

Assessment of the natural radioactivity and its radiological hazards in some Egyptian rock phosphates

A Abbady

Physics Department, Faculty of Science, South Valley University Qena, Egypt

Received 30 August 2004; revised 11 January 2005; accepted 13 May 2005

Radioactivity of terrestrial radionuclides ^{226}Ra , ^{232}Th and ^{40}K in phosphate rock samples has been measured using gamma ray spectrometry system with an HPGe detector. The samples showed fairly high mean concentrations, 410 ± 39 Bq.kg^{-1} of ^{226}Ra , 7.9 ± 0.95 of ^{232}Th and 37.6 ± 4 of ^{40}K . Mean absorbed gamma dose rate of 181.9 ± 20 nGy.h^{-1} was measured in air. Assessment of radiological hazards was made by calculating radium equivalent activities with external and internal hazard indices. The calculations showed radium equivalent activity ranging from 317 to 520 Bq.kg^{-1} . External and internal hazard indices ranging from 0.4 to 0.7 and 0.1 to 0.4, respectively. The results of the analysis have been found to be in a good agreement with the data obtained by others.

Keywords: Radioactivity, Radiological hazard, Phosphate, Radionuclides

IPC Code: G21F9/00

1 Introduction

Gamma radiation from radionuclides which is characterized by long half-lives comparable to the age of the earth, such as ^{40}K and the radionuclides from the ^{238}U and ^{232}Th , and their decay products, represent the main sources of external irradiation to the human body. The absorbed dose rate in air, have been carried out in the last few decades in many countries of the world. The average values range from 18 to 93 nGy h^{-1} . A typical range of variability for measured absorbed rates in air is from 10 to 200 nGy h^{-1} (Ref. 1). External exposures from outdoors arise from terrestrial radionuclides occurring at trace levels in all ground formations. Therefore, the natural environmental radiation mainly depends on geological and geographical conditions². Higher radiation levels are associated with igneous rocks, such as granite, whereas lower levels with sedimentary rocks. However, there are exceptions, as some shales and phosphate rocks have relatively high content of radionuclides¹.

The distribution of naturally-occurring uranium, radon, and other radioactive elements, depends on the type of rocks from which they originate and the processes which concentrate them. Rock phosphate contains high level of uranium-235 and radium-226. Up to 1974, the US-phosphate industry has mined about 37% more U_3O_8 than the US-uranium mining

industry³. In the production of fertilizer, recycling of by product phosphogypsum as a building material causes lung exposure to public member about 15 times higher than normal levels⁴. Personnel engage in handling, packing and transport of fertilizer do receive additional external gamma exposure at dose rates up to 0.8 $\mu\text{Gy.h}^{-1}$ (Ref.5,6).

Phosphate ores contain significant amounts of natural radioactive elements, specially ^{238}U and descendants, in concentrations depending on its geographical and geological origin, up to 300 ppm for uranium. Sedimentary and igneous phosphate ores are used as raw material for the production of phosphoric acid and consequently fertilizers. Large-scale production of phosphoric acid results in the redistribution of huge amounts of natural radioactivity.

The Egyptian phosphates are widely distributed in many localities on the Red Sea coast, Nile Valley and Western Desert. The deposits in the first two districts are relatively rich in phosphate and exploited at several mines, but those of the western Desert are of low grade except at Abu-Tartur mine. In the present study, the results from gamma radiation measurements in samples of a variety of natural phosphate rocks extracted from Eastern Desert (G.Anz, G.Duwi), Western Desert (G.Abu Tartur) and Nile Valley (G.Saria, G. Abu Saboun, G.Owina) in Upper Egypt, are presented.

2 Experimental Details

Environmental radiation is measured to identify the radioactive sources in the environment, quantify their concentration and estimate their dose contributions. This is most readily done by spectrometric technique, however simpler measurements can often yield relevant information. In both the cases, the essential part of the system is the methodology associated with detector's calibration, measurement procedure, and analysis and interpretation of data.

In the present paper, a high-resolution gamma spectrometric system is used for the measurement of the energy spectrum of the emitted gamma ray in the energy range 90-3000 keV. The system consists of a high purity germanium (HPGe) detector coupled to a signal-processing units including a spectroscopy pulse amplifier and an advanced multi-channel analyzer. The HPGe detector is *p*-type with the following specifications:

- Resolution (FWHM) at 122 keV, ^{57}Co is 1100 eV and at 1.33 MeV, ^{60}Co is 2.00 keV.
- Relative efficiency at 1.33 MeV, ^{60}Co is 20%
- Operation bias voltage is + 2000 V *dc*.

The detector is shielded in a chamber of four layers starting with Plexiglas (10 mm thick), copper (30 mm thick), lead (100 mm thick) and finally cadmium (3 mm thick).

