A preliminary study of gross alpha/beta activity concentrations in drinking waters from Albania

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Abstract A pilot study involving screening measurements of gross alpha/beta activity concentration in drinking water was performed in 12 cities with the highest population density in Albania. The aim of this study was to develop a first insight regarding the radiological quality of drinking and bottled water supplies. The tap and bottled water samples investigated are acceptable for consumption, complying with the WHO recommendations for drinking water. The average gross alpha and beta activity concentrations were 36^{+37}_{-18} and 269^{+337}_{-150} mBq/L, respectively in tap waters. While for bottled water the gross alpha and beta activities were respectively 39^{+55}_{-23} and 220^{+336}_{-132} mBq/L. The data obtained can provide information for authorities regarding the quality of drinking water and a baseline for future contaminations.

Keywords Gross alpha/beta activity · Gas-flow proportional counter · Drinking water · "fracking" waste water

Introduction

Rocks and soils are the primary sources of radioactivity. The principal responsible radionuclides in surface and drinking water are ^{238}U, ^{232}Th, ^{40}K and, to a lesser extent, other trace elements, such as ^{235}U and ^{87}Rb [1, 2]. The primary alpha-emitting radioelements are ^{226}Ra, ^{224}Ra, ^{210}Po and, in certain circumstances, the uranium isotopes ^{238}U, ^{234}U and ^{235}U, as they are rather insoluble in reducing environments but are soluble in oxidising and particularly in acidic conditions [3]. Because thorium is practically insoluble in water, its contribution tends to be rather low. In general, beta activity is primarily due to ^{40}K, ^{228}Ra, and ^{210}Pb. As the world's demand for energy increases markedly and exploration for and production of new sources become increasingly important, the ways in which these activities impact the environment, health, and quality of life have become of increasing concern.

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According to the World Health Organisation (WHO) [4] guidelines, governmental regulatory agencies should report the total indicative dose (TID) parameter of drinking water to ensure that it is safe for consumption. The WHO [4] and European Directive 98/83/EC [5] established a TID of 0.1 mSv/year. This specified TID excludes tritium, ^{40}K, radon and radon decay products. In Albania, the health criteria of water intended for human consumption were published in governmental mandates [6, 7]. These mandates made compulsory the monitoring of gross alpha and beta activity concentration and specific radioelements at various sampling frequencies. A practical approach used is a screening procedure that involves the determination of the total radioactivity present in the form of alpha and beta radiation (gross alpha/beta activity concentration). This procedure is a simple radioanalytical screening [8] that is performed without regard to the identities of specific radionuclides. Various other methods are used to measure the gross alpha/beta activity. Two of these methods are based on evaporation [9] or co-precipitation [10] of the sample, using either a gas-flow proportional counter or a solid scintillation detector [11–13]. Another alternative, based on the total evaporation of the sample and measurement by liquid scintillation counting [14], has been increasingly used.

Exploration for oil and gas in Albania began in 1918 and was initially centred along the eastern and southeastern edges of the Peri-Adriatic depression in sandstone reservoirs of Oligocene flysch deposits (Tortonian–Messinian) and afterward in oil- and gas-bearing rocks of the Cretaceous–Palaeogene carbonate section (Tortonian–Messinian). Improper disposal of production water can lead to contamination of ground and surface water due to dissolution of certain constituents together with naturally occurring radioactive materials (NORMs), particularly radium [15]. New unconventional methods of shale gas extraction have become economically viable, particularly the application of horizontal drilling and hydraulic fracturing known as "fracking". Extraction of shale gas requires 50–100 times more water (85 % fresh and 15 % saline) than does the extraction of conventional gas, and NORMs can be brought to the surface as part of the flowback and production brine (typically 25 % of the water used). The prevailing hypothesis is that the increased concentration of salts in the flowback brine is due to dissolution of constituents in the shale by the water injected during the hydraulic fracturing [16]. Major concerns include possible precipitation of $BaSO_4$ and, to a lesser extent, $SrSO_4$ and $CaCO_3$ with radium isotopes in the shale formation, particularly the shale rich in uranium. The concentration of ^{226}Ra may be hundreds of times higher than the drinking water guidance level (1 Bq/L, according to WHO [4]) and therefore should be rigorously monitored.

