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Radionuclides and the uranium isotope fingerprint of globally produced phosphate rocks, mineral fertilizers, and phosphogypsum waste and its potential effect on the environment

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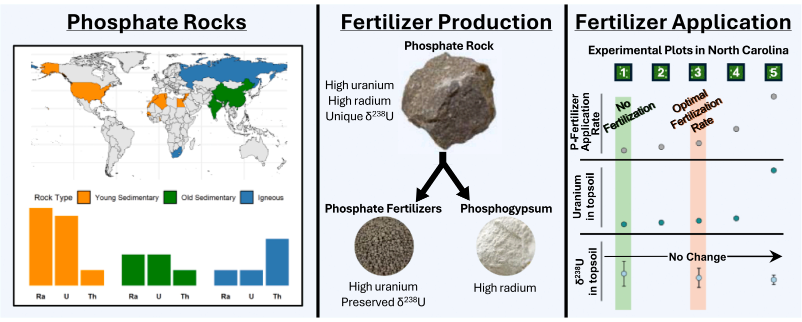
HIGHLIGHTS

- Global dataset of phosphate rocks, fertilizers, and phosphogypsum is presented.
- Processing phosphate rocks enriches U in fertilizers and Ra in phosphogypsum.
- The $\delta^{238}U$ fingerprint of phosphate rocks are retained in the produced fertilizer.
- High flux and percolation of fertilizer-derived U on soils may impact groundwater.

GRAPHICAL ABSTRACT



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ABSTRACT

The occurrence of uranium (U), thorium (Th), and radium (Ra) in phosphate rocks (PRs) leads to enrichment of these radionuclides in produced fertilizers and phosphogypsum waste byproducts. This study presents a global dataset of U- and Th- series radionuclides (^{238}U, ^{232}Th, ^{226}Ra, and ^{228}Ra) and uranium isotope composition ($\delta^{238}U$) in PRs, fertilizers, and phosphogypsum. Results reveal systematic variations in U and Ra content and $\delta^{238}U$ signatures based on geological age and phosphate rock type (igneous vs sedimentary). Sedimentary PRs are in ^{238}U-^{226}Ra secular equilibrium, with younger rocks (Miocene-Permian) exhibiting higher ^{226}Ra activities (up to 1837 Bq/kg) than older rocks ($\leq$241 Bq/kg). Fertilizers tend to be concentrated in ^{238}U and depleted in ^{226}Ra relative to source rocks, while phosphogypsum retains ^{226}Ra. In a survey of P- and NPK-fertilizers (i.e., fertilizers that contain a mix of nitrogen, phosphorus, and potassium), we find that fertilizers preserve the U isotope signature of the source phosphate rocks, with $\delta^{238}U$ ranging between −0.28 ‰ and −0.15 ‰ for young PRs and −0.70 ‰ and −0.34 ‰ for older PRs, which are different from the average continental crust (-0.29 ‰). Field data from an agricultural research station in North Carolina demonstrate that, despite high U and Ra input rates from P-fertilizer application, surface sandy soils retain little of these radionuclides. Fertilization did not significantly alter soil $\delta^{238}U$ and the U and Ra levels were low, below environmental safety thresholds. These findings highlight the potential of fertilizer-derived radionuclide contamination in adjacent or underlying water resources

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and offers the $\delta^{238}U$ fingerprints of fertilizers as a potential geochemical tracer for identifying fertilizer-derived U contamination in the environment.

1. Introduction

Phosphate rocks have been shown to contain elevated concentrations of uranium (U) and radium (Ra) nuclides, commonly defined as naturally occurring radioactive materials (NORMs) [1]. Uranium is incorporated into phosphate rocks primarily by substituting for calcium (Ca) in the crystal structure of carbonate-fluorapatite ($Ca_5(PO_4,CO_3)_3F$), which is the primary phosphate-bearing mineral in marine sedimentary phosphate rocks, via reduction of U during early diagenesis [2]. However, only phosphate rocks deposited after the Paleozoic Oxygenation Event (POE) contain elevated U concentrations (i.e., > 40 ppm), due to changes in the depositional environment of phosphate rock diagenesis and the greater availability of U in the ocean from widespread ocean oxygenation following the POE [3]. In contrast, older phosphate rocks deposited, prior to the POE, contain lower U concentrations (< 20 ppm) [3]. In igneous phosphate rocks, U content is generally low (< 20 ppm) due to the differentiation of apatite ($Ca_5(PO_4)_3(F,Cl,OH)$), the primary phosphate-bearing mineral in igneous phosphate rocks, incorporating minor amounts of U, with primary differentiation of U-containing minerals from melt (e.g., uraninite) occurring in later-stage differentiation [2, 4, 5]. Nonetheless, phosphate rocks have concentrations of U that are typically higher than the average continental crust (2.8 ppm) but not as high as a low-grade uranium ore (1000 ppm) [3, 6].

