

Sequential separation and determination of plutonium, americium-241 and strontium-90 in soils and sediments

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(Received September 1, 1997)

A sensitive and reliable method for the sequential separation and determination of plutonium, ^{241}Am and ^{90}Sr in soil samples was developed. Plutonium was separated by a Microthene-TNOA column. Then ^{90}Y (for ^{90}Sr determination) was separated from americium by a HDEHP column after elimination of large amounts of interfering stable or radioactive nuclides (iron, ^{210}Bi and ^{210}Po etc.) by an oxalate precipitation and a Microthene-TNOA column. Finally americium was purified by another HDEHP column and a PMBP-TOPO extraction. A special attention was paid to the decontamination of Pu and Am from ^{210}Po and of ^{90}Y from ^{210}Bi ; the relevant decontamination factors resulted greater than 10^5 , 10^6 and 10^4 respectively. The detection limits were 1.2 mBq/kg for Pu and 1.7 mBq/kg for ^{241}Am and 0.32 Bq/kg for ^{90}Sr . The procedure was checked by analyzing three certified samples supplied by IAEA. Some Italian soil samples were also analyzed giving average yields of $84.9 \pm 7.2\%$ for Pu, $57.8 \pm 3.2\%$ for Am and $96.7 \pm 1.6\%$ for Y; the $^{239+240}\text{Pu}$, ^{238}Pu , ^{241}Am and ^{90}Sr contents (Bq/kg) ranged from 0.347 to 1.53, from 0.013 to 0.048, from 0.126 to 0.556 and from 2.89 to 11.6 respectively and the average ratios were 0.037 ± 0.017 for $^{238}\text{Pu}/^{239+240}\text{Pu}$, 0.357 ± 0.040 for $^{241}\text{Am}/^{239+240}\text{Pu}$ and 7.0 ± 1.2 for $^{90}\text{Sr}/^{239+240}\text{Pu}$.

Introduction

Interest in the levels of the artificial radionuclides in the environment has been elevated in recent years by the Chernobyl accident, as the accumulation of these hazardous radionuclides in the environment started after the first nuclear weapon test and has continued ever since. A particular attention has been given to $^{239+240}\text{Pu}$, ^{238}Pu , ^{241}Am , ^{137}Cs and ^{90}Sr , which have longer half-lives and represent the greatest hazard for mankind. The purpose of the present study is to develop a method to sequentially separate and determine plutonium, ^{241}Am , and ^{90}Sr in soil samples. ^{137}Cs was excluded as it can be measured easily by γ -ray spectrometry without any chemical separation. Although $^{239+240}\text{Pu}$, ^{238}Pu , ^{241}Am and ^{90}Sr levels from fallout can be detected in soil and other environmental samples, their contents are often so low that large sample quantities are needed together with very selective radiochemical procedures. Unless these radionuclides are isolated in a very pure form, one or more of the natural alpha or beta emitters (U, Th, ^{210}Pb , ^{210}Bi and ^{210}Po etc.) can lead to spurious results. The majority of the methods described in the literature can be applied only for individual radionuclide or element analysis.^{1–4} Based upon our previous work,^{4,5} further experiments have been carried out for a sequential separation procedure. By using the present method some environmental soil samples and three IAEA reference materials have been analyzed to check the suggested procedure. The results showed that the technique can give reliable figures; moreover it is sensitive and selective and it can save manpower and reduce the analytical costs.

Experimental

Apparatus and reagents

The plutonium and americium sources were counted by alpha spectrometry (Silena, Italy) with a counting efficiency of 31% and a background of $< 6 \cdot 10^{-6} \text{ s}^{-1}$ over the interested energy regions. The ^{90}Y sources were measured by a low background ($6.2 \cdot 10^{-3} \text{ s}^{-1}$) β -counter (C Canberra-2404, USA) with a counting efficiency of 23%. The electroplating apparatus and the chromatographic columns were described previously.¹

