu as spike material, which will allow the determination of the environmental ^{242}Pu concentration, too. This extended isotopic fingerprint will provide strong clues on the origin of plutonium in environmental soil or sediment samples.

Figures:

Fig. 1 Time-of-flight vs. Energy spectrum obtained with the mass AMS tuned for $^{242}Pu^{5+}$ (a), $^{240}Pu^{5+}$ (b) and $^{239}Pu^{5+}$ (c). In (b), $^{144}Nd^{3+}$ is indicated with a dashed frame.

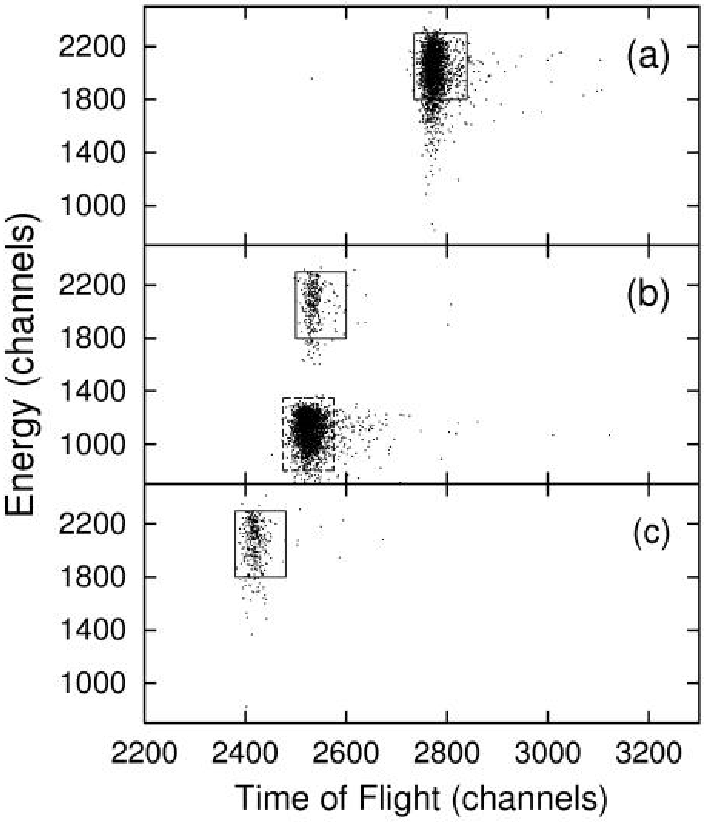


Fig. 2: Comparison of alpha spectrometry and AMS. The ordinate shows decay rates which were calculated from the plutonium concentrations measured with AMS for comparison with alpha spectrometry. The error bars plotted represent 1 σ uncertainties.

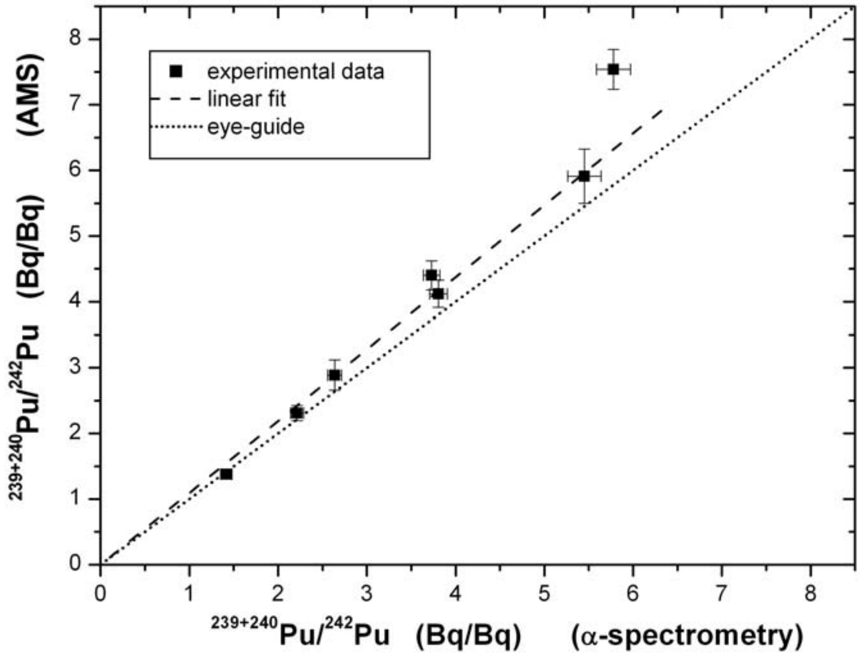
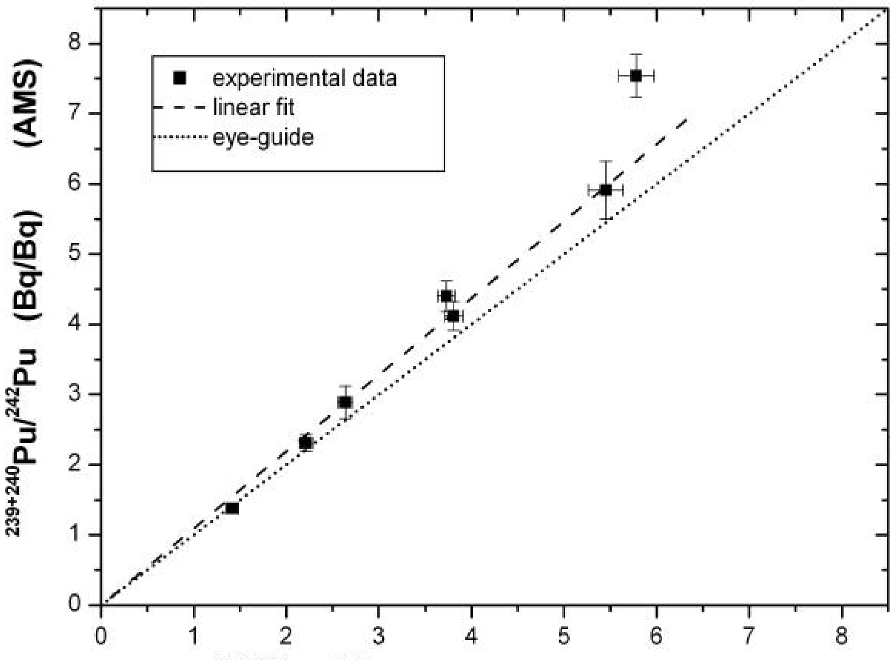