Phosphate rocks are mined to primarily produce phosphate-containing fertilizers [7]. Following possible calcination and physical separation, phosphate rocks are dissolved with sulfuric acid to produce phosphoric acid and consequently generates phosphogypsum as a waste byproduct [1]. The phosphoric acid is used to formulate phosphate fertilizers such as single- and triple- super phosphate (SSP and TSP) or mono- and di- ammonium phosphate (DAP and MAP). As a result of this process, many trace elements that are enriched in phosphate rocks, including U, become enriched in the phosphate fertilizer products [8]. Due to the mixing of phosphorus (P) with nitrogen (N) and potassium (K) sources to produce NPK fertilizer, NPK fertilizers contain relatively lower concentrations of trace elements [8].

The radiological survey of phosphate rocks and subsequently manufactured materials have been documented with respect to the primordial radioisotopes of U (^{238}U), thorium (^{232}Th), and potassium (^{40}K) in addition to ^{226}Ra, the decay product of ^{238}U [9–43]. In these studies, it has been observed that phosphate fertilizer products usually contain high concentrations (expressed as radioactive decay or activities) of ^{238}U and low activities of ^{226}Ra, relative to the concentrations in source phosphate rocks. In contrast, these studies show that the phosphogypsum waste byproduct retains some of the ^{226}Ra but is often depleted in ^{238}U. In addition to the radioactive isotopes of the U-decay series, the stable isotope ratio of ^{238}U and ^{235}U (expressed as $\delta^{238}U$) can be useful to understand geological processes related to redox and terrestrial flux of U into the ocean and in relation to U ore deposits [44, 45]. While the $\delta^{238}U$ in phosphate rocks was used to reconstruct the evolution of phosphate rocks over geological time and the evolution of ocean redox [3], the $\delta^{238}U$ has never been measured in phosphate fertilizers or used to detect the occurrence of phosphate fertilizers in U-contaminated soil or groundwater.

In this study, we present ^{238}U, ^{232}Th, ^{226}Ra, ^{228}Ra, and $\delta^{238}U$ data of phosphate rocks, fertilizers, and phosphogypsum from a global dataset. While most previous studies have focused on local case studies, here we present a global perspective using dataset that covers a wide range of phosphate rocks aged from the Precambrian to the Miocene as well as associated produced P-fertilizers. As far as we are aware, this is the first study to integrate the variations of radionuclides from the U- and Th-decay series with stable U isotopes of ^{238}U and ^{235}U for a global phosphate rocks and fertilizers dataset. This data is leveraged to identify global trends in radionuclide enrichment among these materials and applied to an agricultural research field site to observe the effects of radionuclide-rich fertilizer application on soil quality.

2. Material and methods

2.1. Sample collection

Phosphate rocks (n = 69), fertilizers (n = 36), and phosphogypsum (n = 10) were collected directly from phosphate mines, fertilizer manufacturing plants, and archives at the Julius Kühn Institute in Germany [46–48]. Five 10-point composite surface soils (up to 10 cm depth) and the utilized P fertilizer were collected from the Tidewater Research Station in Plymouth, North Carolina, an agricultural research station that contains several plots with varying amounts of P fertilizer application. Surface soils were collected from plots FTS1, FTS2, FTS3, FTS4, and FTS5, with most P fertilizer application rates of 0, 11.2, 22.4, 67.2, and 168.1 kg P_2O_5 ha^{-1}/year, respectively, and a uniform K-fertilizer application rate of 16.8 kg K_2O ha^{-1}/year [8, 49, 50]. The soil type is a poorly drained Portsmouth series fine sandy loam and is manage under minimum to no tillage and long-term fertilization since 1966 [49].

2.2. Radionuclide analysis

Samples were crushed and homogenized, and all samples were packed into hinged-lid plastic containers “pucks” (6.5 or 2.5 cm in diameter by 1.5 cm in height, based on available sample mass). The pucks were sealed with electrical tape and wax, then allowed to incubate for at least 27 days to reach secular equilibrium in respect to ^{222}Rn ($t_{1/2} = 3.8$ days). Samples were then counted for 1–3 days on one of two Gamma Spectrometers at Duke University: an Ortec GMX Series GAMMA-X high purity germanium detector (HPGe) or a Canberra Broad Energy 5030 germanium (BEGe) detector. ^{228}Ra was measured by analyzing its progeny ^{228}Ac at 911 keV and ^{226}Ra was measured by analyzing its progeny ^{214}Pb at 351 keV. The measurements of ^{226}Ra were corroborated by assessment of its other progeny, ^{214}Bi, at 609 keV which yielded similar activity values to that of ^{214}Pb.

