

2024-01-09

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<https://doi.org/10.3390/su16041496>

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Review

Uranium Dissemination with Phosphate Fertilizers Globally: A Systematic Review with Focus on East Africa

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Abstract: Growing concern has been expressed about uranium (U) accumulation in agricultural soils caused by the long-term application of mineral fertilizers. More than 80% of naturally occurring U transfers from phosphate rock (PR), the raw material used in mineral fertilizer production, to phosphorus (P) fertilizers. These fertilizers are then distributed on agricultural soils, where the U could accumulate over time and become a risk to the environment. The objective of this work was to review the reported content of U in P fertilizers, its potential dispersion in soils, and its uptake by plants in different countries in the world as reported in the literature. The articles for this systematic review were selected from the Scopus database published between 2003 and 2022. The preferred reporting items for systematic reviews and meta-analyses (PRISMA) protocol were used. A total of 54 articles were assessed based on the standard inclusion and exclusion criteria. U concentrations in P fertilizers, agricultural soil dissemination, and plant uptake for available data were obtained and assessed. In order to compare a set of related data from the collected articles, box and whisker plots showing the distribution of U in P fertilizers are presented by region. The results from the reviewed articles show that the U concentrations in P fertilizer were in the range of 0.1–653 mg kg^{−1}. Interestingly, Minjingu P fertilizers from Tanzania, which are used in six East African countries, showed the highest U concentrations (159 to 653 mg kg^{−1}, average 390 mg kg^{−1}). The reported U concentrations for these fertilizers are, in fact, comparable to those of conventional low-grade uranium deposits mined in Namibia and elsewhere. Additionally, approximately 96% of the reviewed articles indicate that fertilized soil has higher U concentrations than non-fertilized soils, hinting at a measurable effect of mineral fertilizer use. The review recommends U extraction during mineral fertilizer production so that potential environmental risks can be reduced and U resources that would otherwise be lost can be recovered and used to substitute conventional U mining elsewhere.

Keywords: mineral fertilizers; uranium; environmental pollution; cleaner production



Citation: Mwalongo, D.A.; Haneklaus, N.H.; Lisuma, J.B.; Mpumi, N.; Amasi, A.I.; Mwimanzi, J.M.; Chuma, F.M.; Kivevele, T.T.; Mtei, K.M. Uranium Dissemination with Phosphate Fertilizers Globally: A Systematic Review with Focus on East Africa. *Sustainability* **2024**, *16*, 1496. <https://doi.org/10.3390/su16041496>

Academic Editor: Sean Clark

Received: 22 November 2023

Revised: 12 January 2024

Accepted: 22 January 2024

Published: 9 February 2024



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1. Introduction

1.1. Background Information

Natural uranium (U) is an impurity in phosphate rock (PR), the ore used for mineral fertilizer production. Phosphorus (P) fertilizers are the second most often used fertilizers in the world after nitrogen fertilizers [1]. During P fertilizer production, it is estimated that over 80% of the U contained in the PR ends up in the fertilizers [2–5], with the rest transferring to phosphogypsum tailings [6,7]. Most P fertilizers contain at least low U concentrations. Thus, long-term applications of P fertilizers can increase U loads in

agricultural soils. The concentration of U in P fertilizers depends on the type and origin of the PR used in fertilizer production and, to a lesser degree, the production process that decides how much of this U transfers to the final fertilizer product [8]. In addition, impurities in the used industrial sulfuric acid can introduce additional impurities. For instance, according to Kouzbou et al. [9], the wet phosphoric acid (WPA) process that is used in the production of P fertilizers frequently results in a product which contains more than 80% of the U impurities found in the PR [10]. In sedimentary PR, for instance, U concentrations are in the range of 80–200 mg kg^{−1} [11]. Sedimentary PR accounts for approximately 80% of the PR mined globally, while in igneous PR, the U concentrations are typically much lower (around 30 mg kg^{−1}) [11–14]. The application of P fertilizers can disseminate considerable amounts of U on agricultural soils that may leach from the soil to ground and surface waters [11,15].

The applications of P fertilizers have been reported to enhance U concentrations in agricultural soils on different continents around the world. For instance, in Asia [15–26], similar in Europe [16–20], in Australia [21,22], in America [23–26], and in Africa [27–31]. This study revealed that from all reviewed articles, the concentration of U from Minjingu P fertilizers from Tanzania ranged from 200 to 600 mg kg^{−1}, which is similar to what is found in open-pit commercial uranium mines in Namibia. Minjingu P fertilizers have been reported to be used in neighboring East African countries such as Kenya, Uganda, the Democratic Republic of Congo (DRC), Burundi, Rwanda, and Zambia, so this issue can hardly be described as a local phenomenon, but does indeed affect millions [32–35].

P fertilizer application is regarded as a major cause of U dissemination in agricultural soils [36]. It is no secret that radiotoxic U is hazardous to human health and the environment [17,37–39]. For instance, Yamaguchi et al. [40] and Yamazaki and Geraldo [41] reported that agricultural soils with a long history of P fertilizer application showed elevated U concentrations compared with unfertilized soils. P fertilizers with high U concentrations are prone to soil contamination and may influence soil-to-plant uptake [42,43]. Furthermore, P fertilizers have also been identified as a major risk for U contamination in drinking water [36,44,45].

Minjingu PR is reported to be suitable for direct application after simple beneficiation and granulation. The produced P fertilizers from the Minjingu mine and processing plant react with acidic, P-deficient soils in humid to sub-humid regions as they are found in various East African countries. According to Banzi et al. [46], Minjingu PR has an average U concentration of 390 mg kg^{−1}, which is significantly higher than that found in other PRs. Minjingu PR is nonetheless used to produce different types of fertilizer products (i.e., Fertilizer Powder, Fertilizer Granules, Nafaka Plus, Golden Leaf Tobacco, and Top Dressing), which are marketed and sold in the East African region [47].

High U concentrations in agricultural soils have also been reported in other regions of the African continent [13,27,48–52]. High U concentrations in P fertilizers increase the chances of plant uptake of this radiotoxic element.

The objective of the following work is to present a comprehensive overview of current knowledge regarding the U concentration in different P fertilizers around the world and its distribution in agricultural soils and plant U uptake. Additionally, we performed a comparison of the U concentrations from other countries or continents with that of the Minjingu material to demonstrate the elevated U concentrations present here. Based on the findings, the paper concludes by providing a sustainable policy recommendation.

1.2. Rationale of the Review

P fertilizers are a source of elemental phosphorus (P), which is one of three essential nutrients, together with nitrogen (N) and potassium (K), that plants require in large amounts for reproduction and growth. It is crucial since it is required by plants, and its deficiencies impede plant development and yields. World population growth has intensified global initiatives to increase the use of P fertilizers in the agriculture sector to increase food production [53].

U contained in P fertilizers is preferentially transferred to mineral fertilizers during fertilizer production using the commonly used WPA process [13]. As a result, U persists in all P-derived fertilizers used on agricultural land. The presence of U in P fertilizers, as well as their continuing applications in agricultural soils, causes U dissemination in agricultural soils [37,54,55].

