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Resources, Conservation & Recycling 207 (2024) 107694

Contents lists available at ScienceDirect

Resources, Conservation & Recycling

journal homepage: www.sciencedirect.com/journal/resources-conservation-and-recycling

Review

Rare earth elements and uranium in Minjingu phosphate fertilizer products: Plant food for thought

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ARTICLE INFO

Keywords:
Minjingu phosphate ore
Rare earth elements (REEs)
Uranium
Circular economy
Tanzania

ABSTRACT

Minjingu phosphate ore is Tanzania's sole domestic supply of phosphorus (P). The ore contains medium to high concentrations of naturally occurring P_2O_5 (20–35 %) and relevant concentrations of uranium and rare earth elements (REEs) are also suspected to be present. Currently, neither uranium nor REEs are recovered. They either end up in mine tailings or are spread across agricultural soils with fertilizer products. This work provides a first systematic review of the uranium and REE concentrations that can be expected in the different layers of Minjingu phosphate ore, the way the ore is presently processed, as well as a discussion on alternative processing pathways with uranium/REE recovery. The study analyzed ten distinct Minjingu phosphate ore layers, four mine tailings, and five intermediate and final mineral fertilizer products from the Minjingu mine and processing plant located in northern Tanzania. The results confirm that the uranium concentrations and to a lesser degree, the REE concentrations are indeed elevated if compared to concentrations in other phosphate ores. The study does not identify a significant risk resulting from this. The development of techno-economic solutions for more

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https://doi.org/10.1016/j.resconrec.2024.107694
Received 3 October 2023; Received in revised form 8 April 2024; Accepted 5 May 2024
Available online 14 May 2024

comprehensive utilization of Minjingu ore is, however, strongly encouraged and suggestions on such processes are provided.

1. Introduction

Phosphate ores that are used to produce mineral fertilizers can show elevated concentrations of heavy metals and particularly cadmium has been identified as a potential health risk (McLaughlin et al., 2021). Radiotoxic uranium (Haneklaus, 2021a) as well as rare earth elements (REEs) (Chen and Graedel, 2015; Gaustad et al., 2021) can also be found in phosphate ores worldwide in relevant concentrations.

Phosphate ores can be of magmatic (or igneous) and sedimentary origin, and these ores tend to show very different concentrations of accompanying trace elements. In the case of igneous phosphate ores REEs can reach concentrations of 0.2 wt%, while the concentrations are usually lower in sedimentary phosphate ores. Globally, a REE concentration of approximately 0.05 wt% for phosphate ore is realistic (Wu et al., 2018a) since there are much more sedimentary than igneous phosphate ore reserves. Uranium can show concentrations of 0.01 to 0.02 % in Moroccan phosphate ores (Qamouche et al., 2021; Ulrich et al., 2014) that present more than 70 % of the currently known reserves (Cooper et al., 2011), while uranium concentrations are usually below 0.005 % in phosphate ores of magmatic origin.

Although the concentrations in phosphate ore can be considered moderate at best, the overall quantities of both REEs and uranium that could theoretically be recovered are impressive given that approximately 220 million metric tons (t) per year, phosphate ore is mined globally. Phosphate ore is among the 5th most mined materials on earth, and Hakkar et al. (2021) estimated that REEs recovered from phosphate ore mining in Morocco alone could cover 7–15 % of global demand. Zhang (2014) further showed that the United States could cover its whole demand if REEs were recovered during phosphate processing in Florida. Haneklaus (2021a) further estimated that uranium recovered during phosphate fertilizer production could have theoretically provided some 16–26 % or 8800–14,000 t of the world's commercial uranium requirements in 2018. More detailed country estimates are available for the United States: approximately 2116 t of 20,386 t uranium requirements could be recovered from phosphates in 2014 (Kim et al., 2016; Steiner et al., 2020), China: 648 t (approximately 10 % of the countries uranium demand) could be recovered during fertilizer production in 2016 (Shang et al., 2021), the European Union: approximately 334 t uranium (approximately 2 % of the demand) could have been recovered from phosphate ore imports in 2017 (Tulsidas et al., 2019), and Argentina: approximately 19 t uranium (some 8 % of the countries demand) could have been recovered in 2017 (López et al., 2019). Besides, Diwa et al. (2023) estimated that 14–26 t of uranium (12–21 % of projected near-term uranium demand) could be recovered from imported phosphate ores in the Philippines. Khan et al. (2023) did similar calculations for domestically mined phosphate ores in Saudi Arabia and concluded that there 413–551 t (11–15 % of the projected near-term, future natural uranium requirements) could be recovered during phosphate fertilizer production. Ramteke et al. (2022) further investigated the uranium concentrations in DAP fertilizers in India and found that approximately 1320 t uranium were distributed on agricultural soils between 2021 and 2022. If recovered, this uranium could significantly contribute to the natural uranium requirements of India.

Currently, both REEs and uranium are not industrially recovered during phosphate fertilizer production. Recovery of uranium from wet-process phosphoric acid (WPA), an intermediate liquid product in phosphate fertilizer production, was done on an industrial scale in Florida in the 1980s and 1990s before decreasing uranium prices made this practice uneconomic (Haneklaus et al., 2017a,b). Ye et al. (2019) pointed out that (for uranium) we will probably not see a choreographed recovery from phosphates at all >400 fertilizer plants around the world starting anytime soon, but maybe recovery operations starting at phosphate mines that show elevated uranium concentrations to a degree that there is a pull-factor, in a way that uranium can be sold at economic profit, but also a push-factor, in a way that not recovering the uranium could result in lower final fertilizer prices as a result of the elevated heavy metal content (Scholz and Wellmer, 2018) or even limitations with regards to its sale as a result of potentially introduced uranium limits in fertilizers.

The Minjingu phosphate ore deposit in northern Tanzania is known for its relatively high concentration of naturally occurring uranium. It was in fact the increased radiation that led to the discovery of the deposit at the edge of Lake Manyara, now a national park and protected area in northern Tanzania, by a South African mining company in 1956 (Mchihiyo, 1991; Van Kauwenbergh, 1991).

Minjingu phosphate ore shows natural uranium levels that would also qualify the deposit as a very low-grade uranium ore under the definition of the World Nuclear Association (WNA) (Haneklaus et al. 2017a). Mwalongo et al. (2023a) recently pointed out that the uranium concentration in Minjingu is indeed higher than at commercial uranium mines in Namibia on the other side of the African continent. Since fertilizer is more important for Tanzania's economy than uranium, the ore is mined for its elevated P_2O_5 content (20–25 % on average) and not its elevated uranium content (0.03–0.04 % on average).

