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The first use of ^{236}U in the general environment and near a shutdown nuclear power plant

F. Quinto [a,b,c,*], P. Steier [a], G. Wallner [d], A. Wallner [a], M. Srncik [d], M. Bichler [e], W. Kutschera [a], F. Terrasi [b,c], A. Petraglia [b,c], C. Sabbarese [b,c]

[a] Vienna Environmental Research Accelerator (VERA), Fakultät für Physik - Isotopenforschung, Universität Wien, Währinger Strasse 17, Wien A-1090, Austria
[b] Dipartimento di Scienze Ambientali, Seconda Universitá di Napoli, via Vivaldi 43, 81100 Caserta, Italy
[c] Center for Isotope Research on Cultural and Environmental Heritage (CIRCE), Italy
[d] Institut für Anorganische Chemie, Universität Wien, Währinger Strasse 42, Wien A-1090, Austria
[e] Atominstitut der Österreichischen Universitäten (ATI), Österreichischen Universitäten, Stadionallee 2, Wien A-1020, Austria

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ABSTRACT

We present a first effort to investigate ^{236}U in the environment near a shutdown nuclear power plant far away from highly contaminated sites, by using accelerator mass spectrometry. The detection limit of about 1 pg ^{236}U allowed us to identify a minimal increase of the ^{236}U/^{238}U isotopic ratio correlated to a peak of ^{137}Cs in river sediments downstream of the nuclear power plant, and to detect anthropogenic ^{236}U also upstream, where it is probably not related to the power plant but to global fallout. The ^{236}U content shoved variations of the ^{236}U/^{238}U isotopic ratio in relation to the chemical–physical characteristics of the sediments. This demonstrates the potential of ^{236}U as an environmental tracer, and as an indicator for releases from nuclear facilities.

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1. Introduction

^{236}U/^{238}U has been demonstrated to be a valuable tool to identify an anthropogenic origin of a radionuclide contamination. Environmental studies were up to now focused on sites affected by high levels of contamination: soil from the Semipalatinsk-21 test site (Beasley et al., 1998); intertidal sediment from the North Irish Sea affected by the Sellafiels nuclear reprocessing plant discharges (Marsden et al., 2001); soil and river sediment from the vicinities of uranium processing factories (Ketterer et al., 2003); several biological and environmental samples from downstream of the Mayak Production Association (Boretzen et al., 2005); soil in the 30 km relocation area of Chernobyl NPP (Mironov et al., 2002) and up to a distance of 200 km from Chernobyl (Boulyga and Heumann, 2006).

The dominant mode of ^{236}U formation in reactors is the capture of a thermal neutron by ^{235}U a secondary contribution from the alpha decay of ^{240}Pu is negligible. Several orders of magnitude of difference between the ^{236}U/^{238}U isotopic ratios in naturally occurring uranium (10^{-9}–10^{-14}) (Steier et al., 2008) and in spent nuclear fuel (10^{-2}–10^{-4}) imply that also a small contamination from irradiated nuclear fuel in a natural sample is able to increase the ^{236}U/^{238}U significantly. The long half-life renders decay-counting techniques inapplicable. The only mass spectrometric technique presently able to measure ^{236}U/^{238}U at natural background levels is accelerator mass spectrometry (AMS). The setup at the Vienna Environmental Research Accelerator (VERA) at the University of Vienna is able to achieve detection limits of 10^{-13} (Steier et al., 2008).

River sediments may be considered as a (at least temporary) sink for much of the material which passes through the various cycles operating on the earth's surface. In this study, we have regarded the sediment of the Garigliano river as a potential environmental archive for the radionuclides released with the liquid effluents of the Garigliano Nuclear Power Plant (GNPP).