There is no global consensus on the regulatory limit for U in P fertilizers today. A growing number of studies have confirmed global U buildup in agricultural soils and contamination of ground and surface waters as a result of prolonged P fertilizer applications [56–58]. Depending on the origin of PR, U concentrations are usually in the order of 80–200 mg kg^{−1} for sedimentary PR, which account for approximately 80% of the PR mined today, and much lower (30 mg kg^{−1}) for PRs of igneous origin, as they are mainly found in Russia and South Africa. U concentrations in PRs in East Africa are known to show higher U concentrations than the world average, which can be in the range of 200–500 mg kg^{−1} [11,59]. Although U has proven to be an environmental and health concern, there are no legal limits to the level of U in fertilizers today [56,57]. Therefore, this review aims to examine the extent of U concentration in P fertilizers worldwide and investigate reported U accumulation in agricultural soil as the result of P fertilizer application.

2. Data and Methodology

In this review, we followed the standard Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) template to gather all required information. The approach is built on six core systematic literature review procedures to eliminate bias and improve information repeatability and reliability [60] as follows:

- Develop research questions;
- Establish search keywords using Boolean operators;
- Choose a reliable search database;
- Limit the article's publication time interval;
- Import the article abstract to Excel and select the relevant abstract;
- Extract, group, and analyze the data.

2.1. Countries Included in the Reviewed Study

We reviewed studies conducted in forty-nine (49) countries on U in P fertilizers and its reported impact on agricultural soil. The countries include 27 countries of the European Union (EU) and United Kingdom, 6 East African countries (DRC, Tanzania, Kenya, Uganda, Burundi, and Rwanda), and 16 other countries from North Africa (Algeria and Egypt), Oceania (Australia and New Zealand), Asia (Japan, Kazakhstan, Serbia, Thailand, Vietnam, Saudi Arabia, Pakistan, India, and Bangladesh), and the Americas (Mexico and Brazil). The list of countries is presented in Figure 1.



Figure 1. Countries/regions (orange) from where articles on U dissemination in agricultural soils as a result of the application of P-derived fertilizers were considered in this review.

2.2. Literature Sources and Search Strategy

The research question used in this systematic review was, “To what extent does prolonged P fertilizer use cause U accumulation in agricultural soils?”. From the research question, keywords were generated and connected using Boolean operators. Suitable keywords based on the research question were used. The search string used in the Scopus database was {Uranium} OR {radioactivity} AND {phosphate fertilizer} AND {Soil} OR {Arable land}. The Scopus database was used in July 2023 to search for relevant articles. The article’s time interval was set at ten years (2003–2022).

We assessed relevant articles on the accumulation of U or radioactivity in agricultural soils around the world as the result of the application of P fertilizers. A total of 89 articles were collected from the Scopus database based on a search strategy developed from the research question and then screened based on preset criteria. Fifty-four ($n = 54$) articles were exported to Microsoft Excel in a comma-separated values (CSV) file. Descriptive data analysis was performed based on the year of publication, type of journal, article contribution of the reviewed subject by country, citation, etc., as indicated in the following analysis. A total of 54 articles were considered for the study, and data were extracted and used in the current study. The PRISMA flow plan of the search strategy and selection criteria for articles is depicted in Figure 2.

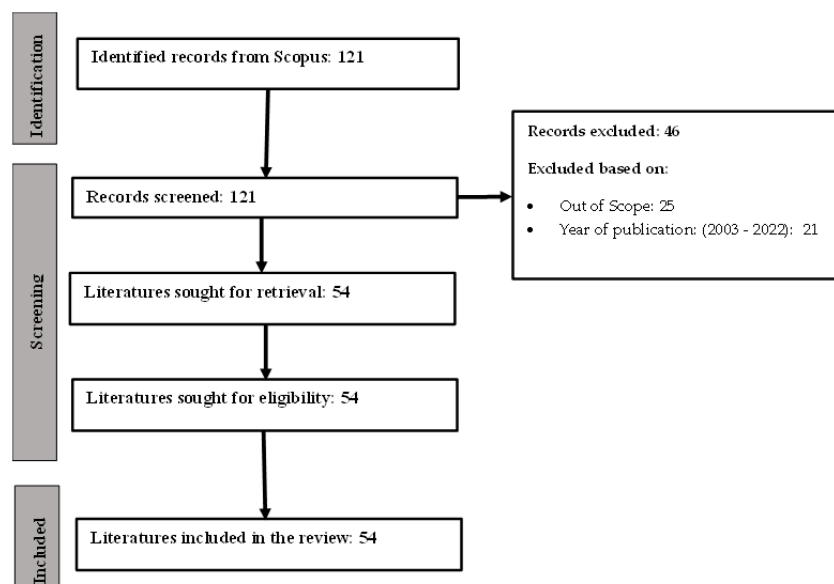


Figure 2. The PRISMA flow sheet of the search and selection of the articles.

Only peer-reviewed articles published in English were considered in this study. The articles were exported to an Excel database, and a review of the abstract was carried out to determine if it fell within the scope of the research question. Finally, article data were extracted and descriptively analyzed.

Essentially, this review addressed the extent of U concentrations in agricultural soils fertilized by mineral P fertilizers versus unfertilized soils. The map (Figure 1) indicates the distribution of articles by country/region. Based on the aim of the review, we performed a thorough article search for availability and accessibility using the Scopus database and Google Scholar. Inclusion and exclusion criteria were also chosen based on the standard PRISMA template, as described in Figure 2. The review answers research on the extent of U accumulation of agricultural soils after the application of P fertilizers.

The summary of the articles’ relevance-based quality evaluation, exclusion, and inclusion criteria is summarized in Figure 1. Based on the criteria for selection, a total of 54 articles were examined and accepted for this review. The scope of the reviewed articles was evaluated by discussing several perspectives on the recovery of U as a soil contaminant

and mineral resources lost during fertilizer application. The outlook for further studies and policy recommendations are also concluded in this review.

2.3. Inclusion and Exclusion Criteria

The Scopus database search led to the discovery of 121 articles. During the first filtering process, the number of possibly relevant publications was reduced to 75. The second filtering process eliminated an additional 21 items that were not relevant. Therefore, 54 papers were used for this review. The inclusion and exclusion criteria used were:

- The full text was available for review;
- Scrutinized uranium or radioactivity in phosphate fertilizers in relation to arable or agricultural soils;
- The document type must be a peer referred or reviewed article;
- The year of publication was restricted to twenty years (2003–2022);
- The subject area was not restricted;
- The language was restricted to English.