Th and U concentrations were measured by inductively coupled plasma mass spectrometry (ICP-MS, Thermo Fisher X-Series II) at Duke University. The sample digestion method is described in detail in previous studies [3, 8, 51]. Briefly, in preparation for analysis, samples were first dried and powdered, then fully digested with a heated nitric, hydrofluoric acid, and hydrogen peroxide digestion sequence.

2.3. Uranium isotope analysis

Uranium isotope analysis was conducted on either a Thermo Neptune multi-collector ICP-MS at the University of New Mexico Albuquerque Radiogenic Isotope Lab or a Nu Sapphire multicollector ICP-MS at the Duke University Shared Materials Instrumentation Facility. Details for analysis completed at the University of New Mexico were previously reported in Hill et al., 2024 [3]. At Duke University, in preparation for uranium isotope analysis, samples were spiked with IRMM-3636a $^{233}U/^{236}U$ double spike to achieve a uranium spike to sample ratio of 3 %. Powdered fertilizers were digested via heated HNO_3/H_2O_2 digestion. Soil samples and standards were digested via heated $HNO_3/HCl/HF/H_2O_2$ digestion. Uranium was isolated by ion-exchange chromatography using eichrom® UTEVA® resin and is reported in detail in Kipp et al., 2022 [52]. The $^{238}U/^{235}U$ ratio is reported in delta notation against CRM 112-A and defined as $\delta^{238}U$ (‰)

= ([(^{238}U/^{235}U)$_{\text{sample}}$ / (^{238}U/^{235}U)$_{\text{CRM112A}}$]-1)*1000. Precision was tested by repeated measures of the Columbia River Basalt (BCR-2) standard yielding a δ^{238}U value of −0.266 ± 0.023 (n = 5), in agreement with previous studies [52].

3. Results and discussion

3.1. U- and Th- series nuclides

Uranium concentration in phosphate rocks varied by location, type, and age, ranging between 1 and 154 mg/kg (17 and 1902 Bq/kg ^{238}U). The lowest activities are observed in sedimentary phosphate rocks from China and India and igneous phosphate rocks from South Africa and Russia (< 35 Bq/kg). The highest U activities are detected in phosphate rocks from the United States (> 1600 Bq/kg) (Table S1). The U activity variation is associated with the age of the rocks; those from China and India are of Devonian to Proterozoic in age and have low U content, while the others (from the United States, Northern Africa, and the Middle East) are of Mid-Miocene to Permian in age and have higher U content. The ^{226}Ra activity in phosphate rocks varied between 19 and 1837 Bq/kg, which is consistent with the ^{238}U activities in the rocks (Fig. 1 and Table S1). Thus, the U-series radionuclides of phosphate rocks are approximately in radioactive secular equilibrium, in which the activity of the decay product (^{226}Ra) is equal to that of the ^{238}U parent nuclide (Fig. 1).

Phosphate fertilizers have variable and, in some cases, higher U concentrations between 2 and 338 mg/kg (26 and 4174 Bq/kg), but with lower ^{226}Ra activities (< 1 – 803 Bq/kg) relative to phosphate rocks from the same origin (Fig. 1 and Table S1). As an exception, P fertilizers from China and India with low U have ^{226}Ra ≈ ^{238}U, such that the ^{226}Ra/^{238}U ≈ 1. In contrast, phosphogypsum are substantially depleted in U (≤ 26 mg/kg, or ≤ 321 Bq/kg) and less depleted in ^{226}Ra relative to the parent phosphate rocks (2 – 894 Bq/kg) (Fig. 1 and Table S1), in line with what reported in previous studies [15, 41, 43, 53]. There has been reported higher concentrations of U in phosphate rocks sources used for fertilizer production, particularly in Tanzania where concentrations can be as high as ~700 mg/kg [54, 55]. However, fertilizer produced from ore originating from Tanzania contained U concentrations comparable to those observed in this study [54].

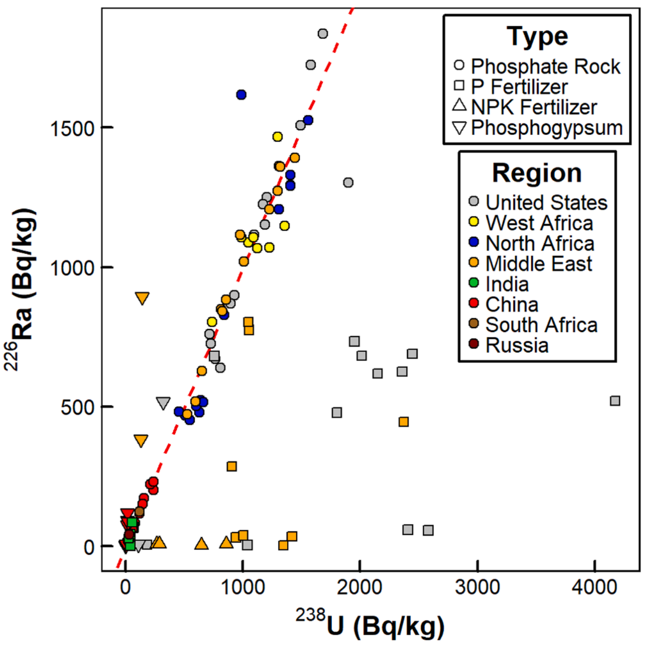

Fig. 1. ^{226}Ra versus ^{238}U activities in global phosphate rocks, fertilizers, and phosphogypsum. The dashed line represents secular equilibrium (i.e., ^{226}Ra/^{238}U activity ratio of 1) between the two radionuclides.

