Journal of Environmental Radioactivity 102 (2011) 336–339

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

The radioactivity measurements in soils and fertilizers using gamma spectrometry technique

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ARTICLE INFO

Article history:
Received 24 July 2010
Received in revised form
12 January 2011
Accepted 25 January 2011
Available online 22 February 2011

Keywords:
HPGe detector
Environmental radioactivity
Gamma spectrometry
Phosphate fertilizers

ABSTRACT

Because of their mineral content, soils are naturally radioactive and one of the sources of radioactivity other than those of natural origin is mainly due to the extensive use of fertilizers. The main aim of this paper is to evaluate the fluxes of natural radionuclides in local production of phosphate fertilizers to determine the content of radioactivity in several commercial fertilizers produced in Algeria and to estimate their radiological impact in a cultivated soil even for the long-term exposure due to their application.

For these purposes, virgin and fertilized soils were collected from outlying Setif region in Algeria and from phosphate fertilizers used in this area.

Gamma spectrometry was exploited to determine activity concentration due to naturally occurring ^{226}Ra, ^{232}Th and ^{40}K in five types of samples (two different sorts of fertilizers, virgin and fertilized soils and well water used for irrigation) taken from Setif's areas.

The results show that these radionuclides were present in an average concentration of 134.7 ± 24.1, 131.8 ± 16.7, 11644 ± 550 Bq/kg for the first fertilizer NPK and 190.3 ± 30, 117.2 ± 10.3, 5312 ± 249 Bq/kg for the second fertilizer (NPKs). For the virgin and the fertilized soils, the corresponding values were respectively 47.01 ± 7.3, 33 ± 7, 329.4 ± 19.7 Bq/kg and 53.2 ± 10.6, 50.0 ± 7, 311.4 ± 18.7 Bq/kg. For well water, the values were 1.93 and 0.12 Bq/kg; however the third value was below the Minimum Detectable Activity (MDA).

The radium equivalent activity (Raeq) and the representative level index $I_{\gamma r}$ for all samples were also calculated. The data were discussed and compared with those given in the literature.

1. Introduction

The natural radioactivity in soil may vary considerably from one type to another. In soils, one of the sources of radioactivity other than those of natural origin is mainly due to extensive use of fertilizers which is rich in phosphates used for agricultural purposes (Abbady et al., 2008). Phosphate ores, especially sedimentary ores, can be significantly enriched with naturally occurring radionuclides, uranium (^{238}U) and the daughter radionuclides coming from the radioactive decay of ^{238}U (Hamdy et al., 2007). There is a direct relationship between uranium and P_2O_5 content of fertilizers (Akhtar et al., 2005a,b), and then several studies have noted that the concentration of uranium follows the concentration of P_2O_5 in various fertilizers.

The ^{232}Th series has only a minor contribution to the radioactivity in phosphate compared with the uranium ones, in addition, soils and phosphate fertilizers contain the naturally occurring ^{40}K (Ahmed and El-Arabi, 2005).

In order to reach high agricultural productivity, the present practice of replacing nutrients in soils and consequently supplying substances is done by the application of chemical fertilizers, mostly compounds commercially named NPK (nitrogen (N), phosphorus (P) and potassium (K)) and NPKs (sulfate based fertilizer).

Their formulas vary widely and the concentrations are chosen according to the need of each soil and cultivation (Becegato et al., 2008). The phosphate rock is used as raw material and it presents in its composition radionuclides of the uranium and thorium natural series.

It has been known since the beginning of this century that phosphate rocks contain substantial concentration of uranium, thorium, radium and their decay products (Skorovarov et al., 2000). Phosphate rock is an important raw material used for manufacturing different types of phosphates' fertilizers. Therefore, when this rock is processed into phosphates' fertilizers, most of the uranium and some of the radium come into the fertilizers. It has also been

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doi:10.1016/j.jenvrad.2011.01.006

estimated earlier that phosphates' fertilizers applied to the fields in recommended amounts could raise radioactivity level in soils (Akhtar et al., 2005a,b).

This study aims to assess the environmental radioactivity level of virgin and fertilized soils which were collected from outlying Setif area (Algeria) and of phosphate fertilizers used in these areas. Well water is also analyzed to estimate the contribution of the radioactivity content in water used for irrigation.

Comparison of the results obtained with those of national and world averages are presented and the studies of this sort are expected to serve as baseline data of natural radioactivity level and will be useful in assessing public doses.

2. Experimental procedures

2.1. Sampling and samples preparation

In order to measure the natural radioactivity in the phosphate fertilizers, the soil samples before and after fertilization have been taken from the region of Sétif – Algeria. In this area, there are several species of green vegetables which are planted such as potatoes, sweet pepper, tomatoes, cauliflower and of large surfaces of wheat. Most of the cultivated areas considered as the main source of food for citizens who live in Sétif. For a complete survey the natural radioactivity of irrigation water was measured as well (Baykara and Dogru, 2006).

