



Dispersion of radionuclides and heavy metals from phosphogypsum stacks in soil and plants at Northwestern Greece

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Abstract

Phosphogypsum is classified into naturally occurring radioactive materials containing radionuclides such as ^{238}U , ^{226}Ra and ^{232}Th , and heavy metals which are dispersed in different form and concentrations and can be dangerous for humans. Determination of the concentration of radionuclides in soils and plants near a phosphate fertilizer production unit and phosphogypsum stacks was carried out by γ - and α -spectroscopy while trace elements were determined by Neutron Activation Analysis and Atomic Absorption Spectroscopy. Transfer and enrichment factors were estimated as well as radiation risks and the results led to the conclusion that the radiation hazard in the investigated region was below than the recommended level.

Keywords Phosphogypsum · Radioactivity · Uranium · Heavy metals · Soils · Plants

Introduction

Phosphogypsum (PG) is a waste by-product of the phosphate fertilizer industry. The composition of the material, generated from the wet-process between calcium phosphate ore, $\text{Ca}_{10}(\text{PO}_4)_2\text{F}_2$, and sulfuric acid, is $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ (96%), P_2O_5 (1–2%), F (1.2%), SiO_2 (1%) and Al_2O_3 (0.2%) [1–3].

The PG production rises globally to 300 million tons per year and most is stock-piled. Approximately 5.2 tons of phosphogypsum are produced per ton of phosphoric acid (PA) in a typical facility [4, 5]. The by-product has high volume, low toxicity and according to International Atomic Energy Agency (IAEA) is classified into N.O.R.M. (Naturally Occurring Radioactive Materials) [6]. It contains radionuclides, as well as heavy metals such as Pb, Zn, Fe, Mn, Cu, Ni, Cd and As. Most of the radionuclides of the uranium and thorium series appear in the phosphoric acid and then in the phosphate fertilizer, whereas radium (and its decay products), favors the phosphogypsum because is chemically like calcium [7–9]. The average uranium concentration in phosphate rocks worldwide range from 25 to 50 ppm while high concentrations up to 600 ppm have been found in local

deposits. The greatest percentage (80–90%) of uranium transfers in the phosphate fertilizer [10, 11].

Circa 15% of world phosphogypsum production is recycled and applied in agriculture (amended soil), constructions (cement and concrete), mine reclamation and Rare Earth extraction [12, 13]. Moreover, the majority is disposed in large stacks, usually placed in regions close to the PA producing factories like in Florida, China, Brazil and Morocco. These stacks are exposed to weathering dispersing NPs into the environment and undergo leaching process. As dust PG is carried by air and water and thus heavy metals can be transported to adjacent surface and ground water resources [14–22].

More than 1.7 billion tons of PG stored in the U.S. and worldwide whereas the IAEA in order to establish standards of safety for protection of health, concluded that PG with 1 Bq g^{-1} or less can be reused without any meaningful restriction, and with higher concentrations can still be reused safely under certain conditions [4, 6, 22]. A significant number of publications can be found in the literature concerning mainly the radiological impact of the phosphogypsum. Up to our knowledge there is lack of information concerning the dispersion of radionuclides and trace elements after long term storage of PG in the surrounding regions and their accumulation, particularly of uranium, by plants [23–26].

The aim of this work was to determine the natural radionuclides and heavy metals dispersed from PG stacks at a phosphate fertilizer production factory, in the surrounding

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ecosystem, to draw an overall estimation of the hazard and radiological impact from phosphogypsum. For this reason, PG samples, soils and plants were collected and analyzed in terms of minor and trace elements, determined by Instrumental Neutron Activation Analysis (INAA) and Flame Atomic Absorption Spectroscopy (AAS), and in natural radionuclides, determined by γ - and α -spectroscopy.

Materials and methods

Study area and sampling

For this study 24 samples of soil and 18 of plants were collected from a region close to the phosphate fertilizer production factory, which is located at Nea Karvali in Kavala gulf in Northwestern Greece. The factory has been operating since 1965 and produces 500,000 tons phosphate fertilizer per year [27]. The samples were collected at sites 1 (next to the fence of the factory), 2 and 3 (at distance 0.5 and 1.5 km from the fence), as indicated in Fig. 1a, (corresponding coordinates, longitude and latitude A: 40° 52' 18.1" N 24° 30' 01.1" E, B: 40° 57' 19.3" N 24° 30' 20.1" E and C: 40° 57' 27.8" N 24° 31' 05.4" E). Furthermore, six PG-samples (spot 1, 2 and 3 respectively) taken from an aged PG stack (more than 15 years) were also analyzed (Fig. 1b).

