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Natural radioactivity and mineralogical composition of different particle size fractions of a river sediment from Calabria, southern Italy: a case study

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Abstract. The natural radionuclides (^{238}U , ^{232}Th and ^{40}K) content and the mineralogical characteristics of three different particle size fractions of a sediment sample from the Amendolea river, Calabria region, southern Italy, were investigated.

High Purity Germanium (HPGe) gamma spectrometry was employed for the radioactivity measurements and specific activity data were reported. X-Ray Diffraction (XRD) measurements were carried out to identify the mineral contents of each particle size fraction.

This case study was developed with the aim of evaluating the specific activities of natural radionuclides and their relation to specific minerals. Data obtained can provide useful information in view of a deep understanding of the adsorption of radioactive elements and the influence of the sediment granulometry on the radionuclides specific activities.

1. Introduction

Natural radioactivity provides the greatest contribution to the dose received by the population. From this point of view, important radionuclides are those belonging to the radioactive families ^{238}U , ^{232}Th and ^{235}U , together with the radioisotope of potassium, ^{40}K [1]. Together with their product of decay, they are commonly found in different concentrations in environmental matrices, especially in water, rocks and soils [2–5].



Among the environmental matrices, river sediments are of course particularly interesting, due to their mineralogical composition, particle size distribution and because they are reliable long-term indicators of river pollution by radionuclides [6]. Furthermore, knowing the distribution of the latter in terms of activity concentration in river sediments is extremely important, as it allows us to evaluate any temporal variations of specific activities within the river [7].

On the other side, geochemical studies of river sediments are also particularly useful, as the geomorphology of the investigated site greatly influences the chemical composition of this type of sediment, as well as the mineralogical composition reflects the geological history of the transport and the sorting process [8–11].

In this article, a case study was taken into account; in particular, natural radioactivity and minerals composition of three different particle size fractions of a sediment sample of the Amendolea river, Calabria region, southern Italy, were investigated with the aim to evaluate the influence of sediment particle sizes on natural radionuclides concentrations and the influence of the mineralogical composition on the ^{226}Ra , ^{232}Th and ^{40}K activity concentration. River sediments can be employed as building materials and thus the knowledge of their granulometric distribution, as well as the radioactivity concentration in each particle size fraction and the mineralogy of the investigated sediment, strictly correlated with its natural radioactive content, is extremely useful from a radiological point of view [12].

The schematic block diagram of figure 1 summarizes what was performed in the present study.

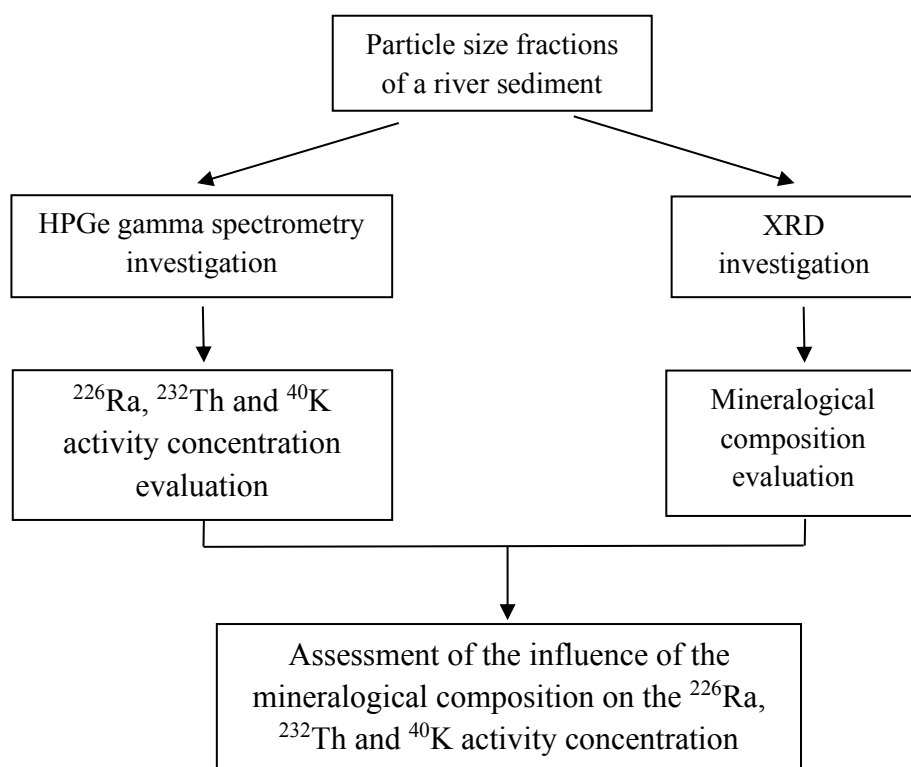


Figure 1. Schematic block diagram.

