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Assessing the impact of phosphate fertilizer application on radionuclide accumulation in soil and *Spinacia oleracea*

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ABSTRACT

Phosphate fertilizers improve phosphorus-depleted agricultural soil for better plant growth; however, overuse may lead to human health issues. Analyzed soil samples were treated with five phosphate fertilizers and *Spinacia oleracea* to quantify the levels of natural radionuclides 232 Th, 226 Ra, and 40 K. The study aimed to investigate how phosphate fertilizers contribute to the accumulation of radionuclides 232 Th, 226 Ra, and 40 K in *Spinacia oleracea* using a high-performance germanium detector. Results revealed the mean activity concentrations in agricultural soil mixed with phosphate fertilizer were 232 Th (66.66 ± 6.39 Bq/kg), 226 Ra (54.78 ± 5.41 Bq/kg), and 40 K (837.17 ± 62.89 Bq/kg); *Spinacia oleracea* were 232 Th (5.79 ± 0.83 Bq/kg), 226 Ra (40.24 ± 5.38 Bq/kg), and 40 K (1268.12 ± 129.95 Bq/kg). The soil-to-plant transfer factors were 232 Th (0.244), 226 Ra (0.572), and 40K (1.756). The mean annual limits on the intake of radionuclides were 232 Th (231.67 ± 33.20 Bq/kg), 226 Ra (1609.53 ± 215.33 Bq/kg), and 40 K (50724.67 ± 5,198.13 Bq/kg). The mean annual effective dose was 0.849 ± 0.103 mSv/year, and the internal radiation index was 0.504 ± 0.059 Bq/kg. The results indicate that the mean activity concentrations of 232 Th, 226 Ra and 40 K are below standards but exceed those of the control sample, suggesting regular monitoring of phosphate fertilizer quantity applied to the soil for cultivation.

Keywords: agricultural soil, radionuclides, activity concentration, radioactivity, *Spinacia oleracea*, phosphate fertilizer.

INTRODUCTION

Agriculture is the primary human activity that serves as a food source and generates raw materials for manufacturing industrial products to fulfil the needs of billions of people globally (Hemathilake and Gunathilake, 2022). It is an essential sector of global food security, economic liberation and environmental preservation (García-Díez et al., 2021). Continuous agricultural practices often degrade soil fertility over time by depleting essential elements for plant growth, thus necessitating their replenishment. Although the application of fertilizers has been used as a method for soil nutrient regeneration in optimal agricultural practices, it poses several challenges, including soil degradation, climate change, and pollution from industrial inputs and agricultural raw materials (Bello et al., 2024). Soil fertility is highly affected by hazardous substances and toxic pollutants like heavy metals, radionuclide materials and pesticide residues, which threaten crop health and food safety over extended periods (Alengebawy et al., 2021). The addition of phosphate fertilizers and other agricultural inputs to the soil is used worldwide to enhance crop production and eventually meet the food and raw material demand gap. Phosphate (PO_4^{2-}) is a naturally occurring chemical compound of phosphorus and oxygen, which has been part of the Earth's crust for millions of years (Fayiga and Nwoke, 2016). It is a key raw material in the manufacturing of phosphate (phosphorus) fertilizer, a vital element for plant growth and thus enhancing agricultural production (Mwalongo et al., 2024). The phosphorus element is involved in numerous critical biological and chemical processes. Therefore, these fertilizers supplement its deficiency in the soil (Yahaya et al., 2023). Although phosphate fertilizers are the primary agricultural raw materials for phosphorus replacement, they are associated with naturally occurring radioactive materials (NORMs) impurities, such as uranium, thorium, and their decay products. Excess and uncontrolled use of these fertilizers result in serious environmental concerns and human health issues (Ahmed et al., 2017). Repeated use of phosphate fertilizers typically promotes the transfer and accumulation of radionuclides in agricultural soil, establishing a channel for radioactive components in farm soils, which may allow background radiation to increase over time (Mwalongo et al., 2022). Depending on the half-lives of the precursors, radionuclides can take a long time to attain stability (decay) after being introduced into agricultural soils along with fertilizers (Sarap et al., 2020). During nutrient uptake, these radio nuclides can be absorbed, with a tendency to accumulate in edible parts of the plant. The introduction of these radionuclides to the food chain can pose severe health risks, including cancer, genetic mutations, and organ damage due to prolonged exposure to ionizing radiation (Semenova et al., 2020). Broader environmental contamination and ecosystem degradation result from either leaching into groundwater or dispersal into the atmosphere. Currently, there is limited information on how radionuclides from phosphate rock-based fertilizers influence agricultural soil and their subsequent uptake by plants. Therefore, this study aimed to investigate the levels of natural radionuclides, thorium-232 (^{232}Th), radium-226 (^{226}Ra), and potassium-40 (^{40}K) taken up and accumulated by *Spinacia oleracea* following the application of phosphate fertilizer.