Surface soil samples (circa 2 kg) were normally collected using a metallic sampler from a certain area (30 × 30 cm²) at 0–15 cm depth). Samples of native wild plants (herbs and radishes) growing around were also collected. The PG-samples were taken from the surface of the piles at different locations, according to the Environmental Protection Agency guidelines [28–30]. The sampling was done for one-year period, in two seasons end of March and September

(wet and dry period), and in each location, three samples were collected.

Concerning the climate at the region of Kavala, is characterized by mild winters and dry, hot summers and the prevailing wind direction is south-east (average annual humidity 71% and annual rainfall 700 mm).

The samples were placed in plastic bags and transferred immediately to the laboratory. The soil samples were cleaned from stones and the plants were cut in small pieces after washing with water and distilled water. Then were dried in an oven at 60 °C until constant weight and pulverized into a fine powder after passing through a standard 1-mm mesh size (ASTM No. 18). The homogenized samples were placed into cylindrical sealed plastic beakers (volume 50 cm³) and stored for at least four weeks before the measurement to attain radioactive secular equilibrium of ²²⁶Ra, ²²²Rn and their short-lived progenies.

Applied techniques

For the γ -ray spectrometry a high-resolution detector was used (HPGe detector, efficiency 20%, energy resolution 2.1 keV for the 1332 keV ⁶⁰Co γ -radiation). Details for the measurements have been described earlier [28]. The minor and trace elements of the samples were determined by Instrumental Neutron Activation Analysis (INAA), at the 2 MW pool-type research reactor (TU-Delft, The Netherlands, neutron dose: 4.5–5.3 × 10¹⁶ n cm⁻² s⁻¹, activation time: 4–5 h) and Flame Atomic Absorption Spectroscopy (AAS) using a Perkin Elmer AA300 spectrophotometer.

The determination of the concentrations of uranium isotopes ²³⁸U and ²³⁴U in plants was performed by alpha spectrometry after digestion and dilution of the sample using an Ortec Dual 576A alpha spectrometer and PIPS detectors (450 mm²). Prior to sample measurement, the background



Fig. 1 a Investigated area and sampling sites, b PG-samples from the PG stack

Table 1 Activity concentrations (Bq kg⁻¹) of ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K in PG-samples

	²³⁸ U	²²⁶ Ra	²³² Th	⁴⁰ K
Spot 1	28.9 ± 1.9	573.5 ± 30.6	2.2 ± 1.6	2.1 ± 0.19
Spot 2	27.6 ± 1.6	253.6 ± 19.2	15.6 ± 3.9	142.3 ± 11.8
Spot 3	25.4 ± 1.4	48.6 ± 3.8	51.9 ± 5.9	562.4 ± 34.9
Worldwide range*	15–60	162–5126	2.1–35.2	15–1410

*[25, 26]

was carefully measured under identical conditions and was found to be about 50 counts per day within the energy range of 3–8 MeV. The efficiency of the separation using the cation exchange Chelex 100-resin was around 75% [28]. The determinations with gamma and alpha spectrometry were performed in duplicate. Radiological health indexes as Radium equivalent activity (Ra_{eq}) in Bq kg⁻¹, external (H_{ex}), internal hazards (H_{in}), and gamma level index (I_γ) in Bq kg⁻¹, absorbed dose rate (D) in nGy h⁻¹, and annual effective dose equivalent, external and internal (AEDE_{in} and AEDE_{ex}), in mSv y⁻¹, were calculate using the following formulas 1–7 [31].

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.07C_K \tag{1}$$

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \tag{2}$$

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \tag{3}$$

$$I_{\gamma} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{1500} \tag{4}$$

$$D(\text{nGy h}^{-1}) = 0.461C_{Ra} + 0.623C_{Th} + 0.0414C_K. \tag{5}$$

$$AEDE_{ex}(\text{mSv y}^{-1}) = D(\text{nGy h}^{-1}) * 8760(\text{h y}^{-1}) * 0.2 * 0.7(\text{Sv Gy}^{-1}) * 10^{-6} \tag{6}$$

$$AEDE_{in}(\text{mSv y}^{-1}) = D(\text{nGy h}^{-1}) * 8760(\text{h y}^{-1}) * 0.8 * 0.7(\text{Sv Gy}^{-1}) * 10^{-6} \tag{7}$$

where C_{Ra}, C_{Th} and C_K correspond to the activities of ²²⁶Ra, ²³²Th and ⁴⁰K in Bq kg⁻¹.

