



Radiological and chemical assessment of phosphate rocks in some countries

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ABSTRACT

In this study, the radiological, structural and chemical characterizations of Mardin-Mazıdađı phosphate rock, which is an important phosphate fertilizer source in Turkey were investigated and compared to those of several different phosphate rocks of Tunisia, Egypt, Morocco, Algeria and Syria using gamma spectrometry, X-ray diffraction (XRD) and X-ray fluorescence (XRF) measurement techniques. Elemental analysis results of phosphate samples showed that they were mainly composed of CaO, P₂O₅, SiO₂, Al₂O₃, SO₃ and Fe₂O₃. Elemental concentrations of U and Th were calculated using ²²⁶Ra and ²³²Th activity concentrations, respectively. As a result of XRD analysis, the main peaks of the samples were found to be Fluorapatite (Ca₅(PO₄)₃F). The radioactivity concentration levels for ²²⁶Ra, ²³²Th and ⁴⁰K in all phosphate samples ranged from 250 to 1029 Bq kg⁻¹ with a mean of 535 Bq kg⁻¹, from 5 to 50 Bq kg⁻¹ with a mean of 20 Bq kg⁻¹ and from 117 to 186 Bq kg⁻¹ with a mean of 148 Bq kg⁻¹, respectively. The computed values of annual effective doses ranged from 0.17 to 0.59 mSv, with a mean value of 0.33 mSv, which is lower than the recommended limit of 1 mSv y⁻¹ by the International Commission on Radiological Protection.

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1. Introduction

Phosphate rock is a general term which refers to rock with high concentration of phosphate minerals, most commonly of the apatite group {Ca₅(PO₄)₃[F, OH or Cl]}. The phosphate rock is used to manufacture phosphate fertilizers and industrial products and, also the only significant global resource of phosphorus used in animal feed supplements, food preservatives, anti-corrosion agents, cosmetics, fungicides, ceramics, water treatment and metallurgy [1]. Composition of phosphate rock largely depends on its type and origin, which is mainly composed of phosphorus and minutely of many other elements. This result supported that the phosphorus released from sedimentary phosphate rock containing high concentration of heavy elements was mainly originating from inorganic phosphorus fraction [2].

In addition, phosphate minerals occurring in the primary environment include: Fluorapatite (Ca₁₀(PO₄)₆F₂), found mainly in igneous and metamorphic environments, e.g., in carbonatites, and mica-pyroxenites, hydroxy-apatite (Ca₁₀(PO₄)₆(OH)₂), found in igneous, metamorphic environments but also in biogenic deposits, for example, in bone deposits, carbonate-hydroxy-apatites (Ca₁₀(PO₄,CO₃)₆(OH)₂), found mainly on islands and in caves, as part of bird and bat excrements, guano, Francolite

(Ca_{10-x-y}Na_xMg_y(PO₄)_{6-z}(CO₃)_zF_{0.4z}F₂). This complex, carbonate-substituted apatite is found mainly in marine environments, and, to a much smaller extent, also in weathering environments, for instance over carbonatites [3].

The structural properties of the natural and physical- and thermal treated-phosphate rock minerals can be characterized using X-ray diffraction (XRD), X-ray fluorescence (XRF), scanning electron microscopy (SEM), infrared methods, microprobe analysis, and chemical analyses [4,5]. For example, X-ray diffractometry (XRD), scanning electron microscopy (SEM), mercury porosimetry as well as BET surface area techniques were employed to enlighten the changes in physical structure and chemical composition of phosphate rock obtained from the phosphate deposits in Mazıdađı region (Mardin, Turkey) as a result of calcinations [6]. However, radiological characterization of phosphate rocks and phosphate fertilizers manufactured from these phosphate rocks, which may contain radionuclides, heavy metals and fluorine, was carried by alpha and gamma ray spectrometers [7,8].

The knowledge of the concentrations of heavy metals and distributions of these radionuclides in these materials provides useful information in the monitoring of environmental contamination by natural radioactivity.

In the present work, the radiological, structural and chemical characteristics of Mardin-Mazıdađı phosphate rock (SE Anatolia, Turkey), which is an important phosphate fertilizer source in Turkey, were investigated and compared to those of several different phosphate rocks (Tunisia, Egypt, Morocco, Algeria, Syria) using

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Table 1
Information related to collected samples.

