

Uranium comparison by means of AMS and ICP-MS and Pu and ^{137}Cs results around an Italian Nuclear Power Plant

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Abstract. Italy built and commissioned 4 nuclear power plants between 1958-1978, which delivered a total of 1500 MW. All four were closed down after the Chernobyl accident following a referendum in 1987. One of the plants was Garigliano, commissioned in 1959. This plant used a 160 MW BWR1 (SEU of 2.3 %) and was operational from 1964 to 1979, when it was switched off for maintenance. It was definitively stopped in 1982, and is presently being decommissioned. We report here details on the chemistry procedure and on the measurements for soil samples, collected up to 4.5 km from the Nuclear Plant. A comparison between uranium (^{238}U) concentration as determined by means of AMS (Accelerator Mass Spectrometry) and by ICP-MS (Inductively Coupled Plasma-Mass Spectrometry) techniques respectively at the ANU (Australian National University) and at the Ecowise company in Canberra, Australia, is reported, as well as ^{236}U and $^{239,240}\text{Pu}$ concentration results detected by AMS. $^{236}\text{U}/^{238}\text{U}$ and $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratios by means of AMS are also provided. A contamination from Chernobyl is visible in the $^{137}\text{Cs}/^{239+240}\text{Pu}$ activity ratio measurements.

1 Introduction

Anthropogenic long-lived alpha-emitting radionuclides have been (and still are) released into the environment by nuclear tests, nuclear accidents and operations of fuel reprocessing and plant decommissioning. Among these, the actinides (e.g. ^{236}U and $^{239,240}\text{Pu}$ isotopes) are the most significant, along with the historical ^{137}Cs γ -emitter. Quantifying the actinides releases and tracing their dispersion in the environment has traditionally been the task of alpha spectrometry or, more recently, of the mass spectrometry technique. Although these are mature methodologies, each has its limitations, which are surmounted by the relatively new technique of AMS [1, 2].

Individual aspects have to be taken into account where a quantification of ^{236}U and $^{239,240}\text{Pu}$ isotopes is required. The chemistry is relatively simple for uranium (substantially the only requirement is that enough uranium has to be extracted) and, the ^{236}U concentration can be determined relative to a ^{233}U spike material and/or to the stable ^{238}U isotope. Detection is the principal challenge confronting the quantification of the ^{236}U concentrations, owing to the presence of interfering ^{238}U and ^{235}U background ions. A complex detection system is required for ^{236}U , e.g. a Time of Flight-Energy system [3].

In the case of $^{239,240}\text{Pu}$ isotopes the chemical separation and purification of plutonium from the natural amounts of uranium is of great importance, and as there

are no macroscopic levels of other plutonium isotopes present, measurements are normalized to a ^{242}Pu spike. In contrast, the detection of the different Pu isotopes can be done effectively with a simple ionization chamber since only a measurement of the total energy is required for the identification.

The work reported here was motivated by the desire to lower the level of risk perception by the surrounding population by showing that the cumulative effect of the operation and decommissioning of the plant on the nearby environment has been negligible. The Center for Isotopic Research on Cultural and Environmental heritage (CIRCE), recently upgraded for actinides measurements [4–6], and the ANU [7] in collaboration with SoGIN (the Nuclear Plant Management Company) started a research program to measure the concentration and isotopic ratios of U and Pu isotopes in and around the Garigliano Nuclear Power Plant (GNPP) [8]. The measurements employed AMS, and were applied to the analysis of both soil (environmental) and concrete (structural) samples to quantify and determine the origin of any ^{236}U or $^{239,240}\text{Pu}$. In this paper we present results on soil samples, collected up to 4.5 km from the plant and a comparison between the uranium concentration as determined by using ICP-MS by Ecowise and by means of the AMS technique at the ANU (the AMS detailed measurement results for ^{236}U and $^{239,240}\text{Pu}$ isotopes for some of the samples are reported in [8]). Evidence of fallout from the Chernobyl accident is present in our ^{137}Cs gamma activity measurements [8, 9].