Previously, some 10–15 years ago, Minjingu phosphate ore was after simple beneficiation that included sorting, sieving and rapid drying at 700–800 °C, applied directly as fine fertilizer powder on agricultural soils in East Africa. The fertilizer produced this way proved to be effective on the acidic soils found in East Africa, and the material was considerably less expensive than imported fertilizers from abroad. It was therefore historically not only used in Tanzania, but also in Kenya, Uganda, Burundi, Zambia and even South Africa as indicated in Fig. 1 (Mkangwa, 2003; Msolla et al., 2005; Nabahungu et al., 2007; Ndeleko-Barasa et al., 2021; Nyambati and Opola, 2014; Sarini et al., 2015;

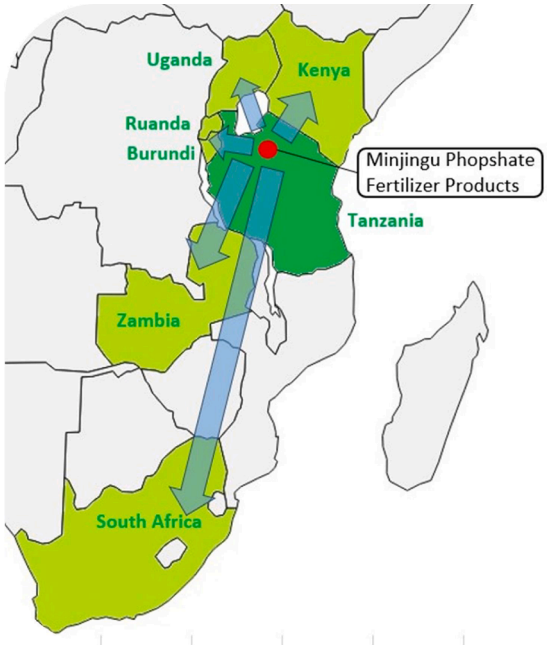

Fig. 1. Location of the Minjingu phosphate rock deposit in northern Tanzania as well as countries to which Minjingu phosphate ore products have historically been exported to.

Szilas et al., 2007a,b, 2008). In all these regions similar acidic soils can be found.

15 years ago, all fertilizer powder has been further processed into granules, which constituted a tremendous improvement since the fertilizer produced this way can be distributed much better by hand on agricultural soils. In the last decade, the granules were further blended with urea, a source of nitrogen, to produce different kinds of compound fertilizers that show a similar agronomic response in East Africa if compared to that of DAP fertilizers (Benson et al., 2012).

At present, the fertilizer plant at the Minjingu mine produces some 50,000 t of fertilizer per year that is distributed to agro-dealers in Tanzania (Rutsaert et al., 2021). The Minjingu Mine and Fertilizer Ltd. intends to double production with a new rotary dryer for mechanical drying of the mined phosphate ore, as well as a larger granulation and blending plant (currently erected) in the next 1–2 years. Even after the Minjingu fertilizer plant extension, the combined output of 100,000 t of fertilizer per year will be far from covering the fertilizer need of Tanzania, which easily exceeds 400,000 t per year right now and may double over the next 5–10 years given the still relatively low crop yields in the country (Harou et al., 2022; Michelson et al., 2021).

Currently, Minjingu products, which are produced locally, receive substantial subsidies in Tanzania. As a result, they are predominantly utilized within the country, with only limited quantities being exported. It is worth noting that agriculture plays a significant role in Tanzania's economy, contributing to over 25 % of the GDP and employing about 75 % of the workforce. A major ammonia and urea fertilizer facility is under construction in the southern Tanzanian port town of Mtwara, aiming to offer more cost-effective fertilizers to local farmers in East Africa. Furthermore, there are several promising studies exploring the utilization of organic fertilizers like goat manure (Mhagama et al., 2023).

Due to the relatively high uranium concentrations at the Minjingu deposit (0.03–0.04 % on average) that are similar to the concentrations found at commercial uranium mining projects, such as the Manyoni Uranium Project (0.011–0.013 % uranium) (Loila et al., 2022) and the Mkuju River Project (0.026 % uranium) (Uranium One, 2023) in Tanzania, an economic case (pull-factor) can also be made for uranium recovery in Minjingu despite the relatively small phosphate ore processing operation of approximately 100,000 t per year. Large WPA units process 2–3 million t of phosphate ore per year, and often multiple units make up a whole phosphate fertilizer plant, as is, for instance, the case in Jorf Lasfar (Morocco), the largest WPA complex in the world (Arhouni et al., 2023). Given the average uranium concentration of 0.03–0.04 % or 300–400 ppm (parts per million), theoretically 30–40 t of uranium could be recovered per year at the Minjingu fertilizer plant, assuming no losses. Given a uranium price of USD 50 per lb of U_3O_8, some USD 3.9–5.2 million in revenue could theoretically be generated annually from selling co-recovered uranium during Minjingu fertilizer production.

The amount of co-produced uranium is obviously tiny in comparison to commercial uranium mines, such as the Mkuju River project in southern Tanzania, which, if started as planned, is expected to produce some 1600 t of uranium annually (Rweyemamu and Kim, 2020). It is noteworthy though, that a potential uranium recovery at the Minjingu fertilizer plant, will ideally result in a cleaner fertilizer product, and does not come with the potential environmental pollution that is presently discussed for in-situ leach (ISL) and open pit uranium mining in Tanzania (Banzi et al., 2015; Loila et al., 2022; Rweyemamu and Kim, 2020). It is further noteworthy that due to the ongoing uranium exploration and mining operations, Tanzania already has a regulatory framework for uranium production in place (Kimaro and Mdoe, 2018; Winde et al., 2017) that could ease the way for byproduct uranium recovery at the Minjingu deposit.

The (literally) million-dollar question is, what are realistic average uranium concentrations? What are realistic uranium recovery rates? Do the final fertilizer products still work as effectively as they do now? And are there other valuable minerals, such as REEs, present in sufficient concentrations so that they could also be co-extracted and sold as well?

This review aims to provide an overview of what is presently known about the REEs and uranium concentrations at the Minjingu deposit and discuss for the first time current and potential alternative processing pathways to answer as many of the previously raised questions as possible. Section 2 of this review provides an overview of the work published on heavy metals in Minjingu phosphate ore. To the best of our knowledge, the REE content of Minjingu ore has not been systematically analyzed yet, and we did our own analysis which is presented here to fill this gap in the literature. The sampling and the methods used for the sample analysis are presented in Section 3. Section 4 presents and discusses the results of the sampling campaign and how the results may support alternative processing pathways for the Minjingu ore. Section 5 presents the conclusions of this review, discusses the limitations of the presented work, and offers policy recommendations based on the findings of this study.

2. Literature review

Radioactivity measurements can be conducted with relatively simple measurement equipment, while determining REEs and other trace elements in phosphate ores is more challenging and requires more sophisticated machines. Usually inductively coupled plasma mass spectrometry (ICP-MS) is used. We believe that this, and the fact that REEs are less relevant for Tanzania's economy, which is largely based on agricultural products, are the reasons why there are a relevant number of studies reporting the uranium content and radioactivity measurements of Minjingu phosphate ore, but we were not able to find a study that systematically analyzed the REE content of the material. There are further considerable differences in the reported uranium content. Interviews with researchers from the Tanzania Atomic Energy Commission (TAEC) who have been monitoring the Minjingu mine for more than 20 years and mining engineers of Minjingu Mining and Fertilizer Ltd. suggest that there is a large difference in the radioactivity of the Minjingu ore depending on the location and depths at which ore samples can be drawn, so that all measurements reported in this literature review, though different, seem indeed to be correct, but simply report on samples taken from different locations of the Minjingu deposit.

Bianconi (1987) documented that Minjingu ore contained a maximum uranium content of 680 ppm, or 0.068 %, when the material was still being transported to the Tanga WPA fertilizer plant for processing. This relatively high uranium concentration in comparison to other phosphate ore deposits was subsequently validated by Meza et al. (2015), who examined a total of 45 Minjingu phosphate ore samples from various locations and identified a peak uranium concentration of 650 ppm. It is noteworthy that, aside from one sample exhibiting this peak uranium concentration, all other Minjingu phosphate ore samples that were investigated showed uranium concentrations half of that value.