MATERIALS AND METHODS

Materials

The agricultural soil samples were collected from the Nelson Mandela Institution of Science and Technology project farms, located at grid references 3°24’08”S, 36°47’45”E and 3°24’14”S, 36°47’47”E. The varieties of phosphate fertilizers investigated were purchased from the Arusha dispensing store, located at coordinates 30°23’08” S and 36°41’58” E. The varieties of phosphate fertilizers with their trade specifications investigated in this study were; Pamba (N: 10%, P_2O_5: 10%, K_2O: 20%, S: 5%, CaO: 13%, MgO: 1%, B: 1%, Zn: 1%); Top dressing (N: 27%, P: 10%, Ca: 15%); Minjingu organic hyper phosphate granules (P: 28%, CaO: 36%); Minjingu organic hyper phosphate powder (P: 28%, CaO: 36%) and Nafaka Plus (N: 9%, P_2O_5: 16%, K: 6%, S: 5%, CaO: 25%, MgO: 2%, Zn: 0.5%, B: 0.1%+). These brands have been selected based on their application for the annual crop cultivars, including vegetables. The plant used for the uptake and accumulation studies was *Spinacia oleracea*, and its seeds were bought from the Arusha market. The ORTEC High Purity Germanium (HPGe) detector, coupled with gammavision software, was used to measure the radioactivity of ^{232}Th, ^{226}Ra, and ^{40}K. A WFX-210 atomic absorption flame photometer (AAS) was used to calculate the cation exchange capacity after digesting the sample in a microwave using analytical-grade hydrochloric acid, nitric acid, and hydrogen peroxide. A polyethene container of approximately 2 liters of capacity was used as a pot to grow *Spinacia oleracea*. Potassium dichromate, ammonium ferrous sulphate, Ferron indicator, and Diphenylamine of analytical grade were used in determining the soil organic matter.

Experimental design

The experiments were arranged in a completely randomized design. Each of the five phosphate fertilizer varieties investigated was planted in double-triplicate (six) pots for growing *Spinacia oleracea*. Every pot contained 2 kg of agricultural soil (AS) homogenized with 200 mg of Pamba (PA) phosphate fertilizer (Jayadi et al., 2023). The same treatment was applied to the soil for top dressing (TD), Minjingu organic hyperphosphate granules (MOHPG), Minjingu organic hyperphosphate powder (MOHPP), and Nafaka Plus (NA). Two to four *Spinacia oleracea* seeds were placed in every pot, controlled in a greenhouse environment at the Nelson Mandela Institution of Science and Technology. The irrigation process was performed frequently to maintain a moisture content of approximately 80%.

Sampling and processing

One hundred kilograms of composite agricultural soil was collected from the field, screened by removing debris and plant remains, and then homogenized. A portion of approximately 8 kg was placed in a polythene bag and transported to the laboratory for further processing and analysis of selected soil properties, including cation exchange capacity, pH levels, soil texture, and organic matter content, which control the uptake of natural radionuclides by plants. Approximately 80 kg of AS was used for every pot, containing 2 kg, to support the growth of *Spinacia oleracea*. Irrigation water (IW) was taken from the Nelson Mandela Institution of Science and Technology water tap. Five varieties of phosphate fertilizers were purchased from the company's Arusha depot: Pamba, top dressing, Minjingu organic hyperphosphate granules, Minjingu organic hyperphosphate powder, and Nafaka Plus. A package containing about 200 g of *Spinacia oleracea* seeds was bought from the Arusha market.

After two months of care, the *Spinacia oleracea* plant leaves were harvested, mixed into a single homogeneous sample, and then washed. The agricultural soil, phosphate fertilizer-amended soil, and *Spinacia oleracea* were air-dried and then oven-dried at approximately 105 °C. Afterwards, the samples were ground and sieved to pass through a 2 mm pore size for particle uniformity. Approximately 250 g of homogenized samples were weighed and packed in airtight steel canisters and then stored for at least 21 days to allow uranium and thorium to attain secular equilibrium with their progeny. Afterward, the samples were loaded into a gamma spectrometer for a radiometry analysis of ^{232}Th, ^{226}Ra, and ^{40}K (Mwalongo et al., 2022).

The samples investigated were agricultural soil, agricultural soil mixed with phosphate fertilizers brands: top dressing, Pamba, Nafaka Plus, Minjingu organic hyperphosphate granules, Minjingu organic hyperphosphate powder, control, and irrigation water. Additionally, samples of *Spinacea oleracea* were grown in the presence of phosphate fertilizers.