Table 1: Results obtained by AMS and Alpha Spectrometry. Errors given are ± 1 σ.

	AMS		Alpha Spectrometry	
	^{240}Pu/^{239}Pu [at/at]	$^{239(40)}$Pu [Bq/kg]	$^{239(40)}$Pu [Bq/kg]	^{238}Pu [Bq/kg]
11.4.1 Fangataufa (Kilo) loose coral rocks	0.049 ± 0.002	51.3 ± 2.6	43.5 ± 1.6	15.0 ± 0.7
11.4.3	0.049 ± 0.003	19.3 ± 0.8	19.8 ± 0.9	6.8 ± 0.4
11.4.4	0.050 ± 0.007	93.6 ± 4.7	71.8 ± 2.4	26.1 ± 1.0
9.3.6 Mururoa (Faucon) top soil	0.018 ± 0.002	175 ± 15	159.1 ± 4.9	0.77 ± 0.18
9.3.18	0.017 ± 0.002	115 ± 5	109.9 ± 3.7	0.59 ± 0.16
9.3.25	0.018 ± 0.003	221 ± 11	204.3 ± 5.4	1.21 ± 0.19
7.2.4.2 Mururoa (Colette)	0.019 ± 0.002	539 ± 39	497 ± 17	2.24 ± 0.46
7.2.4.1 (hot particle)	-[1]	-[1]	31200 ± 1000	150 ± 6

[1] Count rate limit of detection system exceeded



References

[Chiappini et al., 1996] Chiappini, R., Taillade, J.-M., Brébion, S., 1996. Development of a High-sensitivity Inductively Coupled Plasma Mass Spectrometer for Actinide Measurement in the Femtogram Range. J. Anal. Atom. Spectrom. 11, 497-503.

[Chiappini et al., 1999] Chiappini, R., Pointurier, F., Millies-Lacroix, J.C., Lepetit, G., Hemet, P., 1999. ^{240}Pu/^{239}Pu isotopic ratios and $^{239+240}$Pu total measurements in surface and deep waters around Mururoa and Fangataufa atolls compared with Rangiroa atoll (French Polynesia). Sci. Tot. Env. 237/238, 269-276.

[Currie 1968] Currie, L.A., 1968. Limits for Qualitative Detection and Quantitative Determination. Analytical Chemistry 40, 586-593.

[Danesi et al., 2002] Danesi, P.R., Moreno, J., Makarewicz, M., Radecki, Z., 2002. Residual radioactivity in the terrestrial environment of the Mururoa and Fangataufa Atoll nuclear weapon test sites. J. Radioanal. Nucl. Chem. 253, 53-65.

[Donohue, 1998] Donohue, D.L., 1998. Strengthening the IAEA safeguards through environmental sampling and analysis. Journal of Alloys and Compounds 271-273, 11-18.

[Fifield et al., 1996] Fifield, L.K., Cresswell, R.G., di Tada, M.L., Ophel, T.R., Day, J.P., Clacher, A.P., King, S.J., Priest, N.D., 1996. Accelerator mass spectrometry of plutonium isotopes. Nucl. Instr. Meth. in Phys. Res. B 117, 295-303.

[Hrnecek et al., 1999] Hrnecek, E., Irlweck, K., 1999. Determination of Americium and Plutonium in contaminated Soil Samples., In: Newton, G.W.A (Ed.), Environmental Radiochemical Analysis, Royal Society of Chemistry, Cambridge, UK, pp.73-80.

[Hrnecek et al., 2002] Hrnecek, E., Aldave de las Heras, L., Betti, M., 2002. Application of microprecipitation on membrane filters for Alpha Spectrometry, liquid scintillation counting and Mass Spectrometric determination of plutonium isotopes. Radiochim. Acta 90, 721-725.