There is now no legislative restriction on the amount of uranium allowed in fertilizers while such limits exist for cadmium (Samrane et al., 2023; Ulrich, 2019). According to Kratz et al. (2016), the German Commission for the Protection of Soils recommended setting a legal limit of 50 mg of uranium per kilogram of P_2O_5 or 167 ppm for fertilizers containing 30 % P_2O_5. Similarly high values of 9,550 Bq kg^{-1} ^{238}U or 767 ppm eU (uranium equivalent) were reported by Mustonen and Annanmaki (1988) for the topmost phosphate ore layer, while a lower ^{226}Ra activity concentration of 2,850 Bq kg^{-1} or 232 ppm eU was noted for the lower phosphate ore layer. These results support Bianconi's earlier assessment.

Prior to the Tanga fertilizer plant's activities ceasing in the early 1990s, Makweba and Holm (1993) examined one ground phosphate ore sample, two fertilizer (triple superphosphate and single superphosphate) samples, and phosphogypsum samples produced at the plant utilizing Minjingu ore. Using gamma-ray spectrometry and alpha-spectrometry, the researchers observed ore concentrations of 337

and 377 ppm eU and 408 and 481 ppm eU, respectively. Further observations of the radioactivity of surface water, phosphate ore, mine tailings, leaf vegetation, calf meat, poultry feed, and leaf vegetation, as well as background radiation measurements near the Minjingu mine were provided by Banzi et al. (2000). The estimated uranium concentrations for mine tailings and phosphate ores were 4,250 Bq kg^{-1} ^{226}Ra and 5,760 Bq kg^{-1} ^{226}Ra, or 468 ppm eU and 346 ppm eU, respectively. The study explored exposure pathways and risks to the local population. Upon publication, this research even led to a brief suspension of Minjingu phosphate ore processing, before it became evident that not having access to inexpensive domestically produced fertilizer is also an unsustainable solution.

Another relevant study is the work by Semu and Singh (1995) that investigated the long-term accumulation of heavy metals (Cd, Zn, Mn, Cu, Ni, and Pb) in soils and plants after the use of Minjingu phosphate rock. The authors did not consider uranium but found that the relatively low levels of cadmium in Minjingu phosphate ore are an advantage compared to other fertilizers that can show higher cadmium concentrations.

Koleleni and Tafisa (2019) further investigated different vegetables and soil samples taken near Minjingu village using wavelength dispersive x-ray fluorescence. The study did not report the uranium content of the examined samples, but recommended the reduced consumption of several vegetables as a result of relatively high (other than uranium) heavy metal concentrations.

More recently, Mwalongo et al. (2023a) analyzed phosphate ores from Burundi, Kenya, Tanzania, and Uganda, as well as fertilizer products sold in these countries as well as Rwanda. Minjingu phosphate ore was reported to have a uranium content of 446 ppm, and fertilizers produced from Minjingu phosphate ore generally showed the highest concentrations of uranium. In the next step, Mwalongo et al. (2023b) investigated the influence of NPK fertilizers with varying uranium content on the radioactivity of tobacco plants in Kenya, Tanzania, and Uganda (all countries are major tobacco producers in East Africa). Uranium concentrations in Minjingu phosphate ore were not reported, but it was reported that Minjingu phosphate ore products (fertilizers produced with Minjingu phosphate ore) contain relatively high uranium concentrations if compared to other fertilizers, and the tobacco plants grown with these fertilizers also showed slightly higher levels of radioactivity (still within allowed limits) than tobacco plants grown with fertilizers that show lower levels of uranium. More investigations on the heavy metal content of Minjingu ore derived fertilizers were further recommended by Lisuma et al. (2022). The relatively high concentrations of uranium in Minjingu phosphate ore, if compared to other phosphate ores was further confirmed in a recent meta-analysis (Mwalongo et al., 2024).

Naturally occurring radioactivity in Minjingu phosphate ore is not only relevant for the fertilizers that are produced with this ore, but also affects the tailings of the fertilizer plants that need to be managed. Mdachi et al. (2024) recently assessed the Cu, Zn, Al, Mn, Ni, Fe, Pb and As concentrations as well as the radioactivity of Minjiingu processing plant's tailings concluding that the radioactivity is above recommended levels, while heavy metals are of little concern.

The fact that there has been no recent systematic analysis of the present uranium and REEs content in Minjingu ore and related products led to this study, that for the first time aims to shed light on the unique composition of Minjingu ore as well as its products and discuss potential cleaner production pathways.

3. Sampling and sample analysis

Samples from 10 Minjingu phosphate ore layers, 4 Minjingu mine tailings, and 5 Minjingu fertilizer products were analyzed for rare earths and uranium contents using ICP-MS. In addition, Minjingu fertilizer powder was analyzed using X-ray fluorescence (XRF).

Globally, most (>90 %) phosphate ore is processed using the WPA process, during which the majority (>80 %) of the uranium naturally occurring in the phosphate ore transfers to the liquid WPA, from where it can be recovered using industrially proven solvent extraction processes (Beltrami et al., 2014; Singh et al., 2016), while the majority of REEs (>80 %) transfer to the solid phosphogypsum fraction, from which recovery is economically challenging (Bilal et al., 2023; Rutherford et al., 1994; Santos et al., 2006).

The Minjingu phosphate ore undergoes a distinctive processing method illustrated in Fig. 2 that is different from the popularly used WPA process. Following mining, the ore undergoes a concentration process where unwanted materials are manually removed through handpicking. The remaining material is currently spread on the ground for natural sun drying (Fig. 2b). Efforts are underway to implement a coal-fired rotary dryer, allowing for ore drying regardless of weather conditions (Fig. 2c). After two to three weeks of sun drying, the dried ore is fed into an impact crusher, preliminarily sieved, and then passed into a hot air furnace where it is heated to temperatures ranging from 700 to 800 °C (Fig. 2d). The dried ore undergoes another round of sieving before entering gravity classification. This beneficiation process stands out in comparison to other thermal methods employed for phosphate ore concentration (Abouzeid, 2008; Ruan et al., 2019).

Fertilizer powder, already rich in P_2O_5 content, is extracted after final sieving, as indicated in Fig. 2e. Following gravity classification, the fertilizer powder is transported by truck to a granulation plant where it is transformed into fertilizer granules. Finally, these granules are blended to create the final fertilizer products, which are then packaged and transported by truck to agro-traders for final distribution. Fig. 2 offers a concise overview of the Minjingu phosphate ore processing flow, with the green boxes indicating the points where samples were gathered for this study. Samples were obtained from the phosphate ore (prior to handpicking) (see Fig. 2a), the mine tailings (see Fig. 2d), the fertilizer powder (see Fig. 2e), the fertilizer granules, and three final fertilizer products (see Fig. 2f).

Ten 100 g dry samples from each of the nine phosphate ore layers as well as surface samples were collected, crushed, and powdered before being dried in an oven at 100°C for 24 h to a consistent weight. These materials were then homogenized in a pulverizer after being sieved using 150 μm standard sieves. For each sample, the pulverizer was run for 30 min at a speed of 150 rpm. The pulverizer used three spherical balls with a radius of 3 mm each. 50 g of each typical sample (L0-L9) were taken after homogenization and submitted to Morocco's National Centre for Energy and Nuclear Science and Technology (CNESTEN) for ICP-MS analysis.

