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REVIEW

Rare Earths as Emerging Trace Element Contaminants in the Soil

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Abstract

Purpose of Review This review examines recent publications on rare earth elements (REE) in soils, critically evaluating their role as emerging soil contaminants. We emphasized new findings and main gaps using a previous review paper published in 2016 by our research group as a reference point. Three major subjects were prioritized: (1) sources, background levels, and behavior of REE in soils; (2) plant development and metabolism as affected by REE exposure; and (3) environmental and human health risk assessments of REE in the soil environment.

Recent Findings Publications addressing the occurrence and fate of REE in the soil environment have more than tripled in the last decade. Coincidentally, global REE exploration has more than doubled in the past 7 years. Because of their unique features, the global demand for REE is expected to increase by at least 50% in the next 10 years. As soils are the main sink of contaminants, we must continue to investigate the consequences of the unceasing addition of these elements in soil ecosystems.

Summary We highlighted the main sources of REE, their background levels in selected global soils, and their physicochemical behavior. The relationship between REE and plants revealed potential benefits such as environmental stress tolerance. Finally, ecological and human health risk assessment data for REE in soils were carefully discussed in terms of their potential adverse effects on biota. We conclude with a survey in which prominent authors working with REE answered questions about challenges and opportunities for innovative research on REE in soil-plant-animal/human systems.

Keywords Rare earth elements · Ecological risk assessment · Human health risk assessment · Agriculture · Environment

Introduction

Rare earth elements (REE) are used in several modern industries, including chemicals, electronics, clean energy, transportation, health care, aviation, defense, and agriculture [1••]. One of the consequences of their growing use is the need for increasing the exploration of REE worldwide. In fact, the world mine production of REE has almost tripled in the last 7 years (*i.e.*, from 2017 to 2023), when the production changed from 132,000 to 350,000 metric tons of rare-earth-oxide (REO) equivalent [2, 3]. China alone accounted for nearly 70% of the total global production in 2023. In spite of the high abundance of REE in the Earth's crust, their minable reserves are more scarce than for most other minerals [3]. The total world reserves are estimated at 110,000,000 metric tons REO equivalent, with more than 95% of that occurring in China (44,000,000), Vietnam (22,000,000), Brazil (21,000,000), Russia (10,000,000), India (6,900,000), and Australia (5,700,000) [3].

The escalating exploration and use of REE worldwide have also raised an ever-increasing concern about the effects of cumulative concentrations of these elements in the environment. Indeed, publications examining the occurrence and fate of REE in the soil environment have more than tripled in the last 10 years, yet the highest advance occurred after 2016

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when the number of peer-reviewed documents concerning this topic increased from < 100/year (up to 2016) to > 200 in 2022. A search on August 20, 2023, within "*Article title, Abstract, Keywords*" on Scopus using the terms "rare earth elements" AND "soil" revealed 2122 documents published since 1929. About two-thirds of those documents were published in the last 10 years and 50% came out in the past 5 years. When classifying these documents by subject area, Earth and Planetary Sciences and Environmental Science comprised 25.1% each, followed by Chemistry (11.0%), Agricultural and Biological Sciences (10.5%), Medicine (5.4%), and Materials Science (3.6%).

This review paper investigates relevant literature published recently on the occurrence and fate of REE in soil-plant-animal/human systems, critically evaluating documents assessing the role of REE as emerging soil contaminants. We highlight new findings, as well as major gaps on this subject, revisiting a review published by our research group (> 200 citations since 2016) [1••] as a reference point. Briefly, the procedure used to select the most prominent publications (with a focus over the past 3–5 years) was as follows: a Scopus search was conducted using the following keywords: "rare earth elements", "lanthanide", "lanthanum", and "cerium" in combination with "soil", "plant", "agriculture", "fertilizer", "ecotoxicology", or "environment". With a few exceptions, only publications from 2017 or later were considered. To facilitate the screening process for selecting pertinent publications within each section of this review, a semantic search was executed. This involved reordering the publications based on the semantic similarity (measured by cosine similarity) of their abstracts to each section of the mentioned review [1••]. The embedding model "text-embedding-ada-002" from the OpenAI API was employed for this purpose.

Three comprehensive topics are covered by this review, as follows: (1) recent data concerning sources, background levels, and behavior of REE in soils; (2) REE in plants: development and metabolism; (3) environmental and human health risk assessments for REE in the soil environment. We conclude with a summary of information gathered from a survey in which some of the most prominent authors working with REE worldwide ($n = 40$) kindly answered selected questions about challenges and opportunities for innovative research on REE in soil-plant-animal/human systems.

Sources, Background Levels, and Behavior of REE in Soils

Anthropogenic Sources of REE

Important advances have been made regarding the investigation of anthropogenic REE sources to the soil in recent years. For instance, a major concern about the large use of REE in the technology industry was the threat of REE contamination from electronic waste. However, despite the high content of REE in electronic devices, these elements exhibited very low mobility under natural soil physicochemical conditions (< 1% of the total REE leached after a 30-day period), except in very acidic media (pH < 2); therefore, electronic waste (fragments of electronic devices) disposed of in the soil environment poses a negligible risk source of REE contamination [4].

Several studies have also explored the role of mining activities on REE contamination. REE mining, for instance, can result in severe contamination of surrounding soils, but this effect decreases sharply with distance from the mine pit [5]. On the other hand, REE mining can result in diffuse contamination of a much larger area when the mine tailings are rich in exchangeable REE, favoring REE leaching to freshwater reservoirs and leading to contamination of farmlands irrigated with the contaminated water [6]. Conditions that favor REE leaching include acidic media, low cation exchange capacity (CEC), low organic matter (OM) content, and competition for adsorption sites [7, 8].

Coal mining and combustion have also been pointed out as important sources of REE contamination because of the elevated REE concentrations found in coal and their corresponding ashes. Nonetheless, in Southern Brazil, only Gd and Ho showed significant accumulation among all REE in soils close to coal mine areas [9], while a slight to moderate soil contamination was found in South Africa, with a mean enrichment factor of each REE higher than 1.5 [10]. The recovery of REE from coal combustion products [11, 12] or from coal mine drainage [13] has been proposed to reduce the REE contamination threat from coal use and mining, as well as an environmentally friendly source of REE.