2. Materials and Methods

2.1 Sample collection and treatment

The investigated river sediment sample, around 1 kg, was collected in the sampling point indicated in figure 2 (GPS coordinates: latitude 37.9283; longitude 15.8878).

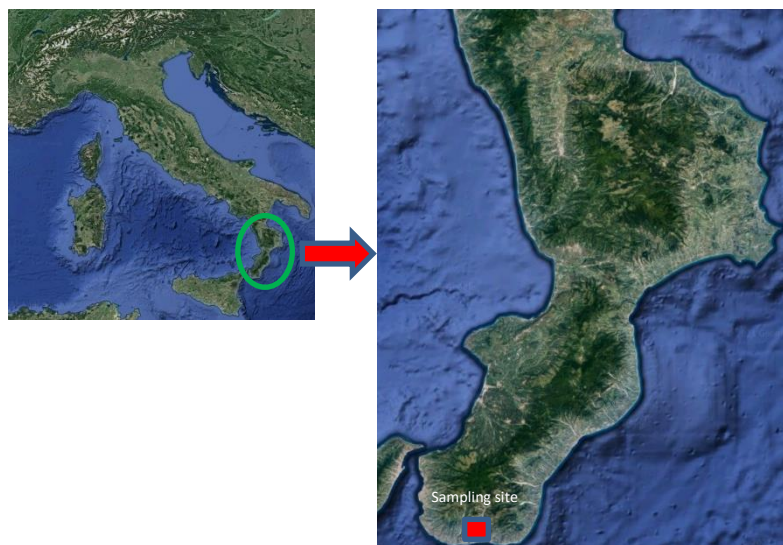


Figure 2. Map of the sampling site.

The sampling procedure is described in [10].

In laboratory, the sample was dried at 105° C in an oven and sieved to obtain the various particle size fractions: pelite (< 65 μm), fine sand (65 μm - 250 μm) and coarse sand (250 μm - 1 mm). Each size fraction was packed in a hermetically sealed 20 mL polyethylene plastic vial. After 40 days, the secular radioactive equilibrium between ²²⁶Ra and their daughter products was attained and samples were ready for gamma spectrometry counting [13].

2.2 Radioactivity analysis

For the gamma spectrometry analysis, the pelite, fine sand and coarse sand fractions were counted for 70000 s. The ²²⁶Ra and ²³²Th activity concentrations were obtained by the 295.21 keV and 351.92 keV ²¹⁴Pb, the 1120.29 keV ²¹⁴Bi and the 911.21 keV and 968.97 keV ²²⁸Ac γ-lines, respectively. For the ⁴⁰K specific activity evaluation, its γ-line at 1460.8 keV was employed.

The experimental setup was composed by an Ortec HPGe positive biased detector (GEM), with FWHM of 1.85 keV, peak to Compton ratio of 64:1 and relative efficiency of 40 % at 1.33 MeV (⁶⁰Co), and integrated digital electronics.

Eckert and Zigler Nuclitec GmgH traceable multinuclide radioactive standard, number AK-5901, was employed for efficiency and energy calibrations [14].

The ANGLE 4 code was employed for the efficiency transfer factors calculations [15].

The Gamma Vision (Ortec) software was used for data acquisition and analysis [16].

The activity concentration of each identified radionuclide was calculated using the following formula [17]:

$$C = \frac{N_E}{\varepsilon_E t \gamma_d M} \quad (1)$$

where N_E , ε_E and γ_d account for the net area, efficiency and yield of a photopeak at energy E , respectively, M is the mass sample (kg) and t is the live time (s).

The quality of the gamma spectrometry experimental results was certified by the Italian Accreditation Body (ACCREDIA) [18].

2.3 X-Ray Diffraction (XRD) analysis

A Malvern Panalytical Empyrean Diffractometer, equipped with a solid-state detector, PIXcel, was employed for the XRD tests on about 1 g of finely powdered sample for each investigated fraction [19].