[IAEA 1998] International Atomic Energy Agency, The Radiological Situation at the Atolls of Mururoa and Fangataufa, STI/PUB/1028, Techn. Rep. Vol. 1, IAEA, Vienna, 1998.

[Irlweck et al., 1998] Irlweck, K., Wicke, J., 1998. Isotopic composition of plutonium immissions in Austria after the Chernobyl accident. J. Radioanal. Nucl. Chem. 227, 133-136.

[Irlweck et al., 1999] Irlweck, K., Hrnecek, E., 1999. ^{241}Am concentration and ^{241}Pu/$^{239(40)}$Pu ratios in soils contaminated by weapons-grade plutonium. J. Radioanal. Nucl. Chem. 242, 595-599.

[Kilius at al. 1997] Kilius, L.R., Zhao, X.-L., Litherland, A.E., Purser, K.H., 1997. Molecular fragment problems in heavy element AMS. Nucl. Inst. Meth. in Phys. Res. B 123, 10-17.

[Mietelski et al., 1999] Mietelski, J.W., Dorda, J., Was, B.,1999. Pu-241 in samples of forest soil from Poland. Appl. Radiat. Isot. 51, 435-447.

[Mulsow et al., 1999] Mulsow, S., Coquery, M., Dovlete, C., Gastaud, J., Ikeuchi, Y., Khanh Pham, M., Povinec, P.P., 1999. Radionuclide concentrations in underground waters of Mururoa and Fangataufa Atolls. Sci Tot. Env. 237/238 287-300.

[Muramatsu et al., 1999] Muramatsu, Y., Uchida, S., Tagami, K., Yoshida, S., Fujikawa, T.,1999. Determination of plutonium concentration and its isotopic ratio in environmental materials by ICP-MS after separation using ion-exchange and extraction chromatography. J. Anal. Atom. Spectrom. 14, 859-865.

[Muramatsu et al., 2001] Muramatsu, Y., Yoshida, S., Tagami, K., Uchida, S., Rühm, W., 2001. ICP-MS analysis of environmental plutonium, In: Kudo A. (Ed.), Plutonium in the Environment - Edited proceedings of the Second Invited International Symposium. Elsevier, Amsterdam, pp.63-77.

[Oughton et al., 2001] Oughton, D., Day, P., Fifield, K., 2001. Plutonium measurements using accelerator mass spectrometry: methodology and applications. In: Kudo A. (Ed.), Plutonium in the Environment - Edited proceedings of the Second Invited International Symposium. Elsevier, Amsterdam, pp. 47-62.

[Priest et al., 1999] Priest, N. D.; Pich, G. Merlo; Fifield, L. K.; Cresswell, R. G, 1999. Accelerator mass spectrometry for the detection of ultra-low levels of plutonium in urine, including that excreted after the ingestion of Irish sea sediments. Radiation Research 152(6), S16-S18.

[Povinec et al., 2001] Povinec, P.P.,. La Rosa, J.J, Lee, S.H., Mulsow, S., Osvath, I., Wyse, E., 2001. Recent developments in radiometric and mass spectrometry methods for marine radioactivity measurements. J. Radioanal. Nucl. Chem. 248, 713-718.

[Solatie et al., 2002] Solatie, D., Carbol, P., Hrnecek, E., Jaakola, T., Betti, M., 2002. Sample preparation methods for the determination of plutonium and strontium in environmental samples by low level liquid scintillation counting and α-spectrometry. Radiochim. Acta 90, 447-454.

[Vockenhuber at al., 2003] Vockenhuber, Ch., Ahmad, I., Golser, R., Kutschera, W., Liechtenstein, V., Priller, A., Steier, P., Winkler, S. Accelerator Mass Spectrometry of heavy long-lived radionuclides. International Journal of Mass Spectrometry 223-224 , 713-732.

[Wallner et al. 2000] Wallner, C., Faestermann, T., Gerstmann, U., Hillebrandt, W., Knie, K., Korschinek, G., Lierse, C., Pomar, C., Rugel, G., 2000. Development of a very sensitive AMS method for the detection of supernova-produced longliving actinide nuclei in terrestrial archives. Nucl. Instr. Meth. in Phys. Res. B 172, 333-337.

[Winkler et al., 2004a] Winkler, S., Ahmad, I., Golser, R., Kutschera, W., Orlandini, K.A., Paul, M., Priller, A., Steier, P., Vockenhuber, C., 2004. Anthropogenic ^{244}Pu in the environment. New Astronomy Reviews 48, 151-154.

[Winkler et al., 2004b] Winkler, S., Ahmad, I., Golser, R., Kutschera, W., Orlandini, K.A., Paul, M., Priller, A., Steier, P., Valenta, A., Vockenhuber, C., 2004. Developing a detection method of environmental ^{244}Pu. Nucl. Instr. Meth. in Phys. Res. B, in press.

[Yamamoto et al., 1996] Yamamoto, M., Tsumura, A., Katayama, Y., Tsukatani, T., 1996. Plutonium Isotopic Composition in Soil from the Former Semipalatinsk Nuclear Test Site. Radiochim. Acta 72, 209-215.