The Th concentrations in phosphate rocks are relatively low, between < 1 and 20 mg/kg (< 1 and 120 Bq/kg), are not age-dependent, and varied by location, with an anomalous Th activity of 487 Bq/kg in an igneous phosphate rock from South Africa (Fig. 2 and Table S1). Th content in phosphate rocks, fertilizers, and phosphogypsum from the United States are within the same range of < 10–37 Bq/kg (n = 31). Samples from North and West Africa only include sedimentary phosphate rocks, of which those from Togo (n = 3) and Algeria (n = 3) contain markedly higher Th activity (mean of 67 and 44 Bq/kg, respectively) while other phosphate rocks have Th activities < 18 Bq/kg (n = 12). With the exception of two phosphate rock samples from Jordon with a Th activity of 12 and 22 Bq/kg, all phosphate rocks, fertilizers, and phosphogypsum from the Middle East have Th activities below 8 Bq/kg (n = 28). Similarly, phosphate rocks and fertilizer from India had Th activities below 8 Bq/kg (n = 6) with the exception of one anomalous fertilizer with a Th activity of 76 Bq/kg. The Th activity in phosphate rocks from China varied from < 16 Bq/kg (n = 12), phosphate fertilizer from 8 to 13 Bq/kg (n = 5) and were < 4 Bq/kg in phosphogypsum (n = 5). Due to the relatively low concentrations of ^{232}Th, and subsequently its progeny ^{228}Ra, secular equilibrium cannot be exemplified for Th as well as for U (Fig. 2). Furthermore, there are no observable trends in the enrichment of ^{232}Th or ^{228}Ra in the manufactured fertilizer or phosphogypsum waste byproduct.

As a result of the substantially greater enrichment of U than Th in phosphate rocks and subsequently manufactured materials, the Th/U activity ratios observed are primarily < 1. This characteristic is also observed in the ^{228}Ra/^{226}Ra, in which most samples (including phosphate rocks, fertilizers, and phosphogypsum) have a consistent ^{228}Ra/^{226}Ra < 1 (Fig. 3). In contrast, igneous phosphate rocks (n = 3) have ^{228}Ra/^{226}Ra > > 1, and it is likely to occur in P-fertilizer produced from this type of igneous phosphate rocks. When compared to the mean U.S. soil [56], which has a ^{228}Ra/^{226}Ra and ^{226}Ra/^{238}U ≈ 1, phosphate fertilizers produced from Mid-Miocene to Devonian aged sedimentary phosphate rocks have lower ^{226}Ra/^{238}U and, in most cases, lower

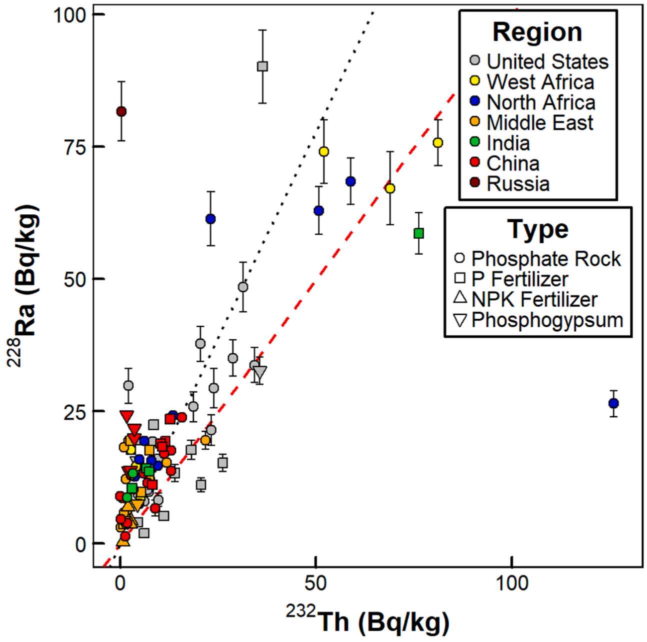

Fig. 2. ^{228}Ra versus ^{232}Th activities in global phosphate rocks, fertilizers, and phosphogypsum. The dashed line represents secular equilibrium between the two radionuclides (i.e., ^{228}Ra/^{232}Th activity ratio of ~1). The dotted line represents the average ^{228}Ra/^{232}Th detected in investigated phosphate rocks of 1.6. Not depicted is an igneous phosphate rock from South Africa with a ^{228}Ra = 516 ± 17 and ^{232}Th = 487.