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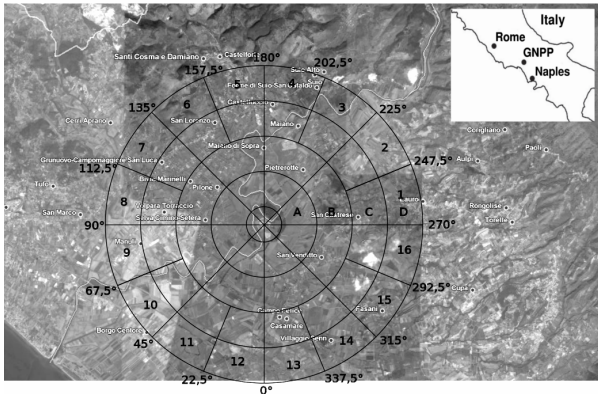


Figure 1. Grid around the Garigliano NPP that is in the center of the crown, see text.

2 Actinide measurements

The sampling program has been described in [8]. Briefly, the area surrounding the GNPP has been divided into circular crowns each of 1 km radius, from an internal radius of 500 m from the NPP out to 4.5 km (Figure 1). The two internal crowns were divided into 8 areas and the outer two crowns into 16 areas, for a total of 48 sampling points. The crowns are identified by a letter of the alphabet, Table 1. The soils are a black loam. The top 15 cm is quite friable. At 7 cm, there is a small pocket of light-coloured material that is more sandy/fine gravel. Below 15 cm, the soil gets harder and denser, and has more pebbles.

A sampling point (BSC2) in the plain of Sele, Salerno province, which is remote from, but in an area geologically similar to the Garigliano NPP, was also analysed for comparative purposes. From the 48 sampling locations 9 were selected for the AMS measurement.

2.1 Sample preparation

The soil preparation chemistry was performed at ANU [10]. Briefly, the soils were accurately weighed and dried in an oven at 110 °C for 24 hours, then homogenized and sieved at 2 mm diameter. The sieved part was then weighed. The 2 mm diameter fractions were combusted at 450 °C for 16 hours in order to remove the organic components of the soil.

For each sample about 20 g of the combusted fraction was weighed and known amounts of ^{242}Pu (4pg, equivalent to 10^{10} atoms of ^{242}Pu) and ^{233}U (1 pg, equivalent to $3 \cdot 10^9$ atoms of ^{233}U) spikes were added and then placed into the oven to dry at ~ 80 °C overnight. The sample was then transferred to a glass beaker, and approximately 60-70 ml of 8M HNO_3 added and the beaker was placed on a hotplate, at 80-90 °C, to leach out U and Pu isotopes for 24 hours.

The sample was then transferred to a centrifuged tube and centrifuged for 5 minutes at 3000 rpm. The supernatants were transferred to a fresh centrifuge tube and 15

ml 8M HNO_3 was added to the residues. The tubes containing the residue were then shaken thoroughly and re-centrifuged at 3000 rpm for 5 minutes. The supernates were combined and placed in a metal block and dried on a hot plate. Unfortunately three of the samples (A910, C5 and D16) lost material as a result of overheating during the drying phase and as a result of trying to recover loose material the possibility of some level of U contamination cannot be ruled out. About 15 ml of 3M HNO_3 was then added to dissolve the dried samples.

Each solution was weighed and a 1 ml aliquot set aside for ICP-MS analysis. The aliquot was diluted with 28 ml 18M Ω MQ water and the uranium concentration determined using ICP-MS at Ecowise. The remaining supernatant was aside for column separation.

Uranium separation using UTEVA resin

Each solution was passed through a pre-conditioned column containing 2 g of UTEVA resin. About 5 ml 9M HCl was then added to the column. A small amount of any Np(IV), if present, will be eluted at this step. The waste beaker under the column was replaced with a clean 30 ml Teflon vial and about 20 ml 5M HCl/0.05M $(\text{COOH})_2$ was added to the column. This step removes Th/Pu/Np from column, and *the eluent solution is retained for plutonium separation*. A new clean 30 ml Teflon vial was placed under the column and 30 ml of 0.01M HCl was added to the column and left to drain. *This elutes the uranium*. The solution was allowed to dry at 70-80 °C in the heating block. *The uranium was ready for target preparation*. Once the yellow U material is dry about 1.2 mg of Fe was added as $\text{Fe}(\text{NO}_3)_3$, in order to increase the mass of the sample and dried down again at 70-80 °C. The solution was baked at 800 °C for 8 hrs and then mixed with Ag-powder roughly at a ratio of 1 to 4 by weight and pressed into an aluminium sample holder.