Determination of soil properties (cation exchange capacity, organic matter, pH and particle size)

Soil sample digestion was done in the microwave for cation exchange capacity (CEC) analysis. Approximately 5 g of soil samples were mixed with 10 cm^3 of concentrated HCl and 10 cm^3 of concentrated HNO_3, followed by the addition of 5 cm^3 of H_2O_2 to release metallic ions. The solution was filtered through Whatman paper and diluted with distilled water to a final volume of 100 cm^3. The filtrate was analyzed using a WFX-210 atomic absorption spectrophotometer (AAS) at wavelengths of 285.2 nm for Mg, 422.7 nm for Ca, 766.5 nm for K, and 589.0 nm for Na. The estimation for CEC of a soil analyzed by AAS was based on Equation 1, as described in Ross and Ketterings (Bankaji et al., 2023),

$$CationExchangeCapacity\ (CEC)\left(\frac{meq}{100}g\ soil\right) = \frac{CM(mg/g)}{AM\ x\ CPWS\ (meq/g)} \quad (1)$$

where: *CM (mg/g)* – concentration of metal ion, *AM x CPWS (meq/g)* – atomic mass x charge per weight of soil (meq/q)

To measure the soil pH, 1:5 (w/v) of agricultural soil and distilled water were used (Barrow and Hartemink, 2023). The soil texture was determined using the hydrometer method (Sikora and Moore, 2014).

To measure the amount of soil organic matter, a solution of 10 cm^3 containing 1.000 N potassium dichromate and 20 cm^3 of concentrated sulfuric acid was added to agricultural soil samples weighing 1 gram, contained in a 250 cm^3 Erlenmeyer flask. The mixture was shaken to achieve uniformity and then left to stand for 20 minutes. Afterwards, distilled water was added to a 100 cm^3 volume. The excess dichromate was titrated with 0.2042 mol/dm^3 ammonium ferrous sulfate using a ferron indicator. The percentage of total organic carbon (TOC) was determined using Equation 2 (Gerenfes et al., 2022):

$$\%\ TOC = 1.724\left(N\,x\,\frac{V_1 - V_2}{w}\,x\,0.39\,x\,cf_m\right) \quad (2)$$

where: *N* is normality of ferrous sulphate solution (from blank titration), V_1 is the volume of ammonium ferrous sulphate solution titrated with a blank in mL, V_2 is the volume of ammonium ferrous sulphate solution titrated with a sample in mL, *w* is weight of air-dry soil sample in g, 0.39 is $3 \times 10^{-3} \times 100\% \times 1.3$ (3 is equivalent weight of carbon), cf_m is moisture correction factor and 1.724 is empirical factor.

Determination of activity concentration

The gamma spectrometric technique, utilizing a high-purity germanium (HPGe) detector, was employed to measure and analyze the sample. The detector was coupled with Gammavision®

software for data processing. Before measurements, the HPGe system was energy-calibrated using gamma lines at 59.5 keV from ^{241}Am, 661 keV from ^{137}Cs, and two gamma lines at 1.333 keV and 1.172 keV from ^{60}Co, all of which are present in the multi-nuclide standard (MBSS2). The system was calibrated using standard reference materials (MSSB2) to ensure accurate quantification of radionuclides present in the samples, based on the photo peak efficiency (Table 1)

The activity concentrations of the radionuclides in each sample were calculated using Equation 3 (Sawe, 2023):

$$A = \frac{N_s}{m\,P_\gamma\,t} \quad (3)$$

where: A is the individual radionuclide activity concentration, Ns is the net count per second (NCPS) = (sample CPS – background CPS), ε is the photopeak efficiency, $P\gamma$ is the gamma emission probability, m is the mass of the sample in kilograms, and t is the counting time in seconds. The results were used to determine the amount of natural radionuclide absorbed by the plants from phosphate fertilizer using a well-established model, as shown in Equation 3.

Transfer/uptake factor (TF) estimation

The transfer factor (TF) assesses radionuclide accumulations in the environment, predicting their release into the soil and uptake by crops to estimate human dose intake. It is the ratio of radionuclide concentrations in vegetation at the time of harvest to those in the soil at the time of planting. Equation 4 (Oladele et al., 2023) estimated the TF of radionuclides from the potted soil to the *Spinacia oleracea*.

$$TF = \frac{RC\ DC}{RC\ DF} \quad (4)$$

where: $RC\ DC$ – radioactivity concentration in dry crops(Bq/kg), $RC\ DF$ – radioactivity concentration in dry fertilizeramended soil (Bq/kg)

Estimation of annual limit on intake for radionuclides

The annual limit on intake for radionuclides (ALI) ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K resulting from the consumption of the *Spinacia oleracea* was estimated based on Equation 5 (Adedokun et al., 2019) (Table 2).

$$ALI = A\ x\ A_{CR} \quad (5)$$

where: A is the mean activity concentration of a specific radionuclide in Bq/kg, and A_{CR} is the annual consumption rate of vegetables in kg.

Annual effective dose (AED)

Equation 6 was used to estimate the annual effective dose (AED) from consuming vegetables grown using phosphate fertilizer (Adedokun et al., 2019).

$$AED = \sum A\ x\ D_{cf} C_R \quad (6)$$

where: A is the activity concentration of radionuclide (Bq/kg), D_{cf} is the dose conversion factor for radionuclide (Sv/Bq) with values, 6.2×10^{-9}, 2.3×10^{-7} and 2.8×10^{-7} for, ^{40}K, ^{232}Th and ^{226}Ra respectively, C_R is the annual consumption rate of the vegetables (UNSCEAR, 2000).

