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Assessment of natural radioactivity in NPK fertilizers

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Résumé

L'ajout d'engrais inorganiques pour traiter les sols des cultures et augmenter leur production est devenue une nécessité et un moyen commun dans le monde entier. Ces différents engrais, tels que les engrais NPK (azote, phosphate et potassium), provoquent une augmentation du contenu des radionucléides dans les nutriments essentiels en les déplaçant du sol à différentes parties des plantes. Par conséquent, la concentration d'activité de radioéléments naturels de certains engrais utilisés dans le domaine de l'agriculture dans la région de Constantine a été mesurée par spectrométrie gamma à haute résolution, afin d'évaluer le risque des rayonnements et son effet sur la santé humaine.

Mots clés : **Engrais inorganiques, Engrais NPK, Concentration d'activité, Spectrométrie gamma.**

Abstract

The addition of inorganic fertilizers to treat crop soil and increase its production has become a necessity and tool around the world. These various fertilizers, such as NPK fertilizers (Nitrogen, phosphate and potassium), cause an increase in the content of radionuclides in the essential nutrients by moving them from the soil to different parts of plants. Consequently, the activity concentration of some fertilizers used in the field of agriculture in the province of Constantine was measured using high resolution gamma ray spectrometry, in order to evaluate the radiation risk and its effect on human health.

Keywords: **Inorganic fertilizers,** ***NPK Fertilizers, Activity concentration, Gamma spectrometry.***

ملخص.

إن إضافة الأسمدة غير العضوية لمعالجة تربة المحاصيل الزراعية وزيادة انتاجها أصبح أمرا ضروريا وشائعا في مختلف أنحاء العالم. هذه الأسمدة بمختلف أنواعها مثل الأسمدة NPK (الآزوت, الفوسفور و البوتاسيوم) تسبب زيادة في محتوى النويدات المشعة في العناصر الغذائية الأساسية وذلك بانتقالها من التربة إلى مختلف أجزاء النباتات. لذلك تم قياس تركيز النشاط الاشعاعي لبعض الأسمدة المستخدمة في المجال الزراعي في ولاية قسنطينة باستخدام المطيافية غاما عالية الدقة بهدف تقييم خطر الإشعاع الناتج عن النويدات المشعة، ومدى تأثيره على صحة الانسان.

الكلمات المفتاحية: الأسمدة غير العضوية، الأسمدة NPK ، تركيز النشاط الإشعاع ، المطيافية غاما

I - INTRODUCTION :

According to the source of radiation, there are two types of radiation sources, natural and artificial. The natural radiations also classify into cosmic and terrestrial radiation, all of them, cause internal and external exposure to the environments.
People are exposed to ionizing radiation from the radionuclides that are present in different types of natural sources [1], of which Nitrogen, phosphate and potassium fertilizers are one of the most important sources.
Exposure of workers and the public to radiation from phosphate rock and fertilizer is therefore not unlikely [2] Because application of fertilizers, particularly phosphates, significantly increases the level of radioactive activity of the cultivated soil compared to the soil of the arid land [3]. Many researchers have found the high value of radionuclide content in fertilizers, especially phosphates produced from phosphate fertilizers [4-5-6] rich in uranium-238 [7].
Fertilizers are chemical compounds that provide necessary chemical elements and nutrients to the plants as plant growth depends mainly on nitrogen, phosphate, and potassium. Relatively large concentrations of natural radionuclides present in phosphate fertilizers contaminate the environment and agricultural lands during cultivation [8-4].
The aim of this work was to determine the concentration of uranium (^{235}U and ^{238}U),^{226}Ra,^{232}Th and ^{40}K in tree inorganic fertilizers (NPK) used in the field of agriculture in the region of Constantine in Algeria.
The measurement were performed at the Laboratory of Low Activity (LBA) of LPSC specializes in the measurement of Low-activity radionuclides. This laboratory also carries out measures to monitoring radioactivity in the environment, in industrial processes, building materials, ground water, ...

II- DESCRIPTION OF THE LBA/LPSC LABORATORY :

The measurement had been carried out in the laboratory of low activities in Grenoble (LBA/LPSC) equipped with two low noise HPGe detectors. Each detector is surrounded by two cm of archaeological lead exempt from natural radioactivity and 15 cm of purified lead. The two detectors and their shielding is positioned at the center of a cube of two meters square of side surfaces, each face of the cube, with the exception of the ground, is in fact a liquid scintillation detector. These detectors act as veto prohibiting the acquisition of data during the passage of a cosmic ray. The InterWinner software (ITECH INSTRUMENTS) was used for both the acquisition and treatment of the data collected by detectors.