2.4. Data Collection Process

The articles were carefully selected based on the inclusion and exclusion criteria listed; further evaluation was based on the abstract. The chosen articles were independently reviewed by separate reviewers, and data were collected and recorded in the data collection form with the following items;

- The names of the authors and the year of study;
- The country in which the study was carried out;
- Article publication year;
- Sampling environment and study areas;
- Measurement and analytical techniques used;
- Uranium activity concentration (Bq kg^{-1}) or uranium concentration in milligrams per kilogram (mg kg^{-1}) or parts per million (ppm);
- Quantitative reported data (mean, standard deviation, sample size, and uncertainties) from review articles were used. For the sake of uniformity and easy comparisons of various quantitative data, all measurements were converted to ppm (mg kg^{-1}) using the relation: $1 \text{ mg kg}^{-1} \text{ }^{238}\text{U}$ in soil $\sim 12.4 \text{ Bq kg}^{-1} \text{ }^{238}\text{U}$ [61] and the relationship between the activity concentrations of ^{238}U (Bq kg^{-1}) and the concentrations of ^{238}U (mg kg^{-1}): $1 \text{ Bq kg}^{-1} = 8.1 \times 10^{-3} \text{ mg kg}^{-1}$ [62].

3. Results and Discussion

3.1. The Focus of the Review

The focus of this review is based on the effect of the dissemination of U with P fertilizers on agricultural soils. It is historically known that P fertilizers are the most significant source of P, which is a critical macronutrient for soil fertility and crop production. U is present as an impurity in PR, which is the raw material used for P fertilizer production. U contained in P fertilizers is an environmental contaminant, yet it is not recovered during P fertilizer production at the moment [63]. U in P fertilizers is disseminated on agricultural soil with fertilizers leading to potential accumulation of U in the soil. U loads in agricultural soil may cause plant uptake from the soil so that this element could, in trace concentrations, even enter the food chain.

The availability and uptake of non-essential substances like U are influenced by the availability and uptake of important macro and micronutrients like nitrogen (N), phosphorous (P), potassium (K), and zinc (Zn), among others, by plants [64]. The uptake of U by plants may pose both chemical and radiological damage to human and animal tissues [65,66]. It is imperative to review the current state of U in P fertilizers, its dissemination in agricultural soils, its potential uptake by plants, and its possible entry into the food chain.

This is very important, particularly in areas with the highest average U concentration in P fertilizers, such as East Africa.

3.2. Geographical Distribution of U in P Fertilizers According to Research Studies

The articles that represent the geographical distribution (country/region) of U in agricultural soil as the result of the application of P fertilizers around the world are illustrated in Figure 1. It should be noted that the articles considered in this review representing reporting studies in the highlighted countries do not necessarily report results for the whole country/region. Figure 1 illustrates the general geographical distribution of the literature used in this review.

3.3. Descriptive Statistics of the Selected Articles

The distribution of articles per year relevant to the reviewed topic during the last 20 years is displayed in Figure 3. According to descriptive statistical inferences, the number of articles published annually during 2003–2022 varied from 1 to 10 (average 2.7). The gradual change in the number of articles per year may be attributed to a lack of interest in or scientific relevance of the subject due to the lower U concentration in P fertilizers used in agricultural soils in many countries. Additionally, in 2004, no relevant article was published.

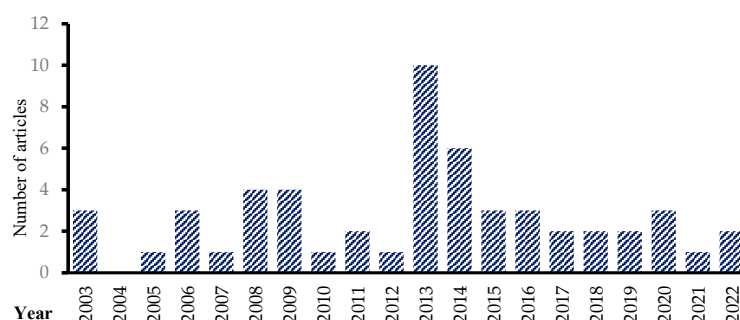


Figure 3. Articles published during 2002–2022 on U contamination in agricultural soils.

From the reviewed articles, we examined the trend of individual country–region article contributions (Figure 4). The region with the most significant article contributions was Europe, which is not surprising given that the EU is actively discussing the implementation of legal U limits in fertilizers. In Europe, Germany had the highest number of articles (10), followed by Switzerland (3) and Turkey (3). Additionally, two studies were carried out for all countries in the EU. Indian researchers published the second-highest number of articles (10) after the EU [16,67].

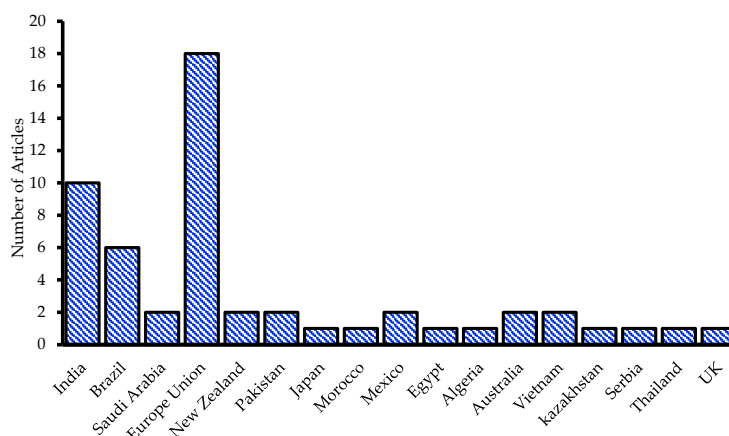


Figure 4. Number of articles published on U in agricultural soils in research in the 10 most featured countries and regions.

3.4. U in P Fertilizers and Agricultural Soils

In this review, we extensively examined reported U concentrations in P fertilizers used in arable soils in different countries around the world in comparison to the countries of East Africa. The reviewed literature involved short and long-term field experiments where fertilized and unfertilized (control or forest) agricultural fields were compared. A comparison between U concentration in fertilized and unfertilized soils was made to determine if there was a statistically significant difference. Out of the 54 selected articles, 10 data points from 40 countries (27 EU, 5 East Africa, and 8 others) were plotted in box and whisker plots for easy data visualization (Figure 5), while the remaining articles' data were presented using range and central tendency (mean, median, maximum, and minimum). A discussion was conducted concerning different continents. Almost all articles (52 out of 54 or 96%) reported that the application of P fertilizers significantly ($p < 0.05$) increases U concentrations presented as specific activity (Bq kg^{-1}) or mass fraction (mg kg^{-1} or parts per million, ppm). However, two articles (approximately 4%) found a nonlinear relationship between U in P fertilizers and U enrichment in agricultural soils of Germany, Denmark, and Greece [68,69]. The results show that different countries have reported varying U concentrations in P fertilizers and in agricultural soils. Uranium in P fertilizer and its accumulation in agricultural soil from various continents are presented below.