Contamination factor (CF) and contamination degree (C_d)

Contamination Factors (CFs) of the specific heavy metals for a specific sampling site were determined, which can be described as follows [32]

$$CF = \frac{(\text{Metal concentration}) \text{ Sample}}{(\text{Metal concentration}) \text{ Background}} \tag{8}$$

Values of CF < 1, 1 ≤ CF < 3, 3 ≤ CF < 6 and CF ≥ 6, are of evidence of low contamination, moderate contamination, considerable contamination and very high contamination.

Table 2 Hazard Indices determined for PG-samples

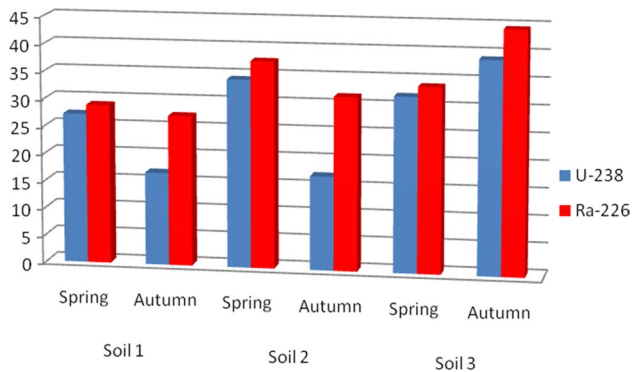
	Ra _{eq} (Bq kg ⁻¹)	H _{in} (Bq kg ⁻¹)	H _{ex} (Bq kg ⁻¹)	I _γ (Bq kg ⁻¹)	D (nGy h ⁻¹)	AEDE _{ex} (mSv y ⁻¹)	AEDE _{in} (mSv y ⁻¹)
Spot 1	576.8	3.11	1.56	3.11	230.9	0.283	1.132
Spot 2	286.9	1.46	0.78	1.53	117.1	0.144	0.574
Spot 3	164.9	0.58	0.45	0.83	74.6	0.092	0.368

Table 3 Concentrations (mg kg⁻¹) of metals in PG-samples

	As	Cr	Ba	Cd	Sc	Co	Zn	Fe	Sb	Sr	U	Th
Spot 1	0.61	15.8	38.3	6.67	0.39	0.51	12	12,800	0.20	470	2.24	0.59
Spot 2	4.68	68.6	166	4.01	4.24	5.76	46.5	10,400	0.34	336	2.21	4.27
Spot 3	17	153	331	3.88	13.1	17.4	123	31,900	0.84	172	2.03	10.1
	Hf	Lu	Yd	Td	Sm	Nd	Ce	La	Eu	Cs	Ta	W
Spot 1	0.68	0.25	2.28	0.66	3.53	21.5	19.2	30.5	0.94	0.09	0.07	0.60
Spot 2	2.15	0.26	2.15	0.70	3.79	17.7	30.1	24.9	0.85	1.49	0.25	0.79
Spot 3	5.04	0.31	2.72	0.74	5.31	23.9	60.7	28.8	1.08	4.82	0.84	2.22

Table 4 Activity concentrations (Bq kg⁻¹) of ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K in soils

	²³⁸ U	²²⁶ Ra	²³² Th	⁴⁰ K
<i>Spot 1</i>				
Spring	27.2 ± 0.9	28.9 ± 0.6	33.2 ± 1.7	500 ± 35.1
Autumn	16.8 ± 0.4	27.3 ± 1.5	31.2 ± 1.6	537 ± 37.6
<i>Spot 2</i>				
Spring	34.2 ± 2.4	37.6 ± 2.6	55.6 ± 3.9	550 ± 38.5
Autumn	17.1 ± 1.2	31.6 ± 2.2	47.8 ± 3.3	546 ± 38.2
<i>Spot 3</i>				
Spring	31.94 ± 2.2	33.8 ± 2.4	83.8 ± 5.9	527 ± 36.9
Autumn	38.9 ± 2.7	44.4 ± 3.1	68.1 ± 4.8	359 ± 25.1
Min	16.8 ± 0.4	27.3 ± 1.5	31.2 ± 1.6	359 ± 25.1
Max	38.9 ± 2.7	44.4 ± 3.1	83.8 ± 5.9	546 ± 38.2
Average	25.4 ± 2.4	32.3 ± 2.3	64.9 ± 4.6	503 ± 36.2