Although less considered, road traffic represents a relevant source of REE. In urban park areas, this source of contamination has been pointed out as the most important anthropogenic source of REE, together with solid wastes from industrial activities [14]. REE can be found in asphalt (ΣREE: 38 ± 7 mg kg^{-1}), brake pads (ΣREE: 50.8 ± 17.7 mg kg^{-1}), and tires (ΣREE: 7.52 ± 2.84 mg kg^{-1}), with most of the REE released into the environment from road traffic due to asphalt abrasion [15]. This type of REE enrichment decreases significantly with increasing distance from the road (up to about 25 m) [16] and increases with traffic intensity [15]. Even close to mining sites, asphalt abrasion and wear on vehicle parts can represent significant sources of REE contamination in urban areas [5].

Finally, agricultural activities are among the most important diffuse anthropogenic sources of REE to soil due to the presence of these elements as contaminants in fertilizers (especially phosphate fertilizers) and amendments [17]. However, REE concentrations in phosphate fertilizers and correctives are highly variable depending on the source, with reported variations of up to two orders of magnitude. In this sense, for samples of single superphosphate marketed in Brazil, one of the largest food producers in the world, Ce mass fractions reported in the literature are as low as 35 mg kg^{-1} [18] or as high as 3934 mg kg^{-1} [17], a 100-fold difference for the same type of phosphate fertilizer. The same is true for soil amendments like gypsum, with reported ΣREE concentrations as low as 4.6 mg kg^{-1} [18] or as high as 5000 mg kg^{-1} [17], a difference of about 1000 times. Liming may also result in REE contamination but reported ΣREE concentrations in limestone used in Brazil are relatively low (average of 27.1 mg kg^{-1} and standard deviation of 6.6 mg kg^{-1} across 10 samples) [18]. Data on the bioavailability of REE from agricultural inputs are still scarce, but the solubility of REE in phosphate fertilizers seems also to depend on the raw material, with fertilizers derived from phosphorite having soluble fraction of LREE (light REE) and HREE (heavy REE) as high as 52% and 50%, respectively; whereas those derived from carbonatite have LREE and HREE solubility as low as 15% and 30%, respectively [18].

Despite the high potential of unintended REE addition to farmland soils due to the REE richness in agricultural inputs [19, 20], the maintenance of these elements in the surface layer of the soil depends on several factors. REE may have a significant potential for leaching, especially HREE, from the upper to the deeper soil layers [21]. Evidence of REE mobility in the soil was provided by Neves et al. [22], who studied the REE accumulation in several layers of a cultivated and a non-cultivated soil and evidenced that REE contents were around five-fold higher in deeper layers (> 10 cm) in the cultivated soil.

Soil Background Levels of REE

The soil environment is recognized as either a source or a sink of contaminants, especially potentially toxic elements (PTE) both from geogenic origin as a result of weathering of parent materials and by the dispersion promoted by anthropic activities (*e.g.*, mining, transportation, industrial, and agricultural practices), as well as by the disposal of domestic and industrial wastes [23–27]. Soil is also referred to as a crucial medium in the ecosystem since it serves as a dispersion agent of PTE to water reservoirs, biota, and atmosphere [28]. Thus, given the recent trend of increased use of REE in product manufacturing experienced in modern society, the availability of reliable data from soil background levels of these elements is a topic of utmost relevance.

The establishment of background levels is essential to evaluate and monitor soil contamination. However, some points must be carefully considered as they are important analytical pitfalls that can produce unreliable data. For example, the lack of certified reference materials, proper choice of instrumental operating conditions, matrix effects, and spectral interferences must be circumvented in order to produce such accurate data by following rigorous QA/QC protocols. In this sense, the most used spectroanalytical methods in recent investigations that have dealt with the determination of REE concentrations in worldwide soils [29–35, 36•, 37–40] are Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and Inductively Coupled Plasma Optical Emission Spectrometry (ICP OES). In the case of ICP OES, the challenges in the REE quantification are well-documented, especially in the analysis of samples enriched with Ca, Fe, K, and Na [41], which can be found at very high levels in the soil matrix. Thus, to achieve reliable results, a meticulous selection of the emission lines, the application of internal standardization for compensating non-spectral interferences, and even inter-element correction measures must be adopted [42]. On the other hand, ICP-MS has distinguished features that qualify it as the method of choice in most REE studies, such as the superior analytical sensitivity (reaching detection limits in the ng L^{-1} range), wide linear calibration range, and modern strategies to correct isobaric interferences [43–46].

Table 1 depicts background levels of REE in selected worldwide soils encompassing different methods of calculation. A particularly relevant initiative on this subject was conducted by a European consortium of research agencies named GEMAS (Geochemical Mapping of Agricultural Soil) [36•]. GEMAS established geochemical background and threshold levels for 53 elements (Ce, La, and Sc included) in soils from 33 European countries ($n = 2108$ samples), representing 5.6 million km^2 (one sample per 50×50 km grid). Some important recommendations highlighted by the GEMAS project can be of interest to the REE research community, such as (i) increase the sample density in sites at the neighborhood of relevant emission sources (*e.g.*, major cities, power plants, industries, and smelters); (ii) incorporate the data produced by the several high-density local soil geochemical surveys to the GEMAS dataset — this strategy can be adapted and directly applied by "continental countries", such as Russia, Canada, China, the USA, and Brazil; (iii) monitoring of diffuse pollution at the continental scale should be focused on selected sites presenting low natural levels; (iv) in order to improve the understanding of the effects resulted from natural conditions *versus* anthropic impacts and the biogeochemical cycling of the target elements, a holistic approach must be adopted including sampling of different matrices (rock, soil, water, plants) in sites that can represent the variable natural conditions of the continent.