The gamma spectra from standard source of known gamma emission rate, usually with few gamma lines, were recorded as a function of two parameters: efficiency-energy and efficiency-sample volume. As the background radiations, interface with the measurement of minute quantities of radionuclides, data reduction procedures was used with analytical system which involves, a correction for background in the standard source, to appropriate powdered samples. The recorded gamma spectrum was analysed by Maestro-programme developed by (ORTEC).

The activity concentration of radium-226 was calculated from the counts recorded by gamma detector in the energy window corresponding to bismuth-214. This technique assumes that uranium and its decay products are in secular equilibrium. Identical technique was used to estimate K-40, and equivalent thorium-232 (^{212}Pb , ^{228}Ac). The activity concentrations of the radionuclide in the samples were given by⁷:

$$A_i = \{C(E_i) - B(E_i)\} / (mftP(E_i)) \quad \dots (1)$$

where, A_i is the activity concentration of radionuclide $C(E_i)$ the net-counts above continuum at the characteristic energy E_i , $B(E_i)$ the background counts at E_i , m the sample mass in kg, f , the branching ratio of the gamma-emission at the energy considered and $P(E_i)$ is the absolute efficiency at energy E_i .

2.1 Calculation of radiological effects

Exposure rates at 1 m above ground level for distributed sources of gamma emitters in soil were extensively calculated during the last forty years by many researchers. Radiation hazards due to the specified radionuclides of Ra, Th and K were assessed by two different indices. The most widely used radiation hazard index Ra_{eq} is called the radium equivalent activity^{8,9}. The radium equivalent activity is the weighted sum of activities of the above three radionuclides based on the estimation that 370 Bq.kg⁻¹ of Ra, 259 Bq/kg of Th and 4810 Bq.kg⁻¹ of K produce the same γ -ray dose rates. Ra_{eq} activity is given by:

$$Ra_{eq} = A_{Ra} + (A_{Th} \times 1.43) + (A_K \times 0.077) \quad \dots (2)$$

Another radiation hazard index called-representative level index - $I\gamma_r$, is defined as follows^{10,11}.

$$I\gamma_r = A_{Ra}/150 + A_{Th}/100 + A_K/1500 \quad \dots (3)$$

where A_{Ra} , A_{Th} and A_K are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K , respectively in Bq.kg⁻¹.

The external hazard index H_{ex} considers only the external exposure risk due to γ -rays and corresponds to a maximum Ra_{eq} activity of 370 Bq kg⁻¹ for the material. The internal exposure to radon and its daughter products is quantified by the internal hazard index H_{in} .

The total air absorbed dose rate (nGy.h⁻¹) due to the mean specific activity concentrations of ^{238}U , ^{232}Th and ^{40}K (Bq.kg⁻¹) was calculated using the following formula^{12,13}:

$$D = 0.427 S_U + 0.662 S_{Th} + 0.0432 S_K \quad \dots (4)$$

where S_U , S_{Th} and S_K are the mean activity concentrations of ^{238}U , ^{232}Th and ^{40}K in rock samples.

3 Results and Discussion

Although the radium contents in phosphate are quite small, but it still pose radiation hazard as the gypsum in the form of a fine powder may result in dust blowing when stockpiled in the exterior. Radium

is the most dangerous decay product of uranium due to gaseous nature of its daughter radon, whose half-life is 3.8 days. During inhalation of gypsum dust, the alpha particles emanating from radon induce radiation damages to lungs.

The specific activity in (Bq.kg⁻¹) measured in area of Eastern desert, Nile valley and Western desert for ²²⁶Ra was 440±45, 387±39 and 420±41. Identical values for ²³²Th and ⁴⁰K 1.75±0.6, 15.4±3, 12.8±2.7 and 10.6±1.0, 62±7, 81±9, respectively. These results indicate an increase in radium concentration, which is attributed to increase in concentration of natural uranium in samples under test in contrast to ²³²Th concentration showing an appreciable decrease. The increase of the natural uranium concentration can be attributed to leaching effects. Actual radionuclide concentrations will vary in location because of varying geological characteristics of phosphate ores in different regions as well as variations in processes used for phosphate mining and production.

The uranium concentration in the sampling region is more dependent upon the climatic effects, seasonal variability and the effects of evapotranspiration and the concentration of suitable complexing agents, which can increase the solubility of uranium. Increased rainfall may result in more effective leaching and/or transport of uranium and lead to an increase in uranium concentration.

Thorium is also usually more abundant in the suspended load than in the bottom sediments. This is interesting as the chemical behaviour of thorium in natural waters is quite different from that of uranium. Thorium is essentially insoluble in normal surface waters, and so thorium transport is within particulate matter rather than in solutions.