Country	Location	Number of sample
Tunisia	Gafsa	4
Egypt	East Sabaiya	5
Morocco	Youssoufia	5
Syria	Hims-Palmyra (Alsharqia)	4
Algeria	Djabel Onk	4
Turkey	Mazıdađı	8
	Şemikan-1	
	Şemikan-2	
	Arisu-1	
	Arisu-2	

gamma spectrometry, X-ray diffraction (XRD) and X-ray fluorescence (XRF) measurement techniques.

2. Materials and methods

2.1. Sampling

Various phosphate samples in different amounts were collected randomly from each station and measurements were taken after the samples were mixed as homogenous group. Some information related to the sampling points is given in Table 1. The samples were crushed thoroughly dried at room temperature to constant weight and later crushed to pass through a 2 mm mesh sieve to homogenize them. The samples were then dried using ovens at 105 °C for 24 h to ensure that moisture is completely removed. The dry samples (170 ± 2 g) were sealed in gas tight, radon impermeable, cylindrical polyethylene plastic containers (5.5 cm diameter and 5 cm height). These samples were then left for 30 days to allow ²²⁶Ra and its short-lived progeny to reach radioactive equilibrium.

Table 2
Elemental analysis results of phosphate samples in some countries.

Compound	Unit	Tunisia	Egypt	Morocco	Algeria	Syria	Turkey
Na ₂ O	g/kg	BDL	BDL	BDL	19	BDL	BDL
MgO	g/kg	BDL	BDL	BDL	20	BDL	BDL
Al ₂ O ₃	g/kg	25	BDL	31	BDL	BDL	32
SiO ₂	g/kg	24	138	38	16	57	31
P ₂ O ₅	g/kg	261	306	300	267	324	335
SO ₃	g/kg	27	14	8	23	7	6
K ₂ O	mg/kg	1039	724	1086	1129	BDL	748
CaO	g/kg	395	393	440	400	414	427
TiO ₂	mg/kg	300	582	444	245	BDL	312
V ₂ O ₅	mg/kg	BDL	297	425	BDL	96	236
Cr ₂ O ₃	mg/kg	284	158	398	286	135	315
MnO	mg/kg	BDL	851	2	BDL	1	22
Fe ₂ O ₃	g/kg	3	24	3	4	1	3
CuO	mg/kg	16	17	43	14	14	82
ZnO	mg/kg	410	340	383	213	203	689
As ₂ O ₃	mg/kg	4	17	21	7	1	19
SeO ₂	mg/kg	14	1	BDL	59	BDL	BDL
Br	mg/kg	12	BDL	BDL	9	BDL	BDL
SrO	g/kg	3	2	1	3	2	2
Y ₂ O ₃	mg/kg	211	102	370	333	71	57
CdO	mg/kg	100	BDL	BDL	BDL	BDL	105
I	mg/kg	BDL	BDL	101	BDL	BDL	122
BaO	mg/kg	BDL	1441	227	BDL	267	289
La ₂ O ₃	mg/kg	306	154	333	488	107	BDL
CeO ₂	mg/kg	383	175	139	664	BDL	BDL
Pr ₂ O ₃	mg/kg	BDL	BDL	BDL	96	BDL	BDL
Nd ₂ O ₃	mg/kg	173	66	153	308	BDL	BDL
PbO	mg/kg	2	5	BDL	1	5	2
U ^a	mg/kg	20	57	83	36	31	33
Th ^a	mg/kg	7	4	1	3	12	2

BDL: below detection limit.

^a Calculated values.

2.2. Structural and chemical analysis

The X-ray diffraction (XRD) analysis of phosphate samples was carried out using a Rigaku D/Max-IIIC X-ray diffractometer with CuK α radiation over the range 10–70° with a scan speed 0.2° min⁻¹ at room temperature.

EDXRF spectrometer (Epsilon5, PANalytical, Almelo, the Netherlands) was used for chemical analysis of phosphate samples. Before doing any other handling, the powder was dried for 4 h at 105 °C to evaporate any remaining water. The pellets were pressed with a hydraulic press applying a pressure of 7 tones during 20 s. The resulting pellets had a diameter of 40 mm and a uniform mass of 500 ± 3 mg.

