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Terrestrial and fallout radionuclide fingerprints of sediments from highway stormwater retention ponds

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Abstract The activity concentrations of terrestrial and fallout radionuclides are investigated in soil and sediments from highway stormwater retention ponds. The increased activity of ^{40}K, ^{238}U and ^{232}Th in sediment respect to soil samples (approximately 20% higher) is found to be correlated with the erosion of clay minerals. Therefore, it can be a useful fingerprint to assess the highway bank stability in long-term measurements. Fallout radionuclides, especially ^{7}Be gives a clear indication of the well operation of the drainage system and the retention ponds, with activity concentrations in sediment samples up to 131.4 ± 7.8 Bq kg^{-1}. Finally, these results are used to assess the potential external exposure to gamma radiation of the general population and worker due to maintenance operations.

Keywords Natural radioactivity · Fallout radionuclides · Beryllium-7 · Runoff retention pond · Gamma-ray spectrometry

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Introduction

The drainage systems and retention ponds installed on highways are widely used to reduce and prevent the pollution of the surrounding environment by permitting the runoff water loads settle down and accumulating them as sediments. It is recognized that road runoff waters carry significant of pollutants, including hydrocarbons, pesticides and heavy metals [1–3]. However, there is a lack of information regarding the characterization of natural and artificial radioactivity concentration because of runoff water treatment in highway retention pond sediments.

The natural radioactivity present in rocks and soils is mainly due to ^{238}U and ^{232}Th decay series and to the ^{40}K radionuclide with concentration varying considerably depending on the geological characteristics of the area [4]. The concentration of radionuclides in river sediments primarily depends on the accumulation of clay mobilized from the upstream catchment [5]. Moreover, in many studies of the dynamics of soil and sediments (in river, lake and sea environments), fallout radionuclides (^{137}Cs and ^{7}Be) are widely used as tracers [6]. ^{7}Be (with half-life 53.22 days [7]) is a cosmogenic radionuclide continuously produced by spallation reaction through interaction of cosmic rays with atmospheric molecules. It is known that ^{7}Be is absorbed by aerosols and distributed in surface air by various transport mechanisms and then is replenished principally by wet deposition (secondarily dry deposition) on the Earth's surface [8]. Therefore, it can accumulate providing a fingerprint for sediments. Indeed, high activity concentrations of ^{7}Be are reported (3.6 kBq kg^{-1}) in certain conditions [9]. On the other hand, ^{137}Cs (with half-life 30.05 years [10]), originating mainly from atmospheric nuclear weapon tests and the Chernobyl nuclear power

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plant accident [11] has also shown to provide a useful tool for fingerprinting processes involving sediments and soils.

The main objective of this study is to characterize the radionuclide concentration of sediments in highway retention ponds and investigate possible fallout radionuclides fingerprints. This baseline data will give useful information for planning further long-term studies in correlation with rainfall events in order to evaluate the highway bank stabilization and efficiently maintain the highway retention ponds. These data will be used to calculate the radiological hazard, and safe disposal of accumulated sediments in routine maintenance operation of the retention ponds.

Materials and methods

Study area and sampling

This study focuses on the "Morinë-Merdarë" highway, approximately 80 km long, located in the central part of the Republic of Kosovo (Fig. 1). Six retention ponds were accurately chosen coupled at distances of approximately 15 km apart in order to have more local information and at the same time independent catchment areas assuring their representativeness.

The study area is part of the Dukagjin Plain, and contains a variety of formations ranging from Paleozoic to Quaternary in age. The younger ones are the alluvial deposits of Holocene, covering the lake sediments of the Pleistocene. The Quaternary deposits are lying over the deposits of the Upper Pliocene, which consist principally of clastic sediment sand, some marl and tuff. The bedrock is characterized by a Cretaceous sedimentary sequence, consisting of limestone with chert, sandy limestone with rudists, and flysch deposits; a Jurassic succession of volcanoclastic rocks and peridotites/serpentinites, associated with a magmatic event of the Middle/Upper Jurassic; in the end there is a Paleolozoic metamorphic basement, covered by local outcrops of metacarbonate rocks (Fig. 1).