The ^{242}Pu , ^{243}Am , ^{210}Pb , ^{210}Bi and ^{210}Po standard solutions, Microthene-710 (microporous polyethylene, 50–100 mesh) and KL-HDEHP [Di(2-ethylhexyl)phosphoric acid] resin (60–100 mesh, 50% HDEHP) were supplied by Amersham (G.B.), S.I.C. (Italy) and C.N.I.C. (P.R. China), respectively. $\text{Sr}(\text{NO}_3)_2$, $\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$, $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ and TeCl_4 were used to prepare the carriers for ^{90}Sr , ^{90}Y , ^{210}Bi and ^{210}Po carriers separately. Tri-n-octylamine (TNOA), tri-octyl-phosphine oxide (TOPO), 1-phenyl-3-methyl-4-benzoyl-5-pyrazolone (PMBP), diethylene-triaminopentaacetic acid (DTPA), L-(+)-Lactic acid (LA) 88–92% and all the other reagents were analytical grade (Carlo Erba, Italy).

Column preparation

Columns A and B: Both column A and B were Microthene-TNOA columns. 20 ml 20% TNOA in toluene (v/v) were added to 16 g Microthene; the mixture was stirred for several minutes to get a

homogeneous product, which was then placed in an oven at 40–50 °C to evaporate toluene and to obtain a porous powder. 3.5–4.0 g of the Microthene-TNOA powder was mixed with 3 ml concentrated HCl and a small quantity of water and transferred into a column. Column A was conditioned with 20 ml 7.5M HNO₃ for plutonium extraction and column B with 20 ml 2M HCl for iron, bismuth and polonium extraction.

Columns C and D: Columns C and D were both KL-HDEHP resin columns. Each contained 3 g KL-HDEHP resin which was immersed into 0.1M HNO₃ for 24 hours. Column C was used for yttrium and americium extraction and separation and column D for americium separation from the rare earth elements.

Preliminary tests

The preliminary tests for sequential determination of plutonium, ²⁴¹Am and ⁹⁰Sr in soil were mainly based on the procedures reported in References 4 and 5.

Leaching: It is confirmed that leaching of Am and Y from soil by a single 7.5M HNO₃ or 6M HCl is complete;⁴ on the contrary this is not true for Pu leaching, especially as far as sediment samples of low salinity were concerned. For instance, when the IAEA-368 sediment was analyzed, the experimental results were about 25% lower than the recommended value (31 Bq/kg). The loss of plutonium could be attributed to the incomplete leaching. According to the literature,^{6,7} joint leachings with concentrated HNO₃ and HCl are thought to be more effective, but they introduce more interfering agents to the leachate, raising new problems for the next separation step. Later on, it was tried to leach by a 7.5M HNO₃ + 0.2M HF solution and satisfactory results were obtained (Table 3). In such a condition, the HF solution is not very harmful to the glassware.

Leachate filtration and Pu decontamination from ²¹⁰Po: The leachate filtration seems to be only a simple physical step and many analysts often ignore its importance for any separation and purification of the concerned radionuclides. During our experiments, it was found that in a simulated solution the decontamination factor (DF) of Pu from ²¹⁰Po with a Microthene-TNOA column was higher than 10⁵, but this value was not confirmed with the leachate. According to our observation and results, any insoluble silicate present in the leachate is very toxic for the Microthene-TNOA column and it does make a great challenge for a radioanalytical procedure: as a matter of fact it can block the column, lower the column capacity and the Pu yield, adsorb small particulates and ions and make the separation incomplete. Sometimes silicate can also play a role in the third phase formation in the successive Am extraction with 0.05M PMBP + 0.025M TOPO in cyclohexane.

Suitable procedures to eliminate the insoluble particulates are either the centrifugation of the leachate by using high speed centrifuge ($\geq 1 \cdot 10^4$ rpm) or the filtration. Good results were obtained by filtration with Millipore filter paper (VCWP-04700: pore size of $\leq 0.1 \mu\text{m}$). Table 1 and 2 show the experimental conditions and the relevant DFs of Pu from ²¹⁰Po (added activity: $2.47 \cdot 10^4$ dpm). The results illustrate that after leachate filtration the DF increased for at least one order of magnitude reaching the value of $1.55 \cdot 10^5$. Moreover this effort also increased the plutonium yield.

The decontamination of Pu from ²¹⁰Po can be further improved by adding 2 mg Te⁴⁺ as the ²¹⁰Po carrier because of its similar chemical behavior to polonium. In such a condition, the results show that nearly all the ²¹⁰Po can be eliminated.