3.4.1. Europe

Bergen et al. [16] reported the trace metal accumulation in agricultural soils, including U from mineral phosphate fertilizer applications in long-term field studies in 27 European Union countries (Austria, Belgium, Bulgaria, Croatia, Republic of Cyprus, Czech Republic, Denmark, Estonia, Finland, France, Germany, Greece, Hungary, Ireland, Italy, Latvia, Lithuania, Luxembourg, Malta, Netherlands, Poland, Portugal, Romania, Slovakia, Slovenia, Spain, and Sweden). The results showed that the average U concentration in unfertilized soils ranged between 0.18 and 0.95 mg kg^{-1} , and fertilized soil ranged between 0.29 and 4.88 mg kg^{-1} , which was statistically significant. The findings showed that long-term use of mineral P fertilizer enriches agricultural soils with U. The concentration of U was attributed to the application of P fertilizer, which has no limitation in EU policy compared to the Cd threshold of 60 $\text{mg Cd kg}^{-1} \text{P}_2\text{O}_5$. While an estimated 90% of P-derived mineral fertilizers applied in the EU met the recommended regulatory policy limit of Cd in fertilizers before the regulatory limit was enforceable [67], it was recommended that the environmental impact of fertilizer-derived U need attention equal to that of Cd in mineral fertilizers [16,70,71]. In Greece, Servitzoglou et al. [69] investigated U content in common P fertilizers. The results showed that U ranged from 3.3 to 57.1 mg kg^{-1} (average 32.1 mg kg^{-1}). Additionally, U contained in P fertilizers did not change its concentration in fertilized soil compared with unfertilized soils and hence did not enhance the natural radioactivity of wheat grain.

Among the EU countries, U in P fertilizers and its effects on agricultural soils have been extensively studied in Germany (Figure 4). Ten articles reported U concentrations in P fertilizers and their impact on German agricultural soils. Kratz et al. [55] investigated U concentrations in P mineral fertilizers commonly used in German agricultural soils and found that U concentrations ranged from 14.3 to 141 mg kg^{-1} . The results further showed that all P fertilizers had higher U concentrations than the proposed legal limit of 50 $\text{mg U kg}^{-1} \text{P}_2\text{O}_5$ [55]. Similarly, soil-to-plant uptake bioaccumulation was calculated, and then U (radioactivity) uptake from soil-to-plant was also reviewed based on the available quantitative data in the searched articles. Kratz et al. [55] further investigated the impact of the long-term (3–78 years) application of P-derived fertilizers on U concentration in Europe from 218 soil samples. The results showed that the long-term application of mineral P fertilizers enriched soils with U. The results further suggested the concentration of U in soils in all sites was directly proportional to applied P (increase of 1 ton P ha^{-1} linearly increased the means concentration of the surface soil (0–23 cm depth) by 0.11 mg U kg^{-1}).

In the same country, Schnug and Haneklaus [72] reported that P-derived fertilizers were found to be a major source of U pollution and buildup in agricultural soils and also enriched U concentrations in groundwater. The article proposed the recovery of U as an environmental contaminant as well as a raw material for nuclear power plants for the generation of electricity. They further argued that U recovery from PRs during fertilizer manufacturing processes would reduce environmental pollution, since the recovered U resources could be used for greenhouse gas lean energy production. Their study was also complemented by work from Liesch et al. [73], who reported that P fertilizers can be a major source of U in agricultural soils together with background concentrations. In addition to U buildup in agricultural soils, Campos et al. [15] investigated the possibility of U mobility in vineyard soils in Germany's Rhineland–Palatinate region. The findings demonstrated that U in fertilized soils varied from 0.48 to 1.26 mg kg^{−1}, which was slightly higher than nearby non-agricultural soils (0.50 mg kg^{−1}), very homogeneous along slope positions, and slightly higher in top soils.

Sun et al. [18] complemented the study by investigating long-term P fertilizer application in Rothamsted and P fertilizer-derived U accumulation in topsoil (0–23 cm) from 1876 to the 2010s. According to the findings, the broadbalk and park grass total U accumulation rates, which ranged from 2.8 to 6.1 µg U kg^{−1} yr^{−1}, were comparable to those noted forty years prior. Additionally, the results showed that in comparison to the soil from forests (the control), soil obtained from fertilizers did not significantly enhance U levels [68]. Rogasik et al. [74] examined, using data from long-term field trials conducted in Germany, the relationship between P fertilizer application rates, fertilizer products, soil parameters, and fertilizer-based U accumulation in soils. The overall long-term field results demonstrated that surface soils receiving continuous mineral P fertilization had greater U contents than unfertilized soils.

3.4.2. Australia and New Zealand

In Australia, Lottermoser [75] conducted a study in Northern Queensland on the concentration of U in P fertilizers and its possible enrichment in P-fertilized sugarcane soils after long-term fertilizer application. The result found that U concentrations in P fertilizers and agricultural soils ranged from 6.0 to 183 mg kg^{−1} and 1.3 to 6.3 (average 3.7) mg kg^{−1}. The U concentration in the control (forest) soil ranged from 1.3 to 2.3 mg kg^{−1} (average 1.83). The study indicated that the average U concentration of the control soil (1.83 mg kg^{−1}) was half of that of sugar cane fertilized soils (3.7 mg kg^{−1}). The results suggested that U enrichment in sugar cane soils was influenced by the application of P fertilizers.

New Zealand is an intensive P fertilizer utilization country. The country uses locally available reactive P fertilizers after simple beneficiation and granulation. Directly applying P fertilizer offers cost reduction to farmers; therefore, it is the most preferable fertilizer in the country [11]. However, the major setback is that it contains more radioactive radionuclides compared to WPA processed P fertilizers. In contrast to the WPA route, only U migrated to fertilizers and U daughters are left in phosphogypsum [76]. In the direct application of P fertilizers into the soils, all U and its progeny left in fertilizers end up in soils [77]. New Zealand has natural PRs, which are reactive and are directly applied in New Zealand's agricultural soils. The most common P fertilizer used in New Zealand is single superphosphate (SSP). Similarly, the country uses reactive phosphate rock (RPR) directly in agricultural soils, which also contains a high ²²⁶Ra concentration. The concentration of U in P fertilizers in New Zealand ranges from 12.1 to 129.5 mg kg^{−1}, while in agricultural soils, it is 2.1–7.1 mg kg^{−1} [78]. The reviewed results show that the concentration of ²²⁶Ra in fertilized soils was higher compared with the mean concentration of it in the control (forestry soils). Therefore, the concentrations of U in New Zealand's agricultural soils displayed a linear correlation with the concentration of U in P fertilizers [78].

Taylor [77] reported soil U enrichment after long-term P fertilizer application in New Zealand. The study analyzed samples that were collected and preserved from four sampling

sites between 36 and 43 years ago to determine whether there was a statistically significant variation in U accumulation in the same four sites in New Zealand. The results showed that for soils collected roughly 40 years ago, mean levels of total U increased by 1.30 mg kg^{-1} , or 0.033 mg kg^{-1} , annually. Agriculture soil U enrichment was again linked to the use of P fertilizers.