Internal hazard index (H_{in})

According to UNSCEAR (2000) the internal hazard index (H_{in}) is estimated using Equation 7. Consideration of the safety of consumed material

Table 1. Comparison of activity concentrations (Bq/kg) of natural radionuclides in *Spinacia oleracea* with literature

Country/Location	Mean activity concentration of radionuclide (Bq/kg)			Reference
	^{232}Th	^{226}Ra	^{40}K	
Tanzania	140.26	105.70	1540.77	Present study
Tanzania	10.83	*	686.86	(Mohammed and Haule, 2018)
Turkey	BDL	0.8	9.84	(Canbazoğlu and Doğru, 2013)
Malaysia	*	1.6	208	(Asaduzzaman et al., 2015)

Note: * Not applicable, BDL is below detection limit.

is likely below the value of unity; otherwise, it poses a health risk. The activity concentration is assumed to be unity when the H_{in} is half of the normal ^{226}Ra, which equals a radiation limit of 185 Bq/kg for ^{226}Ra, 259 Bq/kg for ^{232}Th and 4810 Bq/kg for ^{40}K (Wanjala et al., 2015).

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \qquad (7)$$

where: A_{Ra}, A_{Th}, and A_K are the activity concentrations of radium, thorium, and potassium in *Spinacia oleracea*, respectively.

Statistical analysis

Data analysis was performed statistically using one-way ANOVA, which indicated that the activity concentration of ^{232}Th and ^{226}Ra differed significantly across fertilizers with $p < 0.05$, whereas no significant difference was found for ^{40}K with $p = 0.240$. This is supported by the strength of the group difference (effect size), where ^{232}Th and ^{226}Ra display a near total effect, with an Eta-square of ≈ 1.00, while ^{40}K shows a moderate effect, with an Eta-square of 0.396. Multiple comparisons between fertilizers were assessed using post-hoc analysis. For ^{232}Th, significant differences were observed with $p < 0.05$, except for TD and MOHPP, with CO frequently exhibiting a higher activity concentration than the others. For ^{226}Ra, significant differences were also found, with NA showing higher values than the other samples. The post-hoc results do not reveal a consistent significant difference for ^{40}K, attributable to the non-significance of the ANOVA results. Consequently, the choice of fertilizer significantly affects ^{232}Th and ^{226}Ra, as evidenced statistically. However, despite the high variation of ^{40}K among fertilizer types, it did not demonstrate statistical significance. The TF of ^{232}Th, ^{226}Ra, and ^{40}K shows significant variation in treatment with phosphate fertilizer, $p = 0.01$, with NA leading to higher TF and CO was very minimal, with TD and PA showing similar effects. This was agreed with the ANOVA effect size tests, approximately one, as there was a significant difference statistically and practically.

RESULTS AND DISCUSSION

Soil properties

The soil and water were analyzed for cation exchange capacity, yielding results of 3286.901 meq/100 g in soil and 2280.632 meq/100 g in water. The particle size analysis indicated that the soil consisted of 42% sand, 6% clay, and 52% silt, classifying it as silt loam. The recorded pH value of the soil in this study was 6.24. Additionally, the soil contained a total organic carbon content of 21.65 ± 0.73 g/kg. Integrations of the percentage of sand, clay and silt based on the triangle corresponding to general soil classification as silt loam

Cation exchange capacity (CEC) has been studied to correlate with the pH to regulate soil acidity. The total CEC availability presents the concentrations of Ca, K, and Mg ions in the soil (3286.901 mg/g) and in irrigation water (2280.632 mg/g). Soil pH of 6.24 indicates either precipitation or displacement, as 99% is contributed by potassium and calcium to CEC rather than 1% of magnesium proportions for free ions of radionuclide ^{232}Th, ^{226}Ra and ^{40}K available for plant uptake. It has been studied that the soil with

Table 2. Radiological hazard for natural radionuclides on the consumption of *Spinacia oleracea*

