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Levels of Rare Earth Elements in Food and Human Dietary Exposure: A Review

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Abstract

Rare earth elements (REE) are a group made up of these 17 metals: Ce, Dy, Er, Eu, Gd, Ho, La, Lu, Nd, Pr, Pm, Sc, Sm, Tb, Tm, Y and Yb. In the current century, the applications of REE have significantly increased, being used as components in high technology devices of great importance industrial and economic. However, information on human exposure to REE, as well as the potential toxic effects of these elements is still limited. In general, for metals the dietary intake is the main route of exposure for non-occupationally exposed individuals, which should be also expected for REE. The current paper was aimed at reviewing the studies conducted over the world, focused on determining the levels of REE in food samples, as well as the dietary intake of these elements. The results of most studies do not suggest potential health risks for the consumers of the analyzed freshwater and marine species of higher consumption, or derived from the intake of a number of vegetables, fruits, mushrooms, as well as other various foodstuffs (honey, tea, rice, etc.). Although the current estimated daily intakes (EDIs) of REE seem not being of concern, taking into account the expected wide use of these elements in the next years, to assess periodically the potential health risks of the dietary exposure to REE would be clearly recommendable. In fact, this is already being done with well-known toxic elements as As, Cd, Pb and Hg, among other potentially toxic metals.

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1. Introduction

Rare earth elements (REE) are a group of metals, located in the middle of the periodic table, which belong to the category of emerging contaminants associated with new technologies. According to the International Union of Pure and Applied Chemistry (IUPAC), it is a group made up of 17 elements that include lanthanum (La) and the 14 lanthanides: cerium (Ce), dysprosium (Dy), erbium (Er), europium (Eu), gadolinium (Gd), holmium (Ho), lutetium (Lu), neodymium (Nd), praseodymium (Pr), promethium (Pm), samarium (Sm), terbium (Tb), thulium (Tm) and ytterbium (Yb), as well as scandium (Sc) and yttrium (Y) (USGS, 2023a, b). REE are usually divided into heavy rare earth elements (HREE): Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Sc and Y, and light rare earth elements (LREE): La, Ce, Pr, Nd, Pm, Sm and Eu. The current uses of REE are multiple and diverse. They are used in a variety of industrial applications, which include electronics, clean energy, aerospace, automotive and defense. Manufacturing permanent magnets is the single largest and most important end use for REE, accounting for 43% of demand in 2021 (USGS, 2023a, b).

In the past century, some REE were already being used in corporate and industrial research. For example, battery research led in the 1970s and 1980s to the development of the nickel–metal hydride battery, which used La and Nd. Other REE were also used as catalysts, in magnets, as well as in steel alloys. However, until recently, REE were being familiar only to a relatively small number of people, such as chemists, geologists, specialized materials scientists, and engineers. Notwithstanding, in the current century REE have gained visibility through many media. It has been mainly due to the fact that the world society has recognized the critical, specialized properties of REE, which contribute to the modern technology. It is now well known that REE have particular physical and chemical properties, such as unique magnetic and optical properties. It means that REE have a considerable number of applications that involve many aspects of the modern life. In our globalized world, another important issue related with these elements is the dominance of China in the production and supply of REE, which means an international dependence on that country for the majority of the world's REE supply (USGS, 2023a, b). Since the late 1990s, China has provided 85–95% of the world's REE. Since 2000, the uses of REE have significantly increased, being currently used as components in high technology devices, including smart phones, digital cameras, computer hard disks, fluorescent and light-emitting-diode (LED) lights, flat screen televisions, computer monitors, and electronic displays. Other applications of REE include the making of microphones, headphones, lasers, satellites, radar, and sonar. In addition, due to their luminescent properties, some REE help to make LCD screens, which go from smartphones to large television sets. Although using REE in electronics contributes a small part to the final product, this cannot function without them (Versa Electronics, 2023). Nowadays, the use of REE in electronics is essential, not having a close substitute, while their use in versatile industries makes them even more crucial.

In spite of the great and increasing interest that REE are having in our modern societies, the global environmental health effects, the potential environmental risks associated with the development of REE production, the REE toxicities, or the human health effects or REE are issues of considerable importance on which information is rather scarce (Brouziotis et al., 2022; Gwenzl et al., 2018; Oladipo et al., 2023; Pagano et al., 2015, 2019; Yin et al., 2021). For non-occupationally exposed populations, it is well established that the diet is -in general- the main source of human exposure to metals. For that reason, to know the dietary intakes of REE should be an issue of notable interest. The objective of current paper was to summarize the results reported in the scientific literature of the studies on the concentrations of REE in food, as the dietary intake when data were published. Those studies conducted by regional, national or international agencies and/or

food safety authorities are not included in the present review. The databases Pubmed (<https://pubmed.ncbi.nlm.nih.gov/>) and Scopus (<https://www.scopus.com/>) were used. The terms for the search were the following: “rare earth elements”, “REE”, “food”, “dietary intake” and “human exposure”. Information on the available studies and their results are next summarized.