Table 1 Background levels of REE in selected worldwide soils

Background levels*	Calculation method/ number of samples	Ce	La	Pr	Nd	Sc	Y	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Continent/Country	Reference
GB	Mean ($n = 5$)^{a}	8.10	3.40	0.90	3.70	0.60	8.60	1.10	0.30	1.40	0.20	1.50	0.30	1.00	0.20	1.40	0.30	Brazil (State of Pará, Amazon region)	[35]
GB	Mean ($n = 33$)^{b}	24.2	15.2	5.03	17.1	-	-	4.40	0.03	0.15	1.21	1.31	0.55	2.36	0.021	1.86	0.77	Cuba (13 out of the 15 Cuban provinces)	[29]
GB	Median ($n = 70$)^{a}	86.05	53.45	11.85	36.15	-	19.25	7.50	1.50	5.65	1.00	5.35	0.90	2.55	0.40	2.70	0.30	Romania (Eastern)	[32]
QRV	90th percentile of background variation ($n = 11$)^{a}	145.63	66.06	11.67	73.0	20.88	43.83	8.58	1.72	8.35	0.79	3.11	0.44	1.22	0.13	0.77	0.13	Brazil (Solimões river basin, Amazon region)	[31]
QRV	90th percentile of background variation ($n = 13$)^{a}	41.9	15.85	2.94	7.85	7.38	4.36	1.92	0.40	2.39	0.29	1.16	0.21	0.56	0.07	0.35	0.05	Brazil (Negro River basin, Amazon region)	[31]
GB	Mean ($n = 6$)^{a}	85.0	43.3	8.87	33.5	-	13.15	5.46	1.35	4.85	0.60	3.24	0.61	1.76	0.23	1.54	0.22	Brazil (northeast)	[30]
GT	98th percentile of background variation ($n = 2108$)^{a,b}	81.0	42.0	-	-	7.40	-	-	-	-	-	-	-	-	-	-	-	33 European countries	[36•]
GT	Cumulative probability diagram ($n = 2108$)^{a,b}	150	60.0	-	-	8.00	-	-	-	-	-	-	-	-	-	-	-	33 European countries	[36•]
GT	Tukey's inner fence ($n = 2108$)^{a,b}	118	57.0	-	-	15.0	-	-	-	-	-	-	-	-	-	-	-	33 European countries	[36•]
QRV	75th percentile of background variation ($n = 100$)^{b}	57.26	27.21	10.49	24.29	3.70	10.65	4.75	0.90	4.22	0.82	1.54	0.38	1.23	-	1.07	0.24	Brazil (northeast)	[37]
GB	Mean ($n = 12$)^{a}	36.8	17.83	4.41	16.60	3.34	8.57	3.51	0.61	2.89	0.38	1.98	0.39	0.98	0.16	0.81	0.14	Russia (Central Forest State Nature Biosphere Reserve)	[47]
GB	Mean from data of the most superficial soil profiles ($n = 4$)^{a}	64.4	31.5	7.56	30.0	-	-	5.70	1.15	4.94	0.79	4.85	0.95	2.74	0.40	2.63	0.43	Argentina (Central)	[34]
GB	Geometric mean (Nearly 10% of all samples, $n = 12{,}400$)^{b}	65.0	37.0	6.70	25.0	10.5	22.0	4.90	0.98	4.40	0.58	3.90	0.83	2.40	0.35	2.30	0.35	China (all provinces along the mainland country)	[48]

*Geochemical background (GB) indicates the concentration associated with natural processes and not influenced by human activities [49]; geochemical threshold (GT) represents the outer limit of the geochemical background variation [49]; quality reference value (QRV) is based on natural (background) variation and is typically used for legal purposes [50]

[a] ICP-MS

[b] ICP OES

Behavior of REE in Soils

Soil attributes such as clay content, pH, OM, redox potential, and mineralogy have a strong influence on the REE behavior in soils [40, 51–53]. Organic matter may have a high potential for REE retention at negative surface charges, which varies depending on its type and amount. On the other hand, OM can modify the leaching solution pH, favoring the mobilization of REE [54]. The bioavailability of REE is strongly controlled by soil pH and high REE bioavailability is commonly found in acidic soils [9, 40].

Clay minerals, oxyhydroxides, phosphates, and OM are also key factors governing the fractionation and chemical speciation of REE in the soil, thus playing a critical role in the risk assessment of REE in contaminated sites [55, 56]. For example, a geochemical speciation model predicted that dissolved organic matter in solution complexed a significant proportion (6–73%) of total REE and reduced the bioavailability of Nd and Gd [57]. Contrariwise, Yb was predicted to precipitate in the form of $Yb(OH)_3(s)$ [57]. Regarding fractionation, studies suggest that REE mainly existed in the residual phase in alluvial soils in China [55], tropical soils of the Brazilian Amazon [58], and natural soils and mine tailings in China [59]. It is worth mentioning that some activities, such as mining, can modify the distribution and chemical speciation of REE [59].

REE behavior in soils is strongly related to clay type and content [1••] and important mechanistic insights can be indirectly derived from studies on REE adsorption to lateritic clay minerals, especially kaolinite and halloysite. These two minerals have been extensively studied because of their relevance on the formation of regolith-hosted ion-adsorption deposits and for their economic importance [60–62]. A recent comprehensive review on metal ions (including REE) retention in clay-rich materials can be found in the study of Liu et al., 2022 [63]. An investigation revealed that REE are preferentially adsorbed at the basal surface of kaolinite, as easily leachable outer-sphere hydrated complexes, instead of inner-sphere or interlayer complexes, explaining their high extractability (> 50%) from those minerals by ion exchange [64]. On the other hand, at basic conditions ($pH = 10$), REE adsorb to kaolinite primarily as hydroxides, instead of hydrated ions, but the kaolinite structure is also not altered after REE adsorption, indicating that REE hydroxides are likely attached to the surface of kaolinite by electrostatic interaction [61]. In addition, Fe oxides have a high affinity with REE, especially HREE [65]. When associated with these oxides, the release of REE is dependent on changes in environmental conditions, such as the reduction of the corresponding ions of these elements in humid environments [65].