The GNPP, presently in the decommissioning phase, started operation in 1964, and was shut down in 1978 after a defect at a secondary vapor generator. With a thermal power of 506 MW, the plant reached an electric power of 160 MWe, producing a total of 12.5×10^9 kW h. It incorporated a boiling water cooled and moderated nuclear reactor. The nuclear fuel was made of uranium dioxide, UO_2 enriched, on average at 2.3% ^{235}U (also mixed oxide fuel, UO_2+PuO_2, was used). During the operation phase and the later decommissioning activities, the airborne effluents were filtered and discharged into the atmosphere through a chimney 100 m high. Liquid effluents were purified and drained into a channel which connects the plant to the river (Fig. 1). Between 1978 and 2000, the airborne and liquid releases of fission and

* Corresponding author at: Dipartimento di Scienze Ambientali, Seconda Università di Napoli, via Vivaldi 43, 81100 Caserta, Italy. Tel.: +39 0823 274814; fax: +39 0823 274814.
E-mail addresses: francesca.quinto@unina2.it, francesca.quinto@univie.ac.at (F. Quinto).

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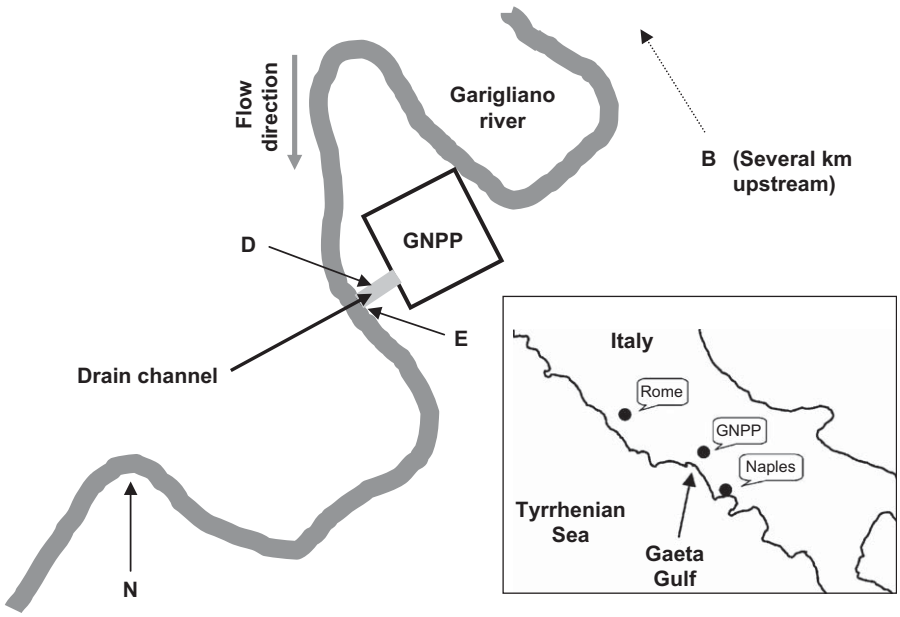

Fig. 1. Schematic representation of the sampling points; the insert shows the location of the Garigliano Nuclear Power Plant in Southern Italy.

activation products, and alpha emitters from the GNPP, were estimated to be always below the radiological dose limits (Documento Nr. 0001, 2001). In an investigation on sediments from the Mediterranean Sea (Delfanti et al., 1995), the authors speculate that the

power plant could be responsible for a twofold increase of the plutonium content in the Gulf of Gaeta (Fig. 1) relative to the average global fallout concentration, and mention a small radioactivity leak in the period from 1969 to 1976.