The aim of this study was to perform an initial screening for gross alpha and beta activity concentrations in drinking water (tap water) supplied in 12 Albanian cities. This screening was based on gas-flow proportional counting (GPC) using the evaporation method. The gross alpha/beta efficiency calibration is discussed in the details. The gross alpha and beta activity concentrations in 26 drinking water samples (four bottled mineral water samples) were investigated over a two-year time span, and the results were used to compile a national database of the radiological content of waters in Albania. Based on the results, clear recommendations were made for future detailed analyses for specific radionuclides.
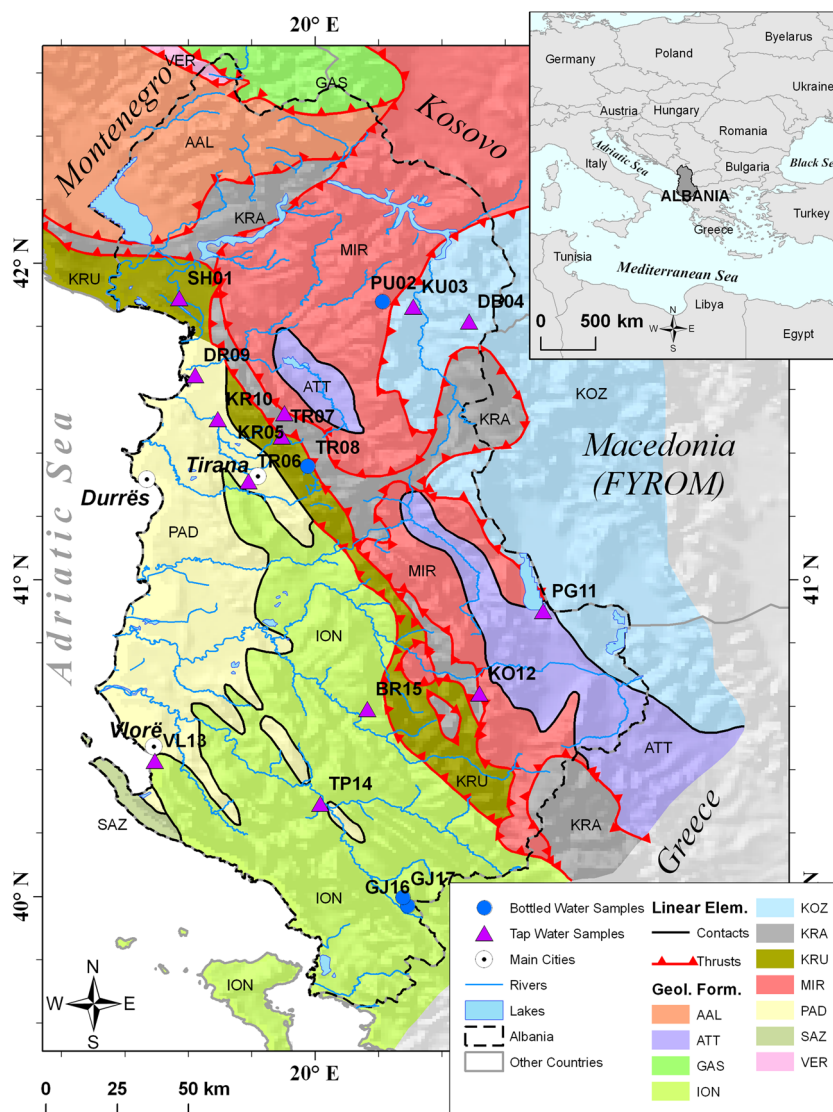
Materials and methods

Study area: water sampling and pretreatment

Albania is located in the western part of the Balkan Peninsula. The nation measures 28,748 km^2 and has a population of approximately 3.2 million. From a geological standpoint, the country is part of the Alpine–Mediterranean mountain belt, and its rocks may be subdivided into several predominantly NNW-SSE trending geotectonic units ("unit" in Fig. 1). These units are of *Palaeozoic* through *Mesozoic* to *Neogene* age. The presence of igneous rocks is the principal feature used to divide these units into the inner and outer units of the region. The inner units are characterised by a *Lower Cretaceous* discordance and an abundance of igneous rocks. The outer units are characterised by nearly continuous sedimentation from the *Triassic* to the *Palaeogene*. The inner and outer geotectonic units of Albania, each described in succession from northeast to southwest, are as follows:

- The inner units consist of the Korabi unit (low-grade metamorphic sedimentary and igneous rocks overlain by conglomerates and carbonate rocks); the Mirdita and Krasta–Cukali units, which consist of an ophiolite belt and flysch; the Albanian Alps (northernmost Albania), composed of terrigenous and platform-type carbonate rocks overlain by flysch; the Vermoshi unit, which is exposed along the boundary between Albania and Montenegro and includes a single formation, the Vermoshi Flysch, characterised by turbidite deposits consisting of arenites, shales and marls; and finally, the igneous rocks of the Gashi unit (northeast Albania), which are represented by volcanic rocks (basalts to rhyolite) within sedimentary-volcanic and volcanic-sedimentary sequences and by granodioritic rocks.
- The outer units consist of the Kruja unit, which is composed of neritic carbonate rocks overlain by flysch;

Fig. 1 Simplified geological map of Albania, modified from Meco and Aliaj [19]. Geological formation labels are as follows: *AAL* Albanian Alps unit, *ATT* Albanian–Thessalian trough, *GAS* Gashi unit, *ION* Ionian unit, *KOZ* Korabi unit, *KRA* Krasta unit, *KRU* Kruja unit, *MIR* Mirdita unit, *PAD* Periadriatic depression, *SAZ* Sazani unit, *VER* Vermoshi unit. Topographic shading layer is from the European Environment Agency, 2013 (http://www.eea.europa.eu/data-and-maps/data/digital-elevation-model-of-europe). Geographic coordinate system: WGS 84

the Ionian unit, which is composed of evaporitic rocks and is overlain by carbonate sediments succeeded by flysch; the Sazani unit, composed of neritic, carbonate and calcareous–clastic rocks; the Albano–Thessalian Neogene molasse basin, also referred to as the Adriatic trough; and finally, the Periadriatic depression, a basin between the external Albanides thrust belt (Ionian and Kruja units) and the Sazani unit.

An extensive investigation of drinking water was performed in 12 cities of Albania in 2008 and 2009. Twenty-six samples were collected from sources of potable water (Fig. 1): Berat, Laç/Durrës (Fushë Kuqe), Fushë Krujë, Krujë, Korçë, Kukës, Peshkopi, Pogradec, Shkodër, Tepelenë, Tiranë (Bovillë artificial lake and Selitë) and Vlorë. In addition, commercial bottled waters from four suppliers were investigated: Glina, Kristal, Lajthiza and Spring. To check for possible seasonal variations the drinking water

samples were collected during summer and winter seasons characterised respectively by dry and wet weather conditions. The locations of the sampling sites were distributed across much of Albania, which is diverse in terms of its geologic, hydrologic and soil characteristics. Therefore, deriving a link between the surface and groundwater sources and the quality of drinking water derived from them is a complex process that requires detailed information supported by extensive field data. We based our assessments on field surveys and published maps, such as the Geological map of Albania [17] and Hydrogeological map of Albania [18]. Data from the hydrogeological aspects are summarised in Table 1.

Albania is mostly mountainous, with a mean elevation of 764 m above sea level (a.s.l.). There are many peaks above 2,000 m a.s.l. located in the northern, eastern and southern parts of the country. The highest peak reaches an

Table 1 Approximate locations, reservoir type of drinking water sources and the gross alpha/beta activity concentrations of tap and bottled water samples with their 1σ uncertainties