Decontamination of ⁹⁰Y from ²¹⁰Bi: The determination of ⁹⁰Sr in environmental samples requires the isolation of ⁹⁰Sr or its daughter ⁹⁰Y from several interfering ions. In recent years, a variety of methods have been described for effecting the necessary separations, such as precipitation,^{8–10} liquid-liquid extraction,^{11,12} ion-exchange^{8,9,13} and chromatography.¹⁴ Most of these procedures, however, suffer from various limitations. Precipitation, for example, is harmful and tedious and it must often be repeated several times to obtain adequate recoveries of strontium. Liquid-liquid extraction is too cumbersome for use with large numbers of samples and often requires the use of toxic organic solvents. Ion-exchange procedures typically require careful pH control, as satisfactory separations are achieved only with a narrow pH range. In a previous paper,⁴ a chromatographic method was developed for the rapid determination of ⁹⁰Sr in soils, with a special attention to the separation of ⁹⁰Y from the most critical natural interfering radionuclide ²¹⁰Bi. The advantage of this method is that ²¹⁰Bi can be eliminated as Bi₂S₃ giving accurate results. It was found that Y can also be separated from Bi either by TIOA extraction in 1–6M HCl or by tri-n-butyl phosphate extraction in concentrated HNO₃.¹⁵ The former technique creates a possibility to combine the ⁹⁰Sr determination method with that for Pu and Am determination in which a special Microthene-TNOA column was used for elimination of ²¹⁰Po and iron. Table 2 shows the decontamination factors of ⁹⁰Y from ²¹⁰Bi (added activity: $2.47 \cdot 10^4$ dpm) at different experimental conditions. From these results two important conclusions can be drawn: (1) by using the Microthene-TNOA column instead of Bi₂S₃ precipitation the DF ($1.94 \cdot 10^4$) is one order of magnitude higher than that in the previous procedure⁴ and (2) the yttrium precipitation as oxalate in a HCl medium is also an effective technique to improve the decontamination effect.

Table 1. Conditions of decontamination experiments

Condition No.	Column A	Column B	Column C	Column D
1	Washing with 30 ml 10M HNO ₃ , 120 ml 10M HCl, 130 ml 7.5M HNO ₃	Feeding solution in 50 ml 4M HCl	Feeding solution pH 1; the column was washed with 40 ml; 1M HNO ₃ and 50 ml 1M HCl and eluted with 50 ml 6M HNO ₃	Feeding solution pH 2; the column was washed with 60 ml 0.1M HNO ₃ and eluted with 20 ml 0.07M DTPA-1M LA
2	Washing with 40 ml 4M HNO ₃ , 120 ml 10M HCl, 100 ml 10M HNO ₃	Feeding solution in 50 ml 2M HCl	Feeding and washing solutions were same as condition 1, but the column was eluted with 6M HCl	Same as condition 1
3	Feeding solution was filtered with VCWT-04700 millipore filter paper (0.1 μm); the column was washed with 30 ml 10M HNO ₃ , 150 ml 10M HCl and 130 ml 7.5M HNO ₃	Same as condition 2	Same as condition 1	Same as condition 1
4	Same as condition 3	Same as condition 2	Same as condition 1, but after elution, 4 ml conc. HCl were added	Same as condition 1

Table 2. Decontamination factors of Pu and Am from ²¹⁰Po and of ⁹⁰Y from ²¹⁰Bi at different experimental conditions (50 g soil)

Condition No.	Pu yield, %	DF _{Pu/Po}	DF _{Am/Po}	Y yield, %	DF _{Y/Bi}
1	46.9	8.10·10 ³	1.28·10 ⁶	86.4	3.25·10 ²
2	81.3	4.00·10 ³	1.67·10 ⁶	89.4	1.24·10 ⁴
3	89.7	1.01·10 ⁵	3.05·10 ⁶	79.1	1.47·10 ⁴
4	93.0	1.55·10 ⁵	3.32·10 ⁶	97.7	1.94·10 ⁴

In the liquid-liquid extraction, lead is not very strongly extracted by TIOA (distribution coefficient ~4.5).¹⁵ In the case of extraction chromatography the situation should be much better in general. In order to observe the possible interference of ²¹⁰Pb passing through the column B to the effluent, a 30 g soil sample containing 0.957 Bq ²¹⁰Pb was analyzed, but the effluent after column B was stored for 6 days. The results show that the source prepared in this way is also following the ⁹⁰Y decay and it is confirmed that no detectable amount of ²¹⁰Bi remains in the effluent. As the ²¹⁰Pb contents in soil vary from sample to sample, it is still suggested that after ²¹⁰Bi and ²¹⁰Po retention by column B the effluent should be passed through the column C in good time to separate the ⁹⁰Y from ⁹⁰Sr in order to avoid any possible ²¹⁰Bi build-up from ²¹⁰Pb decay, especially when the ⁹⁰Sr activity is low.