3.4.3. Middle East

The world's greatest PR deposits are found in the North African and Middle Eastern belt. The belt encompasses all of North Africa as well as several Middle Eastern nations, such as Jordan and the Kingdom of Saudi Arabia. In the Kingdom of Saudi Arabia, U dissemination in agricultural soil as the result of fertilizer application has been studied. The reported U concentration ranged from 1.9 to 315 mg kg^{-1} (average 80.4 mg kg^{-1} [79–81]. Different types of P fertilizers indicated different U concentrations; for instance, the triple superphosphate (TSP) and NPK fertilizers had the highest U concentrations compared to Di-ammonium phosphate (DAP) and mono-ammonium phosphate (MAP). TSP and NPK lead to higher annual effective doses for farmers and members of the general public [79]. The study even recommended that NPK and TSP were unsafe for use as fertilizers in agriculture in Saudi Arabia. Similarly, Latif et al. [80] reported a higher U concentration for imported phosphate fertilizers compared to local P fertilizers. The overall studies indicated that the application of P fertilizers with elevated concentrations of U may significantly increase agricultural soil radioactivity and the surrounding environment. Khater and Al Sewaidan [82] reported U concentrations in P fertilizers sold commercially and made the first judgments about radiation exposure from P fertilizer use in Saudi Arabian agriculture. The results showed remarkable and wide variations in the radioactivity contents of the different P fertilizer samples. The mean (ranges) of specific activities for ^{226}Ra was 6.1 mg kg^{-1} (0.2 – 23.0 mg kg^{-1}) and it aligned well with other work from Khater [83].

3.4.4. Africa

In Algeria, the U concentration in commercial P fertilizers and the radiological impact of fertilized agricultural soils against unfertilized soils were determined using gamma-ray spectrometry. The result showed that U concentrations in P fertilizers ranged from 10.9 to 15.4 mg kg^{-1} , while U concentration in fertilized soil was 4.3 mg kg^{-1} compared to 3.8 mg kg^{-1} in virgin soil. The fertilized soils presented significantly higher values than unfertilized soils.

In Egypt, Ahmed and El Arabi [29] investigated U concentration in P fertilizers and agricultural soils in the Qena governorate and Upper Egypt using gamma-ray spectrometry. The results showed that U concentration in fertilizers ranged from 28.3 to 31.2 mg kg^{-1} (average 29.6 mg kg^{-1}). The U concentration in arable soil and Nile Island soil (control) ranged from 0.8 to 1.1 mg kg^{-1} (average 1.0 mg kg^{-1}) and 0.7 to 1.5 mg kg^{-1} (average 1.1 mg kg^{-1}). The result showed that there were no significant differences between fertilized and unfertilized soil. In East Africa, Minjingu phosphate fertilizer is commonly used in Burundi, DRC, Kenya, Rwanda, Tanzania, and Uganda, showing particularly high U concentrations ranging from 200 to 600 mg kg^{-1} [84–86]. Here, P fertilizers are used directly in agricultural soil after a simple beneficiation process.

3.4.5. Asia

In India, U concentrations of P fertilizers have been reported in the literature [87–90], which range from 0.02 to 42.8 mg kg^{-1} . For instance, Hameed et al. [87] investigated the effect of the application of P fertilizers on the U concentration of highly irrigated agricultural soils in Srirangan taluk, India, where paddy, banana, and sugar cane are grown. The review reported that the U concentration in the single superphosphate was 32.1 mg kg^{-1} , and in the triple superphosphate was 23.0 mg kg^{-1} [87]. The average activity of U in P-fertilized soils was 0.68 mg kg^{-1} , which was approximately 25% higher than that of the control soil (0.55 mg kg^{-1}). Therefore, the application of phosphate fertilizers in agricultural soils in

India increased U concentration by 25%. Additionally, according to Chauhan et al. [88], the natural U concentration was below the recommended limits (370 Bq kg^{-1}) in P fertilizers, except in superphosphate fertilizer and potash fertilizer. However, the two fertilizers satisfied the UNSCEAR [62] requirements (limits of 1000 Bq kg^{-1}), although they were above the recommended limits [91]. India is one of the nations that uses chemical fertilizers most frequently around the globe, and its usage has been rising over time. For example, the number of chemical fertilizers consumed per hectare on average grew from 86.7 kg in 2001 to 128 kg in 2018. Punniyakotti et al. [92] investigated the U content in different types and brands of P fertilizers on the market in the Villupuram District in Tamil Nadu State, India, and its influence on fertilized soils compared to unfertilized soils. U concentrations in fertilized and unfertilized soil ranged from 0.8 to 7.3 mg kg^{-1} (average 1.1 mg kg^{-1}) and 0.8 to 8.1 mg kg^{-1} , respectively. It was observed that over 70% of fertilized soil samples had a higher U concentration than virgin soil samples. In the same country, Kant et al. [93] investigated U concentration in fertilizers and its potential radiological contamination in agricultural soils. According to the results, the mixed soil sample from crop fields had a mean U concentration of 1.3 mg kg^{-1} , while the unfertilized soil had a concentration of 0.7 mg kg^{-1} . U measurement for P fertilizers and non-P fertilizers used in West Bengal, India, was performed. Ghosh et al. [94] measured U concentrations in P fertilizers in 2007, namely superphosphate, di-ammonium phosphate, and nitrogen-phosphorus-potassium, and non-P fertilizers such as Suphala-50 and Gromor and found that U concentrations ranged from 11.4 to 210.1 mg kg^{-1} (average 68.6 mg kg^{-1}) and 0.2 to 0.4 mg kg^{-1} , respectively. The P fertilizers were used in agricultural soil. The concentration of U in P-fertilized soil ranged from 8.8 to 53.6 mg kg^{-1} (average 29.35 mg kg^{-1}).

In Bangladesh, the concentration of U in common P fertilizers used in agricultural soils was studied. U contents in compost, TSP, and DAP ranged from 2 to 8 mg kg^{-1} , 68 to 209 mg kg^{-1} , and 98 to 119 mg kg^{-1} , respectively [95]. The study clearly shows that U concentrations were considerably higher in the three most frequently used P fertilizers in the country. However, the U-containing fertilizers were not tested on agricultural farms to also observe their influence on soil contamination and plant uptake.

In Pakistan, Faisalabad is a fertilizer-intensive agricultural city with a population of over 6 million inhabitants. Nasim-Akhtar et al. [96] examined the difference between the content of U in fertilized and unfertilized soil. The U concentration in fertilized soil ranged from 2.11 to 3.9 mg kg^{-1} , while that of unfertilized soil was 0.6 mg kg^{-1} [96,97]. The results showed that fertilized soils had a higher U concentration than unfertilized soil.

The agricultural sector in Vietnam, just like other countries in the world, uses P fertilizers to increase soil fertility. Vietnamese agricultural soils, which are applied with P fertilizers, have been shown to contain uranium in large amounts. In order to understand how the U decay series to the radioactivity of fertilizers is distributed [98], the gross alpha, gross beta, and activity concentration of ^{226}Ra in certain types of fertilizers typically used in the southern region of Vietnam; it was revealed that the U concentration in the P fertilizer ranged from 0.1 to 47.9 mg kg^{-1} . The P fertilizers were tested in agricultural soils in the field experiment to understand their concentration in soil and their influence on plant uptake. The results showed that the U concentration ranged from 3.1 to 56.9 mg kg^{-1} , with a mean value of 30.5 mg kg^{-1} [99]. The U in P fertilizers was similar to that reported previously in the country. According to the findings, phosphate fertilization did not affect the soil's U accumulation compared to the control (non-fertilized) soils. The results showed that the use of fertilizers had no effect on that radioactivity [69].