**Fig. 2** Distribution of activity concentrations (Bq kg⁻¹) of ²³⁸U and ²²⁶Ra in soils

Another index which characterizes the level of heavy metal pollution is the *Contamination degree*, C_d calculated from the Contamination Factors as described by Eq. (9),

$$C_d = CF_1 + CF_2 + CF_3 + \dots + CF_n \quad (9)$$

where CF_1, CF_2, CF_3, CF_n represent the Contamination Factors. Values of $C_d < 7$, $7 \leq C_d < 14$, $14 \leq C_d < 28$ and $C_d \geq 28$ indicate low contamination, moderate contamination, considerable contamination and very high contamination.

Enrichment factor (EF), transfer factor (TF)

To estimate the anthropogenic impact on soil, calculation of a normalized *Enrichment Factor (EF)* was performed according to the Eq. (10). This factor is referred to metal concentrations above uncontaminated background levels. The background concentrations were based on the elemental abundances of continental crust and iron (Fe) was used as the reference element for geochemical normalization because it exhibits a geochemistry like that of many trace metals and its natural mass fraction is almost uniform [33, 34].

$$EF = \frac{(\text{Metal/Fe})_{\text{Sample}}}{(\text{Metal/Fe})_{\text{Background}}} \quad (10)$$

Metal/Fe represents the mass fraction of metal to the mass fraction of Fe in the sample and in the background (reference values) respectively. When $EF > 1$ the element is of anthropogenic origin. Values in the range 1.5–3, 3.01–5, 5.01–10 and > 10 indicate minor, moderate, severe and very severe contamination.

Transfer Factor (TF) is defined as the ratio of the metal concentrations in plant to that in soil according to the Eq. (11) [35].

$$TF = C_{\text{metal(plant)}} / C_{\text{metal(soil)}} \quad (11)$$

Table 5 Hazard Indices determined for soils

	Ra _{eq} (Bq kg ⁻¹)	H _{in} (Bq kg ⁻¹)	H _{ex} (Bq kg ⁻¹)	I _γ (Bq kg ⁻¹)	D (nGy h ⁻¹)	AEDE _{ex} (mSv y ⁻¹)	AEDE _{in} (mSv y ⁻¹)
<i>Soil 1</i>							
Spring	114.9	0.39	0.31	0.62	52.94	0.065	0.259
Autumn	113.3	0.38	0.31	0.63	52.59	0.064	0.258
<i>Soil 2</i>							
Spring	159.5	0.53	0.43	0.78	72.45	0.089	0.355
Autumn	142.0	0.47	0.38	0.72	65.02	0.079	0.319
<i>Soil 3</i>							
Spring	194.2	0.62	0.52	0.86	87.55	0.107	0.429
Autumn	169.4	0.58	0.46	0.74	75.05	0.092	0.368

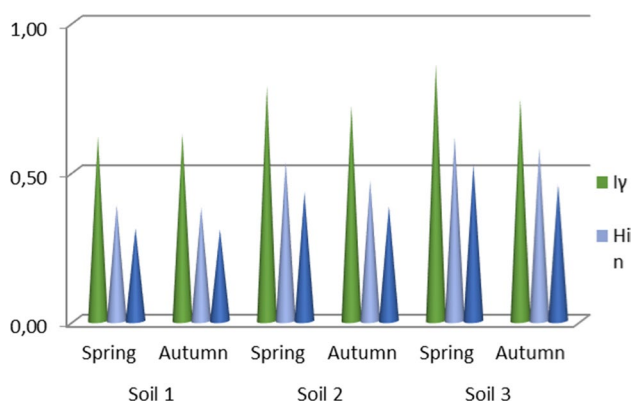


Fig. 3 Values of I_γ factor and Hazard internal and external (H_{in} , H_{ex}) in soils