Sample ID	Sampling site	Geographical coordinates	Type of reservoir	Gross alpha activity (mBq/L)		Gross beta activity (mBq/L)	
				Tap water			
				Summer	Winter	Summer	Winter
BR15	Berat	40°35′30.43″N;20°09′50.17″E	Flysch sand./clay. mixed with calc. rocks	12 ± 3	44 ± 9	94 ± 10	83 ± 10
DR09	Durrës/Lac (Fushë Kuqe)	41°38′44.69″N;19°37′17.14″E	Alluvial and lacustrine deposit	15 ± 3	44 ± 8	674 ± 34	884 ± 57
KR10	Fushë Krujë (Gjole)	41°30′27.02″N;19°41′31.16″E	Alluvial and lacustrine deposit	126 ± 30	42 ± 14	518 ± 49	215 ± 22
KO12	Korçë	40°38′27.47″N;20°31′05.13″E	Flysh calc. sand. surrounded by lim./dol.	24 ± 3	34 ± 10	393 ± 21	295 ± 28
KR05	Krujë	41°31′32.12″N;19°54′07.00″E	Hard lim. altered with lime.	92 ± 14	35 ± 8	348 ± 20	292 ± 22
KU03	Kukës	41°51′42.14″N;20°18′32.64″E	Sand./clay. mixed with calc. congl.	27 ± 7	31 ± 8	74 ± 9	74 ± 10
DB04	Peshkopi	41°48′56.29″N;20°29′08.15″E	Flysch calc. congl.; red sand.	22 ± 5	52 ± 10	134 ± 12	302 ± 19
PG11	Pogradec	40°54′03.41″N;20°43′08.33″E	Hard lim.; flysch red congl. and calc. sand.	121 ± 26	69 ± 15	506 ± 33	321 ± 24
SH01	Shkodër	41°53′21.06″N;19°34′13.38″E	Alluvial deposit	36 ± 9	119 ± 29	326 ± 26	351 ± 30
TP14	Tepelenë	40°17′35.84″N;20°00′56.86″E	Flysch sand. presence of coal deposits	83 ± 21	122 ± 24	79 ± 13	116 ± 19
TR06	Tiranë (Selitë)	41°18′39.95″N;19°47′19.45″E	Hard lim. altered with calc. rocks	33 ± 7	10 ± 3	249 ± 16	29 ± 5
TR07	Tiranë (Bovillë)	41°27′6.50″N;19°53′38.11″E	Derived calc. lim.; dol. over flysch sand./clay.	19 ± 4	28 ± 9	315 ± 24	91 ± 12
VL13	Vlorë	40°25′41.80″N;19°29′37.62″E	Hard lim. mixed with calc. sand./clay.	41 ± 13	30 ± 8	181 ± 23	206 ± 20
				Bottled water			
GJ16	Glina	39°58′14.43″N; 20°17′28.95″E	Gyps.; anh.—evaporate deposit	105 ± 32		151 ± 27	
TR08	Kristal	41°21′32.53″N;19°58′32.57″E	Hard lim. mixed with calc. rocks	53 ± 10		90 ± 10	
PU02	Lajthiza	41°52′43.04″N;20°12′42.94″E	Calc. congl. mixed with ophiolitic sand. and ubas.	13 ± 3		227 ± 15	
GJ17	Spring	39°59′49.42″N;20°16′33.43″E	Gyps.; anh.—evaporate deposit	31 ± 2		781 ± 25	

Sand. sandstone, *Clay.* claystone, *Calc.* calcareous, *Congl.* conglomerate, *Lim.* limestone, *Dol.* dolomite, *Ubas.* ultrabasic rock, *Gyps.* gypsum, *Anh.* anhydrite

elevation of 2,751 m (Mt. Korabi). The west-central part of the country, along the Adriatic coast, is dominated by the Adriatic depression, which consists of hills and plains with elevations below 200 m a.s.l. Albania is in the Mediterranean climate belt, which is characterised by hot, dry summers and mild, rainy winters. The mean annual precipitation amounts to approximately 1,450 mm; more than 3,000 mm falls in the North Albanian Alps, and approximately 650–700 mm falls in the eastern depressions of Korça and Kolonja. These features were taken into consideration when sampling in order to study the seasonal variation of gross alpha/beta activity concentrations during summer/winter seasons.

At each sampling site, 5 L of water was collected in polyethylene containers. When sampling drinking water from taps, the water was allowed to flush for a considerable time before sampling. Each sample was pretreated on site with nitric acid until reaching a pH level below 2. By creating relatively non-polar surfaces, this procedure avoids a loss of radionuclide fractions due to absorption into the walls of the containers. Furthermore, the procedure prevents the formation of precipitates and any biological activity resulting in loss of analytes.

Gross alpha/beta activity measurements using gas-flow proportional counter (GPC) method

According to [9], the total dissolved solids should not exceed a surface density of 5 mg/cm^2 for gross alpha determination and 10 mg/cm^2 for gross beta determination.
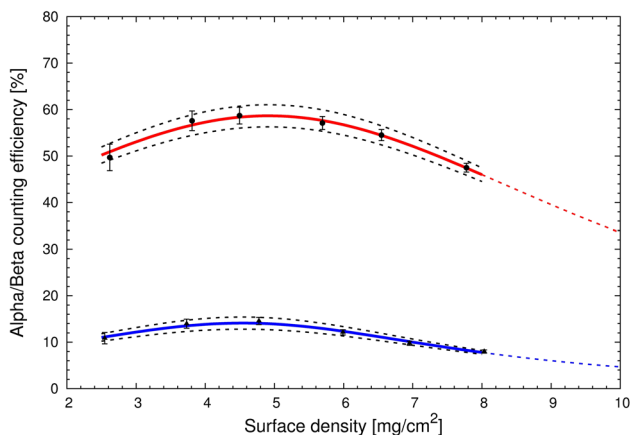
Fig. 2 Gross alpha (*circles*) and beta (*triangles*) efficiency as a function of surface density: the fit is extended up to 10 mg/cm². The *dashed lines* represent the uncertainty of one sigma, respectively less than 5 % for gross alpha and 10 % for gross beta counting efficiency