As a result, there exists no correlation among potassium, thorium and uranium, in phosphate

samples because potassium is a major element in rock-forming minerals occurring mainly in aluminosilicates such as feldspars and micas.

The use of fertilizer for growing crops gives sufficient grounds for the justification. The ALARA-principle implies that reasonable measures must be taken to reduce radiation exposure.

It is important to assess the radiation hazards to personnel associated with phosphate mining. This is done by calculating different radiation hazard indices. The concentration and distribution of ²²⁶Ra, ²³²Th and ⁴⁰K in phosphate and phosphatic rocks are not uniform throughout the world. Uniformity in respect of exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq.kg⁻¹ to compare the specific activity of materials containing different amounts of ²²⁶Ra, ²³²Th and ⁴⁰K.

It is desirable to predict the expected inhalation doses directly by reason of the radionuclide concentrations in mines, due to high contribution of short lived ²²²Rn and ²²⁰Rn daughter radionuclides to natural radiation exposure. It is easy to determine the activity concentrations of radium and thorium by gamma spectroscopic methods in comparison with the more complicated methods, such as the measurement of the actual exhalation rates. The radium equivalent activity, the calculated radiation dose and hazard indices in phosphate samples are listed in Table 1. The values of this index (*I*_γ) must be less than unity for the radiation hazard to be negligible i.e. for the maximum value of *H*_{ex}, *H*_{in} to be less than unity, the maximum value of *Ra*_{eq} must be less than 370 Bq kg⁻¹.

From the calculated values, it can be seen that the phosphate samples have level index above the proposed acceptable level of 1 (370 Bq kg⁻¹) in all areas under study. The total absorbed doses (nGy.h⁻¹)

Table 1—Average radium equivalent, radiation hazard indices and dose rates in phosphate rock samples

Region	<i>Ra</i> _{eq} (Bq.kg ⁻¹)	Dose rate (nGy.h ⁻¹)	<i>H</i> _{in}	<i>H</i> _{ex}	<i>I</i> _γ
Eastern desert					
G. Anz	476.3±22	203.6±14	0.10±0.03	0.6±0.07	3.2±1.8
G. Duwi	410.6±20	175.6±13	0.10±0.03	0.6±0.07	2.7±1.6
Nile valley					
G. Saria	520.5±23	223.1±15	0.2±0.04	0.7±0.08	3.5±1.9
G. Owina	317.6±18	136.8±12	0.3±0.05	0.4±0.06	2.1±1.4
G. Abu-Saboun	378.7±19	162.6±13	0.2±0.04	0.5±0.07	2.5±1.6
Western desert					
G. Abu-Tartur	443.3±21	190.7±14	0.4±0.06	0.6±0.07	3.0±1.7

are very high due to the higher radium contents in phosphate, which consequently leads to an increase in the radium equivalent activity of phosphate fertilizer product. The acceptability of risks from ionizing radiation is decreasing, this tendency leads to more stringent regulation. In the forthcoming years, lower regulatory limits are likely to become obligatory. Precautionary measures and restrictions imposed on workers in factories and farm-yards may become compulsory from view point of radiation protection.

Due to radium content, the disposal of by-product phosphogypsum is thus hazardous. Other environmental effects are only minor. All this might result in a price increase for fertilizers, entailing additional expense for users and the society in general.

The radiological indices, radium equivalent and dose rate are plotted against with sample location in Figs (1 and 2). The results from present analyses were found to be in a good agreement with the data obtained by others¹⁴.

4 Conclusion

The conclusion drawn from the present study is:

- 1 The gamma spectrometric analysis indicate that phosphate samples contain ^{226}Ra , ^{40}K and small amount of ^{232}Th in some samples, while ^{137}Cs could not be detected. The results indicate that ^{226}Ra exhibits highest value of activity concentration as compared with ^{232}Th and ^{40}K .