2.3. Radioactivity measurements

About 170 g of phosphate samples were sealed in gas tight, radon impermeable, cylindrical polyethylene plastic containers (5.5 cm diameter and 5 cm height) for gamma activity analysis. Gamma spectrometry measurements were conducted with a coaxial HpGe detector of 15% relative efficiency and a resolution of 1.9 keV at the 1332 keV gamma of ⁶⁰Co (Canberra, GC 1519 model). The detector was shielded in a 10 cm thick lead well internally lined with 2 mm Cu foils. The detector output was connected to a spectroscopy amplifier (Canberra, Model 2025). The energy calibration and absolute efficiency calibration of the spectrometer were carried out using calibration sources which contained ¹³³Ba, ⁵⁷Co, ²²Na, ¹³⁷Cs, ⁵⁴Mn, and ⁶⁰Co peaks for the energy range between 80 and 1400 keV (calibration sources supplied by the Isotope Products Laboratories) [9]. The energy and absolute efficiency calibration of the spectrometer were described in detail in earlier publications [10–12]. The gamma-energy lines of 351.9 keV (²¹⁴Pb) and 609.3 keV (²¹⁴Bi) were used to represent the ²²⁶Ra series, whereas 911.1 keV (²²⁸Ac) and 583.1 keV (²⁰⁸Tl) were used

Table 3

Activity concentrations, radium equivalent activities, the absorbed dose rates and annual effective doses of phosphate samples.

	^{226}Ra (Bq kg $^{-1}$)	^{232}Th (Bq kg $^{-1}$)	^{40}K (Bq kg $^{-1}$)	Ra_{eq} (Bq kg $^{-1}$)	D (nGy h $^{-1}$)	H_E (mSv y $^{-1}$)
Tunisia	250 ± 6	28 ± 2	167 ± 4	303	139	0.17
Egypt	700 ± 8	16 ± 2	186 ± 4	737	341	0.42
Morocco	1029 ± 13	5 ± 1	119 ± 3	1045	483	0.59
Syria	445 ± 9	14 ± 2	117 ± 3	474	219	0.27
Algeria	384 ± 8	50 ± 4	123 ± 3	465	213	0.26
Turkey	403 ± 8	8 ± 1	173 ± 4	428	198	0.24

to represent the ^{232}Th series. ^{40}K was investigated by gamma lines 1460.8 keV.

The activity concentrations for the natural radionuclides in the measured samples were computed using the following relation:

$$C = \frac{N}{\varepsilon PMt} \text{ (Bq kg}^{-1}\text{)} \quad (1)$$

where C is the activity concentration of a radionuclide, N is the total net count of a specific gamma emission, ε is the detector efficiency for the specific gamma emission, P is the absolute transition probability for that gamma emission, M is the mass of the sample (kg) and t is the counting time.

3. Results and discussion

Elemental analysis results of phosphate samples are presented in Table 2. They are mainly composed of CaO, P₂O₅, SiO₂, Al₂O₃, SO₃ and Fe₂O₃. Also, some oxides and elements are presented in the order of mg/kg. As the U and Th elemental concentrations (mg/kg) are below the detection limit of the system used, the activity concentrations of ^{226}Ra and ^{232}Th (Bq/kg) are measured using gamma spectrometry. Then, the elemental concentrations of U and Th are calculated using ^{226}Ra and ^{232}Th activity concentrations, respectively [13]. Computed values are summarized in Table 2 in units of mg/kg. Representative XRD results of phosphate samples for Algeria, Egypt and Turkey are shown in Fig. 1. The main peaks in all the samples are due to Fluorapatite (Ca₅(PO₄)₃F). The intensity of the peak from Fluorapatite is highest near 33° in all the samples (ranged 300–225 counts). XRD and chemical analysis show that the results of analysis are compatible with each other.

The specific activities of the natural radionuclides of the ^{226}Ra , ^{232}Th and ^{40}K are determined using gamma ray spectroscopy in the phosphate samples collected from different countries. The measured activity concentration values, together with the statistical uncertainty (1σ) and standard deviation (SD), of the above natural radionuclides are presented for the different regions in Table 3. The activity levels for ^{226}Ra , ^{232}Th and ^{40}K in all phosphate samples range from 250 to 1029 Bq kg $^{-1}$ with a mean of 535 Bq kg $^{-1}$, from 5 to 50 Bq kg $^{-1}$ with a mean of 20 Bq kg $^{-1}$ and from 117 to 186 Bq kg $^{-1}$ with a mean of 148 Bq kg $^{-1}$, respectively. Some experimental values presented for the other countries are presented for comparison in Table 4. The results show that the ^{226}Ra activity concentration values of the present study are relatively lower than those of the literature while ^{40}K activity concentration values are relatively higher. The results of the ^{232}Th activity concentrations from this study are lower than those of the literature. Only the results from Tunisia show a good agreement with our data. The mean value of activity concentration of ^{232}Th from phosphate samples of Egypt is 93 Bq kg $^{-1}$ and is higher than our result.