Results and discussion

The activity concentrations (Bq kg^{-1}) of ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K in PG-samples are given in Table 1 with the associated standard deviations. The gamma rays at 92.4 and 92.8 keV of ^{232}Th were used to determine ^{238}U based on the radioactive equilibrium. The natural radionuclides ^{226}Ra and ^{232}Th were determined via the decay of ^{214}Pb (295.2 and 351.9 keV) and ^{214}Bi (609.3, 1120.2 keV) as well as of ^{228}Ac (911.2 and 968.9 keV), ^{212}Pb (238.6 and 300.0 keV) and ^{208}Tl (583.1 keV) respectively. It is shown that the sample in spot-1 is raw phosphogypsum with high activity of ^{226}Ra and low of ^{232}Th while in spot-2 the samples are mixed with soil and in spot-3 the constitution of the samples tends towards that of the soil. Although the activity concentrations for PG-samples, which appeared in the literature, varied significantly (for example for ^{226}Ra different values have been reported; 310 Bq kg^{-1} in Iraq and Syria, 462 Bq kg^{-1} in Greece, 591/811 Bq kg^{-1} in Brazil, 958 Bq kg^{-1} in Florida and 1420 Bq kg^{-1} in Morocco), the determined values in this study were in the worldwide range [22, 25, 26].

Radiological indexes such as Ra_{eq} , D , I_γ , H_{ex} and H_{in} , are given in Table 2. The Ra_{eq} value in spot-1 was higher than the recommended accepted limit of 370 Bq kg^{-1} , for safe use [31]. The I_γ values for the spots 1, 2 and 3 reflecting the dispersion of phosphogypsum and ranged between 0.83 and 3.11 Bq kg^{-1} while the annual effective dose ranged between 0.48 and 1.42 mSv y^{-1} and was always higher than the recommended value 0.46 mSv y^{-1} .

The elemental analysis of the PG samples identified by INAA is presented in Table 3 where can be seen that some metals, such as rare earths, arsenic, zinc and uranium, are present in smaller concentrations than the average values in literature [19]. As it is known the constitution of phosphogypsum depends on the origin of the phosphate rock and the age of the PG-piles. A significant amount (~85%) of the rare earths elements (REE) and mobile metals as uranium and zinc passes in the PG matrix but no significant enrichment of REE has been observed in aged PG-piles [21, 36, 37]. Generally, both concentrations of metals and concentrations of trace elements in PG samples, in this work agree with literature values [20, 23].

The activity concentrations of the ^{238}U , ^{226}Ra and ^{232}Th radionuclides for the soil samples, 9 including minimum, maximum, mean values and standard deviations are presented in Table 4. The values ranged from 16.8 to 38.9, 27.3 to 44.4 and 31.2 to 83.8 Bq kg^{-1} for ^{238}U , ^{226}Ra and ^{232}Th . The differences between cold and warm periods were negligible. Uranium and radium seem to follow the same trend with higher activity concentrations in spring in two sampling sites as it is shown in Fig. 2. The opposite was observed in site 3 likely as consequence of the wind direction in the region. For uranium the difference between the two seasons was higher, probably because this element is transferred from the phosphate rock to the bioavailable fraction in phosphogypsum and finally in the water-soluble phase, being dangerous for the environment [38]. The mobility of the metals is a complex matter depending, among others (pH, speciation, etc.) and on weather conditions. The regional climate, as has been also mentioned, is of a Mediterranean

Table 6 Concentrations (mg kg^{-1}) of selected metals in investigated soils and in literature

This work	As	Cd	Cu	Ni	Pb	Zn
	0.001–1.29	0.06–0.97	1.46–5.63	1.052–33.7	0.699–19.4	161.2–238.7
Voutsas and Samara [45], Greece	27.5–44.4	–	17.9–39.5	–	15.5–37.0	36.0–124
Kicińska and Wikar [43] Poland	15.6–38.6	–	–	13.2–28	37.5–952	153–3014
Folens et al. [44], Belgium	1.97–8.52	–	11.1–107.4	4.07–19.69	14.86–303.3	38.7–1325
Huang et al. [46], China	0.7–1.53	0.0157–0.0168	–	–	23.5–24.98	–
Ahmad et al. 2010, Bangladesh	–	6.21–16.11	31.3–45.2	36.03–74.16	44.31–52.21	103.2–123.5
Mazzilli et al. [23], Brazil	2.4–53	0.8–1.0	21–58	7.7–44	22–100	70–120
Alina Kabata-Pendias [42]	6.83	0.41	113	29	27	70

With bold indicated values higher than those of this study.