The materials were digested at CNESTEN using a microwave SPEEDWAVE4 (Berghof Products + Instruments GmbH, Eningen unter Achalm, Germany) and a solution of 3 mL HNO_3 (60 %) and 5 mL HF (40 %) (Merck, Darmstadt, Germany). After cooling, the liquid was transferred to 50 ml flasks and filled up with high quality water until the appropriate volume was reached. Thermo Fischer Scientific's (Waltham, USA) XSERIES 2 ICP-MS was used to conduct the measurements. The spectrometer was tuned to produce the lowest CeO^+/Ce^+ and $Ba2^+/Ba^+$ ratios as well as the best analysis density. The National Institute of Standards and Techniques in Gaithersburg, Maryland, USA, provided "Western Rock Phosphate" to standardize the correlation coefficient for all calibration curves. Single-component CertiPrep SPEX solutions and lanthanoids from the ASTASOL combination (AN 9088 (MN)) were used for external calibration.

In the same way, two tailing samples and two surrounding soil samples (T1-T4) were prepared and analyzed. Samples from the fertilizer powder, fertilizer granules and final fertilizer products (F1–5) were digested directly and analyzed using ICP-MS. In addition, XRF analysis was conducted for the fertilizer powder (F1) at the Institute of Chemical Technology (ICT) in Mumbai, India.

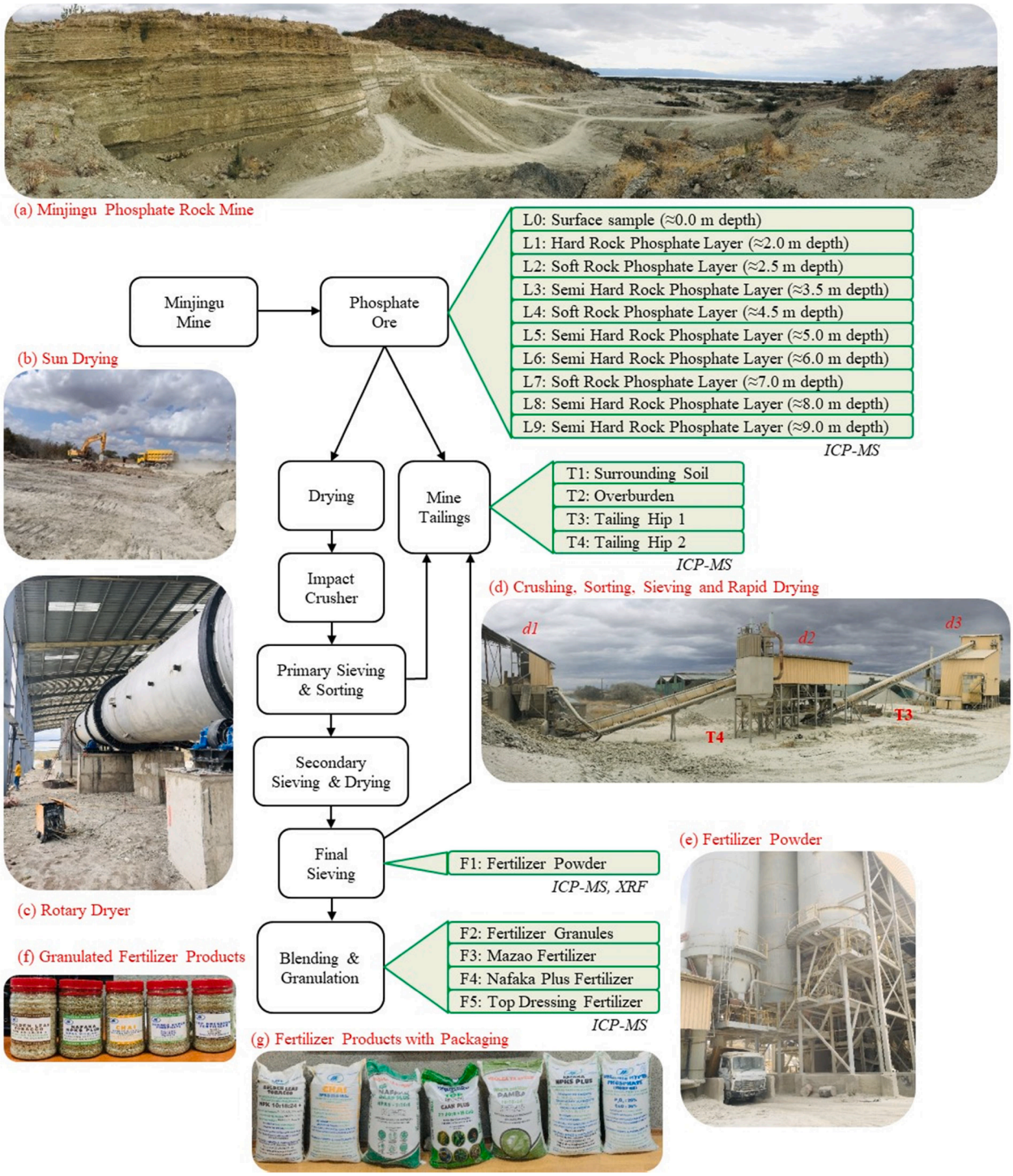

Fig. 2. Overview of Minjingu phosphate ore processing and sample selection.

4. Results and discussion

4.1. Rare earths concentrations

Table 1 shows the REE concentrations in Minjingu phosphate ore (L0-L9), surrounding soil (T1-T2), mine tailings (T3-T4) and fertilizer products (F1–F5) analyzed for this work. With regards to the REE concentration in Minjingu phosphate ore, considerable differences can be observed between the different rock layers from which samples were drawn (see details in Fig. 2). The fact that different rock layers show varying concentrations of elements is not unusual, and at the Minjingu mine, only soft phosphate rock layers L2, L4, and L7 are presently mined and further processed. With the foreseen fertilizer plant extension, it is also planned to mine hard phosphate rock layers that can show even higher P_2O_5 concentrations (>25 %) than the presently mined soft phosphate rock layers (usual P_2O_5 concentrations of 20–25 %). Even the highest concentrations of REEs found in layer L7 (614 ppm) are not exceptionally high if compared to REE concentrations found in other major phosphate rock deposits (see Fig. 3). Measured average REE concentrations of 236 ppm in the investigated rock layers, and average REE concentrations of 374 ppm in the presently mined phosphate ore layers do not encourage the commercial recovery of REEs. The average

Table 1
Rare earth element (REE) concentration in Minjingu phosphate ore, mine tailings, and fertilizer products measured in this study.

	Minjingu phosphate ore samples										Tailings				Fertilizer products				
	L0	**L1**	**L2**	**L3**	**L4**	**L5**	**L6**	**L7**	**L8**	**L9**	**T1**	**T2**	**T3**	**T4**	**F1**	**F2**	**F3**	**F4**	**F5**
Y	22	5	1	7	20	21	27	144	10	31	21	51	10	10	13	27	11	13	46
La	23	8	1	31	12	4	71	52	15	14	36	100	15	14	13	20	9	11	33
Ce	54	22	4	77	30	12	143	86	41	37	82	236	42	38	35	53	26	33	92
Pr	6	2	0	8	3	1	23	13	4	4	8	31	4	3	3	5	2	3	8
Nd	41	19	3	56	22	10	176	95	31	29	60	219	28	25	24	35	19	22	61
Sm	9	4	1	10	5	2	35	20	6	7	12	41	5	5	5	7	4	5	13
Eu	2	1	0	2	1	1	6	4	1	1	2	7	1	1	1	1	1	1	3
Gd	9	5	4	9	7	4	28	22	6	9	12	38	9	6	6	8	5	6	14
Tb	1	0	0	1	1	0	2	2	0	1	1	3	0	0	0	1	0	0	1
Dy	8	4	1	5	7	6	15	43	4	9	9	26	5	4	5	8	4	5	15
Ho	1	1	0	1	1	1	1	9	0	1	1	3	1	1	1	1	1	1	2
Er	5	4	1	2	5	6	5	57	2	9	6	12	3	3	4	7	3	4	11
Tm	1	0	0	0	0	1	0	6	0	1	0	1	0	0	0	1	0	0	1
Yb	6	4	1	1	4	5	3	58	3	12	5	10	3	3	4	8	4	4	13
Lu	1	0	0	0	0	0	0	5	0	1	0	1	0	0	0	1	0	0	1
SUM	**188**	**79**	**15**	**209**	**118**	**74**	**537**	**614**	**125**	**166**	**256**	**776**	**127**	**113**	**115**	**184**	**91**	**108**	**315**
Av.	**236**										**-**		**120**		**163**				