Sample	ALI^a (Bq)				AED^b (mSv/year)	H_{in}^{c} (Bq/kg)
	^{232}Th	^{226}Ra	^{40}K	Total		
TD	116.0 ± 0.001	1706.8 ± 273.60	74876.0 ± 7567.20	76698.8 ± 2720.80	1.014 ± 0.128	0.631 ± 0.076
PA	106.4 ± 0.001	652.8 ± 145.60	75204.0 ± 7612.80	75963.2 ± 2828.00	0.719 ± 0.093	0.489 ± 0.059
NA	728.4 ± 153.60	4404.8 ± 463.20	64924.0 ± 6362.40	70057.2 ± 7792.40	1.842 ± 0.208	1.003 ± 0.110
MOHPG	57.6 ± 0.001	372.8 ± 120.00	29166.0 ± 3051.20	29596.4 ± 2051.20	0.316 ± 0.054	0.208 ± 0.032
MOHPP	284.8 ± 45.60	2243.6 ± 158.00	16738.0 ± 2046.00	19266.4 ± 2738.40	0.808 ± 0.069	0.417 ± 0.036
CO	96.8 ± 0.001	276.4 ± 131.60	43440.0 ± 4549.20	43813.2 ± 4680.80	0.395 ± 0.068	0.272 ± 0.041
Average	231.67 ± 33.20	1609.53 ± 215.33	50724.67 ± 5,198.13	52565.87 ± 5,446.67	0.849 ± 0.103	0.504 ± 0.059

Note: ^{a} Annual limit on intake (ALI) (Bq), ^{b} annual effective dose (AED) (mSv/year) and ^{c} internal hazard index (H_{in}) (Bq/kg). PA is Pamba, NA is Nafaka plus, MOHPG is Minjingu organic hyperphosphate granule, MOHPP is Minjingu organic hyperphosphate powder, and CO is control soil without fertilizer.

higher pH values affects the mobility of radionuclides by forming precipitates with ion complexes of carbonates, hydroxyl, phosphate and sulphides and lower values cause displacement of radionuclides from their compounds with H^+ (Maharn Gafer, 2022). This pH condition regulated the radionuclide uptake from amended agricultural soil to *Spinacia oleracea*, resulting in a radiological hazard index closer to tolerable levels of 1 mSv/y for public exposure.

Activity concentrations of natural radionuclides

Activity concentration refers to the amount of radiation emitted by a specific radionuclide, expressed as the quantity of radioactivity in Becquerels per unit mass of solid material (García-León, 2022). The study evaluated the activity concentrations and their uncertainties of radionuclides ^{232}Th, ^{226}Ra, and ^{40}K in the soil, as well as in a mixture of soil with fertilizer and *Spinacia oleracea* samples, presented in Figure 1. According to UNSCEAR (2000), the global mean activity concentration of radionuclides in soil ranges from 17 to 60 Bq/kg for ^{226}Ra, 11 to 64 Bq/kg for ^{232}Th, and 140 to 850 Bq/kg for ^{40}K. Therefore, for agricultural soil, the allowable minimum limits mean activity concentrations for ^{232}Th, ^{226}Ra, and ^{40}K are 30 Bq/kg, 35 Bq/kg, and 400 Bq/kg, respectively. The allowable minimum limits for activity concentration to pose immediate effects are 1.000 Bq/kg for ^{232}Th and ^{226}Ra, but 10.000 Bq/kg for ^{40}K (IAEA, 2023).

Activity concentrations of natural radionuclides in agricultural soil and water

The ^{232}Th had a mean activity concentration in AS (114.30 ± 10.79 Bq/kg). The irrigation water (IW) had a mean activity concentration that was less than the detection limits. The mean activity concentration of ^{226}Ra in AS was 65.23 ± 6.16 Bq/kg, but it was below the detection limit in IW. The results show that ^{226}Ra has low concentrations compared to ^{232}Th. The activity concentration of ^{40}K was 546.76 ± 51.24 Bq/kg in AS and below detection limits in IW. The result shows that the availability of ^{40}K is 9 times higher than that of ^{226}Ra but 5 times higher than that of ^{232}Th. The average of this study was 4 times higher for ^{232}Th, 2 times for ^{226}Ra and 1.4 times for ^{40}K than the allowable limits for agricultural soil. According to the IAEA (2023), the radionuclides ^{232}Th and ^{226}Ra can pose immediate health effects when above 1.000 Bq/kg, but 10.000 Bq/kg for ^{40}K; therefore, the results suggest minimal immediate radiological risk to the environment and human exposure.

Activity concentrations of natural radionuclides in the soil mixed with phosphate fertilizer