Americium elution: The Am elution from column C by 1M HNO₃ gives a good separation from Y¹⁶. Column D is used for Am separation from rare earth elements by using a 0.07M DTPA + 1M LA solution at pH 3.0. The Am elution profile shows that the first 5 ml

of the DTPA+LA solution contain undetectable amount of Am and that 20 ml can elute nearly all the Am.

Americium extraction from DTPA + LA solution: In Reference 5 the DTPA+LA elution solution was decomposed by H₂O₂ before Am extraction with a PMBP + TOPO solution; during this operation a small quantity of vinyl acetate forms which can be burned automatically at high temperature. Therefore, this step is tedious and critic. Actually the decomposition of DTPA is not necessary, as it is a very strong complexing agent only at pH>3. This has been confirmed by the following experiment.

3 ml of 0.07M DTPA + 1M LA solution each having different pH and containing ²⁴³Am (0.7 dpm) were placed in a 10 ml extraction tube and added with 3 ml of 0.05M PMBP + 0.025M TOPO. The extraction was carried out by shaking the tube for 5 min; after washing with 0.1M HNO₃, Am was stripped by 5M HNO₃, electroplated in oxalic and nitric acid medium and counted by alpha spectrometry. The results of Fig. 1 show that more than 80% of Am is extracted by the organic solution at pHs from 0.75 to 2.0.

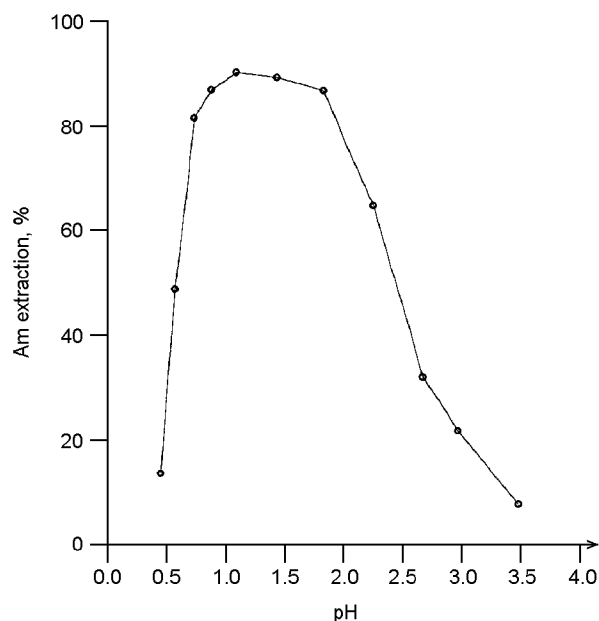


Fig. 1. Effect of pH on the Am extraction by 0.05M PMBP + 0.025M TOPO in cyclohexane from 0.07M DTPA + 1M LA solution

Decontamination of Am from ^{210}Po : Table 2 shows the decontamination factors of Am from ^{210}Po at four different experimental conditions: in any case the DF values were higher than 10^6 .