Despite the important insights provided by studies of clay minerals from regolith at deeper layers, the extrapolation of those findings to clay minerals in soils needs to be done with caution because those minerals can have very contrasting properties depending on where they occur along the vertical profile. For instance, both kaolinite and halloysite tend to have a much lower REE adsorption capacity near the surface in contrast to deeper layers [62]. This is likely due to their higher crystallinity toward the upper layers, where those minerals are well-developed, having lower porosity, specific surface area, and CEC in comparison to those in deeper layers [62]. These remarkably contrasting properties for the same clay mineral along the vertical profile, in addition to the very complex nature of soils (influence of soil structure, role of the soil microbiota, dynamic nature of soil solution, complexity of soil OM) when compared to subsurface clay deposits, reinforces the need for detailed studies under natural soil conditions to better understand and predict REE behavior in this compartment.

The effects of mineralogy on REE levels were observed in a study comparing two soils: an Oxisol representative of tropical regions and a Mollisol representative of temperate regions. This study explored how REY (REE plus yttrium) sorption and speciation are influenced by soil type and pH, using solutions of individual or combined REY [7]. As expected from their mineralogy — Oxisol presenting anatase, Al-Hydroxy interlayed vermiculite, gibbsite, goethite, hematite, kaolinite, muscovite, and quartz and Mollisol presenting albite, anorthite, augite, kaolinite, illite, orthoclase, and quartz — the Oxisol presented a much lower REY adsorption capacity than the Mollisol, because of the lower CEC of the former. The REY adsorption in the Oxisol was more positively correlated with soil pH, in agreement with the known higher dependence of soil CEC on pH in tropical soils due to the variable charges found in kaolinite and oxides of Al and Fe and especially in soil OM, which is responsible for most of the CEC in tropical soils. In the Mollisol, the REE adsorption increased with increasing pH up to the point of zero charge. In the case of the Oxisol, under its natural pH (*ca.* 4.5), the main adsorbed species are REY^{+++} and $REYNO_3^{+++}$. On the other hand, the most prevalent adsorbed REY species in the Mollisol under natural pH condition (*ca.* 6.85) are $REY(OH)^{++}$ and $REY(CO_3)^{+}$. Regarding the REY partitioning with the solid phase, the main controlling mechanism is precipitation instead of adsorption. At pH values above 7–8, REY tend to precipitate, mainly the HREY. In this study, and several other investigations [1••, 7], it was found that under acidic conditions ($pH \leq 6$), hydrated REY ions are the dominant species found in the soil, followed by REY nitrates. Under alkaline conditions, REY hydroxides were the dominant species.

REE in Plants: Development and Metabolism

The application of REE in plants can promote physiological alterations such as increased enzymatic activity, chlorophyll content, and photosynthetic rate [66, 67]. Additional studies also report the effects of REE in the recovery of physiological functions in situations of nutrient deficiency and the mitigation of stresses promoted by environmental factors, such as salinity [68] and PTE exposure [69]. However, the effects of REE on plants depend on the element, applied dose, exposition time, and management conditions [70]. The hypothesis of the hormesis effect (*i.e.*, beneficial responses to low concentrations and inhibitory responses to high concentrations) has been supported by several studies applying REE in different plants [66, 67, 71]. When applied in adequate doses, these elements (alone or in combination) are associated with better plant development, *i.e*., enhancing biomass production and crop yield [70, 72]. REE may also interact with other elements, competing for absorption sites [73], promoting antagonistic effects, either nutritional deficiency or increased nutrient absorption. REE have increasingly been considered biostimulants for plant development and production; however, their effects on plants are still not fully understood.

Effects of REE on Plant Development

Uptake, Distribution, and Contents in Plants

Due to the increased concern about REE contamination, in recent years, several studies have described the REE phytoextraction potential of plants growing in the vicinity of roads [15, 16], mining areas [74, 75], and under natural conditions (without anthropic interference) [76], followed by the determination of translocation and bioaccumulation indices, fractionation, and human health risk assessment. The ability of plants to absorb and transfer REE varies with species and REE concentrations in the soil, besides soil characteristics. For example, acidification and plant-available phosphorus (P) play an important role in REE uptake and accumulation [38, 77].

The effects of REE in plants have been evaluated with soils, hydroponic solutions, in vitro tests, or foliar spray applications and mainly in the form of chlorides and oxide nanoparticles, the last one recurrent in recent studies [69, 78]. It is worth mentioning that results obtained in hydroponic solutions can be very different from those of soil studies. However, hydroponically grown plants have been broadly used to isolate REE effects. Results of studies with REE in hydroponic medium indicate dose-dependent responses on plant physiology and anatomy in different species, avoiding the potential interference with other biotic and abiotic factors naturally present in the soil matrix [78]. These responses are essential to provide insights into the mechanisms behind REE-induced plant stress tolerance and increased plant growth and crop yield.

The physiological mechanisms behind the absorption and accumulation of REE by plants are not fully understood. The correlation between the absorption of REE and other elements by plants suggests the existence of a similar transport mechanism. For example, LREE appears to enter the root of *Phytolacca americana* L. through Ca^{2+} ion channels [79]. Also, a competition between La^{3+} or Ce^{3+} and Ca^{2+} and Mg^{2+} ions for binding to the transport sites on the root surfaces of *Triticum aestivum* has been reported [73]. In the hyperaccumulator fern species, *Dicranopteris linearis*, the NRAMP REE Transporter 1 (NREET1) was recently described as the first REE transporter reported in plants [80]. This gene is mainly expressed in the roots, where it transfers REE from the root cell wall to the cytoplasm [80].

In general, LREE are preferentially absorbed by plants, but it has been reported that either HREE [75] or LREE [81] are preferentially transferred to plant leaves. The higher accumulation of LREE than that of HREE is possibly due to the higher levels of those elements in soils and because HREE generally form stable complexes in the soil solution [1••]. Recent studies investigated where REE accumulate in plant tissues. It is known that after being absorbed, REE tend to preferentially accumulate in the roots compared to the upper parts of the plants, especially due to impediments such as the apoplastic barriers in roots. Also, chelation of Ce^{3+} by citric acid or precipitation by PO_4^{3-} can reduce the translocation and toxicity of CeO_2 nanoparticles [82]. Recently, it has been shown that REE are translocated from the roots and accumulated in the conductive tissues of *Dryopteris erythrosora* fronds [83]. For instance, *Arabidopsis thaliana* exposed to 3000 mg L^{-1} Ce oxide nanoparticles showed a large number of needle-like particle aggregations in the intercellular regions and the cytoplasm of leaf cells [84]. These authors suggest that endocytosis is likely to be a general mechanism that explains the translocation of these Ce compounds, as the up-translocation factor to shoots does not directly depend either on the applied doses or on the supplied forms of Ce [84].