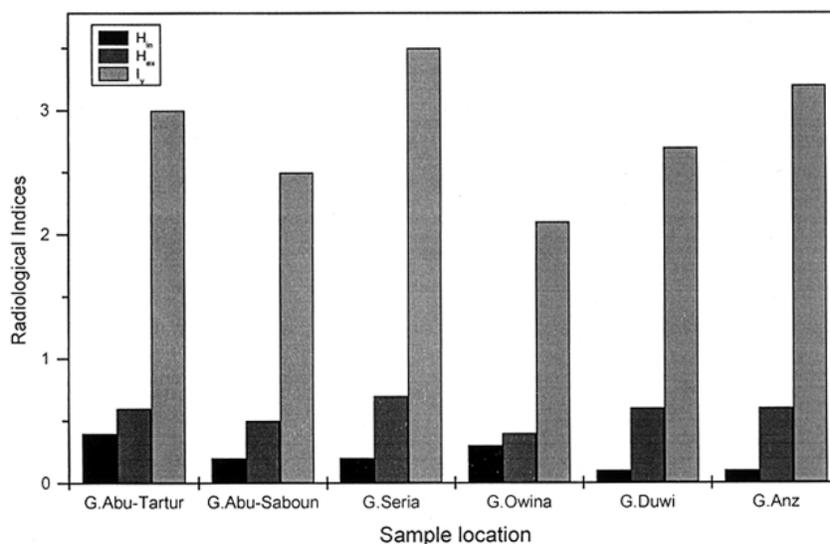


Fig. 1—Radiological indices for phosphate samples from different locations

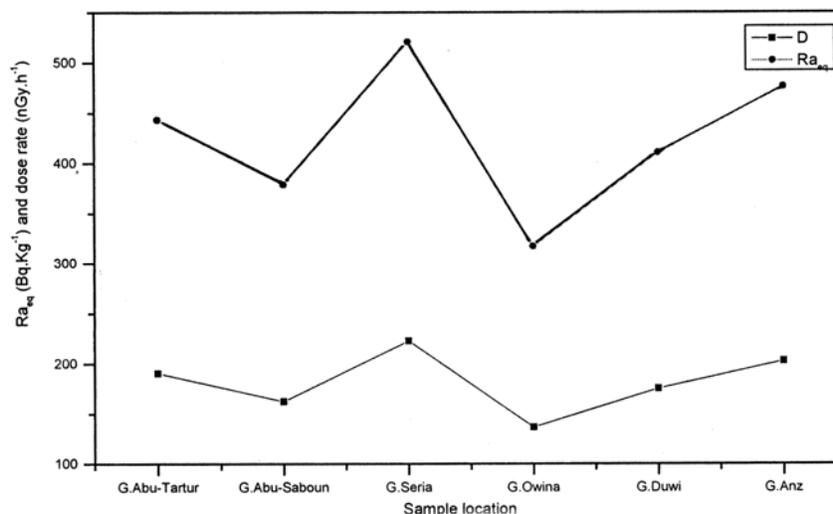


Fig. 2—Average radium equivalent and dose rate for phosphate samples from different locations

- 2 ²²⁶Ra, ²³²Th and ⁴⁰K activity concentrations were in the range between 28.1-1873, 1.5-33.2 and 9.9-258.4 Bq.kg⁻¹, respectively. The lowest value of activity is found in the case of ²³²Th. At the same time the fluctuation in the results of ⁴⁰K is much more than the corresponding values of ²²⁶Ra, and ²³²Th. The activity concentration of the isotopes shows a wide range of values, because of the different location samples.
- 3 From the calculated values, it is found that the phosphate samples have radioactivity above the proposed acceptable level of 1 (370 Bq.kg⁻¹) in all area under investigations.

References

- 1 UNSCEAR *United Nations Scientific Committee on the Effects of Atomic Radiation Sources and Effects of Ionizing Radiation*, New York: United Nations Publication (2000).
- 2 Florou H & Kritidis P, *Radiation Protection Dosimetry*, 45 (1/4) (1992) 277.
- 3 Guimond R J & Windham S T, *USA-EPA, Office of Rad Progr, Techn Note ORP/CSD-75-3* (1975).
- 4 Steinhäusler F & Pohl E, On the “*Biological Effects of low-level Radiation with Special Regard to the Stochastic and non-Stochastic Effects*” paper presented in international Symposium Venice (1983).
- 5 Ahmed N K & El-Arab A M, *Arab J Nucl Sci Appl*, 34 (2001) 245.
- 6 El-Taher A, Mahmoud H M & Abbady G E, *Indian J Pure & Appl Phys*, 42 (2004) 243.
- 7 Chung-Keung Man, Lau S Y, Au S C & Ng W K, *Health Phys*, 57 (1989) 397.
- 8 Krieger R, *Betonwerk Fertigteil technol*, 47 (1981) 468.
- 9 Beretka I & Mathew P I, *Health Phys*, 48 (1985) 87.
- 10 *NEA-OECD, Report by NEA Group of Experts, OECD*, Paris (1979).
- 11 Alam M N, Chowdhury M I, Kamal M, Ghose S & Ismal M N, *J Environ Radioactivity*, (1999).
- 12 UNSCEAR, *Sources effects and risks of ionizing radiation* New York: United Nations Annex A, B (1988).
- 13 Beck H L, Decompo J & Gologak J, New York: *Health and Safety Laboratory AEC; Report HASL*, 258 (1972).
- 14 El-Arabi A M & Yousef A M M, *Nuclear Science and Technique*, 13 (2002).