Extraction of Pu using an anion-exchange resin

Each solution containing Pu was dried to a white (oxalic acid) salt at 70-80 °C in the heating block on the hot plate. About 10 ml 8M HNO_3 was then added to the vials and the salt cake dissolved while the vial was still warm. The sample solution was then added to a pre-conditioned column containing 1.7 g of BioRad AG 1X8 100-200 mesh chloride resin. Once the solution had drained it was followed by 25 ml of 8M HNO_3 (to elute contaminating elements including U, Cl, Mg and Fe), and then 70 ml of 12M HCl to elute Th and Np. A new clean 30 ml Teflon vial was placed under the column. *Plutonium was then eluted from the resin with 25 ml of a freshly prepared solution of 0.1 M $\text{NH}_4\text{I}/12$ M HCl*. The elutant was taken to dryness and a small amount (2 ml) of HNO_3 added to remove iodine. When the solution turned clear 2 ml of HCl was added to remove ammonium nitrate. The solution was then evaporated to dryness. A further 1 ml of concentrated HNO_3 , to evolve chlorine, and approximately 1.2 mg of Fe as $\text{Fe}(\text{NO}_3)_3$ solution was added and the vial contents taken to dryness at 85 °C. The resulting powder was baked at 800 °C for 8 hrs. The sample was mixed with Ag-powder roughly at a ratio of 1 to 4 by weight and was pressed into an aluminium sample holder.

2.2 ANU measurement procedure

In this section a brief description of the various steps of the U and Pu isotope measurements obtained with the 14UD ANU Tandem Accelerator are given [1]. Before the measurement, a tuning of the transport elements up to the final detector is required in order to maximize the ion optical transmission. A Thorium beam is used for this purpose and in order to have a good negative ion yield, molecular negative ions of $^{232}\text{Th}^{16}\text{O}^-$ are extracted from the ion source. The $^{232}\text{Th}^{16}\text{O}^-$ ions are accelerated to injection energy of $E_{inj} = 100$ keV and undergo mass analysis prior to entering the accelerator.

Once in the accelerator the ions are accelerated by the positive high voltage towards the gas stripper, where they lose electrons and gain high positive charge states, leading to full molecular dissociation. The positive atomic ions are then accelerated a second time by the same potential in the high energy tube of the tandem. For $^{232}\text{Th}^{5+}$, this results in an energy of $E = 24.424$ MeV with a terminal voltage of $V = 4.098$ MV. The stripping yield is about 3%. For heavy ion tuning, the object and image slits of the injection magnet are closed to ± 1 mm, the slits of the analyzing magnet are closed to ± 1.25 mm and a collimator of 3 mm is used if high selectivity is required just after the Wien filter. For actual measurements, the object and image slits of the injection magnet are opened to ± 2 mm, the slits of the analyzing magnet are opened to ± 3 mm and the collimator is out.

For uranium measurements, once the setup for the pilot $^{232}\text{Th}^{5+}$ beam is established, the fields of the injection magnet, the terminal voltage of the accelerator and the electric field of the Wien filter are scaled to $^{238}\text{U}^{5+}$ for a fine tuning and then to the other wanted masses. For $^{236}\text{U}/^{238}\text{U}$, the measurement procedure is composed of two loops of three steps. Each loop consists of integration of the $^{238}\text{U}^{5+}$ beam current for 10 s in the Line Cup, counting of $^{236}\text{U}^{5+}$ ions for 5 min in the TOF-E system and a final $^{238}\text{U}^{5+}$ integration. For ^{233}U (tracer), ^{234}U and ^{236}U , the measurement procedure is composed of two loops of four steps. The isotope sequence would usually start with the reference isotope ^{233}U followed by ^{234}U and ^{236}U , and finishing with ^{233}U . All of them are counted with the TOF-E system. The typical counting intervals were 1 minute for ^{233}U , 1 minute for ^{234}U and 5 minutes for ^{236}U .