Accordingly, an aliquot of 100 mL of each sample was evaporated to dryness at 80 °C, and the final concentration of the total dissolved solids (mg/L) was determined. Afterwards, based on the concentration of the total dissolved solids, an appropriate test aliquot of each sample was chosen and was slowly evaporated in a sand bath to avoid spattering. The total dissolved solids were then transferred to a 2-in.-diameter planchet (measurement geometry), using 8 M NHO_3 to carefully wash the beaker. The planchets were then oven-dried at 80 °C and then weighted and stored in a desiccator before analysis. The samples were measured after 30 min to prevent water uptake from the possible presence of nitrates. The samples were measured for 36,000 s using an MPC-9604 ultra-low background gas-flow proportional counter (Protean Instrument Corporation) with passive (lead) and active (guard detector) shielding. The background for gross alpha and gross beta activity concentrations were respectively 0.1 and 0.7 cpm. Calibration of the proportional counter for measuring gross alpha and beta activities involves several steps, such as selecting the appropriate values of the bias, plateau, and discriminants. The operating high voltage of the detector was set at 1,455 V. This proportional counter distinguishes events originating from alpha and beta decays by analysing the pulse height.

To determine the dependence of the gross alpha/beta efficiencies on the mass thickness, two sets of six sources each with surface densities ranging from approximately 2.5 to 8 mg/cm² were prepared using sodium chloride in approximately 8 mL of deionised water solution. The choice of chemical composition of dissolved salts can influence the gross alpha efficiency by up to 20 %, as studied by Timón et al. [20] using Monte Carlo simulation.

The mass variation of standard sources due to hygroscopic properties of NaCl was found negligible within the measurement time interval. The first set was spiked with known activities of [239]Pu in solution, and the second set was spiked with known activities of [90]Sr (^{90}Y) in solution. The final solutions were then evaporated in 2-in.-diameter planchets at a controlled temperature of 80 °C until they reached constant weights, and their mass thicknesses were determined. The gross alpha/beta efficiency absorption curves were fitted to the inverse quadratic function
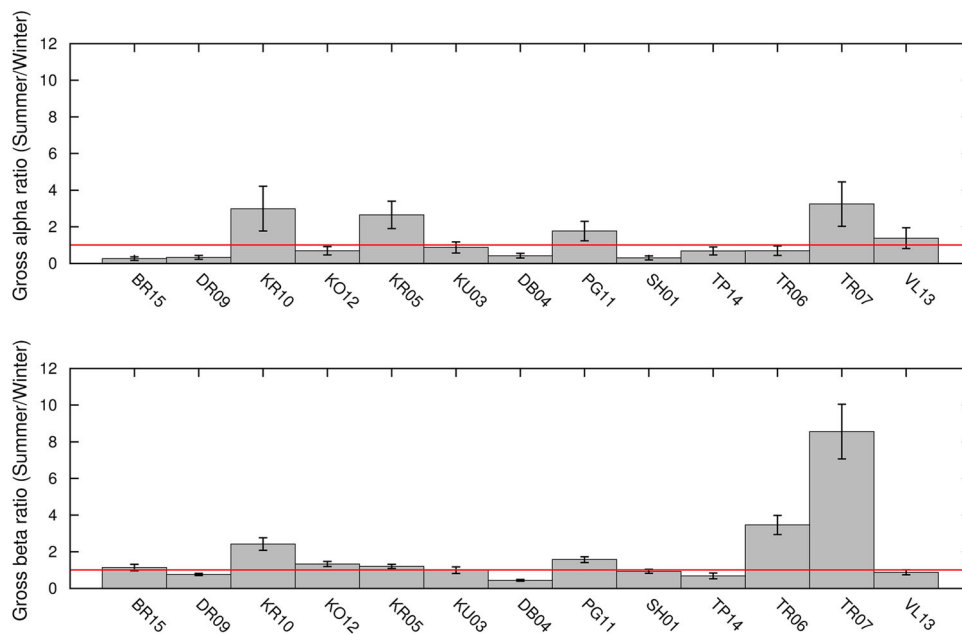
$$\varepsilon_{\text{alpha/beta}} = \left(am^2 + bm + c\right)^{-1} \tag{1}$$