Yamaguchi et al. [40] investigated the effects of P fertilizers containing U in Japan and also assessed the effects of various agricultural practices on the U content of the soil. The results indicated that the long-term application of P fertilizer raised the U concentration in the soil. In the surface soil of agricultural areas, uranium from applied fertilizers was found to be adsorbed, precipitated, or integrated into the soil organic matter with minerals carrying iron or aluminum rock [40]. In grazing land and highland field soil, soil organic matter seemed to be a more significant U pool, but in paddy field soil that experienced

periodic shifts in redox conditions, weakly crystalline Fe minerals seemed to be a more significant U pool [40]. These findings imply that the extra U was cemented to the soil particles to prevent crops from readily absorbing it [100].

3.4.6. Latin America

Intense fertilizer application is frequently required due to the study's findings about the effects of P fertilizer application in Mexico's agricultural sector. In the highlands of central Mexico, where corn is grown, P fertilizers are the most commonly used. U concentrations in P fertilizers in the region are reported to range from 50 to 200 mg·kg⁻¹, depending on the origin of the PR used in their production. U content in fertilized agricultural soils varied between 19.5 and 50.6 mg kg⁻¹, while in unfertilized soils, it ranged between 0.4 and 1.0 mg kg⁻¹. The results showed that adding phosphate fertilizers to agricultural soils enhanced U concentrations in soils.

A study was carried out in Brazil by De Souza et al. [37] on the long-term usage of P fertilizers in citrus, oil palm, and black pepper crops for 26, 10, and 5 years, respectively, and it was found that long-term P fertilizer application enhanced radioactivity in the investigated crops. The study revealed that the concentrations of U in unfertilized soils used to grow oil palm, black pepper, and citrus were 1.51 mg kg⁻¹, 1.71 mg kg⁻¹, and 2.19 mg kg⁻¹, respectively, while in fertilized soils, the concentrations of U were 2.15 mg kg⁻¹, 3.48 mg kg⁻¹, and 2.99 mg kg⁻¹, respectively [37]. The results revealed that U accumulation was greatest in black pepper and oil palm-grown soils compared with citrus-grown soils. Furthermore, the concentrations of U in these crops generally were citrus > black pepper > oil palm, in that order [37]. This demonstrates that various plants have distinct absorption processes for U. The U contents in the fertilized soils grown with black pepper and oil palm in Brazil were slightly higher compared with the world average of 2.8 mg kg⁻¹ (35 Bq kg⁻¹) set by the United Nations Scientific Committee on the Effects of Atomic Radiation [62]. So, all the plants had low bioaccumulation factors and, therefore, did not pose any radiological risks to the environment or human health. The study recommended continuous monitoring of radionuclides in agricultural soils if P fertilizers are continuously used, while the sustainable solution to avoid soil pollution from P fertilizer application is for fertilizer industries to recover or remove U and other contaminants before application into the soils.

Yamazaki and Geraldo [41] investigated U concentrations in the most commonly used P fertilizers applied to Brazilian agricultural land. The results showed U that concentrations ranged from 5.2 to 54.3 mg kg⁻¹. The U concentration was lower than other values reported in the literature. Investigating the influence of U-containing P fertilizer in agricultural soils is important to ascertain its potential entry into the food chain. For instance, Lauria et al. [101] investigated the U concentration in vegetables grown using different fertilizers. The study determined the presence of uranium and radium in chemical and organic fertilizers and farm soil. Then, U isotope uptake by vegetables grown in Rio de Janeiro was assessed in acidic and limed soils. The results showed that U from different NPK fertilizers used to grow lettuce, carrots, and bean crops ranged from 0.3 to 25.9 mg kg⁻¹. There was no statistically noticeable difference in U concentration in vegetables grown under conventional fertilizer application compared to those organic management practices that could be attributed to P fertilizers.

The quantitative studies reported U concentrations in soils after the application of P fertilizers compared to unfertilized soils (forest virgin land). All searched literature examined revealed that the application of P fertilizers enhances U concentration in agricultural soils. However, one study reported the non-accumulation of U after long-term application of P fertilizers [41].

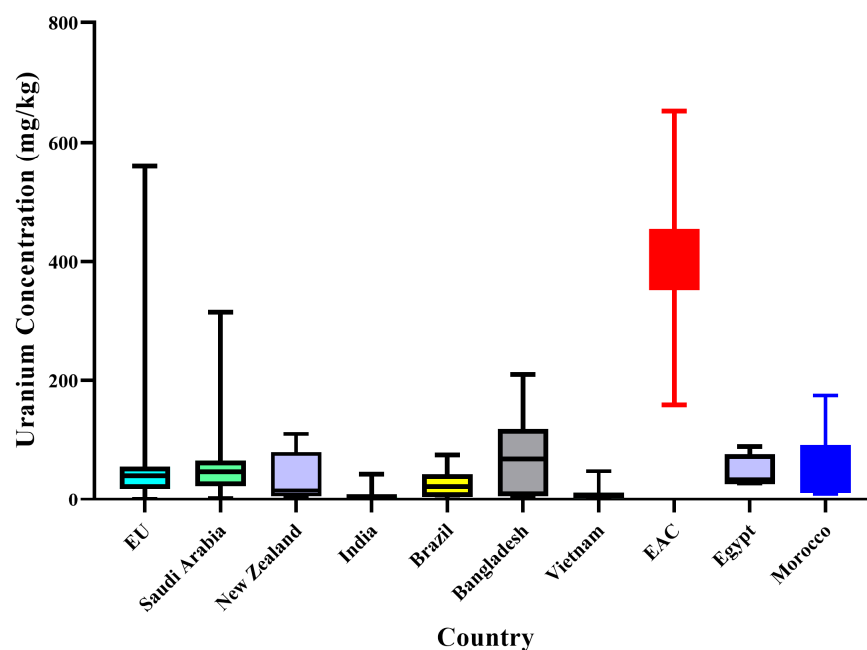


Figure 5. Box and whisker plots of reported U in P fertilizers from 2003 to 2022 from peer-reviewed articles worldwide, irrespective of the type of P fertilizer type and composition. Bangladesh [95], Brazil [37,102], Germany [55,72], Greece [69], India [87,88], New Zealand [78], Saudi Arabia [79,80,82,83], Vietnam [98,103], East African Community (EAC) ([27,47]), Egypt [48], and Morocco [20].