2. Levels of REE in freshwater and marine organisms

It is well known that fish, and very especially marine fish constitute an important part of the human diet. This means that its quality and its safety aspects are issues of notable interest (de Souza et al., 2021; Domingo, 2016; Pinto et al., 2022). People worldwide consume fish and seafood for their health benefits, such as vitamins and minerals, high protein, low saturated fat content, and omega fatty acids among others (Domingo, 2016; Domingo et al., 2007; Mozaffarian and Rimm, 2006; Gowda et al., 2021). In contrast to these benefits, a number of investigations have shown that fish and seafood consumption can lead to humans to be exposed to a variety of chemical contaminants, which can cause harm to the human body (Domingo et al., 2007; Mozaffarian and Rimm, 2006). In recent years, the concentrations of metals in a number of species of fish and seafood have been measured in various areas/zones around the world (González et al., 2021; Guéguen et al., 2011; Kalantzi et al., 2013; Perelló et al., 2015). However, studies regarding the concentrations of REE in freshwater and marine organisms are much more limited. These studies are here reviewed.

Table 1 summarizes the studies focused on the determination of REE in freshwater and marine organisms. Most of these studies have been conducted in China. Zhang et al. (2009) measured the concentrations of Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Er, Ho, Tm and Lu in sixty samples of marine organisms, which were collected at six sampling sites in the Shenzhen coastal region. The mean concentrations of total REE were determined. Values of 2.66 ± 2.28 , 4.69 ± 1.32 , 0.03 ± 0.03 and 0.07 ± 0.02 $\mu\text{g/g}$, for mollusk, barnacle, shrimp, and fish, respectively, were found. An enrichment of the light REE with respect to heavy REE were observed in all marine organisms. Also in China, Yang et al. (2016) carried out a survey aimed at comparing the concentrations of 14 REE in freshwater and marine fish of Shandong Province (Eastern of China). Fourteen REE (Pr, Sm, Lu, Nd, Gd, Dy, Nd, Ce, Ho, Tb, Er, Tm, Yb and Eu) were analyzed in muscle samples of four freshwater fish and six marine fish obtained from local markets and supermarkets within 17 cities of the Shandong Province. The selected fish species were the most commonly consumed by the Shandong residents. It was found that freshwater fish had a relatively higher ΣREE than marine fish ($34.0\text{-}37.9$ $\mu\text{g/kg}$ wet weight vs. $12.7\text{-}37.6$ $\mu\text{g/kg}$ ww). The assessment of human health risks indicated that fish would mean little health risks derived from exposure to REE through fish consumption. On the other hand, in order to assess the potential impacts of the REE industry, Ma et al. (2019) collected samples of water and suspended particles in the Pearl River Delta (China), as well as oysters of the same place. The concentrations of the following elements were analyzed in all samples: La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu. The ΣREE concentrations in the Pearl River Delta oysters ranged between 1.8-9.1 and 0.9–2.6 $\mu\text{g/g}$ for samples collected in December 2016 and June 2017, respectively. It was concluded that in the Pearl River Delta, the dominant REE uptake pathway in oysters was derived from particles. In turn, in the Shandong Province of China, Jiao and co-workers (2021) determined the levels of 14 REE (La, Ce, Nd, Pr, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu) in 120

commercial mantis shrimp (*O. Oratoria*) samples, which were collected from the coast of three sites of Shandong Peninsula. The mean concentration of Σ REE was 25.1 ng/g (range 9.04-75.1 ng/g). The levels of REE found by Jiao et al. (2021) were similar to those previously detected for marine fish: 12.7 to 37.6 ng/g by Yang et al. (2016), but lower than those found for Pearl River Delta oysters (1.8 to 9.1 μ g/g) by Ma et al. (2019). The mean concentrations of the 14 REE in *O. oratoria* followed this decreasing order: La > Ce > Nd > Pr > Gd > Sm > Dy > Er > Yb > Eu > Tb > Ho > Tm > Lu. Recently, Wang et al. (2022) performed a survey aimed at determining the REE concentrations in 14 marine wild fish species from the northern coastal region of the South China Sea, as well as to characterize the distribution patterns and to assess human health risks of 15 REE (Y, La, Ce, Nd, Pr, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu). The concentrations of total rare earth elements (Σ REE) ranged between 1.02 and 178.55 μ g/kg, with a mean concentration of 27.14 μ g/kg. The highest average value of Σ REE was found in *Apogon quadrifasciatus*, while the lowest levels corresponded to *Ariomma indica*. The estimated daily intakes (EDI) of REE through fish consumption were significantly lower than the recommended EDI limit. It clearly indicated that human exposure to REE through fish consumption of that area would be negligible.