where m is the surface density in mg/cm², and a, b and c are fitting coefficients. For the gross alpha (beta) efficiency absorption curve (Fig. 2), the fitting parameters a, b and c are 0.48, −4.35 and 16.96 (0.05, −0.48 and 2.89), respectively, achieved both with a reduced Chi square value close to unity corresponding to an uncertainty of less than 10 % (5 %). The average crosstalk correction was found to be X_{talk} (alpha-to-beta) = 29.6 ± 2.4 %.

Results and discussion

The gross alpha and beta activity concentrations in the drinking and bottled water samples are presented in Table 1. The relative uncertainties in both the gross alpha and beta radioactivity measurements are less than 30 %. As shown in Table 1, the gross alpha activity ranges between a minimum of 10 ± 3 mBq/L (TR06) and a maximum of 126 ± 30 mBq/L (KR10). The gross beta activity ranges between 29 ± 5 mBq/L (TR06) and 884 ± 57 mBq/L (DR09). Because the distributions of gross alpha and beta activity concentrations in the tap water display slightly positive skews (1.3 and 1.13 for gross beta and gross alpha activities, respectively), we did not use a Gaussian distribution to fit them. Instead, the average activity concentrations were evaluated by fitting them to log-normal distributions. The average gross alpha and beta activity concentrations in the tap water are 36^{+37}_{-18} and 269^{+337}_{-150} mBq/L, respectively. The average gross alpha and beta activity concentrations in the bottled water are 39^{+55}_{-23} and 220^{+336}_{-132} mBq/L, respectively. This analysis results in an increase in the incidence of high and low values in the tails of the distributions. The minimum detectable activities (MDA) [21] of the gross alpha and gross beta activity concentrations are 9.7 and 26.8 mBq/L, respectively. These results were comparable to values of gross alpha and beta activities, respectively, in drinking water in various other countries, such as 49.3 and 128.4 mBq/L in southeastern Turkey [22], <3–500 and 27–1,108 mBq/L in northern Italy [23] and 35–85 and 200–330 mBq/L in northeastern

Fig. 3 Summer/winter ratios of activity concentration of gross alpha and beta. The *solid line* shows the case when no seasonal change in radioactivity is observed

Brazil [24]. The results are lower than values measured in the groundwater (26–1,749 mBq/L) and spring water (33–2,015 mBq/L) in Hungary [2].

The gross alpha and beta activity concentrations of the tap and bottled water samples were below the screening level within a 1σ uncertainty, indicating no dose rate exceeding 0.1 mSv/year. In certain cases, the gross alpha activity concentration (KR05, KR10, PG11, SH01, TP14 and GJ16) is at approximately the screening level within 1σ uncertainty. These samples requires a more detailed analysis for the concentration of radioisotopes due to decay chains of ^{238}U, ^{232}Th and ^{235}U to better ascertain and evaluate any possible radiation risk to humans. As discussed by Landsberger and George [25], the underlying geology of the aquifers in the USA, particularly those in Texas rich in uranium and thorium, is responsible for high ^{226}Ra and ^{228}Ra activity concentration in drinking waters.

There are few data in the literature, in particular from the Mirdita ophiolite unit, which is composed primarily of volcanic rocks (basalt, andesite, dacite and rhyolite) and contains <3.1–688.6 Bq/kg of ^{40}K, <1.2–6.4 Bq/kg of ^{238}U and <0.4–4.9 Bq/kg of ^{232}Th [26], quartz diorites, plagiogranites, and gabbroic rocks, which contain <3.1–150.2 Bq/kg of ^{40}K and <0.4–1.2 Bq/kg and ^{232}Th. There are no available data for ^{238}U [27] and ultramafic rocks of mantle and crustal origin with very low radionuclide concentrations (reported MDA values). These values are comparable with those of other ophiolite complexes in the Alps–Apennines (Italy), Pindos (Greece), Samail (Oman), and Troodos (Cyprus), which range between <3.1 and 187.8, 0.1–16.1 and 0.2–11.4 Bq/kg, respectively, for ^{40}K, ^{238}U and ^{232}Th [28] and references therein]. These areas are the locations of samples KU03, DB04, PU02 and

KO12, which appear to reflect the radioactivity content of their corresponding reservoir rocks and exhibit low concentrations of gross alpha and beta activity. Evidence of calc-alkaline rocks are reported in eastern Albania ophiolites, which show high activity concentrations of ^{40}K, ^{238}U and ^{232}Th ranging from 144.0 to 322.4, 14.8 to 51.9 and 24.3 to 49.7 Bq/kg, respectively [29] (PG11).