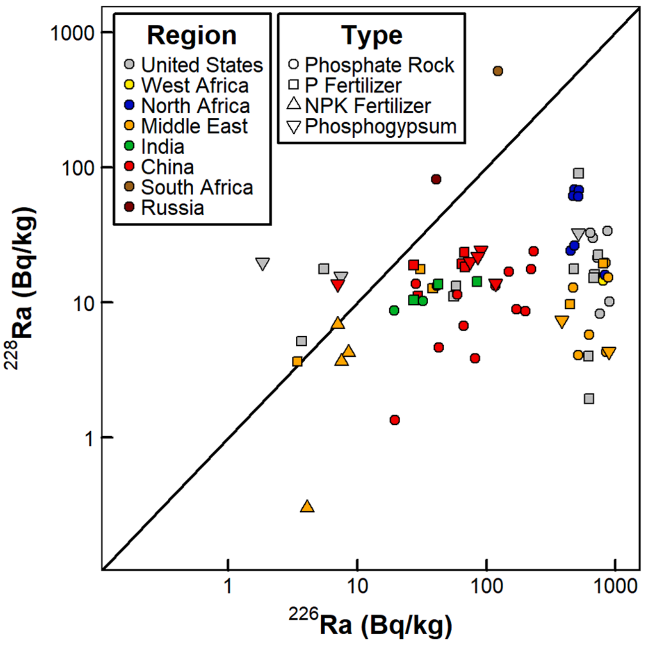

Fig. 3. ^{228}Ra versus ^{226}Ra activities in global phosphate rocks, fertilizers, and phosphogypsum. The line represents a 1:1 ratio between the two radionuclides. The area beneath the line (i.e., ^{228}Ra/^{226}Ra < 1) represents the majority of global phosphate rocks and fertilizers production.

^{228}Ra/^{226}Ra (Fig. 4). Fertilizers produced from Devonian and older sedimentary phosphate rocks (i.e., originating from China and India) also have lower ^{228}Ra/^{226}Ra but similar ^{226}Ra/^{238}U (~1) compared to mean U.S. soil. Although the activity of ^{228}Ra in fertilizer is similar or less than mean U.S. soil (^{228}Ra = 36 Bq/kg), the activity of ^{226}Ra and ^{238}U in fertilizers are about 15 and 65 times higher than the mean U.S. soil, respectively. Thus, the application of P-fertilizer may increase the U and Ra contents and change the nuclide ratios in surface soils in agricultural systems, such that it may be indicative of fertilizer application.

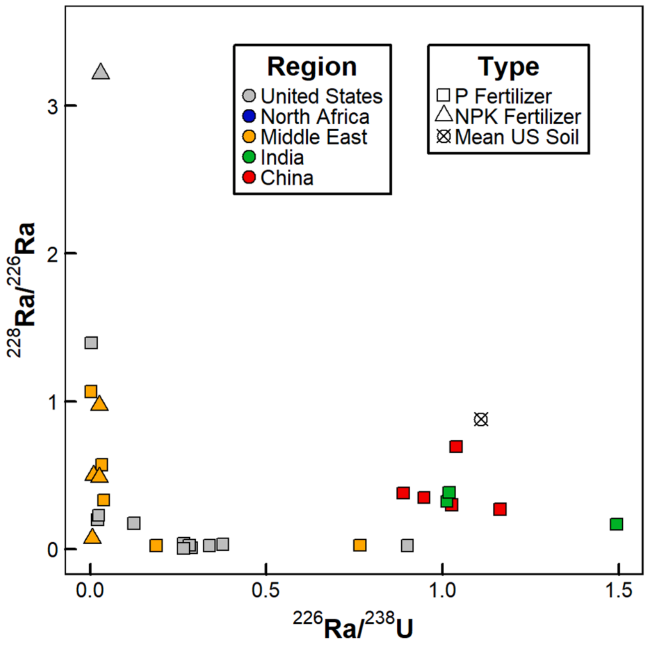

Fig. 4. ^{228}Ra/^{226}Ra versus ^{226}Ra/^{238}U activity ratios of fertilizers analyzed in this study in comparison to the ratios of these nuclides in the mean U.S. soil [56].