Table 1. A summary of levels of REE in freshwater and marine species

Region/country	Analyzed samples	Analyzed REE	Mean values and/or ranges	Remarks	Reference
Shenzen coastal region, China	mollusk, barnacle, shrimp, fish	14	Σ REE: 2.26, 4.69, 0.03 and 0.07 μ g/g, in mollusk, barnacle, shrimp and fish, respectively	Higher levels of LREE than HREE	Zhang et al. (2009)
Shandong Province, China	freshwater (4) and marine (6) species	14	Σ REE: 34.0-37.9 μ g/kg (freshwater species); 12.7-37.6 μ g/kg (marine species)	No health risks derived from fish consumption	Yang et al. (2016)
Pearl River Delta, China	oysters	14	Σ REE: 1.8-9.1 μ g/g (2016 sampling); 0.9-2.6 μ g/g (2017 sampling)	No specific remarks	Ma et al. (2019)
Shandong peninsula, China	shrimps	14	Σ REE: 25.1 (9.04-75.1) μ g/kg	La and Ce showed the highest levels	Jiao et al. (2021)
Northern region of the South China Sea, China	14 marine wild fish species	15	Σ REE: 27.14 (1.02-178.55) μ g/kg	The highest levels of Σ REE were found in <i>Apogon quadrifasciatus</i>	Wang et al. (2022)
Northwestern Mediterranean Sea, Italy	Fish, mussels, oysters	La and Ce	Fish levels < LOD for La and Ce; mussels, 37 and 65 μ g/kg, for La and Ce; oysters, 12 and 20 μ g/kg, for La and Ce	No specific remarks	Squadrone et al. (2016)
Northwestern Mediterranean Sea, Italy (3 sites)	Brown, green and red seaweeds	16	Σ REE: 7.9, 8.5 and 22, for the different 3 sampling sites	No specific remarks	Squadrone et al. (2017)
Ligurian Sea, Italy	fish, bivalves, seaweeds and zooplankton	16	Σ REE: fish (0.21 mg/kg); bivalves (0.16 mg/kg); seaweeds (12 mg/kg) and zooplankton (0.12 mg/kg)	A periodical monitoring of the REE levels in fish and bivalves was recommended	Squadrone et al. (2019)
Italian aquarium	Spotted dogfish (samples of blood, liver, muscle and kidney)	16	Σ REE: 4.1, 30, 15 and 13 μ g/kg, for blood, liver, muscle and kidney	No specific remarks	Squadrone et al. (2022)
Tokyo Bay, Japan	5 species of bivalves (samples of shells and soft tissues)	14	The individual concentrations of the 14 analyzed REE in shells and soft tissues are all reported in the original paper	REE levels in soft tissues were higher than those found in shells	Akagi and Edanami (2017)
Six sampling areas in	12 freshwater fish		Σ REE: 243 μ g/kg (dry weight) (14-3000)	The highest Σ REE levels were	Mayfield and

the state of Washington, USA	freshwater non-species	16	Σ REE: 10 µg/kg (dry weight) (1-1000 µg/kg dw)	found in benthic feeding species of fish	Fairbrother (2015)
14 Canadian lakes	abiotic and food web components	15	Results are available in the original paper	The Σ REE levels in benthic invertebrates was 1000 times higher than those found in fish muscles	Aymot et al. (2017)
31 sampling stations along the French shoreline, France	mussels and oysters	14	Σ REE: 0.18-4.05 µg/g for mussels and 0.21-10.94 µg/g for oysters	The highest concentrations (for oysters) were found in the Gironde estuary	Briant et al. (2021)
Gdansk Bay, Poland and North-East Atlantic, Spain	Baltic herring and sardines	17	Σ REE; samples of muscles: 76 µg/kg for herring and 191 µg/kg for sardine	No specific remarks	Reindl and Falkowska, (2021)
Six locations of the Portuguese coast, Portugal	mussels	15	Σ LREE: 288-2160 µg/kg Σ HREE: 54-230 µg/kg	No specific remarks	Figueiredo et al. (2022)
Black Sea, Crimea	mussels	16	Σ REE: 710 µg/kg (dw)	In soft tissue, La and Ce, followed by Nd, Sc and Gd were the predominant REE	Chelyadina et al. (2023)
Subaé River, Todos os Santos Bay, Brazil	samples of food web (fish, shellfish, phyto- and zooplankton)	15	Σ REE: 6.01, 1.22 and 0.059 mg/kg for bivalves, crustacean and fish	The lowest REE contents were found in fish muscle	Santos et al. (2023)
Southern Caspian Sea, Iran	Golden grey mullet	16	Σ REE: 14.5 µg/g	The relationship between Σ HREE and Σ LREE was 1.53	Bakhshalizadeh et al. (2023)

Italy is another country in which various studies on this same topic have been also performed. Most available investigations regarding the presence of REE in marine species have been conducted in the Istituto Zooprofilattico Sperimentale del Piemonte, Liguria e Valle d'Aosta, Turin. Squadrone et al. (2016) determined the concentrations of 21 trace elements in fish (*Dicentrarchus labrax*), mussels and oysters, which were collected from aquaculture marine ecosystems of the northwestern Mediterranean Sea. Among these 21 elements, the REE Ce and La were included. Neither La, nor Ce, could be detected in fish, while their levels -although rather low- were higher in mussels, Ce (0.065 mg/kg ww) and La (0.037 mg/kg ww) than in oysters, Ce (0.020 mg/kg ww) and La (0.012 mg/kg ww). A subsequent investigation of the same research group (Squadrone et al., 2017) was aimed at identifying patterns and fractionations of REE, as well as to verify the potential use of REE as pollution tracers. The concentrations of La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Yb, Tm, Lu, Y and S were measured in various species (brown, green, red) of marine seaweeds collected at 3 different sites at the northwestern Mediterranean Sea. The Σ REE (mg/kg dry weight) were 22, 8.5 and 7.9, at sites S1, S2 and S3, respectively. Seaweeds showed to be a useful tool for biomonitoring REE, since they may concentrate REE at higher levels than seawater. Squadrone et al. (2019) also measured the concentrations of La, Ce, Nd, Sm, Eu, Gd, Dy, Er, Yb and Lu, in 70 samples of different matrices of marine origin collected from the Ligurian Sea (northwestern Mediterranean Sea). Fish (mullet, redfish, mackerel, and hake) and bivalves (mussels, clams, and oysters) samples were analyzed, while the levels of the REE were also determined in samples of seaweeds (Chlorophyta, Ochrophyta and Rhodophyta) and zooplankton (n = 12) collected from the Ligurian Sea. The highest Σ REE were found in seaweed (mean: 12 mg/kg), followed by fish (0.21 mg/kg), bivalves (0.16 mg/kg) and zooplankton (0.12 mg/kg), being the Σ REE concentration in seaweed significantly higher than the levels detected in fish, bivalves and zooplankton. Taking into account that fish and bivalves are potential sources of human dietary exposure to REE, the authors suggested a periodical