In addition to this local source, the Garigliano river system also received input of radioactive material from global fallout and from Chernobyl fallout which is introduced into the river both directly and through erosion of the soils and sediments of the area. The global distribution of the fallout from atmospheric test explosions reflects the atmospheric transport of debris from the Stratosphere to the Troposphere with maximum transfer at the mid-latitudes (UNSCEAR, 2000). ^{236}U in global fallout has been detected by Ketterer et al. (2007) and recently by Sakaguchi et al. (2009). The Liri-Garigliano basin is situated across the latitude of 41°N, therefore the deposition density expected for this area would be close to the maximum in the Northern Hemisphere, which was estimated to be around 3.3 kBq/m^2 for ^{90}Sr, while in the Mediterranean Sea the global fallout deposition of ^{137}Cs corrected for decay was estimated to be 2.8 kBq/m^2 (UNSCEAR, 2000). The global and the Chernobyl fallout patterns are influenced by the global precipitation pattern (Arnalds et al., 1989). The Liri-Garigliano basin is an area where rather low deposition of radionuclides occurred as a consequence of the Chernobyl fallout with respect to other European areas; an estimation of 10 kBq/m^2 of ^{137}Cs in this area is presented by Cort (1998), in agreement with the measurement of 8.1 kBq/m^2 for the average Chernobyl fallout ^{137}Cs in the close Campania region (Roca et al., 1989). The Chernobyl accident did not contribute significantly to the actinides inventory in the Mediterranean Sea. After the accident, Whitehead et al. (1988) estimated that the deposition of $^{239(249)}$Pu from Chernobyl was 0.01% of the global weapons testing fallout. Most of the plutonium and other actinides, which were released, were deposited close (less than 100 km) to the stricken reactor (Chernobyl forum Expert Group, IAEA, 2006), and the ^{236}U concentration in soils from sampling sites located around 200 km from Chernobyl is $(3–5)\times10^{-13}$ g/g (Boulyga and Heumann, 2006).

We can, therefore, assume that, in the area under study, anthropogenic ^{236}U might originate from global fallout and from the GNPP, while we can expect ^{137}Cs to be detectable from global fallout, from Chernobyl and possibly from the GNPP. We want to emphasize that the present work represents a first study on the applicability of ^{236}U as an environmental tracer. It is not intended to be a thorough study of the releases and environmental distribution of radioactive contamination from the GNPP, which would require well-dated depth profiles, comprehensive ^{137}Cs and ^{90}Sr measurements, and more emphasis on the plutonium isotopes.

2. Materials and methods

Four sediment cores were investigated in the present work (Fig. 1):

D—collected directly in the GNPP's drain channel (41°15′24.6″N; 13°49′55.6″E).
E—just downstream from the mouth of the channel (1 m downstream) (41°15′24.18″N; 13°49′55.28″E).
N—1 km downstream (41°15′07.65″N; 13°49′28.4″E).
B—several km upstream as a background sample for the GNPP, and expected to be representative for global fallout.

The drain channel was built by excavating a soil section and flooring the base with concrete; during the decommissioning, the daily flux of liquid effluents from the plant became much less frequent and also the flow decreased; this allowed the accumulation of sediment due to backflow of the river into the drain channel. The core E was collected just at the edge between the drain channel and the river. For the samples B and N, the sampling sites were chosen in correspondence of point bars (the inner side of a bend in the river), where deposition of sediment mostly occurs. The sampling was performed by driving polyethylene tubes (1 m length and 3.5 cm inner diameter) into the sediment.

No information on the sedimentation rate is available for the river banks investigated.

2.1. Gamma spectroscopy

The strategy for the analyses involved a quick screening measurement to scope out the distribution of ^{137}Cs concentrations. The sediment cores, without any sample preparation, were subjected to gamma spectroscopy scanning at 1 or 2 cm steps. These measurements were normalized for the detection efficiency by measuring a known source of ^{137}Cs of almost the same density and arranged within the same containers as the sediment cores.

This γ-scanning of the core was performed by using a lead shielded hyperpure germanium γ-ray detector (HPGe) (70% efficiency and 1.8 keV resolution at 1332 keV). Typical sample masses were 10 g of wet sediment; counting times varied between 1 h and 3 days according to the counting statistic. The concentration of ^{137}Cs was used to prioritize the AMS analysis of the samples. Where maxima and minima of the ^{137}Cs specific activity could be identified, the cores were cut accordingly.