Seed Germination, Plant Growth, and Product Quality

Positive effects of REE on plant growth, including plant height, number of tillers, root volume, and shoot biomass [70], have been described mainly as a function of the hormesis effect [66, 71]. The application of La, for example, improved the photosynthetic system and promoted higher nutrient uptake, thus increasing the biomass of *Solanum* *nigrum* L., reaching its maximum (89% greater than the control) at 10 µmol La^{3+} L^{-1} [67]. In this sense, plant absorption of N, Mg, and other elements required for chlorophyll synthesis may be enhanced by REE [66]. In addition, in soybean leaf cells, low doses of La activate clathrin-mediated endocytosis (CME), and the intracellular La and La-activated CME disturb intracellular homeostasis, which may stimulate cell growth or induce damage to the plasma membrane of leaf cells [71].

REE can also enhance germination rates and seed vigor due to increased water absorption, hormone content, and hydrolytic enzyme activity. However, the expected benefits of these elements are dependent on the exposure time and dose used. Cerium significantly increased germination and the initial seedling growth of rice (*Oryza sativa* L.) [70]. On the other hand, high concentrations of Y_2O_3 nanoparticles (up to 100 mg L^{-1}) delayed seed germination, but did not affect germination rates of rice [85]. Nevertheless, Moreira et al. [86] observed a significant decrease in the germination percentage of several plant species growing in two typical tropical soils and in an artificial soil under Ce exposure, but no hormesis effect was observed possibly due to the applied doses (from 50 to 2051.7 mg Ce kg^{-1}). It is important to mention that the authors showed that the sensitivity of plants is species-specific.

Effects on the quality of agricultural products are also observed with the application of REE. Cerium nanoparticles application promoted increased fruit quality as demonstrated for strawberries (*Fragaria* × *ananassa* Duch.) [87] by increasing total phenols, ascorbic acid, and other compounds. In addition, Ce (10–20 mg L^{-1}) can promote the accumulation of important organic compounds, such as polysaccharides, polyphenols, flavonoids, soluble sugar, and protein in copper-stressed *Dendrobium nobile* Lindl, a popular plant in traditional Chinese medicine [88].

Tolerance to Environmental Stresses

The metabolic and anatomical plasticity developed in the presence of REE (at the first stage of the hormesis effect) condition the plants to tolerate several environmental stresses (biotic and abiotic). These elements can protect plants against stress by improving their hormone contents, water use efficiency, and plant nutrient accumulation; by protecting the biochemical reactions of photosystems and influencing the activity of important antioxidant enzymes such as superoxide dismutase (SOD), peroxidase (POD), and catalase (CAT) [66–69]. In addition, REE have also been considered in plant nutrition as beneficial elements, reducing the incidence of plant diseases [89]. Some REE, mainly La, at appropriate doses, act as primers (beneficial stressors) reducing the oxidative stress promoted by biotic factors and conditioning the plants to tolerate and control insects infestation and diseases [89].

Oxidative stress control is a fundamental component of tolerance in plants. This acts as an essential player in physiological plasticity, which has been verified with the presence of REE in plants in several studies. High concentrations of CeO_2 nanoparticles (> 200 mg L^{-1}) in marigolds increased the activities of antioxidant enzymes such as CAT, SOD, ascorbate peroxidase, guaiacol peroxidase, glutathione S-transferase, and polyphenol oxidase [90]. Moreover, La and Ce induced an increase in the activity of SOD, POD, and CAT in *H. annuus* [91]. Likewise, the application of Sc alleviated Cr stress in *Lemma minor* by increasing the activities of SOD, NADPH oxidase, ascorbate peroxidase, glutathione reductase, monodehydroascorbate reductase, and glutathione S-transferase, as well as by promoting the maintenance of cellular redox status by regulating antioxidant pathways [92].

Stress tolerance studies are frequent and have been even more important in a scenario of high-tech agricultural systems that can also suffer from the effects of water deficits, which are becoming more frequent due to climate change. Although this is important for food security, it is relevant to emphasize the need for more studies about human health and/or environmental risks that might be associated with the potentially toxic effects of these elements, which is a food safety issue.

Effects of REE on Plant Metabolism

The structure of plants allows the metabolic essential reactions to their life cycle to occur, which at the same time enable the growth and structural development of the plant. Therefore, there is a close dependence and correlation between structure and metabolism in plants. Knowing the effects of REE on plant structure helps to understand their impacts on plant metabolism and vice versa. For example, Ce nanoparticles (250, 500, 1000, and 2000 mg L^{-1}) sprayed in common beans plants led to a decrease in stomatal density (from 337 to 113 per mm^2), enlarged stomatal length (from 12.8 to 19.4 µm), and promoted significant alterations in important biochemical pathways, *i.e.*, in photosynthesis and electron transport chain [93].

Water Use Efficiency and Photosynthesis

There is an intrinsic relationship between water use efficiency in plants and the photosynthetic process since water is required as an electron donor to the electron transport chain of the photochemical phase of photosynthesis. Effects on the physiological traits of common beans under induced drought stress indicate that Ce^{3+} alleviates water stress, due to increased chlorophyll content and water use efficiency, consequently boosting photosynthesis [94]. However, in the same study, no significant effect of Ce^{3+} was verified on plants growing under normal conditions (without hydric stress), reinforcing that REE can attenuate drought stress in plants [94]. Scandium alleviated several symptoms of drought stress (using 5% PEG-6000) in hydroponically grown rice by preventing the inhibition of growth, water content, proline content, and photosynthetic efficiency [95].