The maximum mean activity concentration for ^{232}Th was detected in PA (107.85 ± 10.04 Bq/kg), and the minimum was in NA (24.12 ± 3.21 Bq/kg), but MOHPG had a value below the detection limit. These results align with studies on phosphate rocks that carry elevated levels of thorium due to their geological origin (Mdachi et al., 2024). The contribution of the soil's physicochemical properties could be attributed to the natural retention and adsorption of ^{232}Th to the soil particles. These findings are consistent with other studies, indicating that ^{232}Th is likely to bind strongly to soil minerals under specified soil properties, thereby reducing its mobility in aqueous systems (Tiwari et al., 2024). The highest mean activity concentration of ^{226}Ra was in NA (114.24 ± 9.26 Bq/kg), and the lowest was in MOHPG (24.89 ± 4.77 Bq/kg. The low concentration of ^{226}Ra (Figure 1 (b)), a decay product of uranium, has been attributed to the formation of precipitates with calcium and barium compounds at higher pH values, which limits its mobility in water. The activity concentration of ^{40}K was maximum in NA (2152.78 ± 182.34 Bq/kg) and minimum in TD (546.76 ± 51.24 Bq/kg). The results presented in Figure 1(a) revealed the highest activity concentration of ^{40}K in the soil, as potassium is an essential plant nutrient that is naturally abundant in soil and often sourced from potassium-rich soil minerals and fertilizers (Tiwari et al., 2024). The samples TD, PA, and MOHPP overshoot the recommendation of 30 Bq/kg for ^{232}Th, 35 Bq/kg for ^{226}Ra and 400 Bq/kg for ^{40}K. The records stipulate that NA was below the recommendation for ^{232}Th but above for ^{226}Ra and ^{40}K. The results show that ^{226}Ra has high concentrations compared to ^{232}Th and ^{40}K. The availability of ^{40}K is 15 times higher than that of ^{226}Ra but 12.6 times higher than that of ^{232}Th; therefore, long-term use can pose health effects.

Activity concentrations of natural radionuclides in the Spinacia oleracea

Figure 1 (c) shows that the amount of ^{232}Th recorded in *Spinacia oleracea* grown in the soil

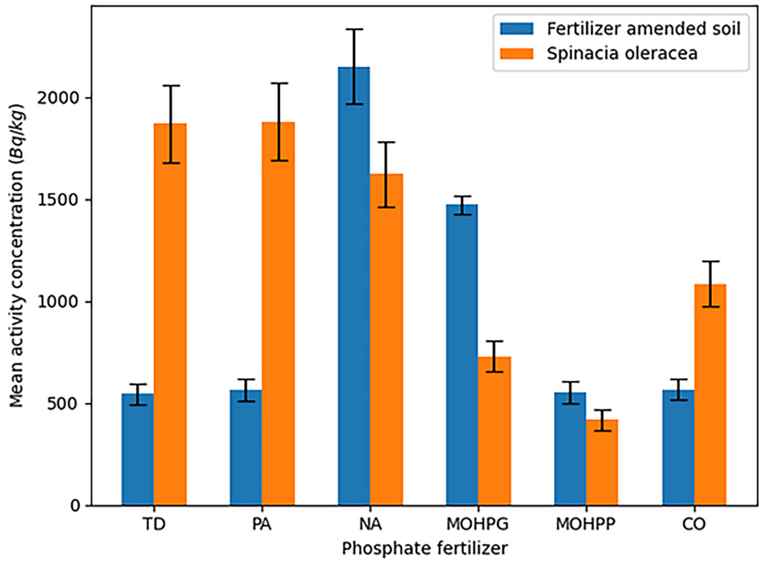

Fig. 1a. Distribution of radionuclides in agricultural soil amended with five varieties of phosphate fertilizer and *Spinacia oleracea* grown in

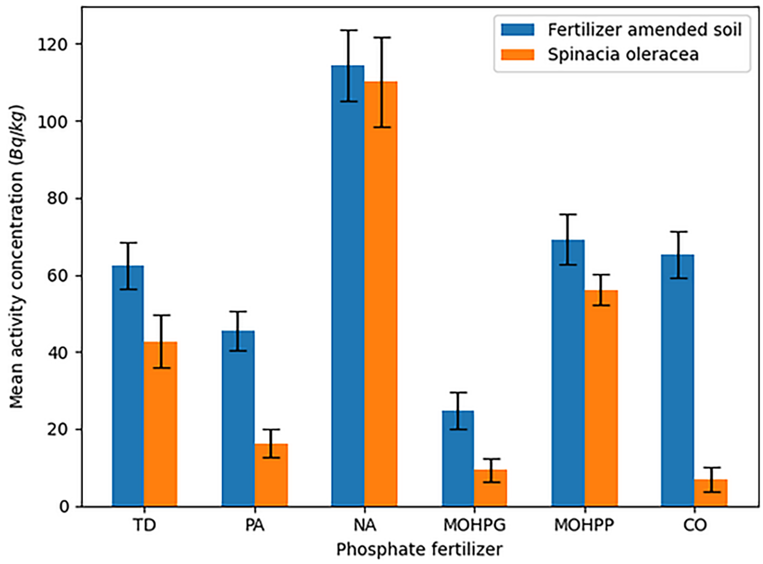

Fig. 1b. Distribution of radionuclides, in agricultural soil amended with five varieties of phosphate fertilizer and *Spinacia oleracea* grown in