2.2. Separation technique

The extraction of uranium was performed following the procedures described in Krtil et al. (1975), adapted to our needs. The sediments were dried in a muffle furnace at 110 °C for 24 h, homogenized and combusted at 450 °C overnight. ^{232}U (86 mBq $= 2.7 \times 10^8$ atoms; $t_{1/2} = 68.94$ years) was added as radiotracer to determine the radiochemical yield of the extraction. The samples were leached in 7.2 M HNO_3+0.5 g $NaNO_2$ for at least 3 h by boiling on a hot plate. The volume of reagents used depends on the sample mass (ranging between 2 and 12 g). In the subsequent description the volumes of reagents relate to 10 g of sample, for which 50 ml leaching solution was used. Uranium was purified through a two step anion exchange extraction. In the first step uranium was separated from plutonium using Dowex 1×2 (6 ml, 100–200 μm particle size, previously conditioned with 7.2 M HNO_3). Uranium was expected to pass through, while plutonium as Pu(IV) was detained on the column. The column was washed three times with 15 ml 7.2 M HNO_3 to ensure all U was collected. Thorium was then eluted as $ThCl_4$ using 30 ml of 32% HCl and discarded. At this point the U fraction contained still a large amount of iron of the original sample material which passed through the ion exchange separation. Therefore, the solution was evaporated to dryness and redissolved in 20 ml 8 M HCl. Iron was eliminated by diisopropyl ether liquid–liquid extraction, and the solution was brought to a second anion exchange column (Dowex 1×2, 100–200 μm particle size, in 8 M HCl). Calcium and residual thorium were washed out with 50 ml 8 M HCl before uranium was eluted with 90 ml 0.1 M HCl.

2.3. Alpha spectroscopy of uranium

Since AMS always measures the isotopic ratio ^{236}U/^{238}U, an independent determination of the ^{238}U content is required to calculate the ^{236}U concentration. The purified uranium solutions were each split into two subsamples for AMS and alpha-spectrometry, respectively. Uranium fractions were co-precipitated with NdF_3 (Hindman, 1983) and deposited on membrane filters suitable for alpha spectroscopy. The measurements were performed using a Passivated Implanted Planar Silicon Detector, Model 7401 VR, Canberra/Packard. The measurement time was between 3 and 4 days. The ^{238}U is expected to originate from the natural uranium content of the sediment.

2.4. Instrumental neutron activation analysis

Extraction by leaching does not recover the complete amount of uranium from the sample. To assess the extraction yield, the total uranium concentration on five layers per core was determined by instrumental neutron activation analysis (INAA). The samples (each sized 100 μg of washed sediments) were irradiated together with international certified reference materials for 2 h in the dry irradiation tube R1 of the TRIGA Mark II-reactor at the Atominstitut of the Technical University Vienna, Austria, at a thermal neutron flux of about 1.7×10^{12} cm^{-2} s^{-1}. After a decay time of 5 days, a γ-spectrum was measured to obtain the activities of ^{239}Np, the decay product of ^{239}U, which had been produced during neutron irradiation. The gamma-spectrometry was performed with a 222 cm^3 HPGe-detector (1.78 keV resolution at 1332 keV; 48.2% relative efficiency).

2.5. AMS measurements of ^{236}U/^{238}U

The AMS measurements were carried out at the 3-MV tandem accelerator facility VERA at the University of Vienna, Austria. The chemical form of the AMS sample was uranium oxide embedded in iron oxide, obtained by iron hydroxide co-precipitation (0.5 mg iron was added for each sample) and subsequent conversion to oxide by combustion at 700 °C. Uranium fractions in the iron oxide matrix were mixed with a similar volume of silver powder and pressed in aluminum sample holders suitable for the cesium sputter ion source of VERA. Uranium was sputtered as UO^-. After acceleration and gas stripping, charge state 5+ was chosen (5% yield for $^{238}U^{5+}$ using O_2 as stripper gas). Particles with different ratios of mass-over-charge are well 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. The efficiency of the TOF/energy detection system is 20–30%.