Recommended procedure

Plutonium determination: 30 g of dried ($110\text{ }^\circ\text{C}$) and ground soil ($\leq 0.154\text{ mm}$) are kept in a muffle at $600\text{ }^\circ\text{C}$ for 2 hours. After cooling, 20 mg Sr^{2+} , 10 mg Y^{3+} , 2 mg Te^{4+} as the carriers and known activities of ^{242}Pu and ^{243}Am (0.01–0.03 Bq) are added and two leachings are carried out with 75 ml 7.5M HNO_3 + 0.2M HF for 20 min each. The leaching solutions are centrifuged and combined. 2 ml of 1M $\text{NH}_2\text{OH}\cdot\text{HCl}$ are added to obtain Pu(III), which is then oxidized to Pu(IV) by adding 5 ml of 8M NaNO_2 . The solution is evaporated at $90\text{--}95\text{ }^\circ\text{C}$ to destroy the excess of NaNO_2 and it is filtered with a Millipore filter paper (VCWP-04700). The 4–6M HNO_3 leaching solution is passed through column A at a flow rate of 0.8 ml/min for the plutonium retention; then the column is washed with 40 ml 7.5M HNO_3 and the two effluents are kept for ^{241}Am and ^{90}Sr analyses. After a further washing with 120 ml 10M HCl and 120 ml 7.5M HNO_3 , plutonium is eluted with 18 ml 0.025M $\text{H}_2\text{C}_2\text{O}_4$ + 0.15M HNO_3 at a flow rate of 0.1 ml/min and it is electro-plated on a stainless steel disk at pH 1.5–2.0 for 90 minutes at 550 mA/cm^2 current density. Plutonium activity, is measured by alpha spectrometry.

Strontium determination: 40–50 g of oxalic acid are added to the collected HNO_3 effluent. After heating, the

solution is adjusted to pH 3.0 with concentrated ammonia to obtain the oxalate precipitation. The precipitate is centrifuged or filtered and washed with 50 ml 1% $(\text{NH}_4)_2\text{C}_2\text{O}_4 + \text{H}_2\text{C}_2\text{O}_4\cdot\text{H}_2\text{O}$. The solution is discarded and the precipitate is transferred into a ceramic crucible and ashed at $800\text{ }^\circ\text{C}$ for 2 hours. 1 mg of Bi^{3+} carrier is added to the residue which is then dissolved by concentrated HCl and 30% H_2O_2 . The solution is evaporated to eliminate the excess of HCl and then diluted with 40 ml 2M HCl. After cooling, the solution is passed through column B at a flow rate of 1 ml/min to retain bismuth, polonium and the remaining iron, then the column is washed with 30 ml 4M HCl. The effluent and the washing solution are joined, adjusted to pH 1 with concentrated ammonia and passed through column C at a flow rate of 0.8 ml/min to retain yttrium. The $^{90}\text{Sr}\text{--}^{90}\text{Y}$ separation time is recorded. The column is first washed with 40 ml 1M HNO_3 , then washed with 50 ml 1M HCl at a flow rate of 0.8 ml/min and finally eluted with 50 ml 6M HNO_3 at a flow rate of 0.3 ml/min. The effluent and the first washing solution are collected for the ^{241}Am determination. The elution solution is evaporated to about 20 ml and, after cooling, 3 ml concentrated HCl and 5 ml saturated oxalic acid solution are added. The pH is adjusted to 1.5–2.0 with concentrated ammonia to obtain the yttrium oxalate precipitation. The precipitate is filtered, dried and counted in a low background β -counter. The counting time is recorded. The chemical yield of yttrium can be obtained either by weighing the yttrium oxalate precipitate or by complexometric titration. The calculation of ^{90}Sr activity in soil through ^{90}Y has been described in detail elsewhere.⁴

Americium determination: The americium portion from column C is adjusted to pH 2 with concentrated ammonia and it is passed through column D at a flow rate of 0.8 ml/min. After washing with 60 ml 0.1M HNO_3 , americium is eluted with 20 ml 0.07M DTPA + 1M LA (pH 3.0) at a flow rate of 0.1 ml/min. The eluate is collected in a 60 ml extraction tube and concentrated HNO_3 is added to adjust the pH to 1.0. Then 20 ml 0.05M PMBP + 0.025M TOPO in cyclohexane are added and the americium extraction is carried out by shaking the tube for 5 min. After centrifuging, the aqueous phase is discarded and the organic phase is washed with a same volume of 0.1M HNO_3 . The organic phase is diluted with 8 ml benzene and americium is stripped twice with 20 ml 5M HNO_3 each. The stripping solution is evaporated to dryness and the organic matter is destroyed with H_2O_2 . The residue is transferred into the electrolytic cell by using 10 ml 0.025M $\text{H}_2\text{C}_2\text{O}_4$ + 0.1M HNO_3 and americium is electroplated as described for plutonium and counted by α -spectrometry.