From the geological point of view the L5 and L6 sites are characterized by Quaternary–Pleistocene lake sediments with heterogeneous granulometry (gravel, sand, silt and clay). The aquifer is mainly composed of unconsolidated rock (gravel, sand and mud/silt) with medium–low porosity permeability. Site L4 is characterized by Upper Paleozoic formations. From the hydrogeological point of view, it is classified as aquiclude without considerable

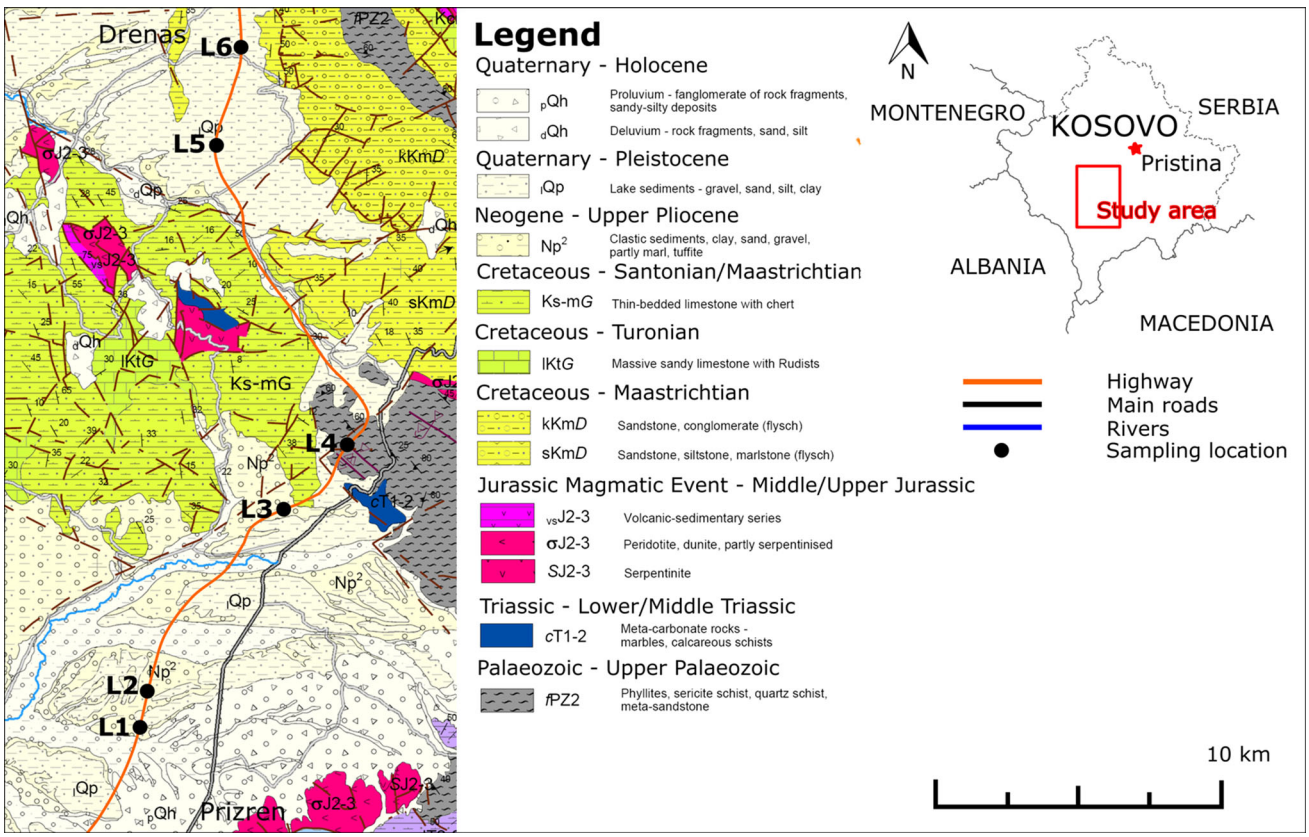

Fig. 1 Geological map (based on [13]) of the investigated area and the approximate location of the retention ponds under investigation along the Morinë-Merdarë highway

