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REVIEW ARTICLE

Natural radioactivity in mineral phosphate fertilizers and its impacts on human health: an overview

Camila Rodrigues e Silva[1] · **Fabiana Monteiro de Oliveira**[1]

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Abstract

Humans are constantly exposed to radioactivity present in rocks, soils, and water, mainly from materials in the Earth's crust that contain chemical elements belonging to the radioactive series of uranium and thorium. An important anthropogenic source of these natural radioisotopes to the environment is fertilizers, widely used to increase agricultural productivity. Exposure to ionizing radiation can become a public health problem worldwide, since it is related to the development of different cancers in humans. The present study aimed to survey research on the radioactive content in different types of mineral phosphate fertilizers used around the world through a comprehensive review of the Scopus and Web of Science databases. About 80 scientific articles fit the purpose of this review. The concentration activity values found varied widely from one country to another, and there is no specific legislation that determines the maximum allowed limits of radioisotopes in these agricultural inputs. In addition, there are still uncertainties regarding the impact of natural radioactivity from fertilizers on human health, highlighting the need for further investigations on the subject.

Keywords Phosphate mineral fertilizers · ^{40}K · ^{226}Ra · ^{232}Th · ^{238}U · Effective annual dose

Introduction

Radiation has always been present around us and life on Earth has evolved by being constantly exposed to it, coming both from space and from radioactive materials present in the Earth's crust. With technological advances, especially in the medical field, there has been an increase in exposure to radioactivity by the general public on a daily basis, such as the greater use of medical diagnostic tests, computed tomography scans, medical nuclear medicine, and medical radiology (Sabol 2020 in Gupta and Walther 2020). Currently, the contribution to total public exposure due to natural and anthropogenic sources is approximately the same, a scenario quite different from that observed not more than 30 years ago, when normally natural radiation was responsible for about 82%, coming from space and the Earth's surface, while the contribution from all other sources such as medical X-rays, medicine, and nuclear industries amounted to 18% contribution (Schauer and Linton 2009; Karam and Stein 2009; Bolus et al. 2013; UNEP 2016 ; Sabol 2020 in Gupta and Walther 2020).

It is known that in general, exposure to natural radioactivity varies based on a number of factors such as geographic location, altitude, geology, and human activities and practices. Cosmogenic radiation, for example, is more intense at higher altitudes, and uranium and thorium concentrations in soils are high in localized areas. And in relation to human influence, we have as an example the use of different construction materials and design and ventilation systems strongly influence indoor levels of radioactive radon gas and its decomposition products, which contribute significantly to inhalation doses. However, it is observed that some exposures are relatively constant and uniform for all individuals everywhere, for example, the dose administered through ingestion of ^{40}K in food (UNSCEAR 2000).

Exposure of human beings to natural radiation occurs through two main routes: external route, characterized by exposure through irradiation from radionuclides in the environment, and internal route, characterized by inhalation and ingestion of radionuclides. The main route of the latter

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✉ Camila Rodrigues e Silva
camilae@id.uff.br

[1] Postgraduate Program in Chemistry, Universidade Federal Fluminense, Outeiro de São João Batista, s/n, Niterói, RJ 24020-141, Brazil

occurs through the food chain with the ingestion of water and foods such as plants and meat with high concentrations of radionuclides (Karam and Stein 2009; ICRP 1993). Exposure for prolonged periods of time can represent a significant risk to human health, since sensitive cells of the respiratory, gastrointestinal, and spinocerebellar tracts will be exposed to ionizing radiation, which can lead to the development of neoplasms or other diseases (Groves-Kirkby et al. 2016).

Among the anthropogenic primary sources of uranium and radium to the environment, mineral fertilizers are of great relevance. The use of these agricultural inputs has been increasing worldwide, as it is necessary to replace natural nutrients that are continually being removed from the soil due to agricultural activities and erosion problems. Other sources include nuclear effluents, uranium mining and milling, and coal combustion (Barišć et al. 1992; Yamazaki and Geraldo 2003; Bjørklund et al. 2017).

The presence of radioisotopes in arable areas is a consequence of the natural occurrence of these elements in phosphate rocks usually used as a source of phosphorus in the production of phosphate fertilizers (Yamazaki and Geraldo 2003). The activity concentration of ^{40}K in the soil is an order of magnitude greater than that of ^{238}U or ^{232}Th, respectively, 400, 35, and 30 Bq kg^{-1} (UNSCEAR 2000).

If the leaching processes is not large enough, this can cause the radioactive elements present in these inputs to continuously accumulate in soils (Liesch et al. 2015; Hegedüs et al., 2016; Bigalke et al., 2017). On the other hand, when uranium is mobilized from the upper layers of the soil, groundwater may be enriched with this chemical element. In regions where the availability of uranium and radium due to geological features is increased in the soil, such concern about human exposure to radioisotopes is particularly serious (Kabata-Pendias and Muherjee 2007; Solodukhin et al., 2015).

The extensive use of mineral fertilizers as a source of radioactive contaminants to the environment has aroused scientific interest since the daily intake of radionuclides can be considered a chronic ingestion. Significant effects of the use of phosphate fertilizers in trials in cultivated areas in France were observed with the increase in uranium content and, to a lesser extent, in thorium, with the application of 52 kg ha^{-1} year^{-1} of fertilizer (Wetterlind et al. 2012).

Similarly, studies on the prolonged application of phosphorus-based fertilizers to agricultural soils in Iceland (IS) have observed a significant risk to the environment and human health in regions where these agricultural inputs have been applied for about 50 years. In Sámstaðir, a town located in the south of Iceland, where phosphate fertilizers were applied at an annual rate of 39.3 kg ha^{-1} y^{-1}, the concentration of uranium in the soil increased from 0.65 mg kg^{-1} in the soil not fertilized to 6.9 mg kg^{-1} in topsoil fertilized. In Geitasandur (IS) with a phosphorus fertilization rate of 78.6 kg ha^{-1} year^{-1}, the uranium concentration in the soil reached 15 mg kg^{-1}. The average annual rates of uranium accumulation were 130 and 310 µg ha^{-1} year^{-1}, respectively. These values were greater, by up to a factor of ten, than any previously reported rates of fertilizer-derived uranium accumulation (Sun et al. 2020).