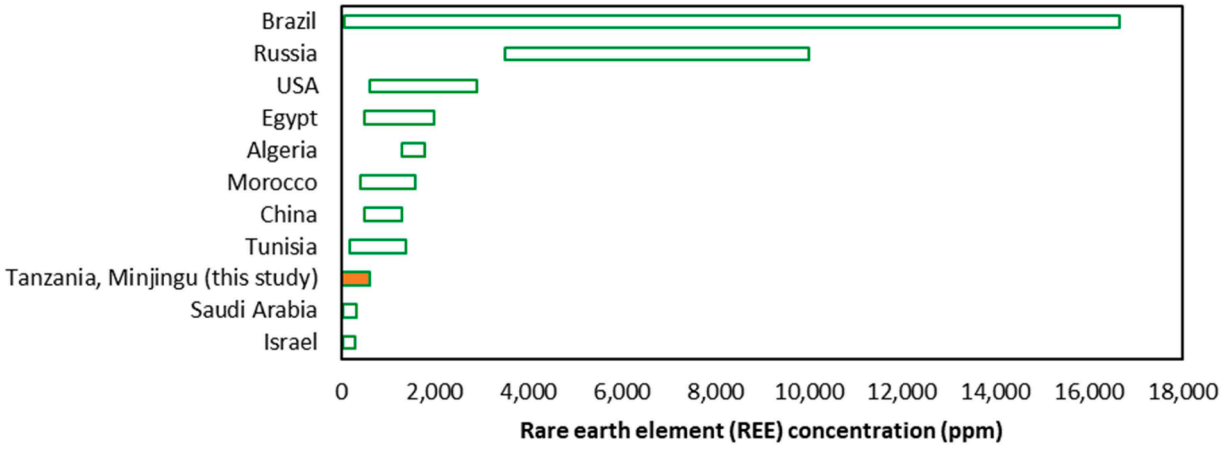

Fig. 3. Rare earth element (REE) concentrations in different phosphate ores. Data sources: Abbes et al. (2020); Aly and Mohammed (1999); Awadalla (2010); Christmann (2014); De Oliveira and Imbernon (1998); El-Zrelli et al. (2021); Habashi (1985); Kandil et al. (2010); Khater et al. (2016); Nie et al. (2013); Orabi et al. (2018); Pavón et al. (2022); Ramos et al. (2016); Roshdy et al. (2023); Soudry et al. (2002); Wang et al. (2010); Wu et al. (2018a); Zaitsev et al. (2015); Zhang (2014); Zielinski et al. (1993).

REE concentration in the mine tailings of 120 ppm is even lower and similarly does not encourage REE extraction, although it should be mentioned that this material is already mined, ground, and stacked so that it is easily accessible and a large portion of the costs for mining are already covered as illustrated by Reitsma et al. (2018). The 776 ppm measured in the overburden (T2) is slightly higher, but does not seem to be a good representation of the overall Minjingu deposit, nor do we expect to find this REE concentration in large amounts of material on site.

In Fig. 3, the REE concentrations found in the Minjingu phosphate ore are compared to the REE concentrations found in other phosphate ores. REE concentrations in phosphate ores vary but are usually not higher than 1 % or 10,000 ppm (Habashi, 1985), with the exception of the Catalão Minerochemical Complex in Brazil for which Ramos et al. (2016) report, supported by independent analysis from De Oliveira and Imbernon (1998), REE concentrations as high as 16,650 ppm in selected samples. This is highly unusual, and most phosphate ores globally (particularly since most phosphate ores are of sedimentary origin) show concentrations of approximately 0.05 % or 500 ppm.

It can be noted that the REE concentrations in Minjingu phosphate ore are not particularly high and will most likely not make REE recovery an economical option. Similar observations were made for the analyzed mine tailings as well as the fertilizer products. In some of the processes for uranium recovery from phosphates, a large share of the REEs, or lanthanides, that are chemically very similar to the actinides, are coextracted together (Habashi et al., 1986; Wu et al., 2018b), so that if such a process is applied REEs in Minjingu phosphate ore might also become a target for recovery (see more discussion on this in Chapter 4.4).

It is noteworthy that besides REEs, other valuable trace elements could potentially be co-recovered, as pointed out by Chen and Graedel (2015). Most notably, we measured vanadium concentrations of 21–201 ppm in the investigated samples. This concentration is again relatively low if compared to the concentration of primary vanadium ores in Africa (Boni et al., 2023), but co-recovery could again be profitable as a large share of the costs for mining and processing is already covered through the sales of fertilizer products.

It is further noteworthy that there is a much lower push-factor for REE extraction from phosphate ores and phosphate fertilizers than there is for cadmium and uranium, with some fertilizers even being enriched with REEs in an attempt to make them work more effectively (Silva et al., 2019; Ribeiro et al., 2021; Tommasi et al., 2020). Furthermore, REE extraction from phosphates (usually REEs are recovered from phosphogypsum) was not successfully practiced on an industrial scale historically (Diwa et al., 2022; Ramirez et al., 2022), as it was the case with uranium recovery (from WPA), so there are considerably higher

techno-economic risks associated with commercially recovering REEs from phosphates.

4.2. Uranium concentration in phosphate ore

Fig. 4 shows the reported uranium concentrations in phosphate ores globally. Here the situation is different, and it can be clearly seen that phosphate ore from Minjingu shows the highest concentrations of uranium. New Zealand, which is listed in Fig. 4, is not a major phosphate rock-producing country, and the reported uranium concentrations are presently more of a geological interest. Adam et al. (2014) reported uranium concentrations of 1055 ppm for the Uro phosphate deposit in Sudan that were not considered since they seem inflated and are not supported by other studies of the deposit (AbowSlama et al., 2014; Elmahdi and Wagialla, 2018). Besides, phosphate-uranium deposits that are not mined such as the Itataia deposit in Brazil that can show uranium concentrations of 816 ppm (Veríssimo et al., 2016) or the Matongo deposit in Burundi that can show uranium concentrations of 632 ppm (Mwalongo et al., 2023a; Songore, 1991) were not considered as it is not clear yet if they can actually be used for fertilizer production.