intergranular or fissured porosity (undivided metamorphic rock). While site L3 is characterized by Neogene–Upper Pliocene clastic sediments, clay, sand, gravel and partly marl. The aquifer is mainly composed of unconsolidated rock (gravel, sand and mud/silt) with medium–low porosity and permeability. Site L2 is characterized by Quaternary–Holocene alluvium sediments (gravel, sand and silt). While L1 is characterized by an equivalent mix of Quaternary–Pleistocene lake sediments (gravel, sand, silt and clay) to Neogene–Upper Pliocene clastic sediments, clay, sand, gravel and partly marl. From the hydrogeological point of view the intergranular porosity aquifer is within an area with confined or artesian groundwater, composed mainly of unconsolidated rock (gravel and sand) with very high-medium permeability.

In general, the land cover types of the catchment areas are characterized by land intensively used for agriculture and meadow, while site L4 is defined by hilly areas with rocky ground and presence of mainly oak vegetation.

From the meteorological point of view the Dukagjin Plain is characterized by a mountainous continental Mediterranean climate characterized by mild winters (average temperature ±0.5 °C and rarely up to −23 °C) and warm summers (average temperature up to 23 °C) [12]. The biggest amounts of rainfall (>1000 mm year^{-1}) are frequent in the peripheral mountainous part (where is located the study area), while the lowest amounts (600 mm year^{-1}) are frequent in the central (plain) part.

In each site are collected undisturbed soil samples representative of the catchments soil type and sediment samples directly from the wetlands. The bulk samples of top soil (5 cm depth) and sediment provide information on the concentration of natural and fallout radionuclides. Samples L1, L4 and L5 were collected immediately after heavy rain events, while the other samples were collected in dry weather conditions. Soil and sediment samples were placed in plastic bags (1 kg of mass) and transported in laboratory for further treatment and analysis.

Measurement of radionuclide concentration in soils and sediments

To remove the moisture content, soil and sediment samples are dried to constant weight in a temperature-controlled furnace at 110 °C, generally for at least 24 h. After cooling in a moisture-free atmosphere, samples are homogenized into fine powders with a particle size of less than 2 mm and then transferred into the measurement containers (with an effective volume of 180 cm^3). The containers are hermetically sealed and weighed prior to be measured.

Regarding the presence of fallout radionuclides (i.e. the presence of ^{7}Be and ^{137}Cs) samples are measured immediately by high-resolution gamma-ray spectrometry, due to the relatively short half-life cosmogenic radionuclide of ^{7}Be. Indeed, for ^{7}Be, the results were further corrected both for the nuclide decay from the time the sample was collected to the start of the measurement and for the decay during the counting period. Then the containers are stored for at least 4 weeks to allow ^{226}Ra and its short-lived decay products to reach secular equilibrium before being again measured by high-resolution gamma-ray spectrometry (HPGe) for determining natural radionuclides. The gamma-ray spectrometry system (described in detail in Xhixha et al. [14]) consists of two 60% relative efficiency, coaxial p-type High Purity Germanium (HPGe) detectors, with an energy resolution of 1.9 at 1332.5 keV (^{60}Co). The gamma-ray spectrometry system is accurately shielded by 10 cm of copper and 10 cm of lead, leaving enough space between the two detectors for the sample container. The absolute full energy peak efficiencies are determined using certified reference materials with an overall combined standard uncertainty of less than 5% [15].