The Ra$_{eq}$ (defined as ^{226}Ra + 1.43^{228}Ra + 0.077^{40}K) is utilized by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) to regulate radioactivity limits in soils and other materials. UNSCEAR recommends a Ra$_{eq}$ limit of 370 Bq/kg for exposure to individuals of the public; and a limit of 40 Bq/kg for ^{238}U and ^{232}Th in soils [57]. Most TSP fertilizers analyzed in this study (n = 7 from the US, and n = 1 from the Middle East) and 1 DAP fertilizer and 1 SSP fertilizer (from the US and Middle East, respectively) exceed this Ra$_{eq}$ limit, with Ra$_{eq}$ values between 459 and 831 Bq/kg. Most fertilizers exceed the US soil limit for ^{238}U, but only one fertilizer from India exceeds the US soil limit for ^{232}Th. As far as we are aware, there is no international standard for limits of U in fertilizer; however, the soil conversation panel of the German national environmental agency (Umwelt Bundesamt) is proposing a limit of 50 mg U per kg P_2O_5 [58], which is exceeded by all fertilizers originated from the young sedimentary phosphate rocks (i.e., Middle East, North Africa, U.S.), but not those originating from phosphate rocks in China or India. While fertilizers are typically applied in mixture with soils, in the absence of specific radioactivity limits for fertilizers, their activity values are contextualized using existing soil and exposure guidelines. Consequently, our data show that global phosphate fertilizers have systematically higher U and Ra activities relative to common soils, and the radiation levels exceed known standards for radiation safety.

3.2. δ^{238}U in phosphate rocks, fertilizers, and phosphogypsum

Similar to U concentration, the δ^{238}U of phosphate rocks vary with type and geological age. The δ^{238}U of igneous phosphate rocks and those sedimentary type of Mid-Miocene to Devonian in age vary from −0.31 to 0.07 ‰, while sedimentary phosphate rocks of Cambrian to Precambrian age have lower δ^{238}U between −0.58 and −0.36 ‰ [3]. Our new data indicates that there is no observed significant difference between the δ^{238}U of phosphate rocks, fertilizers, and phosphogypsum generated from the same region (Fig. 5). Fertilizers from China contain the lowest δ^{238}U of ≤ -0.4 ‰ (n = 2), followed by India with a δ^{238}U of −0.34 ± 0.09 ‰ (n = 1). Both P and NPK fertilizers and

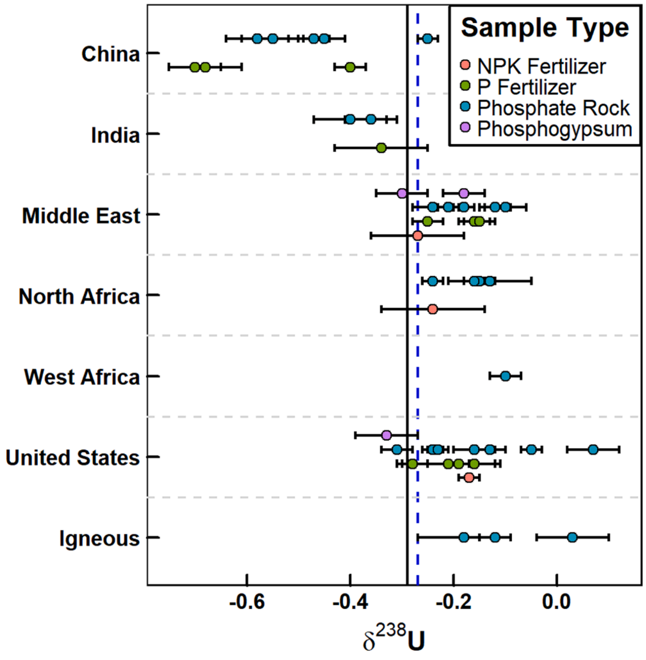

Fig. 5. The δ^{238}U of globally produced phosphate rocks [3], fertilizers, and phosphogypsum. Igneous phosphate rocks include one sample from South Africa (δ^{238}U = 0.03 ± 0.07 ‰) and two from Russia. The solid line represents the value of the average continental crust (δ^{238}U = −0.29 ‰, [59]) and the dashed line represents the value of average riverine waters (δ^{238}U = −0.27 ‰, [60]).

phosphogypsum from the Middle East, United States, and Africa are within a similar range as their originating phosphate rocks, with a δ^{238}U range of −0.33 to −0.15 ‰. The consistent δ^{238}U is likely due to the phosphate rocks being the sole contributor of U into both P and NPK fertilizers. Fertilizers sourced from China and India exhibit a δ^{238}U lower than that of the average continental crust (δ^{238}U = −0.29 ‰, [59]), whereas those from other countries show higher δ^{238}U values (Fig. 5).