4.3. Uranium concentration in phosphate fertilizers

Extended use of phosphate rock and phosphate fertilizer results in uranium build-up on agricultural soils, which is actively discussed by the scientific community (Schnug and Lottermoser, 2013; Sun et al., 2020a, b, Takeda et al., 2006). There is particular concern regarding uranium accumulation since the metal is radiotoxic and can endanger soil fertility, leach into groundwater (Liesch et al., 2015; Mathivanan et al., 2022), and be taken up by crops (Bigalke et al., 2017; Chen et al., 2021; Singh et al., 2005; Yamaguchi, 2009). At present, there is no legal limit for uranium in mineral fertilizers, though some organizations, such as the Soil Protection Commission of the German Federal Environment Agency (UBA), suggest formulating a legal limit. Kratz et al. (2016)

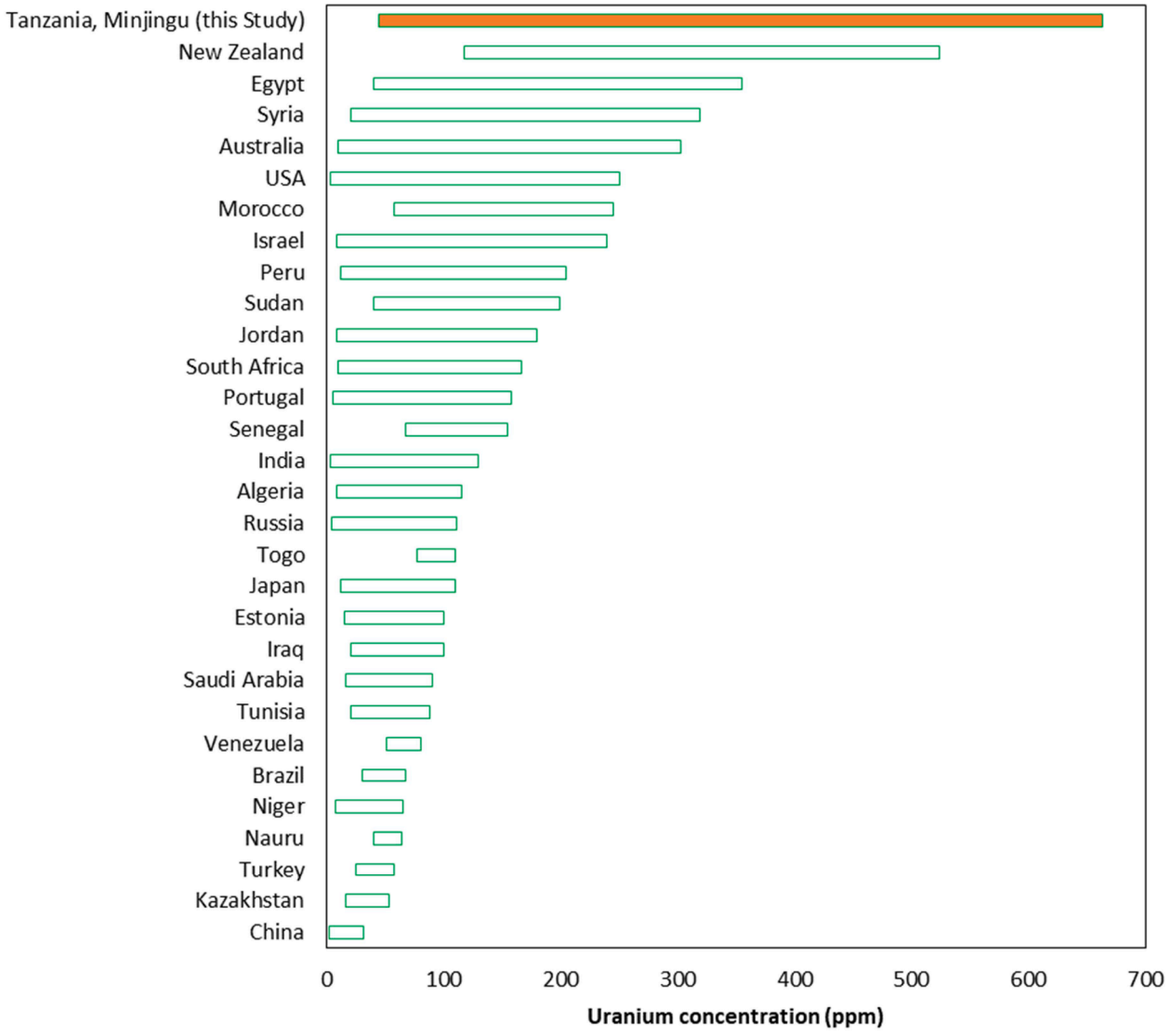

Fig. 4. Reported uranium concentrations in phosphate ore. Data sources: Abbes et al. (2020); Abed (2011); AbovSlama et al. (2014); Al-Bassam (2007); Al-Eshaikh et al. (2016); Altschuler et al. (1958), 1966, 1980; Azouazi et al. (2001); Banerjee et al. (1982); Baturin and Kochenov (2001); Bech et al. (2010); Bouabdallah et al. (2019); Bunus (2000); Cevik et al. (2010); Cook et al. (1990); Cook (1972); Chandrajith and Dissanayake (2009); Dar et al. (2014); El Bamiki et al. (2021); Falck and Wymer (2006); Gnandi and Tobschall (2003); Haneklaus et al. (2015); Hayumbu et al. (1995); Howard and Hough (1979); Jallad et al. (1989); Khater et al. (2016); Krea and Khalaf (2000); Menzel (1968); Onal and Atak (1992); Palattao et al. (2018); Pantelica et al. (1997); Parker (1984); Roselli et al. (2009); Ryszko et al. (2023); Saad et al. (2003); Sattouf (2007); Schnug et al. (1996); Scholten and Timmermans (1995); Shlewit and Alibrahim (2008); Soudry et al. (2002); Sun et al. (2020); Syers et al. (1986); Van Kauwenbergh (1997); Vogel et al. (2020); Weterings and Janssen (1985); Zanin and Zamirailova (2007).

report that the legal limit (50 mg uranium per kg P_2O_5 for P-containing fertilizers) suggested by UBA was exceeded by all 161 phosphate fertilizer samples the researchers investigated. Further analysis of 414 phosphate fertilizers used in Europe (Verbeek et al., 2020), reported similar uranium concentrations but are still somewhat lower (all between 10 and 110 ppm for Germany by Kratz et al. (2016) and 0–242 ppm with a mean concentration of 41 ppm and a median at 23 ppm for the EU-27 + Norway and the United Kingdom by Verbeek et al. (2020)) than the uranium concentrations measured here in Minjingu phosphate ore products, which are roughly by a factor of 1.5 higher compared to the uranium concentrations measured in Europe. It is further noteworthy that the concentrations provided in Fig. 5 are often the result of radioactivity measurements and that the P_2O_5 concentration as well as other important factors of the fertilizers were not reported.

4.4. Alternative processing of Minjingu phosphate ore

Fig. 6 shows a direct comparison between the dry beneficiation process currently used to develop Minjingu phosphate ore (Fig. 6a) and the very different WPA process that is usually deployed to process phosphate ore (Fig. 6b). Globally, less than 0.5 % of mined phosphate ore is used directly on agricultural soil after simple beneficiation (Zhang, 2014). Both processes are simplified here, and the intention is to indicate how different they are and what the challenges of integrating uranium recovery in a dry phosphate ore beneficiation process may be.

It is unlikely that Minjingu phosphate ore will be processed using the WPA process or another wet-chemical process. The WPA- and, by extension, other chemical processes require significant amounts of water in addition to sulfuric or other acids. The Minjingu mine and processing plant are located in a semi-arid region some 550 km away from the closest seaport, making the supply of large amounts of water challenging. In addition, using the WPA process results in considerable amounts of fine-powdery phosphogypsum. If phosphate ore with low radioactivity and heavy metal concentrations is used to produce fertilizers, the resulting phosphogypsum can be utilized as a secondary raw material in construction (Rashad, 2017; Haneklaus et al., 2022) or in agriculture (Alcordo and Rechcigl, 1993). Since the Minjingu ore contains considerable amounts of uranium, this would not be the case, so that the overall environmental footprint of any wet-chemical process (even with uranium recovery) will most likely be significantly larger than that of the current dry beneficiation process (without uranium recovery).