monitoring of their concentrations in order to prevent potential harmful cumulative effects in humans. Related with the levels of REE in fish, Squadrone et al. (2022) also measured the concentrations of 40 elements (24 trace elements and 16 REE) in blood, liver, kidney and muscle of spotted dogfish (*Scyliorhinus stellaris*) reared in an Italian aquarium. Although this fish is only of moderate commercial fisheries relevance, human consumption is not especially rare in the Mediterranean. The mean Σ REE were 4.1 ± 0.51 , 30 ± 1.6 , 15 ± 2.0 and 13 ± 1.3 $\mu\text{g}/\text{kg}$ in samples of blood, liver, muscle and kidney, respectively, being Sc the most abundant REE.

In Japan, Akagi and Edanami (2017) determined the concentrations of 14 REE La, Ce, Nd, Pr, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu in five species of bivalves (Japanese littleneck, blue mussel, trough shell, Stimpson's quahog, and Japanese dosinia), species which were collected from three different sites in Tokyo Bay, being samples of shells and soft tissues specifically analyzed. Although there were considerable differences among the 14 REE analyzed, it was observed that the REE levels in soft tissues were higher (by an order of magnitude) than those found in shells. In turn, in USA, Mayfield and Fairbrother (2015) carried out a study focused on characterizing the concentrations of 16 REE (Sc, Y, La, Ce, Nd, Pr, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu) within several freshwater fish species from a large reservoir (six collection areas) that lies within the State of Washington. REE concentrations were measured in tissues and their relationships with tissue type, size, and trophic group were examined. Samples of the following fish species were included: burbot, kokanee, longnose sucker, largescale sucker, lake whitefish, mountain whitefish, rainbow trout, smallmouth bass, sculpin and walleye. Total concentrations of REE (Σ REE) ranged from 0.014 to 3.0 mg/kg dw and averaged 0.243 mg/kg dw. It was observed that benthic feeding species, which are exposed to sediments, showed higher levels of REE than pelagic omnivorous or piscivorous species. On the other hand, Amyot et al. (2017) conducted a study whose main objective was to characterize the background levels and trophic transfer of REE in natural environments of North America. In order to assess the natural ecosystem fate of 15 REE (Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu), abiotic and food web components were sampled in 14 Canadian temperate lakes, which were unaffected by mines. It was found that individual levels of REE and Σ REE were strongly related with each other throughout different components of lake food webs. The Σ REE were 32, 40 and 275 times higher in whole body than in muscles for brown bullhead, creek chub, and white sucker, respectively. In turn, Σ REE levels in benthic invertebrates were approximately 1000 times higher than those detected in fish muscle (median of 20 nmol/g vs. 0.02 nmol/g, respectively). Nonpredatory benthic invertebrates (mean \pm SD, 60 ± 69 nmol/g) had significantly higher Σ REE than predatory benthic invertebrates (16 ± 14 nmol/g) and zooplankton (13 ± 12 nmol/g). It was concluded that the low concentrations of REE observed in freshwater fish muscle, compared to their potential invertebrate prey, would suggest that fish fillet consumption is unlikely to mean a significant source of human exposure to REE in those areas not affected by mining activities of REE. In France, Briant et al. (2021) determined the concentrations of 14 REE (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu) in mussels (*Mytilus edulis*, *Mytilus galloprovincialis*) and oysters (*Crassostrea gigas*) soft tissues, collected along the French shoreline (31 sampling stations). Σ REE concentrations in bivalves varied among the sampling stations, with a range between 0.18 and 10.94 $\mu\text{g}/\text{g}$. The Σ REE concentrations in mussels and oysters were 0.18–4.05 $\mu\text{g}/\text{g}$, and 0.21–10.94 $\mu\text{g}/\text{g}$, respectively. The highest concentrations were found in the Gironde estuary, with the Σ REE in oysters being up to 10.94 $\mu\text{g}/\text{g}$ dw. On the other hand, Reindl and Falkowska (2021) determined the concentrations of REE in muscles of pelagic fish (Baltic herring (*Clupea harengus*) and sardines (*Sardina pilchardus*) from two European regions: the Gdansk