The analysis of the δ^{238}U in soils is not an occurrent practice; however, it is expected that soil is likely to have little to no fractionation in the δ^{238}U from the originating rock [61]. For example, a USGS in-house reference soil (SoNE-1, U ≈ 3 mg/kg) has a δ^{238}U = −0.32 ± 0.06 ‰, which is similar to the composition of magmatic rocks [59, 62]. In fact, most terrestrial material were measured to have a δ^{238}U between −0.40 and −0.20 ‰ [59] and the δ^{238}U of major rivers is consistently within that range, between −0.31 and −0.13 ‰ (mean = −0.27 ‰, [60]). Consequently, our data indicate that fertilizers from young (Mid-Miocene to Devonian) phosphate rocks and associated fertilizers from the Middle East, North Africa and the U.S. have higher δ^{238}U values, whereas older phosphate rocks and associated fertilizers from China and India have lower δ^{238}U values relative to the average continental crust and riverine water (Fig. 5). While previous studies have utilized the radioactive U isotopes of ^{234}U/^{238}U for detecting the source of U in groundwater resources, in particular the distinction between alpha recoil and leaching processes [63, 64], we hypothesize that the stable isotopes of U (δ^{238}U) in U-contaminated water could mimic the distinctive isotope fingerprints of the different fertilizers sources, and thus could provide a novel methodology for detecting sources of U contamination in water resources. Since no U isotope fractionation is expected from dissolution of fertilizers, one would expect that water contaminated from extensive fertilizer utilization would have a distinctive δ^{238}U fingerprint relative to U leaching from the soil in the unsaturated zone that is commonly assumed to be the major mechanism of uranium contamination of water resources associated with nitrification processes [63–67].

3.3. The radionuclide impact of phosphate fertilizer on soil

The Ra, U, and δ^{238}U data of analyzed surface soils from the experimental site of Tidewater Research Station in North Carolina [49] are presented in Fig. 6. The U and Ra contents of the applied TSP fertilizer were 163 mg/kg and 682 Bq/kg, respectively. Generally, U concentration and Ra activity in agricultural surface soils from the study plots increase with increasing fertilizer application (Fig. 6). However, the increase is only significant, beyond analytical error, for the highest fertilized soil (FTS5). The P-fertilizer application rate of the FTS3 plot is designed to mimic an optimum amount of nutrients to cultivate crops in the region, while FTS5 plot represents a high flux rate, which is a substantially over-fertilized plot in respect to the P-fertilizer application rate. Therefore, within a range of practical P-fertilization rates, no significant changes in soil Ra or U were detected. Similarly, the δ^{238}U of all the three analyzed soils were within the same range (within analytical error; −0.33 to −0.27 ‰). Similarly, there was no significant trend in the Th concentration based on fertilizer application (Table S2). All soils in this study contained U concentrations and Ra activities below the global soil average (2.67 mg/kg and 32 Bq/kg, respectively [57]).

Although this study did not directly measure radionuclide transfer to crops, previous work has demonstrated potential for U and Ra transference to plants, while Th is usually not as plant available [68, 69]. Uranium uptake is generally low and primarily pronounced in the roots of corn rather than the edible grain [70]. Due to its similar chemical characteristics to calcium, Ra may be more readily incorporated into plants, though uptake is likely small due to the preferential uptake of Ca by crops [71].The extent to which U and Ra are retained in soils depends initially on whether these radionuclides occur in labile forms in the fertilizer. In fertilizer, nearly all present U is likely labile, due to the acid treatment used to produce TSP, while Ra is often less labile and tends to be more strongly retained in soils [72, 73].

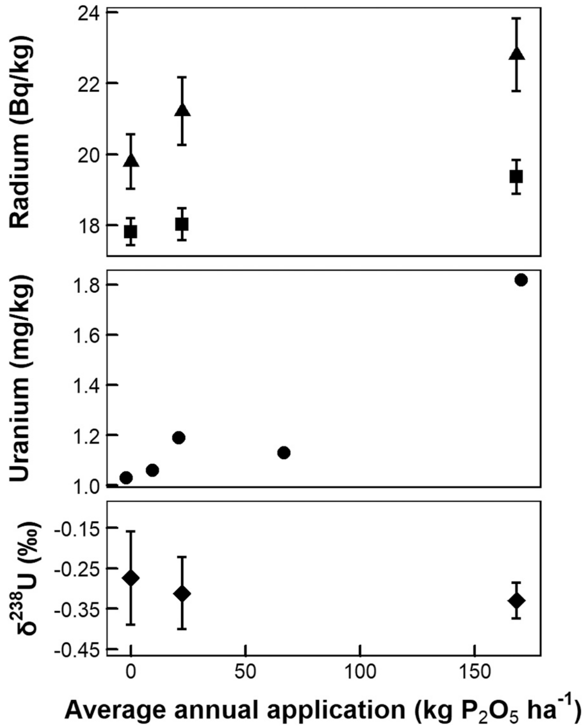

Fig. 6. Variations of Ra (^{228}Ra triangle; ^{226}Ra square), U, and δ^{238}U in surface soils from five different plots treated with varying amounts of P-fertilizer from the Tidewater Research Station in Plymouth, North Carolina. For more details on the research site, see Hu et al., 2024 [49].