During the WPA process with sulfuric acid (Fig. 6b), uranium can be recovered from the liquid WPA using solvent extraction. This process has been used on an industrial scale in the past (Astley and Stana, 2014) and has been well documented in several authoritative reviews (Beltrami et al., 2014; Bunus, 2000; Singh et al., 2016).

Minjingu phosphate ore may profit from alternative dry processing such as electrostatic separation as described by Bittner et al. (2015) as well as Sobhy and Tao (2014), the improved hard process (IHP) presently under development in Florida (Walters, 2011), or other innovative dry concentration processes that have recently been reviewed by Sajid et al. (2021). It is noteworthy that the present process, though simple in its design, works extremely well. Calcination is energy-intensive, so reducing energy inputs, by introducing solar thermal calcination as proposed by Haneklaus et al. (2017c,d, 2021b) and recently investigated to pilot-plant scale by the EU SOLPART project (Esence et al., 2020; Moumin et al., 2019) might be a long term proposition for the Minjingu plant that is located in an area that offers the required high levels of solar radiation.

More importantly, uranium recovery should be considered during the processing of Minjingu phosphate ore as a result of the relatively high occurring concentrations that could make recovery economically profitable. Comparable high concentrations of REEs and other valuable elements could not be confirmed in this work. It is noteworthy, that although the concentrations of uranium can be considered elevated in the Minjingu ore (approximately 3x of more common concentrations), the resulting compound fertilizers do not show significantly higher (approximately 1.5x more common concentrations) uranium concentrations. Since traditionally uranium is recovered from the liquid WPA during wet chemical processing, new innovative solutions for uranium recovery during Minjingu phosphate ore beneficiation should be investigated. Theoretically, the uranium could be directly leached from the phosphate ore after primary sieving and sorting before calcination or from the fertilizer powder after calcination.

Abilash et al. (2009, 2013) and Mäkinen et al. (2019) proposed, for instance, bioleaching of a lower-grade uranium apatite ore in India and Finland. Al-Khaledi et al. (2019) and Roshdy et al. (2023) propose direct leaching of REEs and uranium from Egyptian phosphate ore and Guzmán et al. (1995) were probably the first to propose such an approach for phosphate ores from Mexico with the explicit aim of reducing dissipation of radiotoxic uranium. Gabriel et al. (2013a, b) rightfully point out that direct leaching of uranium from phosphates is presently not economically profitable, and this will most likely also be the case at the Minjingu fertilizer plant, given the relatively small size of the operation, as well as still fairly low uranium prices worldwide.

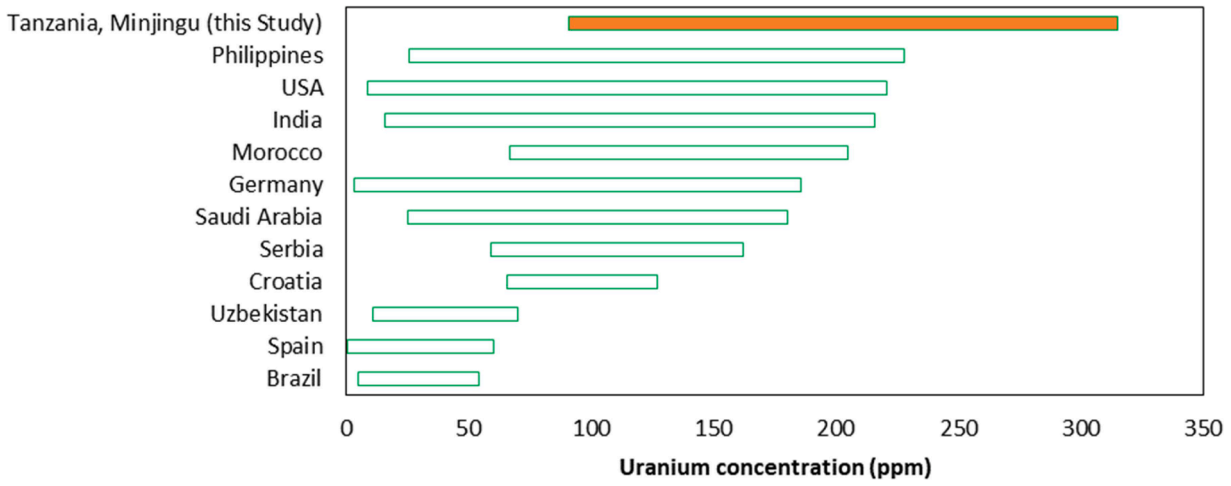

Fig. 5. Reported uranium concentrations in phosphate fertilizers. Data sources: Al-Shawi and Dahl (1995); Barisic et al. (1992); Hamamo et al. (1995); Khater, 2011; Khater and Al-Sewaidan (2008); Kratz et al. (2016); Lal et al. (1985); Otero et al. (2005); Palattao et al. (2018); Pfister et al. (1976); Qamouche et al. (2020); Ramteke et al. (2022); Verbeeck et al. (2020); Vucic and Ilic (1989); Yamazaki and Geraldo (2003).

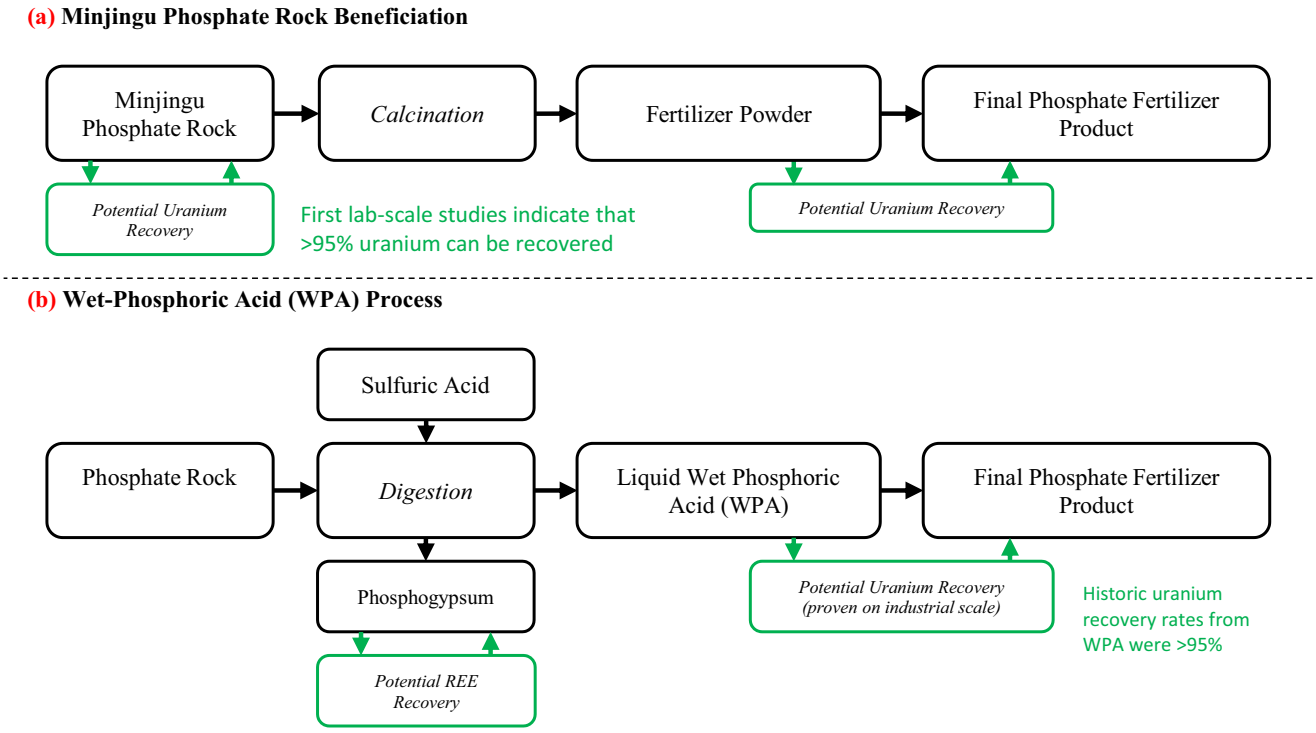

Fig. 6. Comparison of (a) the present Minjingu phosphate rock beneficiation process and (b) the wet phosphoric acid process with sulfuric acid that is used at more than 85 % of phosphate fertilizer plants globally.