Considering this, the aim of this study was to provide an overview of the scientific progress in the utilization of fertilizers in agriculture. We conducted a comprehensive literature review encompassing studies conducted in Brazil and worldwide. Our focus was on investigating the presence of natural radioisotopes, particularly ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K, in commercially available mineral fertilizers, such as single and triple superphosphates, monoammonium (MAP) and diammonium (DAP) phosphates, NPK, and simple phosphates. Thus, evaluating the regional variations of the isotopic contents of these agricultural inputs, the possible radiological impacts that their use can bring and how this radioactivity can affect human health.

It is important to point out that although all these fertilizers mentioned above are phosphate, some studies referenced in this work did not classify the fertilizers used, either as NPK, SSP, or TSP, naming them only as phosphate fertilizer. In order to avoid misclassification in this review, fertilizers called only phosphate fertilizers in the literature will be treated as a separate group.

The different types of mineral phosphate fertilizers

Since the 1950s, the application of agricultural inputs, including phosphate fertilizers, has increased considerably. More than 30 million metric tons of phosphate fertilizers is consumed annually around the world, aiming at maintaining or increasing the productive potential of land (Lambert et al. 2007; Dias and Fernandes 2006). If the soil lacks sufficient amounts of any macro or micronutrients, plant growth and development are impaired, so fertilizers are an essential component of agricultural activities that help increase agrarian production and improve land properties. However, a possible negative effect of the continuous use of phosphate fertilizers is the contamination of arable land by natural radioactive elements (Dias and Fernandes 2006; Lambert et al. 2007; El-Taher and Althoyaib 2012).

Triple superphosphate fertilizers (TSP), single superphosphate fertilizers (SSP), phosphate fertilizers, ammonium phosphate (MAP and DAP), and NPK fertilizers are produced from phosphoric acid, which is the starting raw material. Monoammonium (MAP) and diammonium (DAP) phosphates are obtained by direct reaction of phosphoric acid with different amounts of ammonia. Triple superphosphate, single superphosphate, and NPK are obtained by directly reacting phosphoric or sulfuric acid with phosphate rock and ammonia. During the reaction

of phosphate rock with sulfuric acid, the radioactive balance between uranium, thorium, and their decay products is disrupted and radionuclides migrate according to their solubility and chemical properties of each element (Saueia and Mazzilli 2006; IAEA 2013; El-Taher et al. 2013). In Fig. 1, there is a schematic representation of the production chain of mineral fertilizers, from the raw material to the final composition of the different types of agricultural inputs.

Single superphosphate (SSP) consists of a mixture of monocalcium phosphate and calcium sulfate, with a phosphorus pentoxide (P_2O_5) content of 16–22%, whereas triple superphosphate (TSP) consists entirely of monocalcium phosphate with a content of phosphorus pentoxide between 43 and 48% (IAEA 2013).

Ammonium phosphates such as monoammonium phosphate (MAP) and diammonium phosphate (DAP) are a class of predominantly granular fertilizers that combine the two essential fertilizer ingredients, nitrogen and phosphate, and are generically called NP fertilizers. The addition of potassium during the manufacturing process results in the production of NPK fertilizers. Typically, the composition of MAP is between 10–11% N and 50–52% P_2O_5, whereas in DAP, a content of about 18% N and 46% P_2O_5 is observed (IAEA 2013; El-Taher et al. 2013).

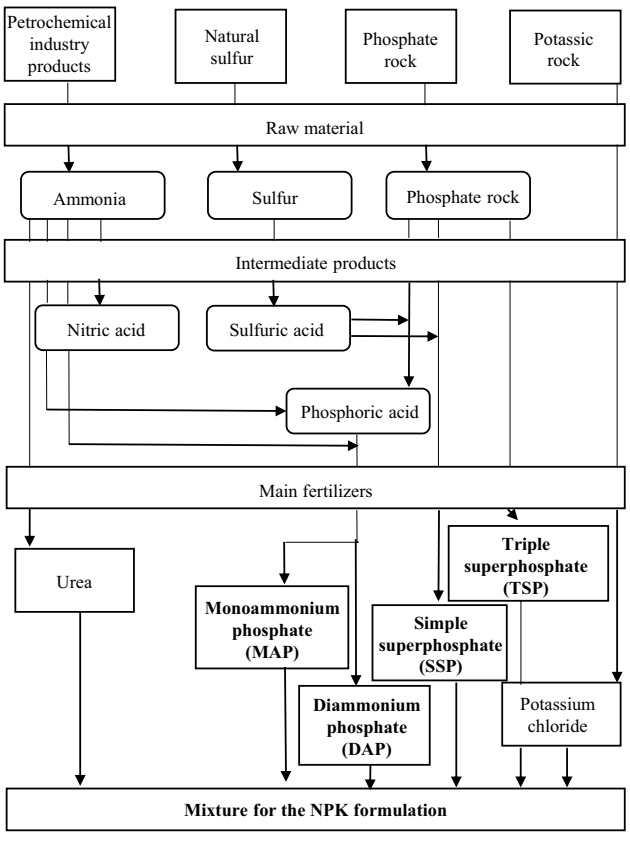

Fig. 1 Schematic representation of the production chain of the main fertilizers. Source: adapted from Dias and Fernandes 2006. COPPE-UFRJ/PETROFÉRTIL

Bibliographic survey

This study consisted of a systematic review covering articles published in the Scopus and Web of Science databases, the largest peer-reviewed databases of abstracts and citations of the literature. Articles were searched that determined the levels of radioactivity in mineral fertilizers in order to evaluate the activity concentrations of the natural radionuclides ^{40}K, ^{226}Ra, ^{232}Th, and ^{238}U in different brands of fertilizers marketed worldwide.

The bibliographic survey approach utilized combinations of keywords related to natural radioactivity in fertilizers found in the title, abstract, and/or body of the text. The obtained results were then filtered according to the study's objectives. Consequently, articles focusing on the determination of natural radioactivity in soils and water bodies in areas where these fertilizers are used, the radiological impacts of fertilizer production industries, and waste disposal, as well as studies investigating radionuclide concentrations in phosphate rocks (the raw material for fertilizer production) and by-products like phosphogypsum generated during manufacturing, were excluded.