3.5. U as a Potential Agricultural Soil Contaminant

The United Nations Sustainable Development Goals (UN SDGs) promote sustainable development goals, which call for responsible mineral extraction and protection of the environment from heavy metals like U. Generally, U is an environmental contaminant as well as a mineral resource lost in agricultural soil [11]. U is a heavy metal and a radioactive element since it emits extremely small amounts of ionizing radiation [104]. The small amount of U-ionizing radiation emitted into the environment can enter the food chain through the soil, which in turn becomes chemically and radioactively toxic to the organisms in the food chain. Thus, the application of phosphate fertilizers to agricultural soils results in an elevation of U concentrations in cultivated soils. Excess U concentrations in agricultural soils, which are due to the application of phosphate fertilizers, have been reported by some scholars to migrate from the topsoil layers into soil bedrocks by leaching and affecting surface and groundwater in oxidative acidic conditions [105–107]. Additionally, Schnug and Lottermoser [36] indicated that U from P fertilizers applied to agricultural soils poses a potential radiological and chemical health risk to human beings.

The amount of U concentrations elevated in agricultural soils depends on the quantity of P fertilizers applied, the concentration of U in the P fertilizers, the type of soils, and the local climatic conditions of the regions [36]. PRs are considered as unconventional U resources [12]. The amount of U present in PRs and P fertilizers is not recovered as a product or co-product because it is not profitable under current market conditions [108].

For instance, the cost of recovering 1 kg of U was reported to range between USD 88 and 122 while the market value was quoted as USD 50 per kg [109,110]. Due to these economic reasons, a huge amount of U is lost to agricultural soils every year. In 2016, Sun et al. [111] estimated that approximately 700 tons of U were dissipated into European Union agricultural soils, while if U was recovered, it would have been sufficient to provide the annual amount U needed to run two nuclear power plants of 1000 MWe- light water reactors with an annual consumption of 310 tons of natural U per year (assuming 5% enrichment with no U losses in the process) [20]. Therefore, the U content in PRs and P fertilizers could be used as the raw material for nuclear power generation. Importantly, it

should not be extracted during fertilizer production based on economic grounds but rather due to environmental considerations [54].

3.6. Reported U Uptake by Plants

Long-term P fertilizer application in agricultural soils increases U build-up and potential plant uptake. The process of minerals moving from the soil to the plant, being taken up by it, and building up therein involves runoff, capillary rise, leaching, sorption, and root uptake. The uptake of U by plants is comparable to how the uptake of vital macro- and micronutrients like zinc, potassium, and nitrogen affects the uptake of non-essential substances. High U concentrations in soils have an impact on agricultural soils and may be absorbed by plants to affect the food chain. The distribution of U in agricultural soils is essentially influenced by a mixture of physical and chemical properties of the soil, such as soil type, texture, pH, redox status, organic matter concentration, mineral composition, climate, the kind of plant, and the plant part in question [112,113]. The presence of additional elements also affects transfer factor values.

A high total U concentration in the soil does not guarantee uptake by plants. The availability of U is fully dependent on aqua regia, which is a measure of U accumulation in soils owing to phosphate fertilizer application compared to the total [114]. U absorption by plants is often limited to the dissolved portion of the soil, implying that U may be less available to plants. Uranyl ions are the sole plant-available and soluble portion of U. Plants absorb all macro and micronutrients from the soil, including uranyl ions, via ion transport from the soil solution to the roots. The translocation of U and other radionuclides is regulated by soil parameters such as soil qualities, climatic circumstances, plant type, plant part involved, the physicochemical form of the elements, and the presence of other elements.

Different types of plants have different uptake characteristics. However, the most consumed crops are of high concern, as many people may be affected no matter how small the amount of uranium is taken up. For instance, wheat (*Triticum* spp.) and maize (*Zea mays*) are the most frequently produced cereal crops, feeding more than 40% of the global population [115]. Monitoring these most consumed cereal crops is important to ensure that the large population is not exposed to U contamination. For example, in Serbia, maize and wheat crops are the two most important cereal crops where P fertilizer application is intensive [116]. Stojanovic et al. [117] reported the assessed U uptake from P-fertilized soils into maize and wheat crops. The results showed that relatively high U concentrations in fertilized soils did not enhance U uptake by wheat and maize crops, which was attributed to both high pH (above 6) and organic matter, which made U unavailable for plant uptake [117].

U in soils has no nutritional value to plants but may pose radiological, chemical, and biotoxicity risks to humans and the environment once it enters the food chain [118]. Studies for naturally occurring radionuclides like U from soils to plants have been reported [118]. In the environment, concentrations of U are enhanced by anthropogenic activities like mining and the application of phosphate ore-derived fertilizers in agricultural soils [119]. U can also accumulate in the environment due to non-nuclear-related activities such as the leaching of waste during PR processing, the application of P-based fertilizers, mining, and processing of raw materials containing high concentrations of naturally occurring materials.

In Thailand, U uptake by common foods and commercial crops such as paddy, maize, sugar cane, and cassava was studied [120]. The application of P fertilizers with high U concentrations, according to the study, raised the concentration in agricultural soils and affected plant absorption. The results show the concentration of U in cassava > paddy field > maize > sugar cane > natural forest (control site sample). Several studies have reported a linear relationship between added U in soils during fertilizer application and the potential uptake by plants. It should, however, be noted that the difference between U concentrations in arable land and grassland has been an important indicator of whether enrichment is possible or not. There are essentially two major possible scenarios when P fertilizers containing U are applied in agricultural soils: either they accumulate in the

soils or they leach to the ground and surface waters. However, few studies have been conducted to evaluate U leaching and speciation in soil solutions, as well as transport in arable soils. Bigalke et al. [121] examined the speciation of U in arable soil solutions and probable U leaching along preferred flow pathways in Switzerland. The result showed that, when compared to manure application, P fertilizer application resulted in larger U input in agricultural soils and effected leaching and accumulation of U in a greater number of carbonate ions.

The U radionuclide's interaction between soils and plants depends on soil properties such as nutrient concentrations, pH, mineral composition, and organic matter content [117]. The mechanism of U radionuclide transfers from soils to crops depends on various factors, such as the physicochemical properties of the U radionuclides of interest, the type of crops, soil management practices, wastes, and the time after the fallout and soil properties [100]. The uptake of U radionuclides from soils to plants follows the natural process of essential nutrient uptake [65]. The analogous chemical properties between essential and non-essential elements can enter the plant's body through similar mechanisms. The factors influencing soil-to-plant nutrient uptake are agroclimatic conditions, pH, soil properties, organic matter content, and soil microbial activity [122]. However, there is little information reporting the transfer of radionuclides from agricultural soils to plants. Therefore, there is a need to investigate the transfer of radionuclides from agricultural soils to plants.

Moreover, U is not an essential element for crops, but it has a detrimental effect on ecosystems, including the health of human beings, due to its carcinogenic effect [123]. U possesses chemical and radioactivity toxicity to organisms in soils. Several studies have revealed the U uptake by plants from the soils, which eventually is transferred into the food chain [124,125]. For example, Nkuba and Mohammed [126] indicated that there is higher natural radioactivity and radiation transfer from soils into maize and bean crops grown near Minjingu phosphate mines. Therefore, this indicates that crops have the ability to take up U from contaminated soils into their tissues. Additionally, other studies reported that sorghum, rice, cassava, sunflower, and vegetables have the ability to increase U uptake from the soils grown in U mineralized sites, which has health effects on humans consuming foodstuffs with elevated U concentrations [127].