2.6. Supportive measurements of $^{239+240}$Pu and ^{240}Pu/^{239}Pu

To assess the source of a viable contamination by nuclear fuel independently, for some of the samples alpha-spectrometry measurements of the $^{239+240}$Pu activity and AMS measurements of the ^{240}Pu/^{239}Pu isotopic ratio were performed. The procedures are similar to those used for ^{236}U, and are more closely described in Srncik et al. (2008). The measurements done for the present work were not intended to represent a thorough investigation of the plutonium isotopes in the Garigliano river system.

3. Results and discussion

3.1. ^{137}Cs

In the drain channel core D, the average ^{137}Cs specific activity was 465 ± 38 mBq/g wet mass. An almost uniform distribution of the ^{137}Cs specific activity along the sediment profile was observed. In the 1 m downstream core E, the average activity was 319 ± 55 mBq/g wet mass. Two peaks can be observed at (21–10) and (33–26) cm depths; in the latter peak a ^{137}Cs specific activity of 1200 mBq/g wet mass is reached (solid line in Fig. 2). In the other two cores, the ^{137}Cs specific activity was below the detection limits.

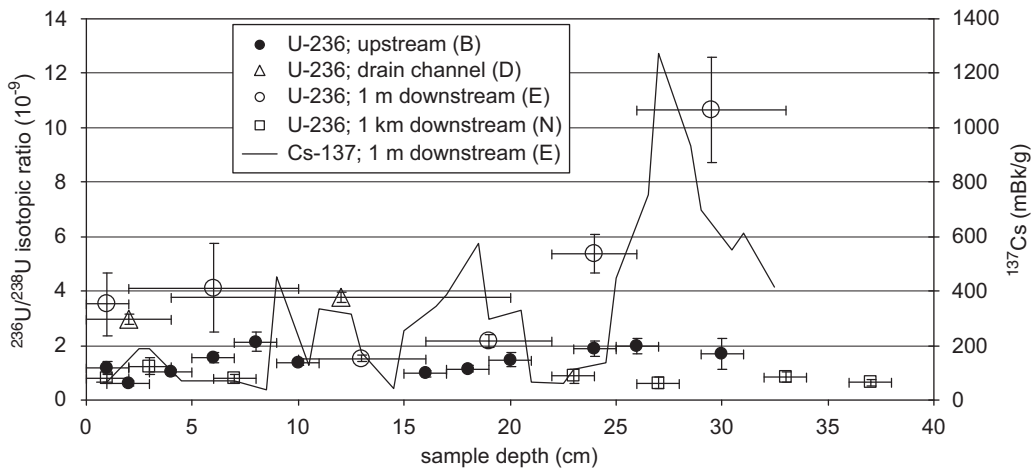

Fig. 2. $^{236}U/^{238}U$ isotopic ratios vs. depth for all four cores. The horizontal error bars indicate the span of the core section, the vertical error bars are the measurement uncertainty. The ^{137}Cs activity is shown for the 1 m downstream core E (solid line).

Table 1
^{238}U average concentration measured by neutron activation analysis and alpha spectroscopy, indicated as total and leachable ^{238}U, respectively.

Sediment core	Total ^{238}U (ppm)	Leachable ^{238}U (ppm)
B (upstream)	2.1 ± 0.2	0.84 ± 0.02
D (drain channel)	2.7 ± 0.1	0.71 ± 0.03
E (1 m downstream)	2.3 ± 0.2	0.77 ± 0.02
N (1 km downstream)	2.2 ± 0.1	0.9 ± 0.3

3.2. Uranium

The total ^{238}U concentration measured by INAA is almost uniform (spanning between 1.8 and 3 ppm for 20 subsamples) for the four cores (Table 1).