4.5. Importance of Minjingu phosphate fertilizer products

It is essential to realize that Minjingu phosphate rock, and its fertilizer products substantially increase crop yield in Tanzania, as may best be illustrated in Fig. 7 using data from the Global Forum on Agricultural Research and Innovation (GFAR, 2018). The domestically produced Minjingu fertilizer products are (especially given the national subsidies in Tanzania) an affordable source of domestically produced fertilizer that works well on the acidic soils found in the country, decreasing the dependency on the international fertilizer market (Mew et al., 2023). If uranium is to be recovered from Minjingu phosphate ore during mineral fertilizer production, it is paramount that the quality of the final fertilizer product is not compromised.

5. Conclusions and policy recommendations

This work provides for the first time a systematic overview of Minjingu phosphate ore processing in Tanzania and puts the REE and uranium concentrations found in the different ore layers, tailings, and fertilizer products in context, by comparing them to concentrations of other phosphates found globally. It was found that the REE concentrations in Minjingu phosphate ore are too low to justify economic recovery. The already suspected elevated uranium concentrations in the Minjingu phosphate ore could be confirmed by the systematic analysis presented here, and elevated uranium concentrations were further found in the final fertilizer products, though in lower relative concentrations than in the ore. It could further be shown that there are

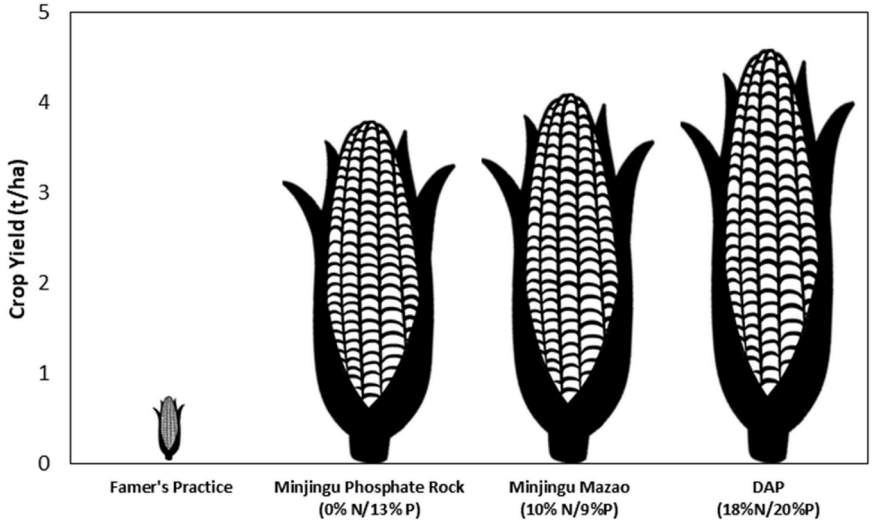

Fig. 7. Comparison of the crop yield in a village in Tanzania with no mineral fertilizer (Farmer's Practice), Minjingu Phosphate Rock, Minjingu Mazao and DAP fertilizers.

considerable differences between the different ore layers analyzed here as well as literature data on Minjingu ore. All this results in considerable uncertainties and limitations of this and other studies. Minjingu phosphate fertilizer products are important for the development and food security of Tanzania, and it is crucial that the final fertilizer products work as effectively as they do now. Despite this, it is still a good idea to look into ways to make the production of Minjingu fertilizer more sustainable, such as solar calcination or reducing dust and uranium recovery without affecting the quality of the final fertilizer products.

Declaration of Generative AI and AI-assisted technologies in the writing process

The authors declare that no generative AI and AI-assisted technology was used in the writing process.

CRediT authorship contribution statement

Nils H. Haneklaus: Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Visualization, Writing – original draft, Writing – review & editing. **Dennis A. Mwalongo:** Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Writing – review & editing. **Jacob B. Lisuma:** Formal analysis, Writing – review & editing. **Aloyce I. Amasi:** Formal analysis, Writing – review & editing. **Jerome Mwimanzi:** Formal analysis, Writing – review & editing. **Tomislav Bituh:** Formal analysis, Writing – review & editing. **Jelena Ćirić:** Formal analysis, Writing – review & editing. **Jakub Nowak:** Formal analysis, Writing – review & editing. **Urszula Ryszko:** Formal analysis, Writing – review & editing. **Piotr Rusek:** Formal analysis, Writing – review & editing. **Ali Maged:** Formal analysis, Writing – review & editing. **Essaid Bilal:** Formal analysis, Funding acquisition, Writing – review & editing. **Hajar Bellefqih:** Formal analysis, Writing – review & editing. **Khaoula Qamouche:** Formal analysis, Writing – review & editing. **Jamal Ait Brahim:** Formal analysis, Writing – review & editing. **Redouane Beniazza:** Formal analysis, Funding acquisition, Writing – review & editing. **Hamid Mazouz:** Conceptualization, Formal analysis, Funding acquisition, Writing – review & editing. **Elizabet M. van der Merwe:** Formal analysis, Funding acquisition, Writing – review & editing. **Wayne Truter:** Formal analysis, Funding acquisition, Writing – review & editing. **Hilda D. Kyomuhimbo:** Formal analysis, Writing – review & editing. **Hendrik Brink:** Formal analysis, Funding acquisition, Writing – review & editing. **Gerald Steiner:** Formal analysis, Writing – review & editing. **Martin Bertau:** Formal analysis, Writing – review & editing. **Raghav S. Soni:** Formal analysis, Writing – review & editing. **Ashwin W. Patwardhan:** Formal analysis, Writing – review & editing. **Pushpito K. Ghosh:** Formal analysis, Writing – review & editing. **Thomas T. Kivevele:** Formal analysis, Writing – review & editing. **Kelvin M. Mtei:** Formal analysis, Writing – review & editing. **Stanisław Wacławek:** Formal analysis, Writing – review & editing.

Declaration of competing interest

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

Data availability

Data will be made available on request.

Acknowledgments

This work was supported by the Tanzania Atomic Energy Commission (TAEC) and Nelson Mandela African Institution of Science and Technology (NM-AIST). This work further received support from the Austrian Federal Ministry of Education, Science and Research (BMBWF) through Austria's Agency for Education and Internationalization (OeAD) [Grant Numbers: Africa UNINET P006, P056, and P058; APPEAR Project 341; CZ 12/2024; FR 12/2024; HR 14/2024; KOEF 01/2019; PL 12/2024]. Khaoula Qamouche further acknowledges BMBWF/OeAD support through Ernst Mach Grant MPC-2023-00562. We thank the participants of the International Symposium on Uranium Raw Material for the Nuclear Fuel Cycle (URAM 2023) for valuable comments that further improved this work.

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