Natural Radiation of Chemical Fertilisers and Radiological Impact on Agriculture Soil

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Research

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Posted Date: October 25th, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-989747/v1>

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Natural radiation of chemical fertilisers and radiological impact on agriculture soil

Akbar Abbasi^{1,*},

Abstract

To investigate, the radiological hazards indices of chemical fertilisers commonly used in agriculture soils of Arasbaran, Iran, the activity concentration of ^{226}Ra , ^{232}Th and ^{40}K in four types of chemical fertilisers were measured by gamma spectrometry system. The concentration range for ^{226}Ra , ^{232}Th and ^{40}K were obtained from 54 ± 2.2 to 1585 ± 29.4 , 6 ± 0.1 to 157 ± 9.1 and 31 ± 1.5 to 1496 ± 32.8 Bq/kg, respectively. The obtained results showed that the average of radiation hazard parameters for the fertilisers were higher than the reference level for the gamma index(I_γ), alpha index(I_α), absorbed dose rate(D), and ambient dose equivalent rate ($H^*(10)$) values. The outcomes give rise to that more parameters values are over the permissible limit, increasing the likelihood of adverse health consequences.

Keywords: concentration, radiological hazards, chemical fertilizer, dose rate, reference level

1. Introduction

Natural radiation sources of terrestrial origin can be found in varying degrees in all media of the environment, including soil(Abbasi et al., 2020a; Ghods et al., n.d.), building materials(A. Abbasi, 2013; Abbasi, 2017; Akbar Abbasi, 2013; Abbasi and Hassanzadeh, 2017; Abbasi and Mirekhtiary, 2013), sediment(Abbasi, 2019; Abbasi et al., 2020b; Abbasi and Mirekhtiary, 2020a), water(Abbasi, 2018; Abbasi and Bashiry, 2016; Abbasi and Mirekhtiary, 2020b, 2019, 2017), air(Lopes et al., 2018), and even the human body(UNSCEAR, 2000). One of the causes of radioactivity in soils other than those of natural origin is the widespread use of phosphate-rich fertilisers for agricultural reasons(Boukhenfouf and Boucenna, 2011). There is a direct relationship between the ^{238}U series and the phosphorus pentoxide (P_2O_5) content of fertilisers(Akhtar et al., 2005).

Phosphate rock is used as a raw material for fertilisers, and it contains radionuclides from the natural ^{238}U (^{226}Ra) and ^{232}Th series and ^{40}K . Chemical fertilisers mostly compound commercially known as NPK (nitrogen (N), phosphorus (P), and potassium (K)) and NPKs, are currently used to replace nutrients in soils and thus supply substances to achieve high agricultural productivity (sulfate-based fertilizer). Their formulae differ greatly, and the concentrations are selected based on the requirements of each soil and cultivation(Becegato et al., 2008). Excessive fertilizer usage may lead to an increase in radionuclides in soil and groundwater, which are exposure sources for food crops and drinking water. Gamma radiation from natural radioactivity accounts for 85 % of total global annual average ionizing radiation(Radiation, 2001). Higher exposure to such radionuclides is hazardous to human health and may result in a variety of health issues, including lung, pancreatic liver, hepatic, bone, and kidney cancer(Abbasi et al., 2020b).

In general, fertilizer use has grown globally over the last decade, with demand for all fertilisers rising from 161 million tons in 2008 to more than 200 million tons in 2018, with nitrogen fertilisers being the most widely used(FAO, 2017, 2015; Heffer and Prud'homme, 2014). As a result, during the last ten years, the use of chemical fertilisers, particularly nitrogen, has grown, which has typically been to enhance crop production, the most significant of which was wheat and corn(Altieri and Nicholls, 2001). As a result of the increased harvesting due to the use of artificial fertilisers, environmental expenses are incurred, as well as soil and ecosystem degradation.

Recently, some researchers have been carried out and reported on the subject of natural radiation and radiological hazards from chemical fertilisers(El-Bahi et al., 2017; Faweya et al., 2018; Korany et al., 2021; Loan et al., 2018). The objectives of this study were to evaluate natural radioactivity concentration in chemical fertilisers used in the Ahar area, Iran, and calculate

radiological risk due to chemical fertilisers using for agriculture purposes.