Bay (South Baltic Sea) and the Iberian Peninsula (North-East Atlantic). These marine species, in various forms (fresh, frozen, processed), are available in the European market. The levels of the examined REE (lanthanides: light REE (LREE, La–Eu) and heavy REE (HREE, Gd–Lu and Y) and Sc) in the ova and seminal fluid were also analyzed. REE were detected in the muscles of the Baltic herring (Σ REE = 0.076 ± 0.047 mg/kg) and the sardine (Σ REE = 0.191 ± 0.163 mg/kg), being the heavy REE the dominant in both species. For Baltic herring and sardine, the Σ REE in muscles were lower than those observed in ova and seminal fluid. In Portugal, Figueiredo et al. (2022) measured the levels of REE in mussels (*Mytilus galloprovincialis*) from six locations of the Portuguese coast. The analyzed REE were divided into light rare earth elements (LREE: La, Ce, Pr, Nd, Pm, Sm and Eu) and heavy rare earth elements (HREE: Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu). The concentration of Σ LREE varied between 288 ng/g in Aljezur (the southernmost point on the Portuguese coast) during autumn, and 2160 ng/g in Porto Brandão (south bank of the Tagus estuary) during spring, while Σ HREE ranged from 54 ng/g in Aljezur during autumn, and up to 230 ng/g in Porto Brandão during spring. In Crimea, Black Sea, Chelyadina et al. (2023) measured in samples of mussels (*Mytilus galloprovincialis*) the concentrations of 16 REE (all the 17 REE excepting Pm). The levels of REE were analyzed in soft tissues, byssus, and shell liquor of the mussels collected in three sampling locations of Crimea. Σ REE in mussels were on average 0.71 mg/kg dw. In soft tissue and byssus, La and Ce were the dominant elements at the 3 sampling stations, followed by Nd, Sc and Gd. In turn, in shell liquor, the dominant REE were Tb, Gd and Nd, while La was also among the dominant elements at Stations 2 and 3. According to the values of target hazard quotients based on two estimates of the threshold oral reference dose, in terms of REE intake, the authors concluded that musssel consumption should not mean dietary risks to human health. In Brazil, Santos et al. (2023) recently studied the fate of REE, their incorporation in biota, and transfer along a food web of the Subaé River, which is one of the main tributaries of the Todos os Santos Bay (northeastern of the country), a heavily contaminated estuary. The levels of 15 REE (Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu) were determined in sediments, suspended particulate matter, as well as important biota (shellfish and fish) for the local community. The Σ REE in phytoplankton and zooplankton were 45.7 and 68.5 mg/kg, respectively, values considerably lower than those found in bivalves, crustaceans and fish: 6.01, 1.22 and 0.059 mg/kg, respectively. The lowest values corresponded to fish muscle. It was concluded that although seafood consumption would be unlikely to be an important source of exposure to REE for humans, taking into account that fish muscle is the main tissue consumed, the levels of REE in fish and seafood should be periodically monitored. In Iran, also recently, Bakhshalizadeh et al. (2023) measured the concentrations of 16 REE (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Sc and Y) in 20 golden grey mullets caught in the south of the Caspian Sea (Bandar Anzali Coast). The levels were compared with the normal values of these elements in the earth's crust. The Σ REE was 14,550 pg/g, with a relationship between Σ HREE and Σ LREE of 1.53. Although all mullet samples contained much lower levels of REE than the concentrations reported on the earth's surface, it should be highlighted that Tb, one of the scarcest elements in the world, was found to be the third most abundant REE. It would suggest that the notable Tb presence was the result of rather recent anthropogenic activities.

3. Levels of REE in vegetables and fruits

Table 2 summarizes the studies focused on the determination of REE in vegetables and fruits. In Hetian Town (Fujian Province, China), Li et al. (2013) conducted a study aimed at measuring the levels of REE in soil and vegetables collected from farms (6 sampling sites) near mining sites of that place. The health risks of human REE exposure due to vegetable consumption were evaluated. The levels of 15 REE (all the 17 REE, excluding Sc and Pr) were determined in samples of the following vegetables: Chinese white cabbage, taro, Chinese radish, water spinach, lettuce, long bean, pakchoi, and eggplant. The Σ REE in vegetables ranged between 0.06 and 64.42 $\mu\text{g/g dw}$ (average: 3.58 $\mu\text{g/g dw}$). The mean contents of REE in vegetables collected in the vicinity of the mining sites were significantly higher than those found in samples collected in a control site, which indicated that REE from rare earth mining was an evident source of REE pollution. The daily intakes of the 8 analyzed vegetables decreased in the following order: taro > water spinach > lettuce > parkchoi > long bean > eggplant > white radish > Chinese cabbage. According to the authors of that study, the consumption of these vegetables should not result in daily intakes of REE that could mean health risks (safe values: 100–110 $\mu\text{g/kg/day}$) for adults and children. In a subsequent Chinese study, Zhuang et al. (2017a) measured the concentrations of REE in 301 samples of a number of vegetables (edible parts of Chinese cabbage, scallion, white gourd, long bean, eggplant, tomato, potato, towel gourd, pumpkin, red pepper, radish and carrot), which were collected from 10 sampling sites (5 from a mining area in Shandong and 5 control sites) in Weishan County. The health risks of human exposure to REE through vegetable consumption were also assessed. Fourteen REE (Sc, Pr and Y were excluded) were analyzed. The average concentration of REE was 66.47 $\mu\text{g/kg}$, being 94.08 and 38.67 $\mu\text{g/kg}$ the average levels found for vegetables collected from mining and control areas, respectively. The difference was significant. La, Ce, Pr, and Nd were the most abundant REE, which accounted for over 91% of total REE in both mining and control sampling sites. On the other hand, the levels of Σ REE in the analyzed vegetables decreased in this order: leaf vegetable > taproot vegetable > alliaceous vegetable > bean > solanaceous vegetable > tuberous root vegetable > gourd vegetable. The EDIs of the total REE were 0.69 and 0.28 $\mu\text{g/kg/day}$, respectively, for samples collected in the mining and control areas. These values were significantly lower than the established EDI (70 $\mu\text{g/kg/day}$). Although those EDIs would not mean health risks in adults, the authors recommended to pay attention to the potential risks in children derived of a continuous exposure to low levels of REE. Also in China (Xinfeng County, Jiangxi province), Cheng et al. (2015) investigated the transfer characteristics of 15 REE (Ce, La, Nd, Y, Pr, Gd, Sm, Dy, Er, Yb, Eu, Ho, Tb, Tm and Lu) from soils to navel oranges. The effects of these REE on the internal quality of that fruit were also examined. The concentration of REE in navel orange pulp was low, ranging between 0.106 and 0.829 mg/kg dw (average: 0.341 mg/kg dw). The LREE accounted for approximately 84% of the total contents of REE, with Ce being the predominant element. The REE level in navel orange pulp was 14 times lower than the food safety limit set in China (MHPRC, 2005). Even when total soil REE level was as high as 1038 mg/kg , the consumption of navel oranges was still safe for humans. In a more recent study on the same topic, Shi et al. (2022) determined the levels of REE in fruits (288 samples of pome fruits, drupes, citrus, melon fruits, tropical and subtropical fruits, berries, and small fruits) and vegetables (942 samples of solanaceous, brassica, melon vegetables, stem vegetables, root and tuber vegetables, bulb vegetables, fresh legumes and leafy vegetables). Samples were collected near mining areas of China (Bayan Obo in Inner Mongolia, Weishan in Shandong, Maoming in Guangdong and Longnan in Jiangxi) and in control sampling points. The levels of 16 REE (Sc, Y, La, Ce, Nd, Pr, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu) in fruits were 12.90 and 11.89 $\mu\text{g/kg}$ in the mining and control areas, respectively. In turn, the levels of REE in