From the database, the VOSviewer software version 1.6.18 was used to build a bibliometric map of keywords, co-authorship, and co-occurrence of terms from the last decades, as shown in Fig. 2.

In Fig. 2, it is possible to perceive some grouping patterns. One group, in yellow and green, includes the main techniques for determining radioisotopes in fertilizers, such as alpha and gamma spectrometry. Another, in blue and red, brings the main study topics involving the radioactivity present in fertilizers, such as the determination of the radiological impact, the calculation of the effective annual dose, and the estimation of the external risk index, which deals with evaluating the dose rate of internal radiation due to external exposure to gamma radiation from natural radionuclides.

Finally, there are other themes that are also strongly correlated with the keywords used, which are fertilized land; agriculture, which is one of the activities most impacted by the presence of radionuclides in fertilizers; and the main isotopes studied, such as ^{238}U, ^{226}Ra, and ^{222}Rn, shown in purple in Fig. 2.

Most of the bibliographic production was concentrated in the last decade, with the first study dating back to the 1975s, as illustrated in Fig. 3. The results of research on the topic of interest included articles, articles from magazines and conferences, books, and editorials. In Fig. 4, there is a schematic representation of the geographical distribution of these surveyed

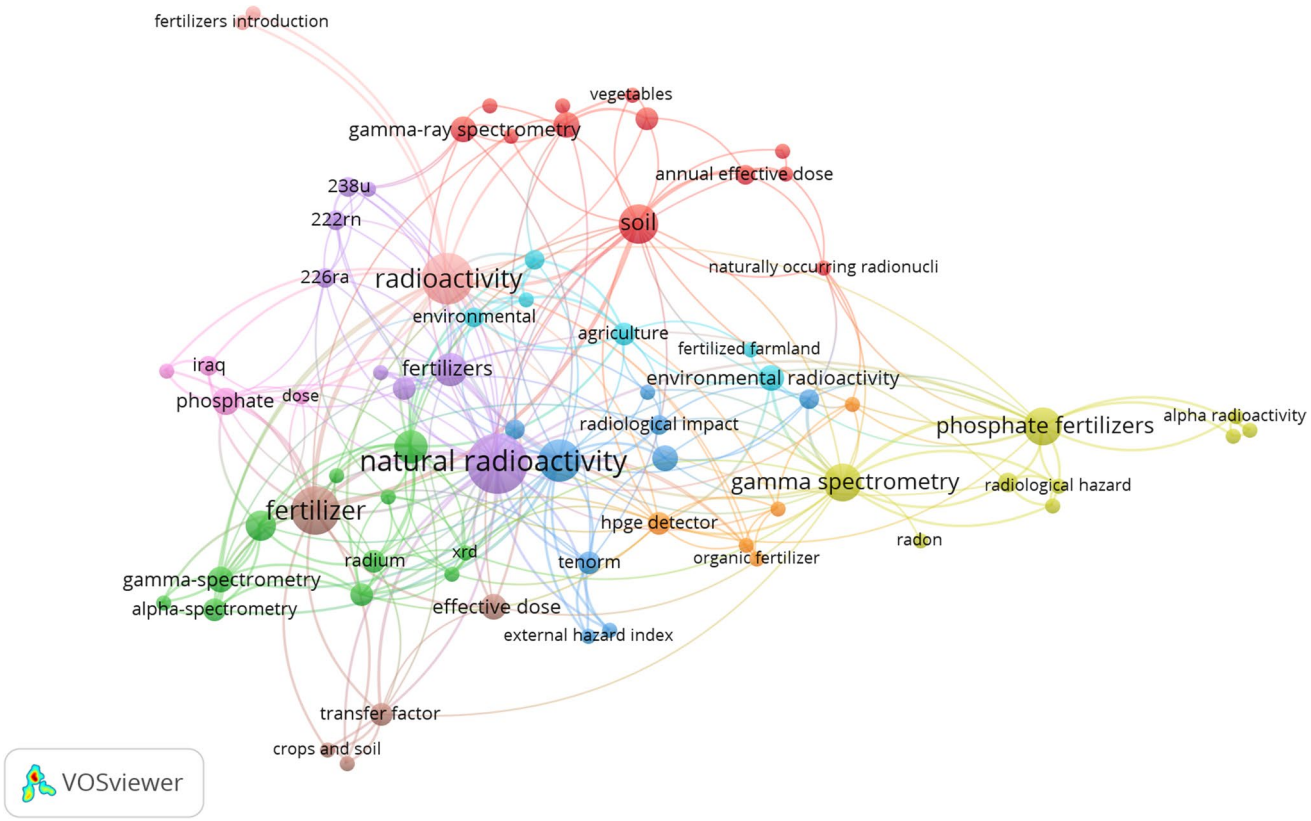

Fig. 2 Cluster graph obtained through bibliometric mapping using VOSviewer and databases for studies involving combinations of keywords related to "natural radioactivity in fertilizers".

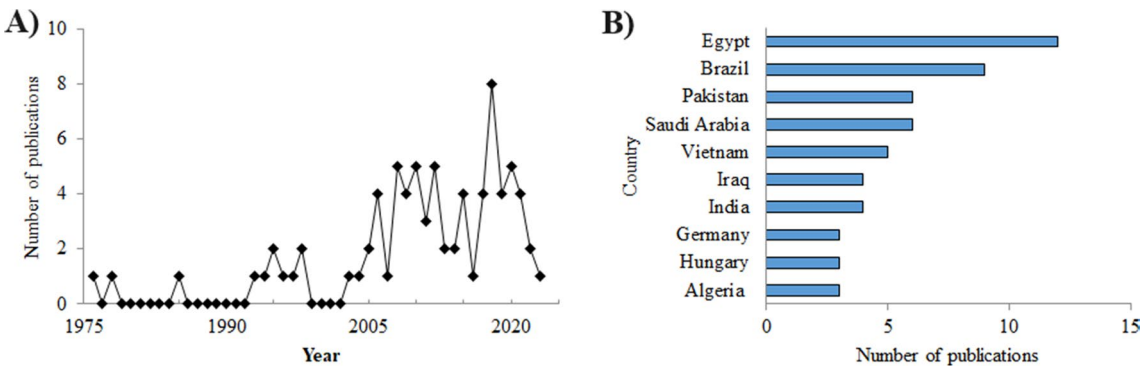

Fig. 3 **A** Temporal variation in the number of publications related to the determination of radioisotopes in fertilizers found in the literature. **B** Number of publications by country in the same time period.

studies, with Egypt (12), Brazil (9), Pakistan (6), Saudi Arabia (6), and Vietnam (5) being the countries with the highest number of papers published in the area, totaling 49% of the scientific production found.