Distribution of ^{226}Ra , ^{232}Th and ^{40}K is not uniform in phosphate samples. In order to determine the specific concentrations of ^{226}Ra , ^{232}Th and ^{40}K in a single quantity, which takes into account the radiation hazards with them, a common radiological index has been introduced. This index is called radium equivalent activity (Ra_{eq})

and can be written as follows [14]:

$$\text{Ra}_{\text{eq}} = A_{\text{Ra}} + 1.43A_{\text{Th}} + 0.077A_{\text{K}} \quad (2)$$

where A_{Ra} , A_{Th} and A_{K} are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in Bq kg $^{-1}$, respectively. It has been assumed that 370 Bq kg $^{-1}$ of ^{226}Ra , 259 Bq kg $^{-1}$ of ^{232}Th and 4810 Bq kg $^{-1}$ of ^{40}K produce the same gamma doses in Eq. (2). The maximum value of Ra_{eq} must be less than the limit value of 370 Bq kg $^{-1}$ recommended by Organization for Economic Cooperation and Development (OECD) [15]. The calculated values of the radium equivalent (Ra_{eq}) activity for the phosphate samples are presented in Table 3. The maximum radium equivalent (Ra_{eq}) activity is calculated for the samples of phosphate from the Morocco

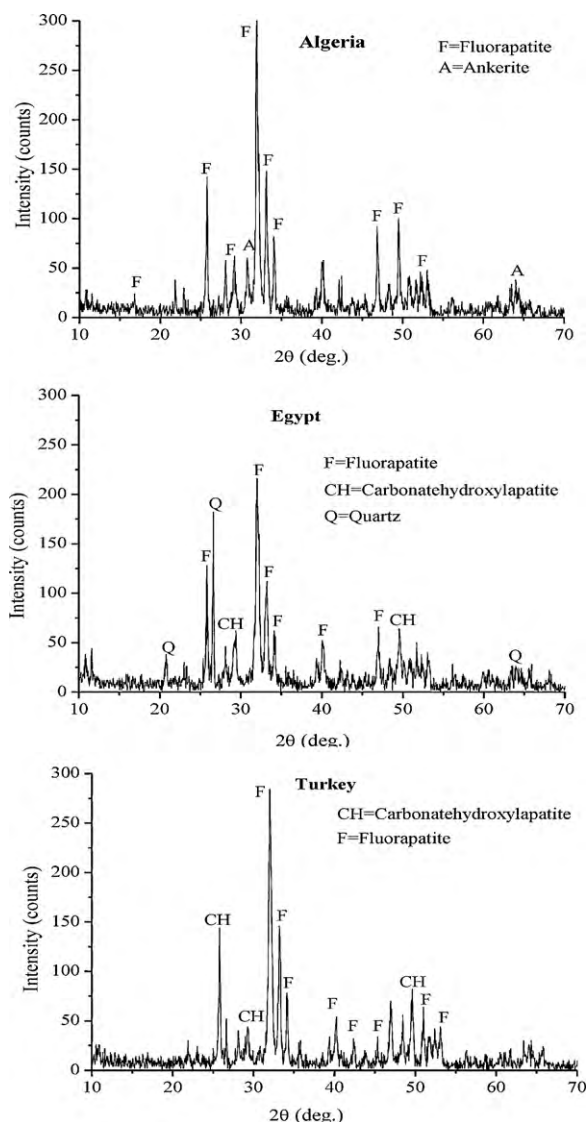


Fig. 1. X-ray diffraction diagram of the phosphate samples.

Table 4
Activity concentration of ^{226}Ra , ^{232}Th and ^{40}K (in Bq kg^{-1}) in phosphate samples from different countries.

Country	^{226}Ra	^{232}Th	^{40}K	Reference
Morocco	1600	20	10	[18]
USA	1600	20	N.F.	[18]
Jordan	1044	2	8	[19]
Tunisia	821	29	32	[19]
Algeria	619	64	22	[19]
Sudan	2263	2.5	52	[20]
Tanzania	5022	717	286	[21]
Egypt (El-Sibaiya)	538	25	N.F.	[22]
Egypt (El-Mahamid)	567	217	217	[23]
Egypt (Abu-Zaabal)	514	37	19	[24]
Pakistan	440	50	207	[25]
Tunisia	250	28	167	Present study
Egypt	700	16	186	Present study
Morocco	1029	5	119	Present study
Syria	445	14	117	Present study
Algeria	384	50	123	Present study
Turkey	403	8	173	Present study

N.F.: denotes to data not found in this reference.