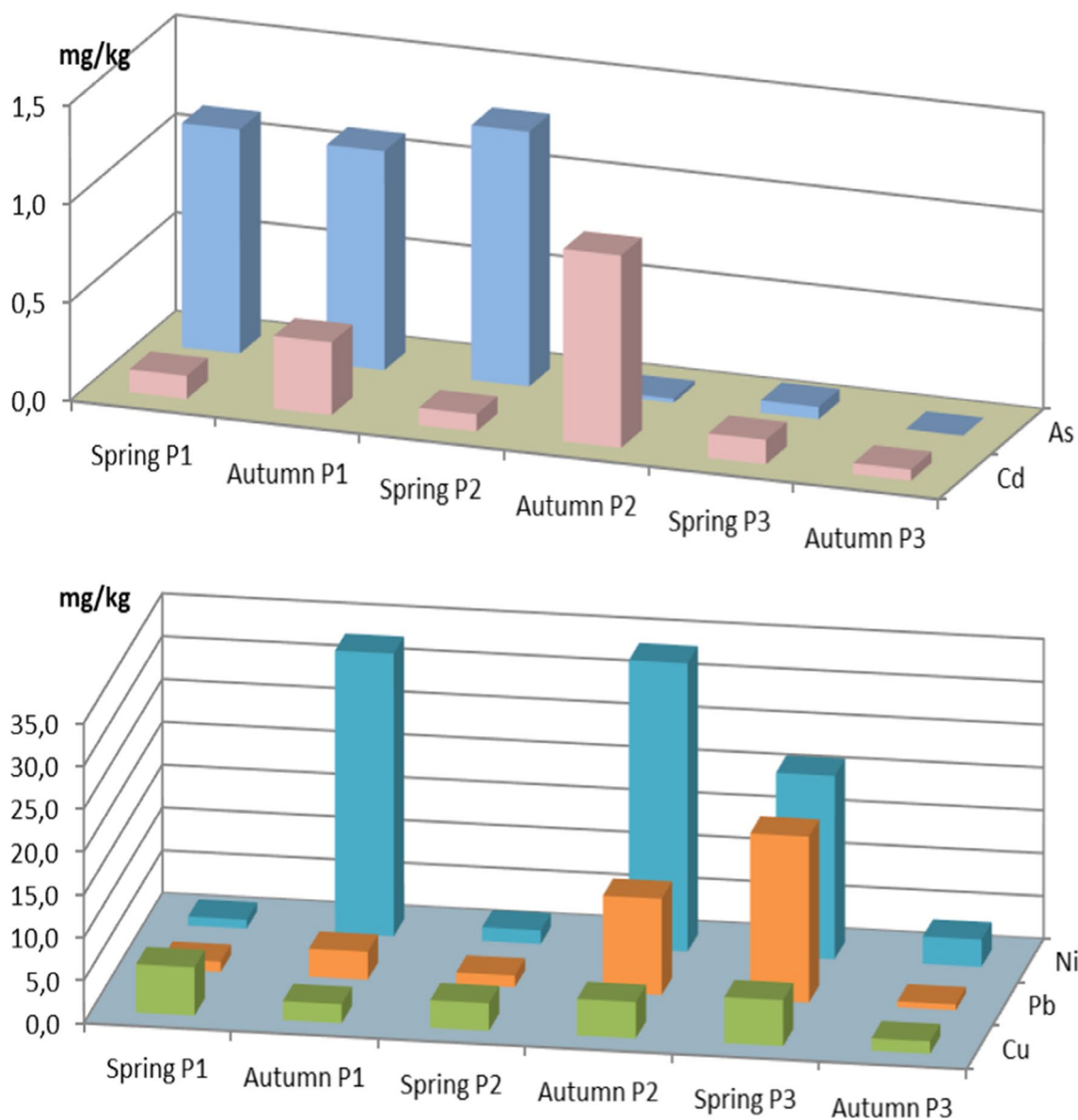


Fig. 4 Distribution of metal concentrations (mg kg^{-1}) in soils

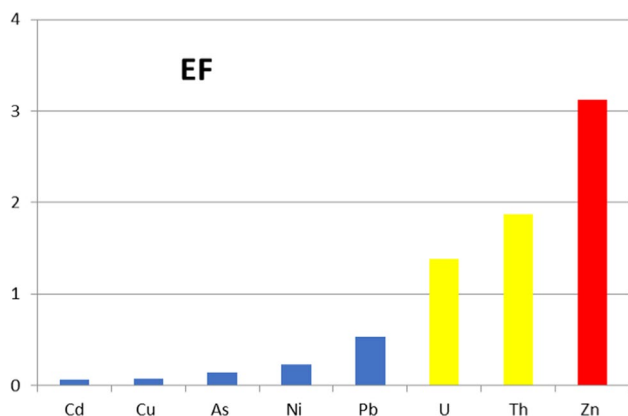


Fig. 5 Enrichment factor (EF) of the metals in soils

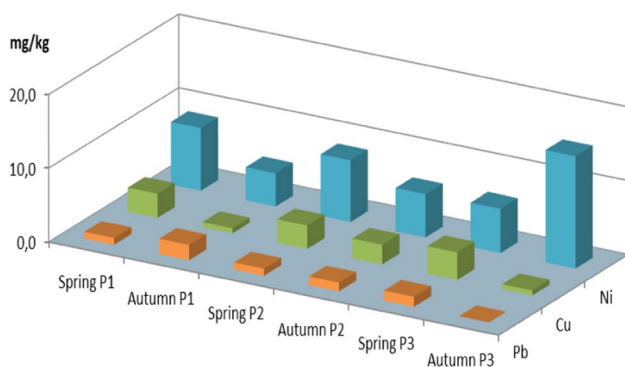
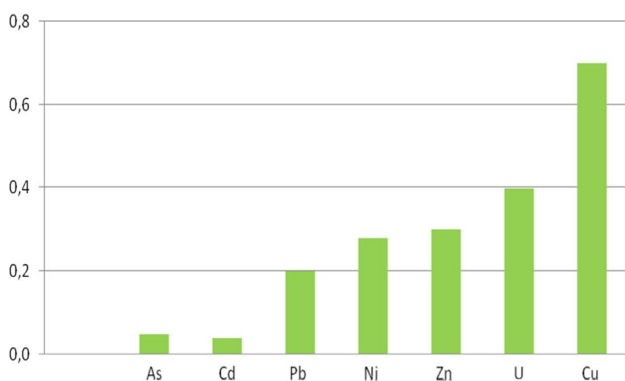
type and characterized by rainy winters and warm-dry summers. This means that in rainy periods, elements bounded to the bioavailable fraction, form soluble phases in rainwater, and released into the surrounding environment [8, 9, 36–39].

The recorded values of the measured radionuclides in soils are in the range of the worldwide activity concentrations but higher than the world mean values of ^{238}U (35 Bq kg^{-1}), ^{226}Ra (35 Bq kg^{-1}), ^{232}Th (30 Bq kg^{-1}) and ^{40}K (400 Bq kg^{-1}), recommended by UNSCEAR 2000 [9, 22, 26, 28, 36–39]. Similar observations have been reported by other researchers in relevant studies in Nigeria, Greece, Tunisia, and Syria [15, 22, 37–39]. The radiological indices are tabulated in Table 5 and illustrated in Fig. 3. The Ra_{eq} activity varied from 113.3 to 194.2 Bq kg^{-1} lower than the