Acquisition conditions were 40 kV and 40 mA. XRD spectra were recorded from 2 to 54 degrees, with step size of 0.007 degrees and counting time of 20 seconds.

The background correction of the Cu Ka2 component with a digital filter and the identification of the crystalline mineral components through the ICDD JCPDS database were also performed [20].

3. Results and Discussion

3.1 Radioactivity analysis

The specific activity of detected radionuclides, ^{226}Ra , ^{232}Th and ^{40}K , in the investigated sediment sample, is shown in figure 3 for pelite, fine sand and coarse sand.

Obtained values are strictly correlated with the sampling point. It is, in fact, well-established that the radionuclides activity concentration in river sediments varies from location to location, considering that the river bottom can exhibit large variations in chemical and mineralogical properties and rare-earth elements [21].

As we can see from an inspection of the figure, experimental values are 1561 Bq/kg d.w. for ^{226}Ra , 691 Bq/kg d.w. for ^{232}Th and 4461 Bq/kg d.w. for ^{40}K , for particle size $< 65\ \mu\text{m}$ (pelite); 41 Bq/kg d.w. for ^{226}Ra , 69 Bq/kg d.w. for ^{232}Th and 1260 Bq/kg d.w. for ^{40}K , for particle size $65\text{--}250\ \mu\text{m}$ (fine sand); 31 Bq/kg d.w. for ^{226}Ra , 13 Bq/kg d.w. for ^{232}Th and 1156 Bq/kg d.w. for ^{40}K , for particle size $250\ \mu\text{m}\text{--}1\ \text{mm}$ (coarse sand).

The adsorption of radioactive elements is then highest in the pelite fraction and it decreases from fine to coarse-grained materials [22].

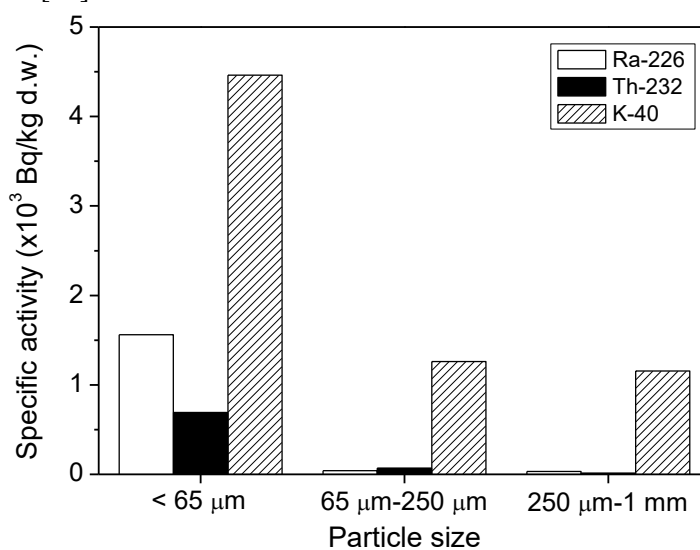


Figure 3. The activity concentration of ^{226}Ra , ^{232}Th and ^{40}K in the various particle size fractions of the investigated river sediment.

Moreover, the decrease in the specific activity values for ^{226}Ra , ^{232}Th and ^{40}K with increasing particle size, seems to indicate that the three particle size fractions are different from a mineralogical point of view and, consequently, have a different radionuclides content. The assessment of the influence of the mineralogical composition on the activity concentration of detected radioisotopes was then performed through the XRD analysis of the three particle size fractions of the investigated sample.

3.2 Mineralogical analysis

XRD spectra of the three particle size fractions of the investigated river sediment are shown in figure 4.

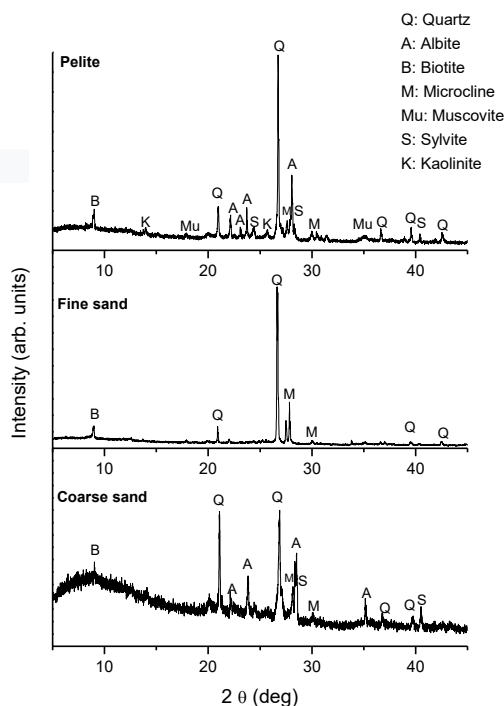


Figure 4. XRD patterns of the three particle size fractions.