The use of CeO_2 nanoparticles increased water use efficiency and the maximum carboxylation rate of Rubisco, which in turn increased the photosynthetic rate in soybean under moisture content above 70% of field capacity [96]. Likewise, foliar-sprayed CeO_2 nanoparticles (10 mg L^{-1}) in sorghum alleviated drought-induced oxidative stress by catalytic scavenging reactive oxygen species, thus reducing 41% of superoxide radical, 36% of hydrogen peroxide levels, and 37% cell membrane lipid peroxidation [97]. Additionally, enhancement of photosynthesis (an increase of 38% in leaf carbon assimilation rates) and seed yield (an increase of 31%) were also observed [97].

The application of La and Ce (both from 1 to 10 µmol L^{-1}) in *Helianthus annuus* resulted in the accumulation of photosynthetic pigments, such as chlorophylls and carotenoids, as well as in increased photosynthetic efficiency, non-photochemical quenching, photosynthetically active radiation, and electron transport rate [91], indicating that REE potentialize the photosynthetic process in the photochemical phase. Contrariwise, nanoparticles of Tb, at high doses (under 250–500 mg Tb L^{-1}), affected the chlorophyll fluorescence and suppressed the trapped energy by the active reaction center of photosystem II while also leading to the accumulation of inactive reaction centers, thus lowering the detected level of electron transport from photosystem II to photosystem I in *Lemna minor* [92]. This indicates that the effect of REE on plant metabolism also depends on the plant species, as well as on the combinations of REE to which they are exposed.

In general, the increased water use efficiency by plants caused by the presence of REE is related to increased compatible osmolytes, which in turn enhance the photosynthetic process and crop yields. Indeed, higher levels of Ce nanoparticles induced the accumulation of compatible osmolytes such as proline in common beans plants [93], as well as proline, soluble sugars, and glycine betaine contents in marigold [90].

Plant Nutrition

As already mentioned, REE interact with other elements, competing for absorption sites or replacing them in organic molecules. For example, Ce can replace Mg under conditions of Mg deficiency, avoiding the malformation of photosynthetic pigments. Due to similar ionic radii, La^{3+} can compete with Ca^{2+} in biological processes and replace this element in the formation of chloroplasts, mitochondria, cytoplasmatic membranes, and oxalate crystals [1••, 98]. This leads to structural modifications, inhibition of enzymes, and destabilization of the cell membrane [1••, 98]. In a study with citrus rootstocks, leaf Ca content was negatively correlated with the La content, indicating that La^{3+} binds to the specific Ca^{2+} site on the plasma membrane, which affects the absorption and transportation of Ca^{2+} and may cause toxicity to plants [99], due to the non-acting of cytosolic Ca as a secondary messenger and/or its replacement in the structure of plants, not maintaining their rigidity and shape. The effect of Tb (< 100 mg L^{-1}) on *L. minor* growth can be explained by the increased contents of N, P, K, Ca, Mg, Mn, and Fe [92]. When Ce was applied to cotton in the form of poly acrylic acid-coated nanoceria, the improvement in tolerance to salinity was explained by a better ability to maintain cytosolic K^+/Na^+ ratio in both the first and second true leaf [100]. Finally, concerning PTE, the foliar application of CeO_2 nanoparticles (200, 400, and 600 mg L^{-1}) in *A. esculentus* reduced Cd toxicity in plant tissues [69].

Environmental and Human Health Risk Assessments for REE in the Soil Environment

Ecological Risk Assessments for REE in Soils

The essentiality of REE for life has not been established yet [101], so it is reasonable to assume that their absorption/assimilation by organisms is unnecessary. Some authors refer to lanthanides as a group of PTE of low to moderate hazard [102], and although several studies show their beneficial effects at low concentrations, adverse effects may occur at high concentrations [78] as observed in the results of several recent studies involving ecotoxicological tests (Table S1). In fact, the effects of REE on organisms often depend on the level of exposure, as evidenced by the Hormetic pattern [103], yet some studies may not reveal this trend due to the range of doses tested.

Most studies on the exposure effects and/or accumulation of REE have been conducted in plant species. However, other organisms have also been tested, such as *Penicillium simplicissimum* (fungus), *Aspergillus japonicus* (fungus) [104], *Porcellio scaber* (isopod), *Eisenia andrei* (earthworm), *Folsomia candida* (springtail), *Oppia nitens* (mite) [105], *Enchytraeus crypticus* (enchytraeid) [105, 106], *Caenorhabditis elegans* (nematode) [107], *Harpalus* (*Pseudoophonus*) *rufipe* (beetle) [20], and *Melanoplus sanguinipes* (grasshopper) [108]. At the ecosystem level, one study assessed the potential for bioaccumulation and trophic transfer of REE in three different ecosystems in the eastern Canadian Arctic [109]. These authors verified a limited potential for biomagnification of REE, with the highest REE levels found in species from the base of the food web [109]. The limited potential for biomagnification has been confirmed in another field-based study of undisturbed ecosystems in subarctic Canada [110]. Also, this accumulation pattern is corroborated by a study that assessed REE concentrations in different food groups in China, with aquatic and food terrestrial samples at low trophic levels generally presenting higher REE concentrations than those at a higher trophic level [111]. According to the authors, a possible explanation for this behavior is related to the trophic dilution effect and the habitat/growing environment of the corresponding species.