The activity concentrations of fallout radionuclides of ^{7}Be and ^{137}Cs are determined using the 477.6 keV (10.44%) and 661.7 keV (84.99%) gamma-rays respectively [7, 10]. The activity concentration of ^{226}Ra is determined through the peaks at the 351.9 keV (35.6%) of ^{214}Pb and at 609.3 keV (45.49%) of ^{214}Bi (the nuclear data are taken from [16]). The activity of ^{232}Th is assessed considering it in equilibrium with ^{228}Ra, which is then determined using the gamma-ray of 911.2 keV (26.2%) of ^{228}Ac, and ^{228}Th, which is determined using the gamma-ray of 583.2 keV (85.0% corrected for the branching ratio 35.94%) of ^{208}Tl (the nuclear data are taken from [17, 18]). The results are corrected for the self-absorption and true coincidence summing effect as described in Xhixha et al. [15]. The activity concentration of ^{40}K is determined using its 1460.8 keV (10.55%) gamma-ray [19].

Qualitative mineralogical analysis

Before conducting the qualitative mineralogical analyses, the samples were dried at a temperature of 45 °C for more than 48 h, and then gradually ground in an agate mortar until having grain dimensions of less than 50 μm. Prepared samples were placed on a glass sample holder for the qualitative XRD (X-ray diffraction) analyses based on θ:2θ Bragg–Brentano geometry. The XRD diffractogramm is recorded using an X'Pert PRO PANalytical powder diffractometer, with Cu–Kα radiation, which operates at 40 kV and 30 mA. The X-ray diffractometry of three samples (L2, L3 and L6) was conducted with the scatter slit set at 1/4° and the receiving slit at 1/2°. The scan step size and the counting time were 0.02° and 2 s/step, respectively. The XRD diffractograms were processed by the HighScore (3.0.d) software. The International Center for Diffraction

Data [20] reference database was used for the mineral identification.

Results and discussion

Fallout and natural radionuclide fingerprints

The activity concentrations of natural and fallout radionuclides in the soil and sediment samples are reported in Table 1. The activity concentrations of the natural radionuclides ^{40}K, ^{238}U and ^{232}Th in the soil samples are found to be comparable with the values given in the report of UNSCEAR [21] (world average values of concentration in soil 400, 35 and 30 Bq kg^{-1} respectively).

The presence of quartz, albite and mica/muscovite minerals (see Table 2) show that the soil samples originate from erosion activities of the rocks forming the geological structure of this region, i.e. metamorphic rocks as phyllites, quartz schist and metasandstone (fPZ2), sedimentary rocks like sandstone (sKmD, kKmD), limestone (kS–mG) and meta/carbonate rocks (CT1-2). Furthermore, the activity concentrations of the natural radionuclides ^{40}K, ^{238}U and ^{232}Th in soil samples show generally comparable results between L5–L6 and L1–L2, while there is a distinct difference between L3–L4, which demonstrate the close relationship with the geological structure of the area. These "major" minerals are found in all sediment samples confirming the representativeness of the catchment area.

In general the activity concentration in sediments respect to soil samples is up to a few ten percent higher (approximately 20%) with exception of sample L6 where the activity concentration in the sediment is lower. Moreover, in Table 2 we observe in the sediments an increase in the presence of clay minerals such as montmorillonite, vermiculite, illite and clinochlore with respect to soil samples. These results suggest that the origin of sediments may be mainly due to the erosion of clay facies from the highway banks, which are the principal contributor to the natural activity concentration increase. The concentration of rock forming mineral in sediments is also confirmed by the ^{232}Th/^{238}U ratios. Indeed, for a specific site, the ^{232}Th/^{238}U ratio in both sediments and soil remains generally constant. This means that the enrichment or removal of specific radionuclides is negligible. A systematic increase in concentration of natural radionuclides in sediments due to clay minerals is confirmed also in Mohery et al. [22] and Suresh et al. [5].