Low retention of radionuclides in these field soils can be attributed to the high permeability and low sorption capacity of sandy soils, where limited clay content does not provide adequate sorption sites for labile species. Hu et al., 2024 showed that U concentrations in plots FTS1, FTS3, and FTS5 at the Tidewater Research Station did not exhibit any significant trends in the soil layers above the groundwater table and highlighted the low clay mineral content of the soil the experimental site, which would induce limited U and Ra retention [49]. Furthermore, under oxidizing conditions, typical of such aerated topsoil, U remains in its soluble U(VI) form, complexed with carbonate ligands, or phosphate ligands derived from fertilizer dissolution, facilitating its leaching into the subsurface [74]. However, soils with high organic matter can substantially reduce the mobility of U and, causing the potential for accumulation in long-term fertilization [75].

Although some accumulation of U may occur in the surface soils, there is potential for the fertilizer-derived U to be removed via runoff or leaching into the subsurface and possibly into the underlying groundwater. Other studies have also show limited accumulation of U in agricultural soils via P-fertilizer application, exhibiting potential impact on groundwater quality [76, 77]. The weak retention of U in surface soils and the vertical soil profile supports their potential for vertical transport into groundwater, particularly in coarse-textured, weakly buffered soils. This underscores the need for ground- and surface water monitoring near agricultural operations.

Previous studies have used the δ^{238}U to trace groundwater contamination in relation only to U ore deposits, with δ^{238}U values of affected groundwater ranging from −0.5 ‰ to as low as −2.5 ‰ [44]. Here we find no significant alteration to the soil δ^{238}U; thus, we suggest that future work should assess the applicability of utilizing the δ^{238}U in water resources near agricultural operations to identify the possible sources of U contamination.

3.4. Broader implications

Phosphorus is an essential nutrient for global food production and phosphate rock is its sole source with no current viable alternative [78]. Although extensive work has went into P recovery from waste streams, P recycling (i.e., recovery of P in a usable form) still remains not economically viable [79]. Therefore, the depletion of high-grade phosphate rock deposits will necessitate increasing reliance on lower-grade deposits, which may have higher concentrations of trace elements [80]. The grade of mined phosphate has already begun to decrease, underscoring the importance of evaluating the environmental pathways of fertilizer derived radionuclides [81]. Our field study at the Tidewater Research Station demonstrates the effects of fertilization on one specific soil type and climate. Although immediate risk for direct crop uptake appears limited here, sustained fertilizer application on soils with, for example, higher clay or organic content may increase the risk of fertilizer-derived radionuclides to crop quality. Alternatively, many methods for U extraction from phosphoric acid have been identified, but remain sub-economically viable in most contexts [82, 83]. Increased consumer awareness and future legislation can force the extraction of radionuclides, specifically U, similar to what has been done for Cd [84], is important to reduce the environmental impact of fertilization.

4. Conclusion

This study provides a comprehensive global assessment of radionuclide activities and uranium isotope signatures (δ^{238}U) in phosphate rocks, fertilizers, and phosphogypsum, with implications for environmental monitoring and agricultural practices. Our results demonstrate that uranium concentrations and δ^{238}U values in phosphate rocks vary with geological age and origin, with younger sedimentary rocks exhibiting higher U enrichment and heavier δ^{238}U signatures relative to their older counterparts. This isotopic fingerprint persists in derived fertilizer products, confirming that the phosphate rock source is the dominant contributor to U content in fertilizers and the ability to use the distinctive U isotope ratio as a tracer for the impact of phosphate fertilizers in the environment. We find that fertilizer often exhibits U enrichment, while phosphogypsum retains much of the Ra content of the original phosphate rocks. While fertilizers may contain high U concentrations, field data from the Tidewater Research Station show that surface soils may not retain the majority of the applied U, with minimal δ^{238}U fractionation, under a wide range of agricultural application rates. This suggests a high mobility of fertilizer-derived U, likely due to the sandy soil characteristics of the research site, underscoring the need for groundwater monitoring in areas of intensive fertilizer use. The consistent δ^{238}U signatures among fertilizers from different regions offer a novel geochemical tool for tracing the source of fertilizer-derived U contamination in the environment. Overall, this research highlights the need for expanded monitoring of U and Ra in both agricultural inputs and adjacent water resources, especially in agricultural regions with intensive fertilizer use.

CRediT authorship contribution statement

Robert C. Hill: Writing – review & editing, Writing – original draft, Visualization, Software, Methodology, Investigation, Formal analysis, Data curation. **Zhen Wang:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Jun Hu:** Writing – review & editing, Investigation, Formal analysis. **Gordon D.Z. Williams:** Writing – review & editing, Writing – original draft, Software, Methodology, Investigation, Formal analysis, Data curation. **Avner Vengosh:** Writing – review & editing, Writing – original draft, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization.

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Declaration of 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.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2025.140033.

Data availability

Data are included in the paper

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