amended with NA was 18.21 ± 3.84 Bq/kg, followed by that of the soils amended with MOHPP (7.12 ± 1.14 Bq/kg). The results of the soil amended with TD, PA and MOHPG were below the detection limits. The activity concentration of *Spinacia oleracea* grown in the soil amended with TD, PA and MOHPG was not detected. For ^{226}Ra, the maximum mean activity concentration in *Spinacia oleracea* grown in the soil amended with NA (110.12 ± 11.58 Bq/kg) and the minimum was in un-amended CO (6.91 ± 3.29 Bq/kg). The *Spinacia oleracea* grown in NA revealed a higher ^{226}Ra activity concentration than the other four fertilizers, with the minimum recorded in MOHPPs. The ^{40}K has shown high accumulation in *Spinacia oleracea* grown in the presence of PA (1880.1 ± 190.32 Bq/kg), and the minimum was MOHPP (418.45 ± 51.15 Bq/kg). *Spinacia oleracea* grown in the presence of PAv was observed to accumulate a higher ^{40}K concentration compared to those grown in soils amended with the other phosphate fertilizers. *Spinacia oleracea* cultivated in the presence of five varieties of phosphate fertilizers revealed that the addition of NA to the soil

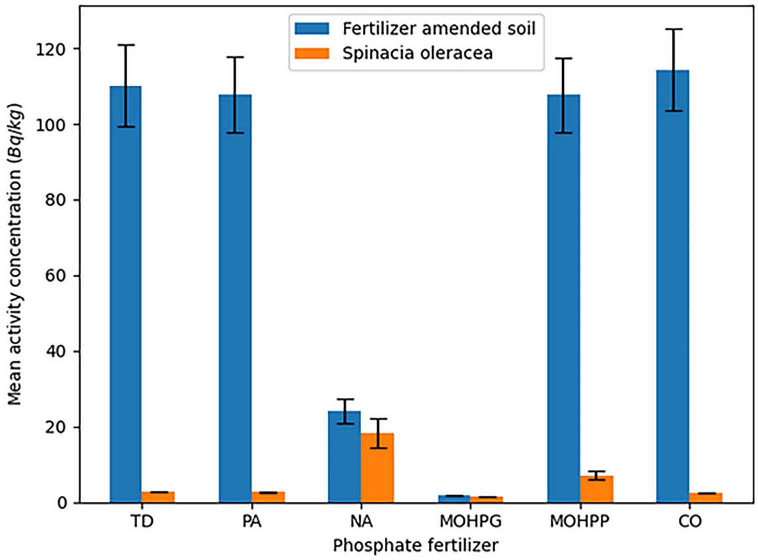

Fig. 1c. Distribution of radionuclides in agricultural soil amended with five varieties of phosphate fertilizer and *Spinacia oleracea* grown in

enhances the bioavailability of ^{232}Th compared to other fertilizers. The activity concentrations obtained in this study are higher than those of *Spinacia oleracea* grown in typical soil, as determined by Mohammed and Haule [33], for natural radionuclides ^{232}Th and ^{40}K, which are approximately 0.6 and 2.4 times higher, respectively, which are relevant to Nenot (2009) results. The sedimentary deposits have been reported to contain a high level of ^{226}Ra, which serves as the original raw material for manufacturing phosphate fertilizer, and this finding correlates strongly with the results obtained in this study (Noli et al., 2024). The higher concentration of radionuclide ^{40}K in *Spinacia oleracea* than in soil amended with phosphate fertilizer was due to the entrance via roots playing a fundamental macronutritional role in plant species (Phuong et al., 2023).

Transfer/uptake factor (TF)

The TF of radionuclides from agricultural soil to *Spinacia oleracea* is a crucial parameter in studying the uptake process, which depends on the soil's physicochemical properties and their interactions with plant roots. The analysis evaluated the transfer and uptake of natural radionuclides ^{232}Th, ^{226}Ra, and ^{40}K from the soil to *Spinacia oleracea* (Figure 1). The transfer/uptake factor (TF) for ^{232}Th was observed in decreasing order. The highest TF was observed in *Spinacia oleracea* grown in soils amended with NA (0.755 ± 1.193), and the lowest was in unamended soil CO (0.020). These results suggest that in CO, a strong interaction with other components of the soil by making them insoluble or creating more competition for root access. Among other sources, it has been reported that adding phosphate fertilizers to agricultural soil as a phosphorus amendment contributes to large amounts of radionuclide contamination worldwide (Gautam et al., 2021). A high ^{226}Ra TF was observed in *Spinacia oleracea* grown on soils amended with MOHPP (1.623 ± 0.963). Minimal TF values were recorded for *Spinacia oleracea* grown with unamended control soil CO (0.103 ± 0.534). An unexpected TF higher than unity for ^{40}K was observed in the soil treated with PA (3.319 ± 3.4249), amended soils with TD (3.305 ± 3.4044), and control soils CO (1.908 ± 2.160), triggering high accumulation in *Spinacia oleracea*. The TF less than unity was observed in the soils amended with MOHPP (0.755 ± 0.984), NA (0.753 ± 0.872), and MOHPG (0.494 ± 1.640), showing less uptake.