vegetables were 92.90 and 62.38 $\mu\text{g}/\text{kg}$ in mining and control areas, respectively. Regarding the EDIs, these were in the ranges 0.02–0.06 and 0.53–1.22 $\mu\text{g}/\text{kg}/\text{day}$ for fruit and vegetables, respectively. These daily intakes were considerably lower than the regular allowable daily intake (60.4 $\mu\text{g}/\text{kg}$ body weight) for all gender/age groups of consumers.

Table 2. A summary of available information on the levels of REE in fruits and vegetables

Region/country	Vegetables/fruits	Analyzed REE	Mean values and/or range	Remarks	Reference
Fujian Province, China	8 species of local vegetables	15	Σ REE, mean: 3.58 $\mu\text{g}/\text{g}$ (dw) (0.06–64.42 $\mu\text{g}/\text{g}$)	The daily intake of the analyzed vegetables was safe	Li et al. (2013)
Weishan County, China	12 species of vegetables from 10 sampling sites	14	Σ REE, mean: 94.08 and 38.67 $\mu\text{g}/\text{kg}$ in vegetables collected in mining and control areas, respectively	La, Ce, Pr and Nd accounted for over 91% of total REE in mining and control areas	Zhuang et al. (2017)
Xinfeng County, Jiangxi Province, China	pulp of navel oranges	15	Σ REE, 0.341 $\mu\text{g}/\text{g}$ (dw) (0.106–0.829 $\mu\text{g}/\text{g}$)	LREE accounted for 84% of the total content of REE	Cheng et al. (2015)
Four mining points (Inner Mongolia, Shandong, Guangdong and Jiangxi), China	228 samples of various fruits and 942 samples of several species of vegetables	16	Σ REE, fruits: 12.90 and 11.89 $\mu\text{g}/\text{kg}$ in fruits collected in mining and control areas. In vegetables, 92.90 and 62.38 $\mu\text{g}/\text{kg}$	EDIs were in the ranges 0.02–0.06 and 0.53–1.22 $\mu\text{g}/\text{kg}/\text{day}$, for fruits and vegetables, respectively	Shi et al. (2022)
La Palma, Canary Islands, Spain	bananas collected from an area affected by a volcanic eruption, and control sites	17	Σ REE, 61.4 (peel) and 14.2 (flesh) ng/g . In control sites: 16.2 (peel) and 5.7 (flesh) ng/g	The intake of REE from consumption of bananas would not exceed 5% of the tolerable daily intake for any of the REE	Rodríguez-Hernández et al. (2022)

On the other hand, following a volcanic eruption that occurred in 2012 in the island of La Palma (Canary Islands, Spain), the concentrations of 50 elements were determined in bananas obtained from the affected area (Rodríguez-Hernández et al., 2022). The REE: Ce, Dy, Er, Eu, Gd, Ho, La, Lu, Nd, Pm, Pr, Sc, Sm, Tb, Tm, Y and Yb were included in the survey. The median values of Σ REE in peel and flesh samples of the fruits collected in the volcano area were 61.4 and 14.2 ng/g , respectively, being 16.2 and 5.7 ng/g the values found to the bananas in the control area. For both peel and for flesh, the differences were significant. Anyhow, these higher levels should not mean a real risk for the consumers. Even the consumption of bananas from the volcano area would be of little relevance in terms of human health. On average, the intake of REE from consumption of bananas would not exceed 5% of the tolerable daily intake of any of the REE. On the other hand, in order to gather information on the occurrence of REE in the daily diet of adults and children, Doulgieridou et al. (2020) published a review on potentially toxic REE, plus thallium and tellurium, in plant-based foods (edible plants). The studies reviewed in that paper have been also included in the current review.