2 Material and methods

2.1 Study Area

The area under study covers the fertile regions of district Arasbaran. Arasbaran located at 38.45138° N, 47.0676° E, is situated on the northern slopes of the Buzghush mountain. The chief river flowing in the west to east of Ahar is Aharchayi. The area is noted for its Moderate mountains. The topography of the area involves rugged terrains, with elevation ranging from 800 to 1800 m a.s.l. the study area is shown in Fig. 1.

2.2 Sample collection and preparation

Four different main types of chemical fertilisers were gathered from farmers and markets. The investigated samples types are ammonium nitrate (AN), ammonium sulfate (AS), ammonium phosphate (AP), and potassium nitrate (PN). For each type of chemical fertilizer, different brands were sampled. Finally, 21 samples were packed in a polyethene container and transferred to the laboratory for measuring. The collected samples were dried in an oven at around 75 ° C for 24 hours to ensure that all moisture was eliminated; the samples were crushed, homogenized, and sieved through a 200 µm sieve. Each sample was weighed and put in a 0.5 kg Marinelli beaker. The beakers were completely sealed for 4 weeks to achieve secular equilibrium between daughter radionuclide ^{222}Rn and parent ^{226}Ra , which occurs when the daughters' rate of decay equals that of the parent. This step is required to guarantee that ^{222}Rn gas is restricted inside the volume and that the daughters remain in the sample (A. Abbasi, 2013).

2.3 Measurement process

A gamma-ray spectrometer with a high-resolution coaxial p-type vertical HPGe detector was used to measure the radioactivity. The detector has a relative efficiency of 80% when compared to the NaI (Tl) (3×3) detector and has a resolution of 1.8 keV at full-width half-maximum FWHM of the 1332.5 keV gamma-ray photopeak from ⁶⁰Co. The system was coupled to a high count-rate Multi-Task 16k MCA card. Commercial software Gamma 2000 from Silena-Italy was used for data analysis. The gamma-ray spectrometer's energy calibration was performed with point sources. The International Atomic Energy Authority supplied reference materials RGU-1 (U-ore), RGTh-1 (Th-ore), RGK-1 (K₂SO₄), and IAEA-375 for full energy peak (FEP) efficiency calibration of the gamma-ray spectrometer. Each of these reference materials was counted in the same geometry with samples until a good counting statistic was achieved. The FEP efficiency (ϵ_γ) for each gamma-ray energy of interest was then computed using the following equation (Abbasi et al., 2020a):

$$\epsilon_\gamma = \frac{Cr}{P_\gamma \cdot A} \quad (1)$$

where CR is the net count rate of the region of interest's gamma-ray photopeak, P_γ is the probability of the gamma-ray of interest, and A is the radioactivity of the reference material in Bq. Each soil sample was placed on the detector's top and counted for about 80,000s. To acquire net counts for the sample, background radiations were measured under the same circumstances as sample measurements and removed. The activity concentrations in the ²²⁶Ra and ²³²Th decay series were averaged from gamma-ray photopeaks at various energies, assuming secular equilibrium. The activity concentration of ²²⁶Ra was determined using the gamma-ray lines of 351.9 keV from ²¹⁴Pb and 609.3 keV from ²¹⁴Bi. The activity

concentration of ^{232}Th was calculated using the gamma-ray photopeaks of 911.2 keV from ^{228}Ac and 583.2 keV from ^{208}Tl . The gamma-ray lines at 1460.8 keV were used to quantify the activity concentration of ^{40}K (Asgharizadeh et al., 2011).

2.4 Uncertainty calculation

Uncertainty assessment was carried out before activity computation, with the activity concentration uncertainty (U_A) computed using the equation (Abbasi, 2019):

$$\frac{U_A}{A} = \sqrt{\left(\frac{U_N}{N}\right)^2 + \left(\frac{U_B}{B}\right)^2 + \left(\frac{U_\varepsilon}{\varepsilon}\right)^2 + \left(\frac{U_M}{M}\right)^2 + \left(\frac{U_{P_\gamma}}{P_\gamma}\right)^2} \quad (2)$$

where U_N is sample counting uncertainty; U_B , background counting uncertainty; U_ε , efficiency uncertainty, U_M , mass measurements uncertainty and U_{P_γ} , gamma line energy uncertainty (Lépy et al., 2015).

2.5 Radiological risk assessment

Some parameters, such as absorbed dose rate, Ambient dose equivalent rate ($H^*(10)$), gamma index, and alpha index, can be used to calculate exposure risk.