The ^{40}K, ^{226}Ra, and ^{232}Th have shown varying uptake efficiencies for *Spinacia oleracea* that strongly correlate with previous studies reported by Oladele et al (2023). The ^{40}K leads to the highest TF, followed by ^{226}Ra, and the lowest was ^{232}Th. The TF of ^{226}Ra from soil to *Spinacia oleracea* is higher than ^{232}Th, showing consistent bioavailability and mobility in the soil-to-plant systems. The soil treated with NA (0.755 ± 1.193) shows a higher uptake of ^{232}Th with a decreasing trend in other fertilizers. The leading high TF on the presence of NA provides insight to facilitate the ionization and mobility of ^{232}Th in the soil compared to untreated soil (CO) with a transfer factor of 0.020. The TF drop in PA (0.0246) and TD (0.0268) suggest that the control of ^{232}Th in the soil occurs either by forming a precipitate with available anions or leaching down to the soil, thereby restricting *Spinacia oleracea* uptake. The values above 1 indicate a high uptake rate of ^{40}K, along with another isotope, suggesting that *Spinacia oleracea* requires potassium as a potential macronutrient for its growth.

Radiological Hazard indices

Annual limit on intake (ALI) of natural radionuclides

The annual limit on ALI is dependent on the consumption of *Spinacia oleracea* leaves in daily diets. The ALI depends on the dose limitation system, the physical properties of parent nuclides and their progenitors, the bioactivity of radiation emitted by the radionuclide, and its metabolic behaviour (Santhanabharathi et al., 2025). For occupations involving the ingestion of radionuclides in *Spinacia oleracea*, the limits should not exceed 30000 Bq for ^{232}Th, 20000 Bq for ^{226}Ra, and 70000 Bq for ^{40}K (Nenot, 2009). The mean activity concentration of radionuclides multiplied by the FAO (Nyanda and Nkuba, 2017) annual consumption rate of vegetables (40 kg) is presented in Table 1, assessing whether radiation exposure remains safe and permissible.

The maximum ALI for ^{40}K was observed in PA (75204.0 ± 7612.80 Bq), while the minimum was in MOHPG (29166.0 ± 3051.20 Bq). The ALI recorded for ^{40}K in soil amended with TD, and PA exceeds the recommended limit of 70000 Bq. The ALI for ^{232}Th was highest in NA (728.4 ± 153.60 Bq) and lowest in MOHPG (57.6 ± 0.0 Bq). These values for ^{232}Th are below the recommendation but approximately 27 times higher than the values reported by Adedokun et al (2019) for leafy vegetables. The ALI for ^{226}Ra was highest in soil amended with NA (4404.8 ± 463.20 Bq) and lowest in unamended soil CO (276.4 ± 131.60 Bq). The ALI values for ^{226}Ra were below the threshold limits, and ^{40}K demonstrated higher levels compared to ^{232}Th and ^{226}Ra. These findings are consistent with previous studies that reported the application of phosphate fertilizer to agricultural soil contributes to higher ALI for natural radionuclides (Hatika and Subekti, 2019).

Annual effective dose (AED)

The AED was estimated based on the average individual activity concentration of *Spinacia oleracea* cultivated in phosphate fertilizer-amended agricultural soil. The highest AED found in NA was 1.842 ± 0.208 mSv/y, and the lowest measured in MOHPG was 0.316 ± 0.054 mSv/y, with an average value of 0.849 ± 0.103 mSv/y. *Spinacia oleracea* grown in the soil amended with TD and NA_v have recorded values above, and others fall below the public safe limits of 1 mSv/y (Dosh et al., 2024), but all are lower than the occupational limits of 20 mSv/y. Similar results have been reported in phosphate fertilizers manufactured from ores that contained a reasonably higher activity concentration of radionuclides, leading to high AED (Hatika and Subekti, 2019). According to Charles (2001), reported to UNSCEAR, the AED limit for ingestion was 0.29 mSv/y. Therefore, all values overshoot the recommendation contributed through the ingestion route.

Internal (H_{in}) hazard

Internal hazard (H_{in}) results from the exposure of internal organs to radionuclides that enter the body through various intake pathways, including ingestion, inhalation, and absorption, as shown in Table 1. The results show that the maximum value of H_{in} was 1.003 ± 0.110 Bq/kg in NA and a minimum of 0.208 ± 0.032 Bq/kg in MOHPG, with an average of 0.504 ± 0.059 Bq/kg. The maximum value recorded in this study is 2.6 times higher than reported by Hossein and Ferdous consumption of vegetables (*Spinacia oleracea*) grown in soil amended with NA in the diet, incorporating radionuclides (^{232}Th, ^{226}Ra, and ^{40}K), results in a value exceeding the recommended public exposure limit of 1 mSv/y. The H_{in} values higher than unity (1 mSv/y) in *Spinacia oleracea* grown in soil amended with NA indicate the need for control of public exposure to internal radiation from radon and its progeny. Frequent consumption of agricultural products can lead to significant internal radiation doses for an individual, highlighting the importance of monitoring and controlling the amount of phosphate fertilizer applied to agricultural soil

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