For plutonium measurements, once the setup for the pilot $^{232}\text{Th}^{5+}$ beam is found, since $^{238}\text{U}^{5+}$ may cause interference for $^{239}\text{Pu}^{5+}$, the fields of the injection magnet, the terminal voltage of the accelerator and the electric field of the Wien filter are scaled to the Pu masses: ^{239}Pu , ^{240}Pu and ^{242}Pu (spike). The measurement procedure is composed of two loops of four steps; the isotope sequence would usually start with the reference isotope ^{242}Pu followed by ^{240}Pu and ^{239}Pu , and finishing with ^{242}Pu . In this case all isotopes are counted with a multiple electrode ionization chamber that is routinely used for measurements of ^{239}Pu isotopes. The typical counting intervals were 1 minute for ^{242}Pu , 5 minutes for ^{240}Pu and 3 minutes for ^{239}Pu .

Table 1. Distance (D) from the GNPP and comparison between the amount of uranium in the cathode, obtained by ICP-MS, and $^{238}\text{U}^{5+}$ beam current obtained by AMS.

Sample	D km	ICP-MS U mass (μg)	AMS $^{238}\text{U}^{5+}$ (nA)	$^{238}\text{U}^{5+}/\text{U}$ nA/ μg
A78	1.2	23.3	0.34	0.014
A910	1.3	9.9	0.12	0.012
B56	1.9	32.9	0.33	0.010
B34	2.0	64.4	0.46	0.007
C7	2.8	55.1	0.54	0.010
C13	3.0	43.5	0.41	0.009
C5	3.1	1.4	0.03	0.022
D16	3.9	32.6	0.57	0.018
D12	4.0	26.7	0.50	0.019
BSC2	131	20.6	0.20	0.010

Table 2. Uranium concentrations as determined by ICP-MS and AMS.

Sample	ICP-MS $\mu\text{g}/\text{g}$ of soil	AMS $\mu\text{g}/\text{g}$ of soil	AMS/ICP-MS
A78	1.23	1.18 ± 0.04	0.96 ± 0.03
A910	0.56	1.95 ± 0.11	3.48 ± 0.20
B56	1.79	2.08 ± 0.07	1.16 ± 0.04
B34	3.79	3.77 ± 0.14	1.00 ± 0.04
C7	3.19	4.13 ± 0.14	1.29 ± 0.04
C13	2.27	2.38 ± 0.13	1.05 ± 0.06
C5	0.08	1.52 ± 0.18	19.0 ± 2.3
D16	1.74	4.95 ± 0.21	2.84 ± 0.12
D12	1.59	1.63 ± 0.04	1.03 ± 0.03
BSC2	1.10	1.00 ± 0.03	0.91 ± 0.03

3 Actinide Results

For the Garigliano NPP environmental samples, gamma spectrometry of natural (^7Be , ^{40}K) and artificial (^{60}Co , ^{137}C) radionuclides, with a high energy resolution, low background germanium detector has been performed [9]. We present the ^{236}U and $^{239,240}\text{Pu}$ isotope results obtained at ANU for samples with the highest ^{137}C activity. A comparison of the amount of uranium (^{238}U) in the cathode, obtained by ICP-MS, and the $^{238}\text{U}^{5+}$ beam currents is shown in Table 1 and exhibit a clear correlation. Results for samples A910 and C5 suggest they have lost a significant fraction of the total uranium, while for sample D16 the uranium content and AMS beam is similar to those from the remaining samples. Excluding samples A910, C5 and D16 the $^{238}\text{U}^{5+}$ current per μg of U ranges from 0.007 to 0.019 nA/ μg .

Since a spike of ^{233}U was added to each sample, it is possible to determine the U concentrations from the AMS measurements independently of the Ecovise analysis. This is achieved by comparing the ^{233}U and ^{234}U count rates and using the $^{234}\text{U}/^{238}\text{U}$ natural abundance ratio of 5.5×10^{-5} . A comparison between the Ecovise ICP-MS values and ANU AMS values is shown in Table 2. Uranium contamination is clearly evident in samples A910, C5 and D16. The agreement for 7 remaining samples is good and ranges from 0.91 to 1.29.

Table 3. Comparison of the $^{236}\text{U}/^{238}\text{U}$ ratios obtained directly from the $^{238}\text{U}^{5+}$ current and from ^{234}U counts.