From this figure we can notice that for each investigated size fraction, quartz (SiO_2) is the most abundant mineral. Biotite, $\text{K}(\text{Mg};\text{Fe})(\text{AlSi}_3\text{O}_{10})(\text{OH})_2$, and microcline feldspar, KAlSi_3O_8 , are also present in all samples. Kaolinite mineral, $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$, was identified only in the pelite fraction, whereas albite, $(\text{Na},\text{Ca})\text{Al}(\text{Si},\text{Al})_3\text{O}_8$, and sylvite (KCl) mineral components were detected only in the pelite and coarse sand samples. Finally, the muscovite mineral, $\text{KA}l_2(\text{Si}_3\text{Al})\text{O}_{10}(\text{OH},\text{F})_2$, was found in the pelite.

Worth of note, kaolinite is detected only for the pelite fraction, not for the fine and coarse sand ones. Now, according to the literature [23], the kaolinite mineral is shown to be responsible for ^{232}Th and ^{40}K concentrations and its highest dispersion is produced by particles of 25 - 35 μm diameter [24]. In the light of this we can hypothesize, then, that kaolinite plays the major role, more than microcline feldspar and quartz, in the observed trend of the specific activity of these radionuclides as detected by radioactivity measurements in the investigated particle size fractions. No specific correlation between some particular mineralogical phase and specific activity of ^{226}Ra was found in literature. Nevertheless, being ^{226}Ra in secular equilibrium with ^{238}U , its decreasing radioactivity concentration with increasing particle could be related to the uranium concentration. An investigation of the elemental composition of each particle size fraction, by means of spectroscopic techniques (like as X-Ray Fluorescence and ICP-MS), with the aim to quantify the uranium content in each of them and thus to verify a possible decrease in concentration by increasing the particle size, can be helpful in clarifying this aspect. It will be planned in the next development of the research.

4. Conclusions

In the present paper, the ^{226}Ra , ^{232}Th and ^{40}K specific activity for three different particle size fractions of a sediment sample of Amendolea River (Calabria region, southern Italy), taken as case study, was

quantified with HPGe gamma spectrometry, in order to investigate the influence of the granulometry on the radionuclides specific activity. The highest concentrations of detected radionuclides were found in the pelite fraction, because the adsorption of radioactive elements has a strong dependence on the sediment nature and decreases from fine to coarse-grained materials.

At the same time, XRD tests were performed to identify the mineralogical composition of each particle size fraction, with the aim of correlating the radioactivity results to specific minerals. From the results, the main role of kaolinite in increasing the radioactivity concentration for ^{232}Th and ^{40}K in pelite fraction with respect to, in the order, fine and coarse sand was hypothesized. As far as ^{226}Ra is concerned, being this radioisotope in secular equilibrium with ^{238}U , the observed decreasing level of its activity concentration with increasing particle size was supposed to be related to the uranium concentration in each of them.

The obtained data can be used as a baseline for future investigations about radioactivity background levels in river sediments samples of the investigated region and they can provide useful information to better understand the adsorption of radioactive elements and the variation of the radionuclides specific activity with the sediment granulometry.