Table 2 Results of the qualitative analysis of mineralogical composition of the soil and sediment samples

Sample ID	Soil			Sediment		
Mineral	L6	L3	L2	L6	L3	L2
Quartz	x	x	x	x	x	x
Albite	x	x	x	x	x	x
Mica/muscovite			x	x	x	
Vermiculite		x			x	
Montmorillonite				x		x
Illite						x
Clinochlore					x	

Table 1 The activity concentrations and the combined standard uncertainties ($k = 1$) of natural radionuclides (^{238}U, ^{232}Th decay chains and ^{40}K) and fallout radionuclides (^{137}Cs and ^{7}Be) in soil and sediment samples

Sample type	Site ID	^{40}K (Bq kg^{-1})	^{238}U decay chain		^{232}Th decay chain		^{137}Cs (Bq kg^{-1})	^{7}Be (Bq kg^{-1})
			^{214}Pb (Bq kg^{-1})	^{214}Bi (Bq kg^{-1})	^{228}Ac (Bq kg^{-1})	^{208}Tl (Bq kg^{-1})		
Soil	L1	412 ± 23	21.6 ± 1.4	22.4 ± 1.9	33.2 ± 2.4	34.1 ± 2.2	3.6 ± 0.3	<1.1
	L2	622 ± 47	23.2 ± 1.8	25.6 ± 2.3	32.6 ± 2.7	32.8 ± 2.9	0.3 ± 0.1	<0.9
	L3	214 ± 16	26.3 ± 2.0	30.4 ± 2.6	35.6 ± 2.9	36.6 ± 3.1	2.5 ± 0.2	2.4 ± 0.6
	L4	475 ± 26	20.4 ± 1.3	20.8 ± 1.7	28.3 ± 2.1	32.1 ± 2.1	<0.6	<0.9
	L5	315 ± 18	33.9 ± 2.0	34.0 ± 2.7	48.9 ± 3.3	48.8 ± 3.0	<0.6	<0.9
	L6	370 ± 28	35.1 ± 2.7	38.4 ± 3.2	52.8 ± 4.4	55.8 ± 4.5	3.7 ± 0.6	3.5 ± 0.7
Sediment	L1	646 ± 35	21.5 ± 1.4	24.1 ± 2.1	36.2 ± 2.7	38.7 ± 2.6	2.0 ± 0.3	131.4 ± 7.8
	L2	526 ± 40	27.2 ± 2.1	30.0 ± 2.6	37.6 ± 3.1	40.0 ± 3.4	0.7 ± 0.1	6.3 ± 0.8
	L3	356 ± 27	31.6 ± 2.4	38.7 ± 3.2	41.2 ± 3.4	44.0 ± 3.7	1.3 ± 0.1	4.7 ± 0.5
	L4	401 ± 22	24.8 ± 1.6	23.7 ± 2.0	42.3 ± 3.0	42.3 ± 2.7	0.8 ± 0.3	124.9 ± 7.4
	L5	423 ± 23	43.4 ± 2.5	42.1 ± 3.4	58.3 ± 3.9	59.3 ± 3.6	1.6 ± 0.3	108.6 ± 6.6
	L6	383 ± 29	29.3 ± 2.2	33.0 ± 2.7	46.9 ± 3.9	49.2 ± 4.0	0.4 ± 0.1	18.8 ± 1.6

Values expressed "<" indicate minimum detectable activity concentrations

The activity concentrations of the fallout radionuclides ^{137}C and ^{7}Be in soil samples range from below the minimum detection activity (mda) up to 3.7 ± 0.6 Bq kg^{-1} for the first and from mda up to 3.5 ± 0.7 Bq kg^{-1} for the second. Low concentrations of ^{137}Cs are also observed in sediment samples varying from 0.4 ± 0.1 Bq kg^{-1} to 2.0 ± 0.3 Bq kg^{-1}. These results are reasonable because the highway banks expose deeper fresh soil horizons which are not previously contaminated with radioactive fallout. Moreover, small amounts of ^{137}Cs are currently deposited from the atmospheric fallout. On the other hand, the activity concentration of ^{7}Be in sediment samples shows a clear accumulation with values ranging from 4.7 ± 0.5 to 131.4 ± 7.8 Bq kg^{-1}. The high variability of the concentrations of ^{7}Be is generally due to different meteorological events prior to the sample collections. Indeed, samples showing the highest activity concentration are being collected after heavy rainfall events. The accumulation of ^{7}Be can be mainly confirmed by the affinity with fine particles (aerosols) [23], which are continuously washed out from runoff waters and then precipitating in the retention ponds. Together, the presence of ^{7}Be and ^{137}Cs are a clear evidence of the well operation of the drainage system and the retention ponds in accumulating polluted runoff water loads as sediments. However, a major effort must be done for the rehabilitation of highway banks and careful maintenance of the retention ponds in order to prevent their overload and consequently to be potential sources of contamination of the surrounding lands.