Table 7 Metals concentrations (mg kg^{-1}) in plants in this work and in literature

This work	As	Cd	Cu	Ni	Pb	Zn
	0.001–0.16	0.001–0.14	0.69–3.75	4.46–15.2	0.001–2.08	0.001–260
Voutsas and Samara [45], Greece	0.12		2.8		11.2	39
Kicińska and Wikar [43] Poland				2.9–5	1–8.9	60–373
Folens et al. [44], Belgium	0.05–2.9				0.3–34.1	
Warming et al. 2012, Denmark	0.21	0.25	8.66	0.62	1.56	77.9
Alina Kabata-Pendias [42]		0.4–2.7	4.2–35	1.0–4.8	0.1–28	59–180
Jolly et al. [33], Bangladesh	0.01–0.08	0.04–0.97	0.22–6.69	0.13–0.94	0.001–0.98	0.43–112.2
Mazzilli et al. [23], Brazil	0.01–0.15	0.008–0.12	1.1–3.9	3.5–12.7	0.2–29	9.9–67

With bold indicated values higher than those of this study

**Fig. 6** Distribution of metal concentrations (mg kg^{-1}) in plants**Fig. 7** Transfer factor (TF) of the metals from soil to plants

recommended value. The values of external (H_{ex}) and internal hazards (H_{in}) and Gamma level index (I_{γ}) were always lower than unity indicating that the soil in this area is safe for use.