Results and discussions

A total of 310 studies were obtained from the theoretical search carried out in all selected databases using combinations of keywords related to "natural radioactivity in

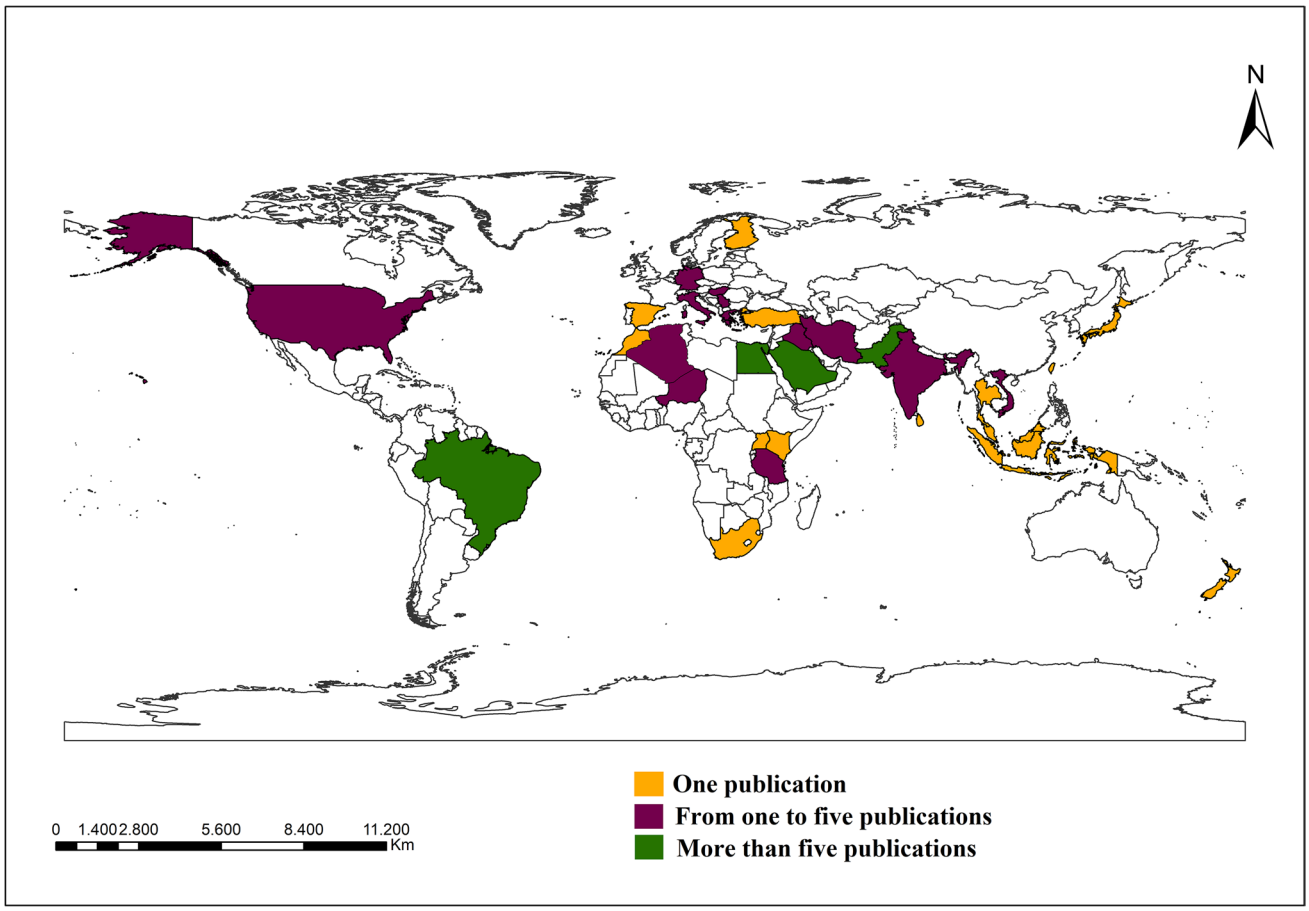

Fig. 4 Countries with at least one publication on the determination of radioactivity in mineral fertilizers published since the 1975s.

fertilizers." After selection, a total of 81 articles were evaluated and the information was then included in the present review.

The most determined element was ^{40}K (78% frequency), followed by ^{226}Ra and ^{232}Th (77%) and ^{238}U (57%). Table 1 describes the values of radionuclide concentrations and radioactivity evaluated in mineral fertilizers of some of the works published in the last 10 years, while Fig. 5 shows the comparison between the activity concentrations in becquerel per kilogram for the different types of fertilizer phosphates found in the literature. The comprehensive table containing the results of the studies can be seen in the Supplementary Material.

Based on Table 1 and the supplementary material, it can be seen that the levels of radionuclides in fertilizers varied widely in different countries, 0.1–19,950 Bq kg^{-1}, 0.1–898.5 Bq kg^{-1}, 0.1–10,000 Bq kg^{-1}, and 0.1–156,066 Bq kg^{-1}, for ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K respectively. In addition, it can be seen that in the single and triple superphosphates, as well as the simple phosphates, the highest concentrations of ^{226}Ra (898.5, 851.0, and 848.3 Bq kg^{-1}) were detected; ^{40}K was found in the highest concentration in triple phosphate fertilizers, 156,066 Bq kg^{-1}; and ^{232}Th and ^{238}U were found in higher concentrations in NPK fertilizers, 10,000 and 19,950 Bq kg^{-1}, respectively.