2.5.1 Absorbed dose rate

The absorbed dose rate (D , nGy/h) due to gamma radiations in the air at one meter above the ground surface by Bq/kg for the naturally occurring radionuclides (^{226}Ra , ^{232}Th , and ^{40}K) is calculated using Eq. (3). The absorbed dosage rate was calculated using conversion factors based on the particular radioactivity levels, A_K , A_{Ra} , and A_{Th} (Bq/kg) of ^{40}K , ^{226}Ra , and ^{232}Th , respectively (Attallah et al., 2019).

$$D \text{ (nGy/h)} = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \quad (3)$$

2.5.2 Ambient dose equivalent rate ($H^*(10)$)

$H^*(10)$ is a quantifiable quantity providing an effective dose assessment that measures the radiation exposure risk to human health. The ambient dose equivalent rate of ^{226}Ra , ^{232}Th , and ^{40}K is calculated at 1 m above the ground surface. The computation of the ambient dose equivalent rate ($H^*(10)$) is as follows (Abbasi et al., 2020a; Lemerrier et al., 2008) :

$$H^*(10)(\text{nSv/h}) = 0.674A_{\text{Ra}} + 0.749A_{\text{Th}} + 0.0512A_{\text{K}} \quad (4)$$

where A_{Ra} , A_{Th} and A_{K} (Bq/kg) are the activity concentration of ^{226}Ra , ^{232}Th and ^{40}K , respectively.

2.5.3 Gamma index

This factor is used to calculate the degree of γ -radiation risk in connection with phosphate fertilisers. The hazard gamma index (I_γ) in Bq/kg is calculated using the following equation (El-Didamony et al., 2013):

$$I_\gamma(\text{Bq/kg}) = \frac{A_{\text{Ra}}}{150} + \frac{A_{\text{Th}}}{100} + \frac{A_{\text{K}}}{1500} \quad (5)$$

To be at the safe level, the value of I_γ must be less than one which is related to an annual effective dose of ≤ 1.0 mSv/y (Ravisankar et al., 2014).

2.5.4 Alpha index

Another significant hazard evaluation aspect of the alpha index (I_α) is the use for the detection of excess alpha radiation from radioactive sources (chemical fertilisers), it is evaluated using the following formula (Joel et al., 2018):

$$I_{\alpha}(Bq/kg) = \frac{A_{Ra}}{200} \quad (6)$$

From literature, if the radiative material activity level of ^{226}Ra above 200 Bq/kg is assumed to contribute to ^{222}Rn concentration $>200 \text{ Bq/m}^3$ of ^{222}Rn exhalation, the recommended activity level of ^{226}Ra is 200 Bq/kg, which gives $I_{\alpha}=1$ (Bangotra et al., 2018).

3. Results and discussions

3.1 Radioactivity concentration

The activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K of different fertilisers types together with the measurement uncertainty (1σ), and average, minimum and maximum values are presented in Table 2. From the show results can be seen that, the values of activity concentrations in the studied fertilisers varied from 54 ± 2.2 to 1585 ± 29.4 , 6 ± 0.1 to 157 ± 9.1 and from 31 ± 1.5 to $1496 \pm 32.8 \text{ Bq/kg}$ for ^{226}Ra , ^{232}Th and ^{40}K , respectively. The minimum and maximum values of ^{226}Ra were obtained in potassium nitrate (PN) and ammonium phosphate (AP) fertilisers. The ^{232}Th minimum and maximum concentration values were found in potassium nitrate (PN) and ammonium sulfate (AS) fertilisers samples. While the sample of potassium nitrate fertilizer (PN) showed the highest amount and the sample of ammonium sulfate fertilizer (AS) showed the lowest amount of ^{40}K concentration. The various sources of raw material and chemical processing of the raw during fertilizer manufacturing might explain the variance in radionuclide concentrations in the investigated chemical fertilisers. The ^{226}Ra average concentrations in all types of chemical fertilisers are higher than ^{232}Th concentrations. While the ^{226}Ra average activity concentrations in all types of chemical fertilisers except for AS and PN are higher than ^{40}K concentrations. It is because of the high potassium present in the NP samples.