The concentrations of heavy metals in the investigated regions are given in Table 6 together with literature data for comparison whereas their distribution is presented in Fig. 4 (except of Zn which had concentrations from 161.2 up to 238.7 mg kg^{-1}). As it is shown the accumulation of metals

in this region is lower in comparison to other reported data [40–45]. Concerning the metal spatial dispersion, no safe conclusion can be drawn because metals due to their high density can be dispersed as dust by wind and deposited near the surrounding area [37]. Moreover, the incidental NPs from the PG stacks contribute to the distribution of REE, toxic elements such as As, Pb and Cd, and natural radionuclides [19].

The average value of the Contamination Factor (CF) was lower than unity for the elements As, Pb, Cu and Ni and between 1 and 3 for Zn, Cd, U and Th, signifying low and moderate contamination respectively which was also proven and by the Contamination degree (C_d). Figure 5 illustrates the Enrichment Factor (EF) of the metals in the soils close to the fertilizer unit where slight enrichment was observed for most of the metals and moderate enrichment for Zn.

The metal concentrations in plants are presented in Table 7 along with published data for comparison. Most of these data refer to different pollution sources because there is a lack of knowledge about the impact of phosphogypsum on plants grown naturally in the vicinity of PG-stockpiles [8, 23, 37]. As it is shown in most of the cases the values in this work were lower than relevant literature data reflecting the moderate contamination in the region. Slight differences were observed in different points and in different season as can be seen in Fig. 6 with higher values in rainy period (spring) [37–46]. The transfer of heavy metals to plants is not only due to the absorption by the plants' roots but also due to the precipitation on the leaves by rain or wind which could not be removed by washing. Generally highest TF values were observed in the literature for the leafy plants while lowest values were observed for grains and the accumulation of metals in plants is connected with the soil pollution [8, 23, 37, 42].

The determined concentrations for uranium ranged between 0.3 and 0.7 (mg kg^{-1}) but general the results referred to uranium in plants are very limited. In Russia they found concentrations up to 20 mg kg^{-1} for native plants. Uranium concentrations between 0.15 and 0.25 and

1–3 mg kg⁻¹ were reported for vegetables in Italy and Vietnam [46–50]. It is worth to be mentioned that usually, the plants investigated in the literature concern vegetables and not native wild plants.

Figure 7 presents the Transfer Factor (TF) of the metals from soil to plants. The values found in this study are according to these reported by other researchers (e.g. Mazzilli et al. in Brazil) and lead to the conclusion that the transfer of toxic metals from the fertilizer unit to the surrounding environment is limited [23]. Of course, there is a large variability among TF data due to many parameters concerning soil, plants group and crop. In this study uranium and copper exhibited the highest TF values, followed the metals Zn, Ni and Pb, while low TF values were exhibited by Cd and As [8, 9].

Conclusions

To assess the hazard from radiological and chemical pollution due to dispersion of radionuclides and heavy metals from phosphogypsum stacks in the surrounding region, soil and native wild plants were investigated. For the soil samples the activities of the radionuclides were comparable to the literature and the values of external (H_{ex}), internal hazards (H_{in}) and Gamma level index (I_γ) were always lower than unity indicating safe use of the soil. The metal concentrations were in agreement with results appeared in the literature and the Contamination Factors and Contamination degree signifying low and moderate contamination. Slight enrichment of the soils was observed for most of the metals and moderate enrichment for Zn according to Enrichment Factor values.

For the plants in most of the cases the metal concentrations were lower than relevant literature data reflecting the moderate contamination in the region. Slight differences were observed in different points and in different season with higher values in rainy period (spring) due to the precipitation by rain. The TF values showed variability due to many parameters concerning soil, plants group and crop.

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Declarations

Conflict of interest The authors declare that they have no competing interest.

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