Radionuclides that occur naturally in the environment affect background levels found in different locations around the world. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has established that the world average concentration values of these radioisotopes are 33 Bq kg^{-1} for ^{238}U, 32 Bq kg^{-1} for ^{226}Ra, 45 Bq kg^{-1} for ^{232}Th, and 412 Bq kg^{-1} for ^{40}K; in addition, it stipulated a world average limit value for an absorbed dose of 59 nGy h^{-1} and a world average effective annual dose of 480 μSv year^{-1} (UNSCEAR 2010). Furthermore, the UNSCEAR 2010 report includes illustrative diagrams of a world map, depicting the average and maximum concentrations of ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K across various countries. These visual representations were created using data from the global survey conducted by UNSCEAR on natural radiation source exposures (UNSCEAR 2010).

Some countries like the USA and China, despite being major world producers of grains, have scarce works that

Table 1 Radioisotopes content (Bq kg^{-1}) present in different types of phosphate fertilizers listed in the literature in the last years.

Country	Fertilizer	^{238}U	^{226}Ra	^{232}Th	^{40}K	Reference
Algeria	NPK		149 ± 6	14 ± 3	3782 ± 250	Boumala et al. 2018
	NPKs		7 ± 1	3 ± 1	3142 ± 222	
	MAP		5 ± 1	15 ± 2	52 ± 11	
	DAP		69 ± 3	8 ± 1	50 ± 7	
	SSP		132 ± 4	12 ± 3	526 ± 39	
	TSP		156 ± 7	16 ± 2	534 ± 43	
Brazil	NPK		3.76–311		1467–3761	Garcêz et al. 2018
	Phosphate		14–22		15.8–53.2	
Egypt	SSP	720 ± 7	1121 ± 10			Salama et al. 2019
Finland	NPK	< 55–610	1.5–110	-	3700–4100	Mustonen 1985
Germany	SSP	749.6 ± 14.7	747.6 ± 7.3	-	-	Römer et al. 2010
	TSP	993.2 ± 8.7	250.1 ± 12.7	20.6 ± 1.8	-	
	DAP	16.2 ± 1.2	4.2 ± 0.3	1.9 ± 0.5	-	
Greece	NPK	< 38–703	< 1–529	< 1–95	238–4483	Servitzoglou et al. 2018
Hungary	Phosphate	1159 ± 10	-	-	65.8 ± 5.0	Srivastava et al. 2017
	NPK	115 ± 1.3	-	-	106 ± 3.6	
India	Phosphate	-	28–374	14.9–29.1	51.1–14,394	Kaliprasad et al. 2021
Indonesia	NPK	311.94 ± 2.50	69.77 ± 0.94	21.60 ± 1.85	82.92 ± 3.27	Hatika and Subketi 2019
	TSP	558.66 ± 5.64	1.14 ± 0.81	4.09 ± 0.72	6418.07 ± 184.22	
Iran	SSP	-	18.83–38.37	17.57–48.84	371.88–652.28	Darabi-Golestan et al. 2019
Iraq	NPK	1.32	-	0.30	188	Kadhim et al. 2021
	SSP	4.15	-	0.25	13.4	
Italy	SSP/TSP	57–1500	34–500	-	-	Ugolini et al. 2020
	MAP/NPK	24–560	1–310	0–9040	-	
Japan	Phosphate	170–750	-	-	37–3400	Michikuni et al. 2008
Malaysia	Phosphate	0.38–112	-	0.81–48	13.4–6003	Ibrahim 1998
Morocco	DAP	2604 ± 162	29.2 ± 5.7	3.2 ± 3.3	63 ± 6	Qamouche et al. 2020
	NPK	826 ± 60	13.4 ± 3.9	0.7 ± 0.7	4710 ± 237	
	TSP	2524 ± 104	650 ± 34	4.0 ± 2.0	22.7 ± 5.6	
	MAP	2478 ± 59	67.7 ± 5.6	1.8 ± 0.6	13.3 ± 5.0	
New Zealand	SSP	-	272	-	-	Pearson et al. 2019
	TSP	-	851	-	-	
	DAP	-	123	-	-	
	NPK	-	3–77	-	-	
	NPK	115 ± 1.3	-	-	106 ± 3.6	
Nigeria	NPK	-	8.6–49.3	71.4–1052.3	225.7–10,051.5	Faweya et al. 2018
	SSP	-	41.5 ± 3.0	506.9 ± 70.5	10,468.8 ± 35.7	
Pakistan	Phosphate	-	307.7–617.5	-	-	Muhammad et al. 2019
	TSP	700 ± 21	-	-	-	
Saudi Arabia	DAP	< 17–498	8.6–87.4	-	856–3686	Alshahri and Alqahtani 2015
	NPK	< 14–1062	< 1.6–35.8	-	1276–11,654	
	SSP	585–1084	7.5–392	-	3405–5029	
	TSP	3299–3900	140–195	-	38.3–67.1	
Serbia	NPK	6–560	1.7–220	2.2–52	2100–11,000	Kuzmanović et al. 2022
South Africa	SSP	85–98	55–90	316–327	-	Louw 2020
	MAP	215–202	36-60	343–410	-	
Spain	DAP	2500	50	-	-	Bolívar et al. 1995
Sri Lanka	TSP		17–72	10–11	103–15,066	Chandrajith et al. 2010
Taiwan	Phosphate	144.3–473.6	25.9–499.5	25.9–395.9	288.6–5801.6	Wu et al. 1978

Table 1 (continued)

Country	Fertilizer	^{238}U	^{226}Ra	^{232}Th	^{40}K	Reference
Tanzania	SSP	3596–3879	-	408–434	-	Makweba and Holm 1993
	TSP	6819–7024	-	632–698	-	
Thailand	SSP	-	16.54–25.72	-	4.02–7.31	Porntepkasemsan et al. 2018
USA	NPK	-	5.1–13.14	0.5–8.6	3024–4092	Billa et al. 2015
Vietnam	DAP	-	18.5 ± 1.6	-	-	Nguyen et al. 2021
	NPK	-	7.6 ± 0.8	-	-	
	SSP	-	85.1 ± 8.3	-	-	