III- EXPERIMENTAL PROCEDURES :

In order to measure the natural radioactivity in NPK fertilizers collected from Constantine, the samples were dried for 48 h to ensure that the moisture is completely removed. Each sample was placed in a plastic in a box with a radius of 50 mm and a depth of 14 mm. Before making the gamma measurement, the recipients were remained hermetically sealed for a minimum of 40 days, in order to establish the radioactive secular equilibrium between the radionuclides of short half-lives [9].
The energy calibration of the detectors was done using standard multi gamma sources. The resolution of the detectors in these measures was 0.85 keV and 1.85 keV at energies 122 keV and 1332 keV, respectively.
The radioactivity sources IAEA-RGU-1 and IAEA-RGTh-1 in powder form, installed in the same plastic box used for the samples, were used to determine the detectors efficiency curves.
Each fertilizer sample was placed directly in the front of the detector and was measured during an accumulating time between 43 and 51 h. The background spectrum was measured under the same conditions of samples during 1 month and was used to correct the calculated samples activities.

IV. DETERMINATION OF ACTIVITY CONCENTRATIONS AND CALCULATION OF RADIUM EQUIVALENT ACTIVITY :

IV.1. Activity concentrations

The activity concentrations of the radionuclides in the measured samples (As) were computed using the following equation [10],

$$A_s(Bq.kg^{-1}) = \frac{N}{\varepsilon * I * m * t} \qquad (1)$$

where N is the net gamma counting, ε is the detector efficiency of the specific γ-ray, I is the absolute gamma intensity of gamma-decay, m is the masse of the sample (kg) and t is the time of measurement (s).

IV.2. Radium equivalent activity

The equivalent radium activity (Ra_{eq}) is a parameter utilized to estimate the external exposure due to the contribution of gamma emitters from the ^{226}Ra, ^{232}Th and ^{40}K . Eq. (2) was used for the calculation of the Ra_{eq}

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

where A_{Ra}, A_{Th} and A_K are, respectively, the radiological concentrations or specific activities obtained for ^{226}Ra, ^{232}Th and ^{40}K, using Bq.kg^{-1} as measurement unit [9].

The weights are based on the estimation 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 γ-ray dosage [11].

V. ESTIMATION OF ABSORBED AND EFFECTIVE DOSE :

The measured activity of ^{226}Ra, ^{232}Th and ^{40}K were converted into doses (nGy.h^{-1} Bq.kg^{-1}) by applying the factors 0.462, 0.604 and 0.0417 for radium, thorium and potassium, respectively. These factors were used to calculate the total absorbed gamma dose rate in air at 1 m above the ground level using the following equation:

$$\dot{D} = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_k \quad (3)$$

Where C_{Ra}, C_{Th} and C_K are the activities (Bq.kg^{-1}) of radium, thorium and potassium in the samples. To estimate annual effective doses, must be taken into account, (a): the conversion coefficient from absorbed dose in air to effective dose and (b): the indoor occupancy factor. The annual effective doses are determined as follows [4]:

$$\text{Indoor annual effective dose (mSv)} = (\text{Absorbed dose}) \text{ nGy.h}^{-1} \times 8760 \text{ h} \times 0.8 \times 0.7 \text{ Sv.} Gy^{-1} \times 10^{-6} \quad (4)$$

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$$\text{outdoor annual effective dose (mSv)} = (\text{Absorbed dose}) \text{ nGy.h}^{-1} \times 8760 \text{ h} \times 0.2 \times 0.7 \text{ Sv.} Gy^{-1} \times 10^{-6} \quad (5)$$

Annual estimated average effective dose equivalent received by a member is calculated using a conversion factor of 0.7 Sv.Gy^{-1} , which is used to convert the absorbed rate to annual effective dose with an outdoor occupancy of 20 % and 80 % for indoors [4].

VI. RESULTS AND DISCUSSION

In Table 1, we present the activity concentration of each radionuclide. The activity of each radioelement was determined from a dominant γ ray, and sometimes from two or three γ rays.