Assessment of radiological hazard

The potential contribution to the total external absorbed dose rate (DR in nGy h^{-1}) in the outdoor air, due to the presence of natural radionuclides in sediments (^{226}Ra, ^{232}Th and ^{40}K), is calculated according to UNSCEAR [11]:

$$DR = 0.462A_{226_{Ra}} + 0.604A_{232_{Th}} + 0.0417A_{40_K} \quad (1)$$

where $A_{226_{Ra}}$, $A_{232_{Th}}$, A_{40_K} are the measured activity concentrations in Bq kg^{-1} for radium, thorium and potassium, respectively. The received DR in soil is found to vary from 44 to 65 nGy h^{-1}, while in sediments from 53 to 73 nGy h^{-1}. These values are comparable with the published world average dose rate of 58 nGy h^{-1} [11].

The DR values are used to estimate the annual effective dose rate (EDR in μSv year^{-1}), considering that the population spent, on average, 20% of their time outdoors, and using a conversion coefficient for the absorbed doses in the air to the effective dose received by an adult of 0.7 Sv Gy^{-1} [11]. The EDR is calculated according to the formula:

$$EDR = \left(DR \times 10^3\right) \times 0.7 \times (8760 \times 0.2) \quad (2)$$

The corresponding average EDR received varied from 54 to 80 μSv year^{-1} in soil and 66 to 89 μSv year^{-1} in sediments. These results are comparable with the world average annual effective dose rate of 70 μSv year^{-1} [11]. The EDR reference values (i.e. the annual effective dose rate is less than 1000 μSv year^{-1}) give a rough estimate of the potential contribution to the annual effective dose rate due to the disposal of sediments.

The contribution to the absorbed dose rate from fallout radionuclides due to the presence of ^{137}Cs is calculated according to Al-Masri et al. [24]. However, its contribution to the absorbed dose rate was found to be negligible. While, for ^{7}Be to our knowledge, there are no practices for estimating their contribution to the absorbed dose rate. This study could be a challenge for future since ^{7}Be is associated with short term variabilities of the activity and inventories in soil and sediments.

Conclusions

There is a lack of attention to the presence of terrestrial and fallout radionuclides in sediments in retention ponds installed on highways. The activity of terrestrial radionuclides (^{40}K, ^{238}U and ^{232}Th) shows a general increase of approximately 20% in sediments relative to the catchment soils. Qualitative mineralogical analysis indicates that clay mineral facies control the concentration of natural radionuclides in sediments. An increase of activity concentration in sediments was observed also for fallout radionuclides (^{137}Cs and ^{7}Be), when ^{7}Be show a clear accumulation in sediments with activity concentration up to 131.4 ± 7.8 Bq kg^{-1} with respect to soil samples where its activity concentration was mda or very low.

This study indicates that terrestrial radionuclides can be used as a useful fingerprint for long-term studies of highway bank stability due to erosion mechanisms. On the other hand fallout radionuclides reveal important information on the efficiency of the drainage system and functionality of retention ponds. However, further long-term studies are needed to verify the effectiveness of radionuclide fingerprinting of environmental processes.

These data are used to calculate the radiological hazard, and safe disposal of accumulated sediments in routine maintenance operation of the retention ponds. The results show an increase of annual EDR from 58–80 μSv year^{-1} in soil to 66–89 μSv year^{-1} in sediments. However, these results are comparable with the world average annual effective dose rate of 70 μSv year^{-1} showing that no significant risk can be attributed to their disposal.

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