In general, the ecological risk has been estimated using the hazard quotient (REE concentration/threshold value) [112], soil contamination indices [113], and hazardous concentration to 5% of the species (HC_5) [86, 105]. For example, the environmental risks associated with Ce and La concentrations in soil were evaluated considering HC_5, estimated by the species sensitivity distribution curve (SSD) [86, 105]. The HC_5 values are very important references because they give an idea of elemental concentrations in the soil at which few toxic effects can be expected for most species (95%) in a similar context for which the SSD was developed [114]. This approach uses toxicity values for different species, which must be obtained from laboratory experiments and compatible matrices, in a cumulative distribution function [114]. For example, SSDs were constructed using EC_{50} values for shoot dry matter of eight plant species (maize, sorghum, rice, wheat, soybean, sunflower, radish, and common bean) to assess Ce toxicity in tropical soils [86] and EC_{10} values of different endpoints (reproduction, growth, and bioluminescence) were used to assess La toxicity to soil organisms (earthworms, enchytraeids, collembolans, mites, isopods, bacteria, and plants) [105]. Based on total REE concentrations, these studies revealed HC_5 of 54 (27–107) mg La kg^{-1} for a LUFA 2.2 natural standard soil, and 262.1, 478.5, and 878.7 mg Ce kg^{-1} for an Oxisol, an Inceptsol, and an Artificial Tropical Soil, respectively. The HC_5 for La is clearly more conservative than those for Ce. Besides the differences in studied soils and elements, this is also probably due to the inclusion of more sensitive species (*e.g.*, *P. scaber* - EC_{10} of 69 mg La kg^{-1} and *Photobacterium phosphoreum* - EC_{10} of 101.3 mg La kg^{-1}), and to the restrictive toxicity values (EC_{10}).

In general, studies devoted to assessing risk based on SSDs are time/resource/labor intensive, considering the implementation of ecotoxicological tests. One of the limitations to the more widespread use of SSDs is the number of studies under similar exposure conditions and the number of species tested [53]. In addition, it seems that researchers are sometimes discouraged from publishing basic studies, such as dose and toxicity evaluations, because of the limitations imposed by some scientific journals, which claim "international" insignificance or lack of novelties. It is likely that if more emphasis were placed on the generation of basic data, a more robust database could be built more quickly to contribute to the risk assessment process.

Establishing guidance values for soil quality by regulatory agencies could be a major challenge because the toxicity response of REE is influenced by several factors such as bioassay and exposure conditions, target organisms, toxicological endpoints, chemical elements, concentrations, speciation, substance/salts, and coexisting cations [53, 73, 103]. For example, results of a wheat root elongation test (4 days), conducted in nutrient solution showed protective effects of Ca and Mg on La/Ce toxicity [73]. These authors observed that $EC_{50}\{La^{3+}\}$ values increased linearly with increasing doses of Ca and Mg, which they attributed to competition for transport/toxicity sites on the roots. In addition, the ecological effects of REE are dependent on soil type, considering that bioavailable concentrations were controlled by mineral phases, soil pH, and OM, among others [53, 103]. Generally, the toxicity values are expressed as total concentrations, which can hinder comparisons between soils with different physical and chemical properties [115]. Furthermore, the use of historically contaminated or naturally enriched soils may provide more realistic toxicity results for environmental assessment than recently spiked soils due to the "aging" effect [116]. The selected endpoints also influence the understanding of the levels of effects caused by REE (*e.g.*, survival, growth, reproduction, behavioral, molecular, and biochemical). It is important to note that results from bioassay using nutrient solution or other exposition media can be very different from those performed with soils, which suggests that high uncertainty factors should be used when extrapolating the results. These and other factors make the establishment of guideline values for REE in soils still controversial and justify the inefficiency/impossibility of having a single value [102].

To address some of the issues related to assessing the toxicity of REE, researchers have used free metal ion activities [73, 117], tissue concentrations [115], $CaCl_2$-extractable concentrations [105], pore water concentrations [105], and the biotic ligand model [73] to estimate toxicity values. Also, REE mixtures have been evaluated to clarify the interactive effects (*e.g.*, additive, synergistic, or antagonistic) on the toxicity [73, 104, 117] and the diffusion gradient in thin films technique to determine the bioavailable contents of REE [118].

The variable toxicity exhibited by REE also contributes to the impracticality of establishing a single threshold value. For example, seedlings of *H. annuus* and *Brassica chinensis* L. when exposed to 268 mg kg^{-1} of La and to 335 mg kg^{-1} of Nd in the growth medium for in vitro testing showed brown, malformed, thick, and underdeveloped roots and significantly reduced shoot growth [119]. In this study, the authors observed that the toxicity of La is slightly greater than that of Nd. In another study, plants of *Solanum lycopersicum*, *Triticum durum*, and *Raphanus sativus* were exposed to commercial black garden soil spiked with increasing Ce, Eu, and Nd doses [115]. In general, these authors observed that *T. durum* was the least sensitive species and *R. sativus* was the most sensitive one. Based on the EC_{10} for REE concentrations in plant tissues, they suggest that Nd is the most phytotoxic REE when compared with Ce and Eu. Also, when the nematode *C. elegans* was exposed to trichloride salts of Nd, Pr, and Sc at sublethal doses, these REE induced similar neurotoxicity [107]. However, these authors verified that Nd had a relatively stronger lethality than Pr and Sc (based on LC_{50} values).

Human Health Risk Assessments for REE in Soils

There are still many gaps for a comprehensive understanding of the effects of REE on human health and studies are needed to better evaluate the impacts of these emerging contaminants [120•, 121]. The major direct routes of REE entry into the human body include ingestion, followed by inhalation and, to a lesser extent, dermal contact. The direct exposure through intravenous contrast agents is also worth mentioning. An investigation revealed deposition of Gd in neuronal tissues of patients submitted to multiple enhanced magnetic resonance examinations using Gd-based contrast agents (GBCA) even in the absence of intracranial abnormalities, raising concerns about the safety of GBCA [122]. In another study, damage to the human renal system due to Gd accumulation has been suggested [123].

Previous studies have indicated memory loss associated with 1.7 times higher levels of REE in the blood of children (aged 7–10 years) residing in the neighborhood of mining areas when compared to individuals living in control sites [124]. The ingestion of REE has been associated with various problems to the health of adults and children. In this sense, studies involving tests on rats showed heart hemorrhage, liver necrosis, as well as congestion of blood vessels and capillaries of kidneys after long-term exposure to multiple lanthanides [125]. In Brazil, a recent study on human and environmental exposure to REE in gold mining areas in the northeastern Amazon showed that potential ecological risk indices ranged from moderate (167.3) to high (365.7) in areas with higher concentrations of REE, but health risks due to soil ingestion for children and adults were low (> 1) in the studied sites [35]. Another study also showed that the potential health risk for soil ingestions from a Beijing urban park was relatively low (118 samples; ΣREE concentrations: 117.19 to 198.09 mg kg^{-1}) [12, 14].