Results and discussion

Quality control

The recommended procedure was checked by analyzing three certified samples supplied by IAEA. The results of Table 3 show that the $^{239+240}\text{Pu}$ and ^{238}Pu values obtained in all the three samples are in good agreement with the recommended values. The ^{238}Pu content found in the IAEA-367 sample is lower than the information value; this could be due to its lower content and greater counting statistic error. The ^{241}Am content in IAEA-368 sample is in good agreement with the information value. A higher ^{241}Am content in IAEA-367 sample has been observed than the recommended value and this could be due to the newly born ^{241}Am from ^{241}Pu . ^{90}Sr contents obtained in IAEA Soil-6 and IAEA-367 samples are also in good agreement with the recommended values. A particular attention should be paid to the obtained ^{90}Sr content (0.56 Bq/kg) in IAEA-368 sample which is much lower than the information value (1.8 Bq/kg). Although we cannot judge the quality of the information value reported by IAEA, this figure is most likely unbelievable due to its too large confidence interval. According to our experience, a lower ^{90}Sr value is more credible as ^{90}Sr determinations often give high values if the analytical procedures are not prepared to eliminate nearly all the ^{210}Bi and other natural beta emitters. After comparing all the obtained results with the IAEA recommended values, the general comment can be made that the recommended procedure for plutonium, ^{241}Am and ^{90}Sr sequential determination can give reliable results.

Applicability of the method

Some uncultivated surface soil samples collected near Urbino (Central Italy) in November 1996 were analyzed by using the recommended procedure. The results of Table 4 show that the mean yields are $84.9\pm 7.2\%$ for plutonium, $57.8\pm 3.2\%$ for americium and $96.7\pm 1.6\%$ for yttrium, respectively. If compared with other reported methods,^{17,18} the recommended procedure for plutonium determination is rather simple, and it gives higher plutonium yields which make the method more sensitive. The yields for americium are not so high, but they are reasonable taking into account the many separation steps for eliminating iron, ^{210}Po and rare earth elements. Yttrium yields for ^{90}Sr analyses in all the determined samples are quantitative; such an advantage makes this method more sensitive and competitive than the others. The $^{239+240}\text{Pu}$, ^{238}Pu , ^{241}Am and ^{90}Sr contents (Bq/kg) in these samples range from 0.347 to 1.53, from 0.013 to 0.048, from 0.126 to 0.556 and from

2.89 to 11.6, respectively. The average ratios of $^{238}\text{Pu}/^{239+240}\text{Pu}$, $^{241}\text{Am}/^{239+240}\text{Pu}$ and $^{90}\text{Sr}/^{239+240}\text{Pu}$ are 0.037 ± 0.017 , 0.357 ± 0.040 and 7.0 ± 1.2 , respectively.

The obtained α -spectra (Fig. 2) show that the Pu and Am sources are quite pure, and that the recommended procedure has good decontamination features also from natural uranium, thorium and other α -emitters.

The purity of ^{90}Y sources was checked by following its radioactive decay ($T_{1/2}=64.1$ h); so far, more than 30 soil and sediment samples have been analyzed and all the sources followed the ^{90}Y decay with the average half life of 64.3 ± 1.3 h (Fig. 3), even at relatively low ^{90}Sr content (0.5 Bq/kg).