4. Mushrooms

It is known that mushrooms are certainly effective to bioconcentrate different chemical elements -including trace elements and REE- from substrate, in which mycelium lives to developed fruiting bodies. In the scientific literature, there are a number of available studies focused on monitoring REE in edible mushroom species. Most of these investigations have been performed in Poland, and more specifically in the Laboratory of Environmental Chemistry & Ecotoxicology, Gdańsk

University. The results are next summarized. Falandysz et al. (2017a) measured the concentrations of La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Sc and Y in composite samples of caps and a whole fruiting bodies of the mushroom *Macrolepiota procera*, whose samples were collected across Poland. The total concentrations of Σ REE in caps (13 sites) and whole fruiting bodies (3 sites) were 0.50 and 0.75 mg/kg dry biomass (db), respectively. Cerium was the most abundant REE, with a mean value in the caps at 0.18 ± 0.09 mg/kg db (range: 0.030-0.34 mg/kg db). In contrast, the lowest levels (0.0011 mg/kg db) corresponded to Lu and Tm. According to the authors, the levels of REE found in *M. Procera* indicated that eating a tasty cap of this mushroom should not mean health risks for the consumers. In another study of the same research group, Falandysz et al. (2017b) investigated the multi-elemental composition and associations between 32 trace elements and 16 REE (the same elements analyzed in the study by Falandysz et al. (2017a)) accumulated in fruiting bodies (caps and stipes) of samples collected at 16 sites from northern and central regions of Poland. Among the 48 elements analyzed, the highest levels corresponded to Cd, Hg and Pb found in edible caps of the fruiting bodies. With respect to REE, as in the survey by Falandysz et al. (2017a), the concentrations of REE detected in *M. Procera* should not mean health risks for the consumers. More recently, Falandysz (2021) carried out a survey focused on extending the data on the levels of 30 metals in samples of the king bolete mushroom, *Boletus edulis* (fruiting bodies). The Σ REE (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu) was determined. For the Σ 14 REE, the median was 0.31 mg/kg dw (range: 0.074-1.8 mg/kg dw). For all the elements analyzed in that survey, including the 14 analyzed REE, their concentrations -even those of the most notoriously toxic metallic elements- in *B. edulis* were considered harmless. In a subsequent survey, Falandysz et al. (2022) determined the concentrations of the REE: La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Sc and Y in parts of the fruiting bodies of *B. edulis* (14 composite samples derived from 261 whole fruiting bodies). Samples were collected from geographically distant locations of Poland. Cerium was the most abundant REE, followed by La and Nd, with median levels of 95, 51 and 32 μ g/kg, respectively. The median REE concentrations in the caps, stipes, and whole fruiting bodies followed this order: LREE > MREE > HREE. The median levels (μ g/kg dw) for these subgroups were 121.4, 12.8 and 5.6 μ g/kg dw in the caps, 218, 19.1 and 9.2 μ g/kg dw in the stipes, and 187.7, 17.6 and 8.7 μ g/kg dw for the whole fruiting bodies. As for other mushrooms, it was concluded that consumption of *B. edulis*, should not mean any known toxicological risk, even for high-level consumers. On the other hand, Medyk and Falandysz (2022) conducted a study focused on exploring the bioconcentration potential and status of La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu and Y in samples of eight different species of wild mushrooms and forest topsoil (0–10 cm layer) collected from 3 countries: Poland, Belarus and China. The Σ REE levels in mushrooms ranged from 15.8 μ g/kg dw in caps of *I. badia* and 443 μ g/kg dw in *C. cibarius*. It was observed that the levels varied between species, with some differences detected also in REE concentration from the same species collected at distantly located sampling sites. Recently, the same research group (Medyk et al., 2023) carried out a new study whose main purpose was to characterize the presence of REE in a relatively large sample (22 composites of 2235 specimens) of *Cantharellus cibarius* collected from 22 locations across Poland. In addition, a pooled sample (153 specimens) of *C. minor* collected at Yunnan (China) was also included in that survey. The mean concentration of Σ REE (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Sc and Y) found in *C. cibarius* was 176 67 μ g/kg dw, while the pooled sample of *C. minor* from Yunnan contained a higher level of Σ REE (2072 μ g/kg dw) than *C. cibarius* from Poland. Anyhow, the amounts of the REE detected in *C. cibarius* mushrooms in that survey would be considered certainly low -and even negligible- from a point

of view of food safety.

Other authors have also determined the levels of REE in wild (edible) mushrooms species growing in Poland. Mleczek et al. (2016) measured the concentrations of platinum group elements (PGEs) and REE in 20 mushroom species (10 above ground and 10 growing on wood). The highest levels of REE in *Suillus luteus* and *Tricholoma equestre* were 5.03 ± 0.50 and 2.18 ± 0.56 mg/kg dw, respectively, while for mushrooms growing on wood bodies it was 4.19 ± 0.78 mg/kg dw in *Ganoderma applanatum* fruiting. The mean concentrations of REE were 1.39 ± 1.21 and 1.61 ± 0.97 mg/kg dw, respectively, in above-ground species and species growing on wood. In turn, Siwulski et al. (2020) examined the trends of the content of REE (La, Ce, Pr, Nd, Sm, Eu, Gd, Dy, Ho, Er, Tm, Yb, Lu, Sc and Y) in four edible mushroom species: *Boletus edulis*, *Imleria badia*, *Leccinum scabrum* and *Macrolepiota procera*, which were collected in forests of Poland between 1974 and 2019. The Σ REE (mean and range in mg/kg dw) in fruit bodies of the examined species were: 2.00 (0.228–4.06), 2.43 (0.879–4.23), 0.607 (0.343–1.71) and 0.593 (0.203–1.46) for *B.edulis*, *I.badia*, *L.scabrum* and *M.procera*, respectively. It was found that the REE content in the examined mushrooms increased between 1990 and 2010, decreasing later, most apparently in *I. badia*.