3.2 Radiation hazard indices

Table 3 shows the gamma index(I_γ), alpha index(I_α), absorbed dose rate (D), and ambient dose equivalent rate ($H^*(10)$) values for all samples under investigation. The average gamma index of fertilisers ranged from 2.1 to 6.7 Bq/kg. It is found that I_γ for all fertilisers is less than 6 except for fertilisers ammonium phosphate type. This value indicates that almost of sample is safe to use following the requirements of the European Commission ($2 > I_\gamma > 6$)(Rafique et al., 2011), as shown in Table 3. The calculated average I_α values for the samples under investigation were between 0.9 to 4.3 Bq/kg. The recommended upper limit of $I_\alpha=1$, which is according to 200 Bq kg^{-1} of ^{226}Ra (Bangotra et al., 2018). The alpha index (I_α) in the sample Ammonium Sulfate was less than the given limit, whereas in the samples ammonium nitrate (AN) and potassium nitrate (PN) was equal to the limit range. Also, the alpha index value in ammonium phosphate (AP) was more than 4 times. These observed values are more than unity and showed that the ammonium phosphate fertilisers are not safe from the environmental radiation hazards and need more investigation. Fig. 3 shows the maximum, minimum and main values of the alpha index in studied samples.

As shown in Table 3 the value of absorbed dose rate (D), in ammonium nitrate, ammonium sulfate, ammonium phosphate and potassium nitrate samples ranged from 48.9-257.4 nGy/h (an average of 137.2 nGy/h), 58.8-216.9 nGy/h (an average of 137.6 nGy/h), 185.9-850.4 nGy/h (an average of 454.3 nGy/h) and 44.8-287.9 nGy/h (an average of 166.0 nGy/h), respectively. The average value of absorbed dose rate (D) in studied fertilisers are higher than the international limit 59 nGy/h(Palomo et al., 2010).

As illustrated in Table 3, the values of ambient dose equivalent rate ($H^*(10)$) vary between (60.9-1214.32) nGy/h, while the maximum and minimum values of $H^*(10)$ were in ammonium phosphate and potassium nitrate fertilizer samples, respectively. The $H^*(10)$ average values in all fertilizer were higher than the average ambient dose equivalent rate value in soil samples (119.16 nGy/h) reported by the author (Abbasi et al., 2020a). Figure 4 shows the maximum and minimum with main values of ambient dose equivalent rate ($H^*(10)$) in four types of studied fertilisers.

4. Conclusion

Low-level gamma spectrometry was used to determine the activity concentrations of natural radionuclides ^{226}Ra , ^{232}Th and ^{40}K in 21 samples from 4 types of fertilizer of the Arasbaran area, Iran. The obtained results have shown that the averages of radiation hazard parameter I_γ for all fertilisers type samples are higher than the acceptable level ≤ 1 . Also, the radiation hazard parameter I_α for all fertilisers samples are higher than reference level ≤ 1 , except the ammonium sulfate (AS) samples. In all investigated samples, the average values of absorbed dose rate (D) due to the fertilizer samples are higher than the reference level of 59 nGy/h (Palomo et al., 2010) and ranged from 137.2 to 454.3 nGy/h.

The computed average ambient dose equivalent rate ($H^*(10)$) values for each group fertilizer were found 1.6 to 5.5-fold in typical soil samples (Figure 4). The use of fertilisers has a major impact on radionuclide concentrations in agriculture soils, especially phosphorus and potassium-containing fertilisers being one of the causes of the existence of high activity of ^{226}Ra and ^{40}K in the soil. The use of these fertilisers causes a buildup of radioactivity in soils, which can be detrimental to the health of farmers, employees, and product consumers.

Declaration

Competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work was partially supported by the Kyrenia research group, we acknowledgements for their support.

Authors ‘contributions

Abbasi contributed to the design and implementation of the research and the analysis of the results.

Funding

This work was supported by Not applicable.

Availability of data and materials

Not applicable.

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Figures

Figure 1

The geographical location of study area (Arasbaran, East Azerbaijan)

Figure 2

Percentage comparison between average concentration values of ^{226}Ra , ^{232}Th and ^{40}K in fertilisers samples.

Figure 3

The maximum and minimum and main values of the alpha index (I_α) in the fertilisers samples.

Figure 4

The range and main value of ambient dose equivalent rate ($H^*(10)$) in the fertilisers samples and comparison of $H^*(10)$ value in previous studied soil samples.