TABLE 1: ACTIVITY CONCENTRATIONS OF NATURAL RADIONUCLIDE IN AZOTE, PHOSPHATE AND POTASSIUM SAMPLES FERTILIZER

Element	A Nitrogen (Bq.kg^{-1})	A Potassium (Bq.kg^{-1})	A Phosphate (Bq.kg^{-1})
^{238}U	4.39±0.32	2.68±1.43	2.21±0.21
^{235}U	0.20±0.01	0.13±0.07	0.10±0.01
^{232}Th	1.32 ±0.20	0.45±0.02	0.007±0.001
^{226}Ra	4.40±0.32	2.68±1.43	0.59±0.05
^{40}K	9.41±2.25	1339.00±89.83	0.02±0.01

TABLE 2: RADIUM EQUIVALENT, ABSORBED DOSE, INDOOR ANNUAL EFFECTIVE DOSE AND OUTDOOR ANNUAL EFFECTIVE DOSE FOR DIFFERENT FERTILIZER SAMPLES

Samples of fertilizers	Ra_{eq} (Bq.kg^{-1})	$\dot{D}$ (nGy.h^{-1})	Indour (µSv/y)	Outdoor (µSv/y)
Nitrogen	7.01±0.46	4. 43 ± 0.21	21.73±1.04	5.43±0.26
Potassium	106.43±7.06	105.28 ± 3.80	516.45±18.66	129.11±4.66
phosphate	0.60±0.01	0.28 ± 0.01	1.39±0.03	0.35±0.01

The use of fertilizers in large extent has affected radionuclide concentration and especially for potassium. It is one of the causes for presence of high activity of ^{40}K in soil [12].

Table 1 show that the activity concentrations of ^{238}U, ^{235}U and ^{226}Ra in the three fertilizers are low but the concentration of ^{40}K is very important in Potassium fertilizer than the two others.

The highest activity concentration of ^{238}U (4.39±0.32) Bq.kg^{-1} was noted in Nitrogen fertilizer (Table1), whereas the lowest value of (2.21±0.21) Bq.kg^{-1}, was recorded in Phosphate fertilizer.

The mean activity concentrations of the three radionuclides ^{40}K, ^{238}U, ^{232}Th are in the order ^{232}Th < ^{238}U< ^{40}K (0.59 Bq.kg^{-1} < 3.09 Bq.kg^{-1} < 449.48 Bq.kg^{-1}).

For ^{226}Ra the concentration is ranged between (0.59±0.05) Bq.kg^{-1} and (4.40±0.32) Bq.kg^{-1}; the concentration trend of this radionuclide is the following: in nitrogen > in potassium> in phosphate.

For ^{235}U is ranged between (0.10±0.01) Bq.kg^{-1} and (0.20±0.01) Bq.kg^{-1}.The results show the low presence of the ^{235}U in the three samples.

The values of calculated Ra_{eq} for the studied samples are shown in the second column of table 2. The

calculated Ra_{eq} values are ranged from (7.01±0.46) (Nitrogen) to (106.43±7.06) (Potassium) $Bq.kg^{-1}$. All the values of Ra_{eq} in the studied samples are found to be lower than the critical limit of 370 $Bq.kg^{-1}$ [13]. The average value for Ra_{eq} obtained in the tree samples was (38.01±2.51) $Bq.kg^{-1}$which is less than the recommended value (370 $Bq.kg^{-1}$) and as such does not pose a radiological hazard to humans.

The absorbed and annual effective dose rates from the samples are calculated as shown in Table 2. The minimum and maximum values of absorbed dose, indoor annual effective dose and outdoor annual effective dose were found to vary from (0.28 ± 0.01) $nGy.h^{-1}$ to (105.28 ± 3.80) $nGy.h^{-1}$, (1.39±0.03) µSv to (516.45±18.66) µSv and (0.35±0.01) µSv to (129.11±4.66) µSv in the samples studied in the present work. The average absorbed dose rate (36.66±1.34) $nGy.h^{-1}$ is found to be lower than the world average value of 60 $nGy.h^{-1}$ [14]. This obtained value is may be due essentially to the elevated level of ^{40}K activity concentration in the study area. Given that the results obtained for the different samples are lower than the world reference values, this implies that the measured samples do not pose any much health hazard for the population.

CONCLUSION :

Based on the obtained results of the foregoing study, the following conclusions can be derived: The samples were analyzed using gamma-ray spectrometry; this technique is considered the best for determination of the activity concentration of these types of radionuclide. The activity concentrations of naturally occurring radionuclide in fertilizer samples were within the world average ranges

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