The inhalation route has been related to pneumoconiosis and fibrosis of the human lungs resulting from the accumulation of several REE (166,500 μg Ce kg^{-1}; 57,750 μg Nd kg^{-1}; 45,600 μg La kg^{-1}; 4,550 μg Sm kg^{-1}; 252 μg Yb kg^{-1}; 230 μg Tb kg^{-1}; 87.5 μg Eu kg^{-1}; 25 μg Lu kg^{-1}) [126]. Regarding the entry of REE through dermal contact, a study assessing the toxicity profiles of REE salts did not verify a significant potential for mutagenicity, skin irritation and sensitization, or endocrine disruption [127]. This study found only exceptional cases of skin irritation or corrosion resulting from the application of anhydrous Tm chloride (III) using a 3D in vitro reconstructed human epidermal model.

REE deserve special attention because they can be released into the environment and absorbed by plants consumed by humans, representing an indirect route of entry of REE into the human body [128]. The main sources of exposure to high REE concentrations are extraction and processing operations in mines, especially in China [129]. However, considering the growing use of REE for various applications in different regions of the world, monitoring of these elements is strongly encouraged. Some human health risk assessments of REE have indicated negligible risks. In this sense, a study demonstrated that the estimated adult daily intake of REE through vegetables (0.69 μg kg^{-1} day^{-1} and 0.28 μg kg^{-1} day^{-1} for individuals from mining and control areas, respectively) were both significantly lower than the maximum acceptable daily intake (70 μg kg^{-1} day^{-1} for vegetables) [130]. Similar findings were also observed for cereals consumption (0.69–1.24 μg kg^{-1} day^{-1}) [131] for individuals living near a mining area in China. Also, studies evaluating the health risk assessment of REE in topsoils of Qatar [113] and due to tea consumption in China [132] demonstrated negligible risks based on reference levels. Studies evaluating REE concentrations in representative food categories in China have verified low/acceptable health risks by daily food consumption [111, 133]. In the survey conducted by Dai et al. [111], the ΣREE levels in food of aquatic origin (174.97 μg kg^{-1}) were ~ six-fold higher than those found in food of terrestrial origin (27.74 μg kg^{-1}). In the study by Yang et al. [133], REE levels varied between 0.04 and 1.41 mg kg^{-1} in 11 food categories and were not associated with health risks.

It is important to mention that the responses of health risk assessments are strongly dependent on several factors, such as the different doses, the peculiar characteristics of the REE sources, the age of the human group under evaluation, and the absorption routes. For a more complete and accurate human health risk assessment, the consumption rate should be updated and different sources should be evaluated, as indicated by Zhuang et al. [130]. In addition, chemical fractionation and bioaccessibility studies are also indicated as they can improve the accuracy of risk assessments.

Survey on Challenges and Opportunities for Innovative Research on Rare Earth Elements (REE) in Soil-Plant-Animal/Human Systems

This survey was conducted by inviting some of the most cited authors working with REE in the literature to answer selected questions about challenges and opportunities for innovative research on REE in soil-plant-animal/human systems worldwide. Although this scientific poll had no intention of being a comprehensive survey concerning the status of research with REE as emerging contaminants in soils, the feedback received was relevant to shed light on the importance of investigating such elements in areas outside of Materials Science. From June 12th till July 21st, 2023, we have contacted > 200 active researchers currently working with REE. They were selected based on a search of articles from the last 5 years, with review format preference, on the Web of Science, Science Direct, and Google Scholar platforms using the following keywords (combinations): “rare earth elements”; “soil”; “agriculture”; “plant”; “lanthanum”; “cerium”; “soil contamination”; “fertilizers”. At Web of Science, we prioritized the areas of knowledge: “Agronomy”; “Soil Science”; “Plant Sciences”; “Ecology”; “Environmental studies”; “Toxicology”; “Biochemistry” and “Molecular Biology” (excluding “Material Sciences”). Also, we have asked authors to forward the survey form to other authors that they judged to be frequent contributors to the scientific literature on REE. Forty researchers from different countries kindly replied to selected questions (see the Supplementary Information). Most respondents (80%) undertake REE studies in Europe and America, with only 12.5% and 7.5% of them in Asia and Africa, respectively (Supplementary Information).

The main methodological challenges in research on REE mentioned are the availability of certified reference materials (33.3%) and interference in quantification methods (35.4%). In addition, the most relevant understudied topics in REE were (i) critical analysis to suggest threshold values in the environment and in biological systems; (ii) lack of general/reliable datasets concerning their occurrence in the environment as well as their physicochemical behavior; improved thermodynamic databases to enable geochemical modeling; (iii) studies concerning separation, fractionation, and removal from primary and secondary sources — including the use of biosorbents and phytotechnologies — while also focusing on life cycle sustainability and circular economy, and (iv) better understanding of their role in plants, including hormesis effects and their interaction with other beneficial/harmful elements.

Conclusions

Rare earth elements have distinct properties that make them vital to many disruptive uses, ranging from digital and clean technologies to agriculture. As these technologies become more widespread, it is likely that the demand for REE will continue to rise significantly, as we have observed especially in the last 5 years. Since soils are the ultimate sink for most contaminants generated by any civilization, it is essential that we continue to foster research for investigating the occurrence and fate of these elements in soil ecosystems worldwide, especially those regarding the physiological effects of REE in biological systems — either negative or positive — which continue to be a major societal concern. This speaks for the need for continuous support and encouragement of studies on the occurrence and behavior of REE in soil ecosystems worldwide.

As a proposal for future research, the most relevant topics that we suggest to be addressed in REE research are (i) establishing threshold values in the environment and in biological systems; (ii) improving the reliability of datasets concerning their occurrence in the environment as well as their physicochemical behavior; (iii) studies concerning separation, fractionation, and removal from their primary sources; and (iv) a better understanding of their role in plants.

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Data Availability Data generated during the survey will be made available on request.

Compliance with Ethical Standards

Competing Interests The authors declare no competing interests.

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- • Of importance
- •• Of major importance

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