In this study, measurements of the activity concentration in samples of limestones from sites KR05, TR06, TR07 and TR08 and evaporates (gypsum) and sites TP14, GJ16 and GJ17 were performed using high resolution gamma-ray spectrometry (HPGe). The HPGe used is described in details in Xhixha et al. [15]. The activity concentrations of ^{40}K, ^{238}U and ^{232}Th are 4.3 ± 0.7, 45.3 ± 4.2 and 1.0 ± 0.5 Bq/kg, respectively, in the limestones and 45.9 ± 5.0, 10.1 ± 1.0 and 2.9 ± 0.3 Bq/kg, in the gypsum. The resulting concentrations are lower than the world average in soils and crustal rocks [30], except for the U in the limestones, which is slightly higher.

However, the highest concentrations of gross alpha and beta activity are found in alluvial and lacustrine deposits, which are frequently replenished with recent deposition by the Mati and Ishëm Rivers. These sites (DR09, KR10 and SH01) are characterised by shallow groundwater that is often at a depth of less than 5 m. A nationwide study of industrial clays [31] and end-products [32] yielded MDA values ranging up to 1189.4, 101.3 and 181.1 Bq/kg, respectively, for ^{40}K, ^{238}U and ^{232}Th. The maximum values were measured near the locations of sites DR09, KR10 and SH01. These high concentrations may be responsible for the elevated values of the gross alpha and beta activity concentrations.

Table 1 presents the gross alpha and beta activity concentrations in tap water measured during the summer and

winter. Figure 3 shows the summer/winter ratios of the gross alpha and beta activity concentrations. No seasonal trend was observed [1], as various water treatment processes may affect the concentrations of radionuclides. However, approximately 60 and 45 % of the sites investigated exhibit no variation in their gross alpha and beta activity concentrations, respectively, within a 95 % confidence interval (i.e. 2σ). The sites showing ratio below unity in Fig. 3 are located in areas of high winter precipitation, in particular DR09, DB04, BR15, SH01 for gross alpha and DR09, DB04 for gross beta. Considering the sites showing ratios above unity are located in areas of low winter precipitation (e.g. KR05 for the gross alpha and KR10, KO12, PG11, TR06, TR07 for gross beta), we observe a weak evidence of seasonal variation of gross alpha/beta activity concentration in drinking water.

Conclusions

This initial screening for gross alpha and beta activity concentrations included the investigation of tap water in the 12 major cities in Albania and four sources of bottled mineral water. The gross alpha activity in these samples varies from 10 ± 3 to 126 ± 30 mBq/L, and the gross beta activity varies from 29 ± 5 to 884 ± 57 mBq/L. The average gross alpha and beta activity concentrations in the tap waters are 36^{+37}_{-18} and 269^{+337}_{-150} mBq/L, respectively. The average gross alpha and beta activity concentrations in the bottled waters are 39^{+55}_{-23} and 220^{+336}_{-132} mBq/L, respectively. In all of the samples, the gross alpha and beta activity concentrations were below or approximately equal to the recommended reference level, and thus they pose no significant contribution to radiation exposure due to water consumption. Since the gross methods estimates approximately the activity concentration, as well documented in Jobbágy et al. [33], we strongly recommend further detailed measurements in particular in the area where we have evidences of high concentrations. This preliminary screening constitutes baseline information for the environmental and health impact due to oil and gas extraction in a scenario when new technologies of unconventional shale gas extraction are of great concern due to contamination of ground and surface water from discharge of wastewater containing NORMs, in particular radium.

No significant differences in gross alpha/beta activity concentrations regarding seasonal variations were observed in this preliminary study. However, a dilution factor due to rainfall in the northern Albania sites, which are located in areas of high precipitation, may be a weak indication for future investigations. Moreover, studies of the radioactivity content of the primary geological reservoirs can help to understand the relationships between radionuclides in groundwater and geochemical environment.

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