References

- [1] United Nations Scientific Committee on the Effects of Atomic Radiation. 2000 *Sources and Effects of Ionizing Radiation: Report to the General Assembly, with Scientific Annexes* vol I
- [2] Navas A, Soto J and Machín J 2002 ^{238}U , ^{226}Ra , ^{210}Pb , ^{232}Th and ^{40}K activities in soil profiles of the Flysch sector (Central Spanish Pyrenees) *Appl. Radiat. Isot.* **57** 579–89
- [3] Caridi F, D'agostino M, Belvedere A, Marguccio S and Belmusto G 2016 Radon radioactivity in groundwater from the Calabria region, south of Italy *J. Instrum.* **11**
- [4] Caridi F, Messina M and D'Agostino M 2017 An investigation about natural radioactivity, hydrochemistry, and metal pollution in groundwater from Calabrian selected areas, southern Italy *Environ. Earth Sci.* **76** 668
- [5] Bayramli G 2020 The environmental problems of Azerbaijan and the search for solutions *WSEAS Trans. Environ. Dev.* **16** 423–33
- [6] Küçükömeroglu B, Kurnaz A, Keser R, Korkmaz F, Okumusoglu N T, Karahan G, Sen C and Cevik U 2008 Radioactivity in sediments and gross alpha–beta activities in surface water of Firtina River, Turkey *Environ. Geol.* **55** 1483–91
- [7] Madruga M J 2008 Environmental radioactivity monitoring in Portugal *Appl. Radiat. Isot.* **66** 1639–43
- [8] Caridi F, Marguccio S, D'Agostino M, Belvedere A and Belmusto G 2016 Natural radioactivity and metal contamination of river sediments in the Calabria region, south of Italy *Eur. Phys. J. Plus* **131** 1–10
- [9] Caridi F, Marguccio S, Belvedere A, Belmusto G, Marcianò G, Sabatino G and Mottese A 2016 Natural radioactivity and elemental composition of beach sands in the Calabria region, south of Italy *Environ. Earth Sci.* **75**
- [10] Caridi F, Bella M Di, Sabatino G, Belmusto G, Fede M R, Romano D, Italiano F and Mottese A F 2021 Assessment of Natural Radioactivity and Radiological Risks in River Sediments from Calabria (Southern Italy) *Appl. Sci.* **11** 1729
- [11] Atzori P, Ferla P, Paglionico A, Piccarreta G and Rottura A 1984 Remnants of the Hercynian orogen along the “Calabrian-Peloritan arc”, southern Italy: a review *J. Geol. Soc. London.* **141** 137–45
- [12] Caridi F, Testagrossa B and Acri G 2021 Elemental composition and natural radioactivity of refractory materials *Environ. Earth Sci.* **80** 170
- [13] Sima O and Dovlete C 1997 Matrix effects in the activity measurement of environmental samples—implementation of specific corrections in a gamma-ray spectrometry analysis program *Appl. Radiat. Isot.* **48** 59–69
- [14] Web site 2020 Eckert & Ziegler Calibration Sources

- https://www.ezag.com/home/products/isotope_product
- [15] Ortec 2016 Angle 4 Software User Manual
- [16] Caridi F, D'Agostino M, Belvedere A, Marguccio S, Belmusto G and Gatto M F 2016 Diagnostics techniques and dosimetric evaluations for environmental radioactivity investigations *J. Instrum.* **11**
- [17] Caridi F, Marguccio S, Durante G, Trozzo R, Fullone F, Belvedere A, D'Agostino M and Belmusto G 2017 Natural radioactivity measurements and dosimetric evaluations in soil samples with a high content of NORM *Eur. Phys. J. Plus* **132** 56
- [18] ACCREDIA 2021 <https://www.accredia.it/>
- [19] Malvern Panalytical. 2013 Epyrean diffractometer user manual
- [20] Caridi F, Santangelo S, Faggio G, Gnisci A, Messina G and Belmusto G 2019 Compositional and Mineralogical Analysis of Marine Sediments from Calabrian Selected Areas, Southern Italy *Int. J. Environ. Res.* **13** 571–80
- [21] Blanco Rodríguez P, Tome F, Lozano J and Pérez-Fernández M 2008 Influence of soil texture on the distribution and availability of U-238, Th-230, and Ra-226 in soils *J. Environ. Radioact.* **99** 1247–54
- [22] McCubbin D, Leonard K S, Maher B A and Hamilton E I 2000 Association of ²¹⁰Po(²¹⁰Pb), ²³⁹+²⁴⁰Pu and ²⁴¹Am with different mineral fractions of a beach sand at Seascale, Cumbria, UK *Sci. Total Environ.* **254** 1—15
- [23] Ramasamy V, Suresh G, Meenakshisundaram V and Ponnusamy V 2011 Horizontal and vertical characterization of radionuclides and minerals in river sediments *Appl. Radiat. Isot.* **69** 184–95
- [24] Yahaya S, Jikan S S, Badarulzaman N A and Adamu A D 2017 Chemical Composition and Particle Size Analysis of Kaolin *Path Sci.* **3** 1001–4