(1045 Bq kg^{-1}), whereas the minimum value is obtained for the samples of phosphate from the Tunisia (303 Bq kg^{-1}). All the R_{eq} values of the samples except for Tunisia are higher than the recommended reference value 370 Bq kg^{-1} recommended by the OECD [15].

The total air-absorbed dose rate D (nGy h^{-1}) due to the presence of natural radionuclides in the phosphate samples is estimated using the following formula [16]:

$$D = 0.0417A_{\text{K}} + 0.462A_{\text{Ra}} + 0.604A_{\text{Th}} \quad (3)$$

where A_{K} , A_{Ra} and A_{Th} are the activity concentrations of K, Ra and Th (Bq kg^{-1}), respectively. The equation is derived for calculating the absorbed dose rate in air at a height of 1.0 m above the ground from measured radionuclides concentrations in phosphate samples. The absorbed dose rates which are calculated from the measured activities in phosphate samples are given in Table 3. These values range from 139 to 483 nGy h^{-1} , with the mean of 266 nGy h^{-1} .

To estimate the annual effective dose (H_{E}), the conversion coefficient (0.7 Sv Gy^{-1}) from the absorbed dose in air to effective dose received by adults, and the outdoor occupancy factor (0.2) proposed by UNSCEAR [16] are used. Therefore, the effective dose rate outdoors, H_{E} , in units of mSv per year , was estimated by the following formula:

$$H_{\text{E}} = DTF \quad (4)$$

where D is the calculated dose rate (in nGy h^{-1}), T is the outdoor occupancy time ($0.2 \times 24 \text{ h} \times 365.25 \text{ d} = 1752 \text{ h y}^{-1}$), and F is the conversion factor ($0.7 \times 10^{-6} \text{ Sv Gy}^{-1}$). In Table 3, the results obtained for annual effective dose in phosphate samples are presented. The computed values of annual effective dose in phosphate samples are found to vary from 0.17 to 0.59 mSv y^{-1} with a mean of 0.33 mSv y^{-1} . It is observed that these values are lower than the recommended limit of 1 mSv y^{-1} by the International Commission on Radiological Protection [17].

Table 5
Pearson correlation coefficients matrix.

	Tunisia	Egypt	Morocco	Algeria	Syria	Turkey
Tunisia	1.00					
Egypt	0.35	1.00				
Morocco	0.79	0.55	1.00			
Algeria	0.94	0.30	0.76	1.00		
Syria	0.18	0.57	0.60	0.16	1.00	
Turkey	0.75	0.57	0.81	0.56	0.52	1.00

Pearson correlation matrix given in Table 5 was created using all the data in Tables 2 and 3. One should understand from the correlation coefficients that in the case of positive correlation coefficients, the elemental and radioactivity concentration values increases or decreases for the corresponding countries while in the case of negative correlation coefficient, the values are inversely proportional. Significant correlations coefficients ($p < 0.05$) are presented in bold. As clearly seen from the table, the most noticeable positive correlations are found between the collected from Tunisia–Morocco ($r = 0.79$), Tunisia–Algeria ($r = 0.94$), Tunisia–Turkey ($r = 0.75$), Morocco–Algeria ($r = 0.76$) and Morocco–Turkey ($r = 0.81$).

4. Conclusion

The specific activities of the natural radionuclides of the ^{226}Ra , ^{232}Th and ^{40}K are measured by gamma ray spectroscopy in the phosphate samples, collected from the different phosphate rocks of Turkey (Mazi Dağı), Tunisia, Egypt, Morocco, Algeria and Syria. The results may be useful in the assessment of the exposures and the radiation doses due to naturally radioactive element contents in the phosphate samples. The mean activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in the phosphate samples are 535, 20 and 148 Bq kg^{-1} , respectively. Obtained values show that the average radium equivalent activity (R_{eq}), air-absorbed dose rate (D) and annual effective dose (H_{E}) for all samples are 575 Bq kg^{-1} , 266 nGy h^{-1} and 0.33 mSv y^{-1} , respectively. These values fall generally in ranges as reported for the world and individual countries in scientific literature. Also, structural and chemical characterizations are investigated in the phosphate samples. The results of the present study would be valuable database for future estimations of the impact of radioactive pollution.

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