The uranium fraction extracted by our chemical separation (Table 1) was determined by alpha spectroscopy; on average $(0.8 \pm 0.2) \times 10^{-6}$ g of uranium was extracted per g of combusted sediment; this corresponds to 35% of the total uranium content. In general, the anthropogenic contamination investigated in this work is expected mainly in the leachable fraction, while the insoluble mineral matrix should contain only natural uranium.

In order to assess laboratory background for ^{236}U measurements, diluted samples of the preanthropogenic in-house standard 'Vienna-KkU' (Steier et al., 2008) uranium (0, 4, 40, 400 and $\sim 10^4$ μg) were prepared for each run. The background correction for the measured isotopic ratios depends on the uranium content of the sample, and is larger for smaller uranium content (Steier et al., 2008). However, all the $^{236}U/^{238}U$ ratios measured are elevated against the blank determined from the dilution series. In Table 2 the background corrected isotopic ratios are listed. All samples are far beyond possible natural levels, and are thus influenced by anthropogenic contamination.

In Fig. 2, the $^{236}U/^{238}U$ isotopic ratios measured are plotted as a function of the depth for all four cores. The values determined for the upstream (B) and downstream (N) cores agree with each other. The average $^{236}U/^{238}U$ and the standard deviation of all samples from these cores are $(1.7 \pm 0.7) \times 10^{-9}$.

The profile of the core B, where we have achieved the best depth resolution and which we expect to be unaffected by the NPP, reveals that the scatter in the data is not random, but that a clear correlation exists between the measured $^{236}U/^{238}U$ isotopic ratio and the water content of the sediment (Fig. 3). We think that this correlation further proves that the ^{236}U detected was already present in the environment and cannot be ascribed to laboratory or detection background.

Table 2
$^{236}U/^{238}U$ isotopic ratios measured by AMS.

Depth interval (cm)	$^{236}U/^{238}U$ (10^{-9})
B (upstream)	
(1–0)	1.2 ± 0.2
(3–1)	0.6 ± 0.1
(5–3)	1.0 ± 0.1
(7–5)	1.5 ± 0.2
(9–7)	2.1 ± 0.2
(11–9)	1.4 ± 0.1
(17–15)	0.98 ± 0.11
(19–17)	1.2 ± 0.1
(21–19)	1.5 ± 0.3
(25–23)	1.9 ± 0.3
(27–25)	2.0 ± 0.3
(31–29)	1.7 ± 0.6
D (drain channel)	
(4–1)	3.0 ± 0.2
(20–4)	3.8 ± 0.2
E (1 m downstream)	
(2–0)	3.5 ± 1.1
(10–2)	4.1 ± 1.6
(16–10)	1.5 ± 1.0
(22–16)	2.2 ± 0.2
(26–22)	5.4 ± 0.7
(33–26)	11 ± 2
N (1 km downstream)	
(2.5–0)	0.79 ± 0.14
(4.5–2.5)	1.2 ± 0.3
(8.5–4.5)	0.81 ± 0.16
(24.5–22.5)	0.87 ± 0.26
(28.5–26.5)	0.65 ± 0.21
(34.5–32.5)	0.85 ± 0.19
(38.5–36.5)	0.65 ± 0.12

Significantly higher $^{236}U/^{238}U$ ratios than in the samples more remote from the GNPP are observed in the drain channel core D and in the Garigliano river sediment 1 m downstream from the mouth of the drain channel (core E). In core E, the ratio of the upmost samples is consistent with the average ratio of the B and N cores, while for the deeper layers, between 16 and 33 cm, the isotopic ratio increases. The highest $^{236}U/^{238}U$ isotopic ratio is found at 33–26 cm depth in correspondence to the peak of ^{137}Cs specific activity.