In agricultural soils, U can enter plants through the roots and be distributed to different parts of the plant. Different plant species have different U uptake behavior. The behavior of U in plants follows the behavior of calcium [128]. It is reported that the uptake and distribution of U radionuclides in plants depend on the type of crop, soil pH, exchangeable Ca and K and organic matter contents, physicochemical properties of the radionuclide, fertilizer application, irrigation, plowing, liming, and climate conditions [128,129].

Little information is reported about U uptake from contaminated soils into crops, which has sparked interest in investigating the uptake of U from soils into crops. Additionally, little information exists on U uptake from the soils into crops in the East Africa region, and it has become important to investigate the concentrations of radionuclides uptake by crops. To our understanding, research studies on screening potential plants and organic materials for removing U toxicity levels from soils have not been studied in the East African Region. Therefore, there is a need to conduct screening for potential crops and organic materials to be aware of the concentrations of the radionuclides transferred from agricultural soils to plants.

3.7. The Fate of U Added to Agricultural Soil

It is important to know what happens to the U that is added to farmland when P is fertilized in order to figure out the chemical and radiological health risks in the environment. However, the assessment of the fate of anthropogenic U in soil is challenging because of the complex nature of its interaction in soil [40,57,130]. In the soil, fertilizer-derived U has been reported to leach out because U is mobile in surface soils as a uranyl complex depending on prevailing oxidation–reduction (redox potential) and acid–base reactions (pH) [36,56]. Therefore, the fate of U in the surface and sub-surface soil environments depends entirely

on U valency states. In soils, U is fairly soluble as a U(VI) cation or (UO_2^{2+}) in an oxidizing environment [118,124,131]. In soil with a reducing environment and a pH between 4.0 and 7.5, uranium is immobile because UO_2 mostly binds to oxidized organic matter (OM) and precipitates on the surface of iron/aluminum (Fe/Al) minerals that are weak or non-crystalline [100]. Iron/aluminum are regarded as significant U sinks [40,57].

U is mostly found as UO_2^{2+} in oxidative environments, and as the pH increases, more of it sticks to negatively charged soil particles [15]. However, the creation of soluble and negatively charged compounds with substances like carbonate improves the mobility of U [132]. According to Ratnikov et al. [125], in soils, U reacts with oxygen to form the hexavalent uranyl ion (UO_2^{2+}), which is stable and soluble in the ambient environment and determines its mobility, bioavailability, uptake, and chemical and radiotoxicity.

Erosion is another important component that affects the fate of U in soils, and it is regulated by a number of variables, including surface slope gradient, infiltration rate, rain intensity, and plant covering [15]. U added to the soil from P fertilizers is the main source of interaction between plants–soil and plants–water. U can also be leached from fertilized soils (anthropogenically added) or from natural sources present in the surrounding soils, tailings, and rocks. Terrestrial plants' roots can take up U from the soil and move it to their shoots. U leachate, through runoff or leaching, can enter water bodies. For instance, in Brazil, Yamazaki and Geraldo [41] reported that prolonged application of P fertilizers was the main source of U contamination of groundwater. Similarly, in Germany, Schnug and Lottermoser [36] reported that U derived from P fertilizer application migrates and contaminates ground and surface waters, posing a potential radiological health risk to humans in Germany. Additionally, Birke et al. [133] reported that the application of P fertilizers was responsible for elevated U concentrations in bottled and tap water in Germany. High concentrations of U in water pose a serious threat to human health and the environment.

Once U is introduced into natural water systems, aquatic plants may begin to accumulate, which poses a risk to both terrestrial and aquatic ecosystems. Estimation of potential U accumulation by anthropogenic and natural U accumulation from different sources in the soils is important to estimate U concentration in the edible parts of plants or those eaten by animals that may fall into the human food chain. The prolonged application of P fertilizers, the substantial persistence of U in soils, and the possibly serious health risks of eating contaminated crops are of great concern.

3.8. Potential Regulatory Limits of U in P Fertilizers

In the environment, U is regarded as a chemical and radioactive element. Along with arsenic and chromium, it is one of the three most dangerous naturally occurring pollutants in groundwater [121]. There is no regulatory limit on the concentration of U in P fertilizer worldwide. However, some countries are considering setting limits on notable risky metals, such as U, in P fertilizers. The European Union has set a regulatory limit for Cd concentration, while U is not yet considered. In Germany, the German Commission for the Protection of Soils advocated setting a limit for the amount of uranium in fertilizers at 50 mg per kg P_2O_5 [55], or 167 ppm for fertilizers with a 30% P_2O_5 concentration. The lack of a regulatory limit for U in P fertilizer has not encouraged P fertilizer producers to recover U from WPA in the past [134], and increasing U prizes could make *U recovery from phosphates great again*, as speculated by Steiner et al. [25].

4. Conclusions and Policy Recommendations

This work evaluated 54 articles from throughout the world that reported on the significant quantity of U in P fertilizers, their enrichment in agricultural soils, and some that explained the possible uptake of P by plants worldwide. The review also examined the fate of U in soil. The results showed that P fertilizers used in different countries worldwide contained varying concentrations of U depending on the type of P fertilizer-derived rock used to manufacture the fertilizers. However, P fertilizer from Minjingu P

fertilizers had a higher U concentration comparable to that of commercial U mines operated in Namibia. Studies have shown that the long-term application of mineral P fertilizers increases U accumulation in agricultural soils. U accumulation in agricultural soils may influence U uptake by plants and could even contaminate surfaces and groundwater. While there are substantial restrictions on the content of Cd in P fertilizer, there are none for U at the moment. Although there is currently no limit on the amount of U allowed in fertilizers, it is still necessary to investigate the viability of recovering U from PR during fertilizer production. This is particularly true for phosphate deposits with relatively high U concentrations such as the Minjingu phosphate deposit in Tanzania. It is also important to evaluate technical and economical extraction methods in order to reduce the amount of U present in agricultural soil and the risk of surface and groundwater contamination.

Author Contributions: Data curation: D.A.M.; formal analysis: N.H.H.; funding acquisition: N.M.; A.I.A. and K.M.M.; investigation: D.A.M.; methodology and review: D.A.M., J.M.M. and N.H.H.; supervision: K.M.M., N.H.H., T.T.K., J.B.L., A.I.A., F.M.C. and N.M. All authors have read and agreed to the published version of the manuscript.

Funding: This publication was made possible thanks to funding from the Tanzania Atomic Energy Commission (TAEC), the Austrian Federal Ministry of Education, Science, and Research (BMBWF), and Austria's Agency for Education and Internationalization (OeAD) [Grant Numbers: Africa UniNet P006; KOEF 01/2019]. Open Access Funding was provided by MDPI.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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