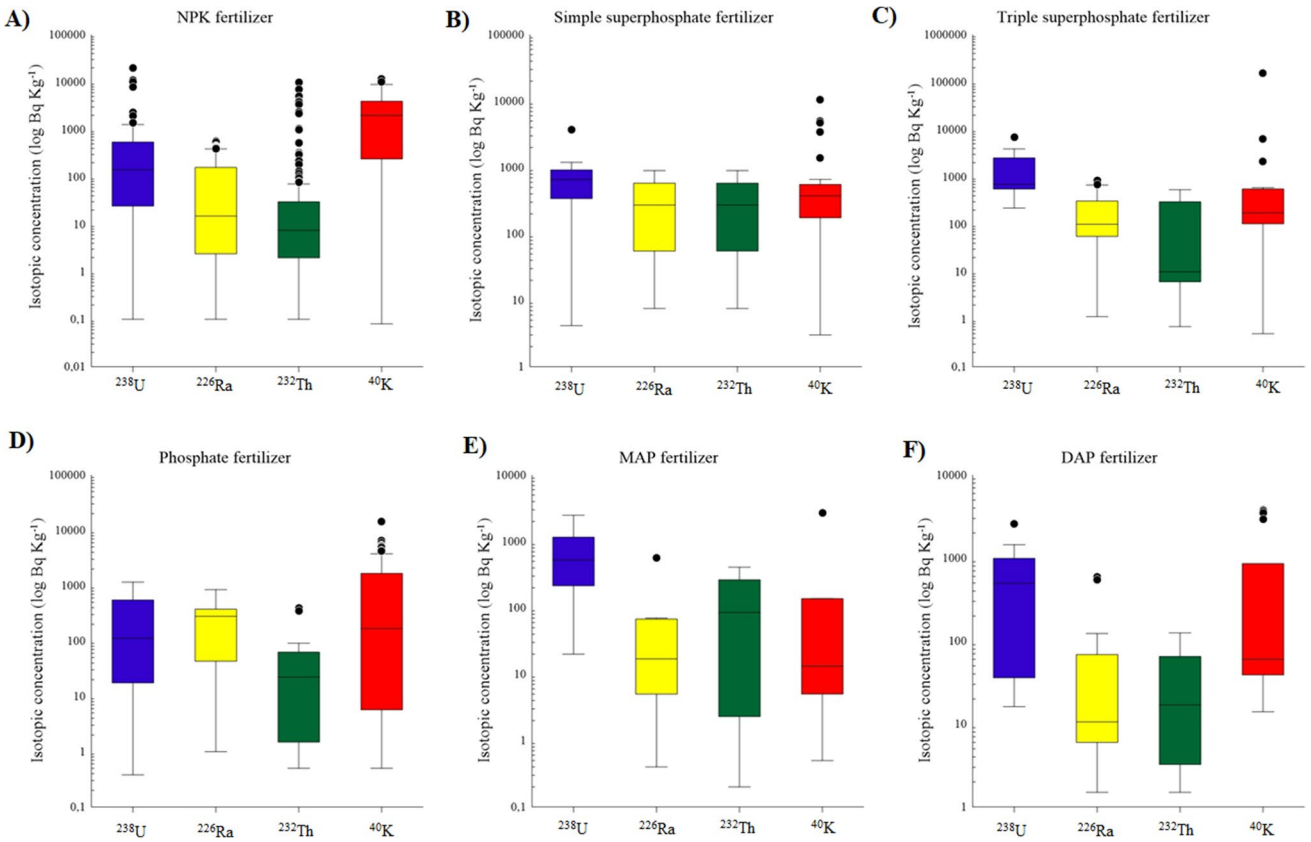

Fig. 5 Variation in the concentration of radium, uranium, thorium, and potassium isotopes present in the different types of mineral fertilizers found in the literature (in Bq kg^{-1}). **A** NPK fertilizers; **B** single superphosphate fertilizers (SSP); **C** triple superphosphate fertilizers (TSP); **D** phosphate fertilizers; **E** monoammonium fertilizers (MAP); **F** diammonium fertilizer (DAP).

address the determination of radioactive elements in their agricultural inputs. The USA, for example, consumed about 5.8 million metric tons of phosphate-based fertilizers between the 1970s and 2007 and then increased to more than 8.5 million metric tons in 2007 (EPA 2022). The use of radioactive materials such as phosphogypsum in this country is even well regulated; however, there is no restriction on the application of fertilizers on agricultural land and farmers are not required to keep any certificates or application records related to the use of fertilizers (EPA 2022).

In general, the amount of radioactive materials added to arable land in the form of fertilizers is not significant. However, the application of these inputs over the years tends to accumulate concentrations of radioactivity in surface soils, resulting in potential health problems when living organisms are exposed (ATSDR 1990). In addition, vegetation has a tendency to absorb nutrients from soils and the presence of isotopes in soils can result in the transfer of isotopes from soils to plants (Billa et al. 2015).

The use of phosphate fertilizers for a prolonged period of time, to recover the land and improve the productivity of the crops, may increase the content of natural radioactive isotopes in the soil, increasing the exposure to ionizing radiation of the population and consequently resulting in an increase in the annual dose of radiation and which can lead to the development of diseases (Righi et al. 2005; UNSCEAR 2010; El-Farrash et al. 2012). According with Drichko (1983), such an increase can exceed the background concentration by 1.5 to 2.0 times. As such, it is important to understand the amount of radionuclides introduced with fertilizers into agricultural land so that regulatory authorities can make some recommendations on fertilizer use.

This fact is relevant mainly since the potassium is more or less uniformly distributed in the body following the intake of foods, and its concentration in the body is under homeostatic control. For adults, the body content of ^{40}K is about 0.18%, and for children, about 0.2%, which corresponds to an annual equivalent dose in tissues in the body to 165 and 185 μSv year^{-1} for adults and children, respectively (UNSCEAR 2000). However, with the enrichment of the soil with radioisotopes and their possible larger entry into the food chain, it may lead to a higher exposure of humans to radiation and no longer maintaining an average of a total effective dose from inhalation and ingestion of terrestrial radionuclides of 310 μSv, of which 170 μSv is from ^{40}K and 140 μSv is from the long-lived radionuclides in the uranium and thorium series (UNSCEAR 2000).