Sample	$^{236}\text{U}/^{238}\text{U}$ $\times 10^{-9}$	$^{236}\text{U}/^{238}\text{U}$ (^{234}U) $\times 10^{-9}$	Ratio
A78	4.61 ± 0.20	5.13 ± 0.34	0.90 ± 0.07
A910	1.97 ± 0.23	1.78 ± 0.25	1.11 ± 0.21
B56	6.03 ± 0.24	7.16 ± 0.43	0.84 ± 0.06
B34	2.18 ± 0.19	2.85 ± 0.21	0.76 ± 0.07
C7	0.94 ± 0.07	0.95 ± 0.08	0.99 ± 0.11
C13	1.30 ± 0.09	1.28 ± 0.19	1.02 ± 0.17
C5	-	7.53 ± 1.62	-
D16	0.45 ± 0.05	0.43 ± 0.06	1.05 ± 0.18
D12	11.70 ± 0.27	13.30 ± 0.60	0.88 ± 0.04
BSC2	4.67 ± 0.27	3.99 ± 0.28	1.17 ± 0.11

Table 4. Concentrations of ^{236}U , ^{239}Pu and $^{240}\text{Pu}/^{239}\text{Pu}$ ratios.

Sample	^{236}U conc. $\times 10^6$ ats/g	^{239}Pu conc. $\times 10^6$ ats/g	$^{240}\text{Pu}/^{239}\text{Pu}$
A78	15.4 ± 0.9	106.8 ± 3.1	0.21 ± 0.01
A910	8.8 ± 1.2	29.5 ± 5.2	0.14 ± 0.10
B56	37.7 ± 1.9	342.5 ± 8.6	0.19 ± 0.01
B34	27.2 ± 1.7	85.5 ± 5.4	0.22 ± 0.05
C7	10.0 ± 0.8	32.1 ± 2.6	0.17 ± 0.05
C13	7.7 ± 1.1	92.3 ± 5.1	0.21 ± 0.02
C5	26.8 ± 3.4	165.7 ± 43.3	-
D16	5.4 ± 0.7	37.2 ± 22.1	-
D12	54.9 ± 2.0	396.4 ± 7.2	0.17 ± 0.01
BSC2	10.1 ± 0.7	95.6 ± 6.2	0.19 ± 0.02

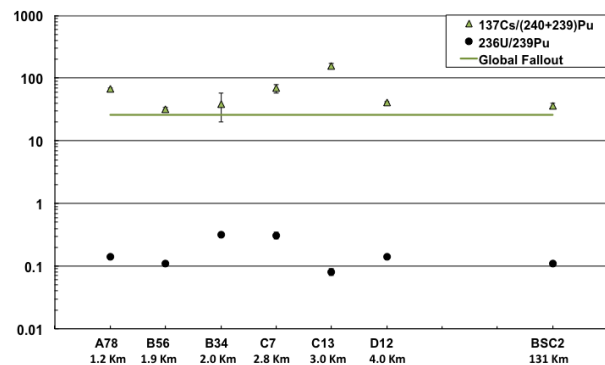
Table 3 shows a comparison of $^{236}\text{U}/^{238}\text{U}$ ratios derived directly from the AMS measurement with that determined from the ^{234}U counting rate and the natural $^{234}\text{U}/^{238}\text{U}$ ratio. Good agreement is clearly evident (the ratio ranges from 0.76 to 1.17) and the $^{236}\text{U}/^{238}\text{U}$ ratios range from 10^{-9} to 10^{-8} . Notably, the ratios are consistent with global fallout; they agree with sediment results from the Garigliano NPP drain channel and Garigliano river, which range from 0.6 to 11×10^{-8} as reported in [11]. Moreover, the values are comparable with the BSC2 sample, from the plain of Sele, Salerno province, which is geologically similar to but ~ 100 km distant from the GNPP.

Plutonium measurements for these samples typically provided ^{242}Pu rates that ranged from ~ 100 to 2500 counts per minute and, at the lower end of the range, are significantly lower than typical rates of ~ 4000 per minute from the blank samples to which only the 4 pg of ^{242}Pu spike had been added. Since the UTEVA column was used to extract the uranium in addition to the anion-exchange column normally used for plutonium, it is possible that some Pu has been lost in the chemistry. Further investigations are required. Nevertheless, sufficient Pu remained for reliable measurements.

The concentrations of ^{236}U and ^{239}Pu in the 10 soil samples, as well as the $^{240}\text{Pu}/^{239}\text{Pu}$ ratios are shown in Table 4. For all of the samples, the ^{236}U concentration has been derived from the relative count rates of ^{233}U and ^{236}U . For the 7 unmelted samples, essentially the same re-

Table 5. $^{236}\text{U}/^{239}\text{Pu}$ atom ratios and $^{137}\text{Cs}/^{239+240}\text{Pu}$ activity ratios from around the Garigliano NPP.