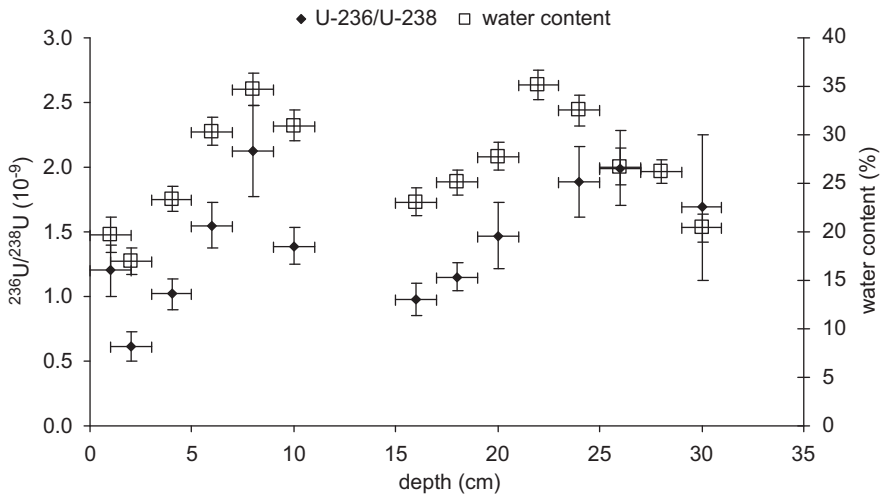

Fig. 3. $^{236}U/^{238}U$ isotopic ratio and water content vs. depth of the upstream core (B).

The ^{236}U concentration is calculated from the ^{238}U concentration determined by alpha spectroscopy, and from the $^{236}U/^{238}U$ measured by AMS. Since the ^{238}U concentration is almost constant for the set of investigated samples, the ^{236}U concentration reflects the $^{236}U/^{238}U$ isotopic ratio. Upstream and 1 km downstream, the ^{236}U concentrations are below the value of 2×10^{-15} g/g dry sediment. Significantly higher ^{236}U concentrations are found in the deeper sections of the cores D and E, where 4×10^{-15} and 9×10^{-15} g/g are reached, respectively.

$^{239+240}Pu$ activities, detected by alpha-spectrometry, did not exceed typical global fallout levels (the total activity of plutonium in soil from global fallout is usually less than 4 mBq/g, (Hardy et al., 1973); in fact, the activity concentrations range between 0.5 and 6 mBq/g for all the samples. The $^{240}Pu/^{239}Pu$ ratios determined by AMS indicated, as well, global fallout origin (i.e. $^{240}Pu/^{239}Pu$ atom ratio about 0.2), for all four cores.

4. Conclusions

All ^{236}U concentrations measured in this work are much lower than those found in known contaminated sites. In the drain channel of the GNPP, and 1 m downstream, the $^{236}U/^{238}U$ isotopic ratio is slightly but significantly higher than several km upstream and 1 km downstream. This finding increases the sensitivity of ^{236}U as fingerprint of releases from nuclear facilities. The highest ratio is found in correspondence to the highest ^{137}Cs specific activity, which might indicate a common origin of these radionuclides in that sediment layer. However, the ^{236}U concentration present in the sediment is still very low. In our opinion, this contradicts the interpretation of Delfanti et al. (1995), that a release from the GNPP might be responsible for an increase of the plutonium concentration in sediments in the Gulf of Gaeta. Also our (sparse) plutonium data show no indication of reactor fuel releases.

Also upstream of the GNPP, anthropogenic ^{236}U was detected, which is unlikely to originate from the GNPP. We think that the source is global weapons test fallout. A correlation between the $^{236}U/^{238}U$ isotopic ratio and the chemical/physical properties of the sediment (as represented by its water content) is observed. This demonstrates the potential of ^{236}U to serve as a tracer for geomorphologic processes like erosion and deposition, similar to the nuclides ^{137}Cs and ^{239}Pu (Everett et al., 2008).

We have demonstrated that the detection of anthropogenic ^{236}U is now possible at levels present in the typical environment. We expect that the study of its distribution, origin and behavior will find many fruitful applications, similar to other environmental radionuclides.

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