The effective dose is used for regulatory purposes worldwide. In practical radiological protection applications, the effective dose is used to manage the risks of stochastic effects on workers and the population (ICRP 2007). For public exposure, UNSCEAR recommends that the limit be expressed as an effective dose of 1 mSv per year. However, under special circumstances, a higher effective dose value may be permitted in a single year, provided that the average over defined 5-year periods does not exceed 1 mSv per year. To estimate the annual effective doses, one must take into account the conversion factor from absorbed doses in the air to effective dose and the external occupancy factor, as shown in Eq. 1 below (Todorović et al. 2015):

$$\text{Effective annual dose } \left(\text{mSv y}^{-1}\right) = \text{absorbed dose } \left(\text{nGy h}^{-1}\right) \times 8760\ \text{h} \times 0.2 \times 0.7\ \left(\text{Sv Gy}^{-1}\right) \quad (1)$$

The main radionuclides of natural origin that are important for the purposes of radiological protection are the decay series of ^{238}U, ^{232}Th, and ^{40}K. The activity concentrations of these radionuclides in natural materials, such as rocks and minerals, are variable. An important step, both for the industry and for the regulatory agencies of a country, is to detect when and where a naturally occurring radioactive material (NORM) material can occur in a process, to identify the places where the highest concentrations of this material are found within a given process and what the concentration of a NORM material capable of producing a potential radiological risk (Mazzili et al., 2016). According to the International Atomic Energy Agency, when the activity concentrations of all radionuclides in the ^{238}U and ^{232}Th decay series are less than or equal to 1 Bq g^{-1} and the activity concentration of ^{40}K is less than or equal to 10 Bq g^{-1}, no waste treatment is required (IAEA, 2013).

Although there is no specific legislation regulating the levels of radioisotopes in fertilizers, they are regulated in soil and water. The WHO recommends concentration values of less than 12.4 mBq L^{-1} of ^{238}U (equivalent to 1.0 μg L^{-1}) of uranium in water (WHO 2012; UNSCEAR 2017), but there are no guidelines regarding their presence in soils. However, the Canadian Council of Ministers of the Environment (CCME) provides soil quality guidelines for uranium for the protection of the environment and human health, recommended threshold values of 23 mg kg^{-1} soil for agricultural and residential use, 33 mg kg^{-1} of soil for commercial use, and 300 mg kg^{-1} of soil for industrial use (CCME 2007).

The US Environmental Protection Agency (EPA) has established a potable water limit of 0.185 Bq L^{-1} for ^{226}Ra and ^{228}Ra (combined). In addition, the EPA has established a soil concentration limit for ^{226}Ra in uranium and thorium tailings of 0.185 Bq g^{-1} in the top 15 cm of soil and 0.555 Bq g^{-1} in deeper soil (ATSDR 1999).

In Brazil, NORM is typically associated with non-nuclear industries, i.e., with conventional ore extraction and processing industries. From a regulatory point of view, uranium mining and processing constitute a monopoly of the union and, therefore, follow the legislation of the nuclear fuel cycle, not applicable to NORM. For the disposal of large amounts of materials with the presence of natural radionuclides, the activity concentration limits correspond to 10 Bq g^{-1} for ^{40}K and 1 Bq g^{-1} for each radionuclide in the radioactive decay chain of uranium or thorium, to meet the dose criterion of the order of 1 mSv year^{-1}, which is compatible with typical doses due to background radiation (Mazzili et al. 2016).

In the European Union, Regulation (EU) 2019/1009 of the European Parliament and of the Council of 2019 establishes the rules for making fertilizer products available in the European Union market. It is foreseen that by 2026, the impacts of the implementation of restrictions on the levels of contaminants should have been evaluated, in which until now the legislation includes elements such as cadmium, chromium, mercury, nickel, lead, arsenic, biuret, and perchlorate, in addition to copper and zinc, and evaluated any recent and relevant scientific information, with regard to the toxicity and carcinogenicity of the contaminants, including the risk arising from uranium contamination of fertilizer products (EUR-Lex 2019).

In countries like Greece, for example, regulations for the management of radioactive waste, in particular naturally occurring radioactive materials, are in line with European Union directives. There are currently no specific waste management regulations regarding NORM waste. These materials, such as phosphogypsum, resulting from waste from the phosphate fertilizer industry, are also included in the general regulatory framework for waste management (IAEA 2006).

In the case of NORM waste management, the basic reference proposed for the general population is an annual dose limit of 300 μSv. In the case, for example, of the reuse of phosphogypsum for agricultural purposes, a maximum concentration of 400 Bq kg^{-1} was established, a value calculated based on a specific situation of exposure to radiation through the consumption of rice produced in soil enriched with phosphogypsum, in which the dose limit would be 10 μSv $year^{-1}$ (IAEA 2006).

Austria, another member country of the European Union, adapted the European radiation protection legislation and included fertilizers as products that must be monitored by government authorities regarding their concentrations of natural radionuclide activity (Dauke et al. 2016). They listed animal feed and fertilizers as products that, due to their radioactivity content, must be inspected by the authorities. Dauke and collaborators in their studies concluded that, even with conservative scenarios, there would not be a great risk in relation to NORM nuclides, despite observing that the storage of large amounts of potassium fertilizers can lead to exposure to levels greater than 1 mSv $year^{-1}$. However, they emphasized that phosphate fertilization is the main source of the distribution of uranium and its decay products in the environment over large areas in Europe (Dauke et al. 2016).

Another example of NORM waste management is the USA where phosphate rock mining is the fifth largest mining industry in the USA, whereas in 2019, total production was estimated at 23 million metric tons, with the majority of phosphate production going to fertilizer manufacturing. Although the use of radioactive materials is well regulated in the USA and fertilizers contain traces of radioactive isotopes such as ^{226}Ra, there is no restriction on the application of fertilizers to agricultural land (Billa et al. 2015; EPA 2022). It is only defined that the activity of the phosphogypsum used for agricultural purposes cannot exceed 0.37 Bq g^{-1}. Similarly, technologically concentrated naturally occurring radioactive material (TENORM) waste and consumer products are regulated in the USA by federal or state government agencies, but there is no single national law that regulates it. Most agencies apply general radiation protection and waste disposal standards designed for all radiation sources to TENORM (EPA 2022; IAEA 2006).