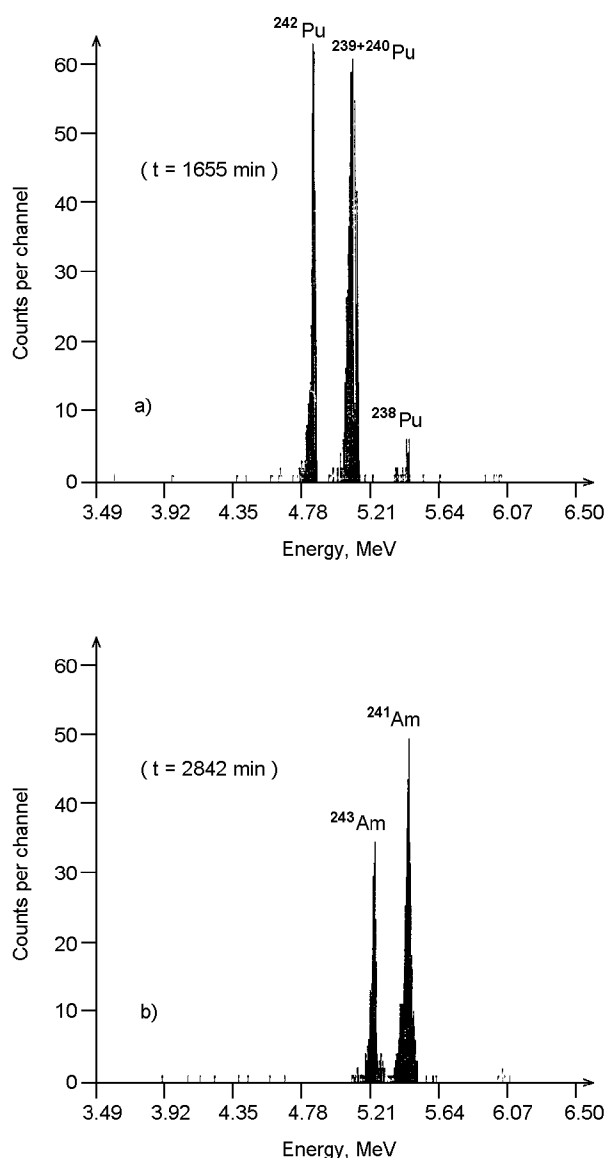


Fig. 2. Plutonium (a) and americium (b) α -spectra for a soil sample collected in Central Italy

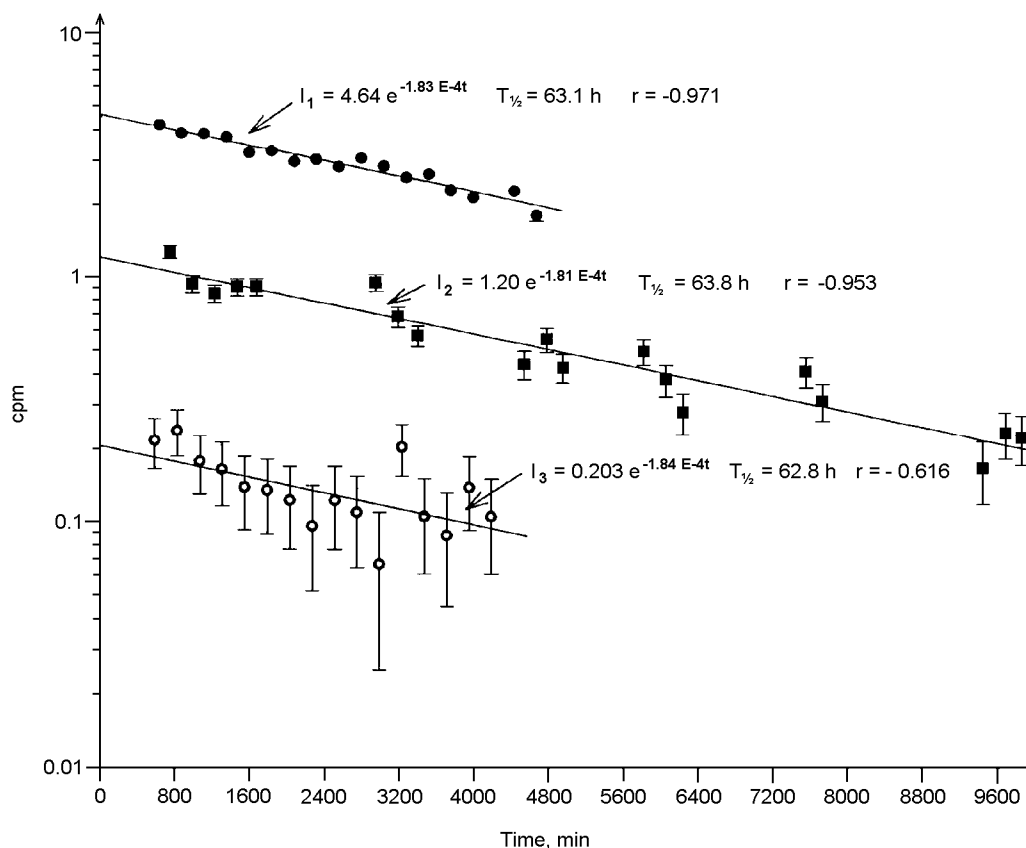


Fig. 3. Decay curves of the ^{90}Y sources obtained for three soil samples

It should also be pointed out that the recommended procedure is applicable only to these samples where ^{90}Y reached the equilibrium with its parent ^{90}Sr . The interference of other yttrium and strontium radionuclides is negligible and the ^{91}Y interference can be eliminated by the procedure described in Reference 4.