Sample	$^{236}\text{U}/^{239}\text{Pu}$	$^{137}\text{Cs}/^{239+240}\text{Pu}$
A78	0.14 ± 0.01	67.4 ± 3.0
B56	0.11 ± 0.01	32.4 ± 1.2
B34	0.32 ± 0.03	38.7 ± 18.8
C7	0.31 ± 0.04	68.9 ± 11.1
C13	0.08 ± 0.01	158.1 ± 13.1
D12	0.14 ± 0.01	40.7 ± 1.1
BSC2	0.11 ± 0.01	36.7 ± 3.5

**Figure 2.** $^{137}\text{Cs}/^{239+240}\text{Pu}$ activity ratio and the $^{236}\text{U}/^{239}\text{Pu}$ atom ratio. The average global fallout value for the $^{137}\text{Cs}/^{239+240}\text{Pu}$ activity ratio is shown as a solid line.

sults are obtained multiplying the $^{236}\text{U}/^{238}\text{U}$ ratio for the ^{238}U concentration of the Ecowise. For the three melted samples, however, only the relative counting rates of ^{233}U and ^{236}U method is applicable. The ^{239}Pu concentrations have been derived from the counting rates relative to ^{242}Pu .

The ^{236}U and ^{239}Pu concentrations (as well as those for ^{137}Cs), are quite variable, but, follow the same trend as a function of distance from the NPP. Furthermore, the average isotopic concentrations are of the same order as the more remote sample BSC2. The $^{240}\text{Pu}/^{239}\text{Pu}$ ratios are consistent with the northern hemisphere average fallout of ~ 0.18 [11]. The $^{236}\text{U}/^{239}\text{Pu}$ atom ratio is also variable, from 0.08 to 0.32 (Table 5 and Figure 2). This may reflect a different depth distribution of ^{239}Pu and ^{236}U as a consequence of different chemical mobilities in the surface soil [12].

Importantly, Table 5 and Figure 2 show that significant variability is also observed in the $^{137}\text{Cs}/^{239+240}\text{Pu}$ activity ratio, which varies between 37 and 158, and is consistently higher than the global fallout ratio of ~ 26 (as corrected for ^{137}Cs decay to October–November 2008, when the gamma-ray measurements were performed) [13]. It is likely that this variability reflects the influence of the Chernobyl accident which deposited significant amounts of ^{137}Cs , but not Pu or ^{236}U , in this part of Italy [11].

4 Summary

The AMS methodology is a powerful technique to measure isotopic ratios and concentrations of actinides and provides measurements over a wide range of isotopic ratios.

The data presented here for soil samples shows very good agreement for the U concentration results between the ICP-MS and AMS methodologies, with ratios that range from 0.91 to 1.29.

The $^{236}\text{U}/^{238}\text{U}$ soil sample ratios measured at ANU are somewhat variable, ranging from 10^{-9} to 10^{-8} , but are close to that measured at the BSC2 site and can most likely be attributed to global fallout. The $^{240}\text{Pu}/^{239}\text{Pu}$ ratios, show clearly northern-hemisphere average fallout values of ~ 0.18 . The values are in agreement with the sediment results from the Garigliano NPP drain channel and Garigliano river reported earlier [11]. Moreover both values are comparable with the BSC2 sample from the plain of Sele, an area geologically similar to that around the Garigliano NPP.

The data presented here indicate values consistent with global fallout and additional ^{137}Cs contribution from Chernobyl. The $^{137}\text{Cs}/^{239+240}\text{Pu}$ activity ratio varies between 37 and 158, but all the samples show higher values than the ~ 26 typical of global fallout. It is possible, although unlikely, that actinides were released from the NPP and deposited only at distances greater than 4.5 km from the plant. Further measurements are needed to ensure that this was not the case and that the soil actinide concentration results are indeed representative of soils at greater distances from the NPP.

This study has demonstrated the potential of $^{239,240}\text{Pu}$ and ^{236}U isotopes to serve as tracers of soil loss and as sensitive indicators of the release of material from nuclear facilities.

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