In general, most of the works covered in this review focused on the determination of radioisotope concentration activity in different types of fertilizers, both locally produced and imported. However, about 35% of these presented data on the effective annual dose, the absorbed dose, and the radiological risks of radium equivalent activity due to the presence of these radionuclides in these agricultural inputs, comparing them with current legislation and with other available results in literature.

The annual effective dose received due to the application of all types of fertilizers studied in the environment varied between 1.1×10^{-5} and 17.3 mSv $year^{-1}$, with an average of 2.4 mSv $year^{-1}$, the highest being contribution due to NPK fertilizers, whose observed average was 3.8 mSv $year^{-1}$. Furthermore, Van et al. (2018), Loan et al. (2021), and Salama et al. (2019) determined the absorbed dose due to the ingestion of food grown with the use of different mineral fertilizers and found a variation between 0.1 and 21.5 mSv $year^{-1}$ and an average of 3 .8 mSv $year^{-1}$. It is important to emphasize that Van et al. (2018) observed that food produced with the application of NPK fertilizers may contribute more to human exposure to radioactivity compared to other types of fertilizers.

In addition, studies developed, for example, by Ahmad (2007), Da Conceição and Bonotto (2009), and Tufail et al. (2010) explored the increase in the activity concentration of some isotopes in the soil due to the use of fertilizers and determined the transfer factors of primordial radionuclides from the soil to cultivated grains, an approach that enriched the discussion on the problem of the presence of radioactive elements in these inputs.

Ahmad (2007) investigated the impact of fertilizer application on background radioactivity level in two newly developed desert areas in Egypt. Their data indicated that the concentrations of ^{238}U, ^{232}Th, and ^{40}K in local fertilizers are comparatively high and pose a considerable risk to humans, ranging from 1.27 ± 0.12 to 950.09 ± 8.63 Bq kg^{-1} of ^{238}U, 0.68 ± 0.06 to 162.16 ± 11.06 Bq kg^{-1} of ^{232}Th, and 10.22 ± 1.02 to 23854.24 ± 11.93 Bq kg^{-1} of ^{40}K. Since the superphosphate fertilizer produced in Abu Zaabal and Kafr El-Zayat (EG) showed activity levels higher than the internationally allowed levels, which may cause concerns about the health of farmers, people who handle the material and the public that consumes food. The soil-plant transfer factors varied widely for the studied isotopes ^{238}U (0.35–1.82), ^{232}Th (0.23–0.48), and ^{40}K (1.95–31.85), showing that this parameter is dependent on a number of factors, such as the composition of plant species, soil type and mobility of radionuclides in it, climate, and even the depth of the roots.

Da Conceição and Bonotto (2009) concluded that if it is assumed that radionuclides are homogeneously distributed in the top 10 cm of soil, the results indicate a maximum increase of about 0.14, 0.16, and 6.24 Bq kg^{-1} of soil for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. Thus, the maximum additional exposure to radiation caused by phosphate fertilizers used in their study in a sugarcane crop

would correspond to 0.42 nGy h^{-1} at 1 m above soil level, a value well below the average outdoor world exposure, determined to be 51 nGy h^{-1} by UNSCEAR (2000).

Tufail et al. (2010) observed that the activity concentrations of ^{40}K and ^{232}Th in the SSP fertilizer are lower than those measured in the soil of the farm in Faisalabad, Pakistan, where the study was carried out, while those of ^{226}Ra are much higher than those of the soil, about 19 times larger. Thus, the application of this fertilizer in arable areas can be a source of radioactivity due to ^{226}Ra, although its contribution was small. They also evaluated the effective annual dose due to wheat consumption, 216.9 μSv $year^{-1}$, concluding that the consumption of these foods can be considered safe from the point of view of radiological protection. Regarding the transfer factor from the soil to the grains, they obtained an average of 0.20, 0.03, and 0.04 for ^{40}K, ^{232}Th, and ^{226}Ra respectively.

Final considerations

This review sought to survey all the information published in scientific journals on the content of radioisotopes present in different types of mineral phosphate fertilizers around the world in the last 50 years. It was seen that there are few studies focused on the determination of environmental radioactivity in phosphate mineral fertilizers, with Egypt, Brazil, and Pakistan being the countries with the highest number of bibliographic productions, mainly in relation to the countries with the highest production and use of these agricultural inputs, which this is the case of the USA, China, Canada, Morocco, and Saudi Arabia.

The distinct characteristics of each fertilizer category were observed, among them single superphosphate, triple superphosphate, monoammonium phosphate, diammonium phosphate, and NPK, and their different concentrations of ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K and the possible radiological impacts of these levels of radioactivity were investigated.

Isotopic concentrations varied in several orders of magnitude in all types of fertilizers studied, 0.1–19,950 Bq kg^{-1}, 0.1–898.5 Bq kg^{-1}, 0.1–10,000 Bq kg^{-1}, and 0.1–156,066 Bq kg^{-1}, for ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K respectively, showing that the radioisotope contents can vary according to the region where the rocks are produced or extracted.

Based on this study, the importance of regulating these agricultural inputs is noted since they can become, over time, a source of radioisotope contamination and contribute to public exposure to ionizing radiation.

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Data availability All data included in this review paper are available in the literature. The software used in the analysis of the results is either open source or duly licensed.

Declarations

Ethical approval This review article does not involve research involving animal or human participants; therefore, an ethics approval is not applicable. The present work seeks to survey the existing literature on the presence of natural radioactive isotopes in mineral phosphate fertilizers and, based on these results, estimates the possible impacts on human health through calculations of the effective annual dose.

Consent to participate Not applicable.

Consent for publication All authors agreed with the content and all gave explicit consent for the submission of this work in the journal. In addition, they obtained the consent of the responsible authorities at the institute/organization where the work was carried out, before submitting the work.

Competing interests The authors declare no competing interests.

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