Detection limits

Taking into account the blank count rate, the counting efficiency of the instrument, the chemical or radiochemical yield and the soil quantity for each analysis, the detection limits are 1.2 mBq/kg for plutonium, 1.7 mBq/kg for ^{241}Am and 0.32 Bq/kg for ^{90}Sr .

Conclusions

In the present method some critic steps have been eliminated or improved. The decontamination factors of Pu and Am from ^{210}Po and of ^{90}Y from ^{210}Bi are $1.55 \cdot 10^5$, $3.32 \cdot 10^6$, and $1.94 \cdot 10^4$, respectively. The application practice shows that it provides a very economic, selective, sensitive and reliable technique for the environmental survey program dealing with the most toxic, long-lived and world-wide contaminated artificial radionuclides.

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Table 3. $^{239+240}\text{Pu}$, ^{238}Pu , ^{241}Am and ^{90}Sr concentrations (in Bq/kg) in IAEA reference materials (20 g and double test)

Sample No.	$^{239+240}\text{Pu}$			^{238}Pu			^{241}Am			^{90}Sr	
	Pu yield, %	Experimental value	Recommended value (confidence interval)	Experimental value	Recommended value (confidence interval)	Am yield, %	Experimental value	Recommended value (confidence interval)	Y yield, %	Experimental value**	Recommended value (confidence interval)
IAEA/Soil-6	79.6	1.02 ± 0.07	1.04 (0.96–1.11)	0.031 ± 0.008	–	55.8	0.354 ± 0.033	–	97.8	28.2 ± 0.8	$30.3 (24.4-31.7)$
IAEA-367 Sediment	82.5	39.4 ± 1.6	38 (34.4–39.8)	0.020 ± 0.007	0.08 (–)*	68.1	28.0 ± 1.5	26.4 (24–27.7)	98.9	120 ± 2	102 (62–129)
IAEA-368 Sediment	85.7	33.9 ± 1.6	31 (29–34)	8.44 ± 0.46	8.5 (7.6–8.9)	67.4	1.23 ± 0.11	1.3 (1.2–1.5)*	99.3	0.56 ± 0.28	1.8 (0.45–6.9)*

* Information value.

** The results have been adjusted to the declared reference date.

Table 4. $^{239+240}\text{Pu}$, ^{238}Pu , ^{241}Am and ^{90}Sr concentrations (in Bq/kg) in soil samples collected in the Urbino area (Central Italy)

Sample No.	$^{239+240}\text{Pu}$			^{238}Pu			^{241}Am			^{90}Sr	
	Pu yield, %	Experimental value	Recommended value (confidence interval)	Experimental value	Recommended value (confidence interval)	Am yield, %	Experimental value	Recommended value (confidence interval)	Y yield, %	Experimental value**	Recommended value (confidence interval)
1	91.8	0.368 ± 0.033	0.014 ± 0.005	61.4	0.126 ± 0.020	95.7	3.00 ± 0.22				
2	83.0	0.347 ± 0.022	0.023 ± 0.004	58.2	0.150 ± 0.018	95.7	2.89 ± 0.21				
3	88.0	1.25 ± 0.076	0.026 ± 0.006	55.2	0.436 ± 0.037	97.5	7.44 ± 0.33				
4	92.9	1.53 ± 0.085	0.048 ± 0.008	61.7	0.556 ± 0.042	95.0	11.6 ± 0.4				
5	76.0	0.657 ± 0.043	0.013 ± 0.004	53.9	0.221 ± 0.024	99.3	4.23 ± 0.26				
6	77.7	0.546 ± 0.043	0.025 ± 0.007	56.3	0.174 ± 0.019	96.8	2.92 ± 0.24				
Mean $\pm \sigma$:	84.9 ± 7.2			57.8 ± 3.2		96.7 ± 1.6					
Range:		0.347–1.53	0.013–0.048		0.126–0.556		2.89–11.6				