The soil under investigation is an argillaceous soil contains 60% clay, 24% loam and 16% sand. The soil textures for all samples were very similar as they are taken from the same region. The soil is slightly alkaline (pH 8).

The field experiment was laid on 12 sites. Before fertilization, the sub-samples were collected from undisturbed areas (the distance between each site was about 2–5 km). Three sub-samples were taken from each 12 sites in which its surface is 1 m^2. The soil sub-samples were collected by a core method in which cores of 15 cm diameter (the sampling area of sub-samples was 177 cm^2) and 25 cm in depth were used to take soil samples. The collected sub-samples were about 2 kg.

Each time, one sub sample has to be taken from each site and then mixed till three composite samples were obtained to make the radioactivity study and their average as well.

The site was fertilized with phosphate fertilizers that contain trace concentrations of uranium and have used since more than six years in quantities of about 800–850 kg phosphate fertilizer per hectare.

Six years after fertilization, other samples were taken in the same way as before and mixed to get their composite samples for making the radioactivity study and their average too.

For the fertilizers, three samples have been taken from each type of fertilizer (NPK and NPKs).

After removing the stones and inorganic materials, the samples were dried in an oven at about 100 °C then crushed and sieved through 0.5 mm mesh sieve. The sieved portion of the samples was transferred into 500 ml Marinelli beakers for gamma spectrometry and sealed for four weeks to reach secular equilibrium between the thorium and radium contents of the sample and their daughter radionuclide's. The average sample weight was 500 g for phosphate fertilizers and soils and 450 ml for well water.

2.2. Experimental setup

Each sample from soil and fertilizers samples were subjected to a gamma-ray spectrometer with HPGe setup and multichannel analyzer 8192 channels. The HPGe detector is p-type with the following specifications: resolution (FWHM) at 122 keV, ^{57}Co is 0.825 keV and at 1332 keV, ^{60}Co is 1.8 keV, relative efficiency is 10%. The detector was situated in a well to shield the measuring station against background radioactivity. The samples were placed over the detector for at least 30 h. The spectra were evaluated by the computer software programme genie 2000 (CANBERRA). The natural radioactivity is calculated manually with the use of a spread sheet (Microsoft Excel).

After measurement and subtraction of the background, the activity concentrations were calculated:

a) ^{226}Ra concentration was calculated as the weighted average of the activity determined using the gamma-ray lines 295.1 (19.2%) and 351.9 (37.1%) keV gamma-rays from ^{214}Pb, 609.3 (46.1%), 1120.3 (15%) keV and 1764.5 keV (15.4%) gamma-rays from ^{214}Bi and 186.2 keV (3.59%) the specific gamma-ray of ^{226}Ra. The peak recorded around 186 keV arises from the emissions of ^{226}Ra and ^{235}U. Therefore, the count rate under the 186 keV peak was treated as the sum. The concentrations of ^{226}Ra and ^{235}U under the photopeak 186 keV were separated (Papachristodoulou et al., 2003).
b) The gamma-ray photopeaks used for the determination of the ^{232}Th contents were 338.4 (12%), 911.2 (29%), 964.6 (5.05%) and 969.0 keV (17%) of ^{228}Ac, 238.6 keV (44.6%) of ^{212}Pb and 583.2 (84.4%) and 860.6 keV (12.4%) of ^{208}Tl.
c) ^{40}K and ^{137}Cs were directly determined using 1460.8 (10.7%) and 661.7 (85.12%) keV g-rays, respectively.

The Minimum Detectable Activity (MDA) for ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs in soil were determined and are given in Table 1. The MDA in groundwater for the radionuclide ^{40}K is 7.35 Bq/l. The gamma-ray spectrum was accumulated for up to 108000 s (30 h) for each sample. In our case, inactive soil and sterilized water were used as blank and the time of collection was 194400 s (54 h). The relative efficiency curve of the detector was determined from soil samples spiked with a ^{152}Eu and ^{133}Ba solution (Lavi and Alfassi, 2005).

Table 1
The Minimum Detectable Activity (MDA) for the radionuclides ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs.

Nuclide	MDA (Bq/kg)
^{40}K	25
^{226}Ra	11.4
^{232}Th	3.6
^{137}Cs	1.4

3. Results and discussion

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K have been determined for the various samples. The average values of activity measured as well as the respective standard deviations, of the above natural radionuclides are presented in Table 2.

The use of fertilizers in large extent has affected radionuclides concentration, and especially for potassium, it is one of the causes

Table 2
Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs for all samples (fertilizers, well water and soils).

Sample name	Activity concentration (Bq/kg)			
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Virgin soil	47 ± 7	33 ± 7	329 ± 19	9.9 ± 0.5
Fertilized soil	53 ± 10	50 ± 7	311 ± 18	7.7 ± 0.4
NPK fertilizer	134 ± 24	131 ± 16	11645 ± 550	11.7 ± 0.6
NPKs fertilizer	190 ± 30	117 ± 10	5312 ± 249	7.5 ± 0.3
Well water	1.93	0.12	*MDA	*MDA