In Greece, Koutrotsios et al. (2018) measured the concentration of 16 REE (Ce, Dy, Er, Eu, Gd, Ho, La, Lu, Nd, Pr, Sc, Sm, Tb, Tm, Y and Yb) in various mushroom production substrates, as well as in the edible fruit-bodies of the species *C. cylindracea* and *P.ostreatus*. As in other studies above reviewed, the REE detected at higher levels in fruit-bodies was Ce (31–81 and 33–75 g/kg for *C.cylindracea* and *P. Ostreatus* respectively), followed by La (13–42 and 15–39 g/kg), Nd (3.1–53 and 3.4–13 g/kg), and Sc (15–27 and 3.4–13 g/kg). It was in agreement with the results observed from analysis of substrates. Regarding health risks, the EDIs of REE were estimated to be in the ranges 0.034–0.098 and 0.046–0.081 g/kg/day for *C.cylindracea* and *P.ostreatus*, respectively. The results suggested that consumption of cultivated mushrooms at the REE concentrations found (even on substrates with a high content of REE), would not be potentially harmful for human health. In Serbia, Vukojevic et al. (2019) determined baseline concentrations of REE (Sc, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu and Y) in soils from two different geographical sites, an unpolluted mountain region (S1) and a forest near the city of Trstenik (S2). The accumulation in the mushroom *Macrolepiota procera* was also estimated. The median values ($\mu\text{g/kg}$) of the Σ LREE were 0.21 (S1) and 0.12 (S2), and 0.47 (S1) and 0.93 (S2), in cap and stipe samples, respectively. In turn, the median levels of the Σ HREE were 0.021 (S1) and 0.012 (S2), and 0.032 (S1) and 0.041 (S2), in cap and stipe samples, respectively. The results showed that wild *M.procera* did not have the ability to accumulate REE from either of the soils (BCF values were lower than 1). On the other hand, while preparing the current paper, an interesting critical review of REE in cultivated macrofungi has been published by Falandysz et al. (2024). These authors have concluded that a significant proportion of the previously reported data show unexplained anomalies in occurrence patterns. This might be unsuitable for assessing human health risks derived from exposure to REE, as well as to evaluate potential increasing trends of REE contamination in foodstuffs.

5. Other foodstuffs

Howe et al. (2005) reported the concentrations of 27 elements in samples of various food that were collected from farms

and markets in central Jamaica. The following food groups were included: fruits, legumes, leafy and root vegetables, and other root crops (being potatoes the item most consumed of this group). Among the 27 analyzed elements, 5 REE (Ce, Eu, La, Sc and Sm) were included. For Ce, most food samples were below the detection limit, with the highest levels observed in “callaloo” (0.24 mg/kg) and red kidney beans (0.006 mg/kg). For the rest of analyzed REE, the concentrations were in the ranges: 0.004 (fruits)-0.061 mg/kg (vegetables leafy); 0.3 (vegetables leafy)-1.09 µg/kg (fruits); 0.4 (fruits)-3.4 µg/kg (vegetable roots), and 0.6 (fruits)-4.6 µg/kg (vegetables leafy), for La, Eu, Sc and Sm, respectively. The authors stated that it would be unlikely that dietary exposure to these 5 REE might mean public health risks. In China, Jiang et al. (2012) determined the levels of 16 REE (Sc, Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu) in samples of fresh vegetables, cereals, fresh meat, eggs, and aquatic products (mollusks, crustaceans, and marine and freshwater fish). The average contents of Dy, Ce and La were between 0.0524 and 0.0289 mg/kg (high level), while the mean levels of Sc, Y, Er, Nd, Gd and Pr were between 0.0184 and 0.0082 mg/kg (middle level). In turn, the mean concentrations of Sm, Yb, Ho, Eu, Tb, Tm and Lu were between 0.0038 and 0.0012 mg/kg. The mean concentration of total REE was 0.2060 mg/kg. In general, the 16 REE were found at low levels in all the examined foodstuffs. Also in China, Zhuang et al. (2017b) carried out a study aimed at investigating the concentrations of 14 REE (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu) in cereals of mining and control areas in Weishan County (near to a large-scale REE mining site in Shandong). The potential health risks derived of the REE exposure through cereal consumption were also assessed. The average concentration of ΣREE for the 327 analyzed samples was 55.79 g/kg, while for the samples of cereals collected from the mining and control areas, the mean concentrations were 74.22 and 47.83 g/kg, respectively. It was found that La, Ce, Pr and Nd were the most abundant REE, accounting for over 90% of the total REE for the mining area. The EDIs of the total REE were considerably lower than the allowable daily intake (70 g/kg bw).

Some data on the concentrations of REE in honeys are also available in the scientific literature. Squadrone et al. (2020a) conducted a survey focused on assessing the occurrence of a number of trace elements and 16 REE in honeys from various countries. Another objective of the study was to evaluate if the metal concentrations in honeys might be permissible for human consumption. The levels of La, Ce, Pr, Nd, Sm, Gd, Eu, Tb, Dy, Tm, Yb, Ho, Er, Lu, Y and Sc were analyzed in samples of multi-floral honeys from the Balkans (n 0), Kazakhstan (n 0), Italy (n 0), South America (n 0) and Tanzania (n). The ΣREE in multi-floral honeys followed this decreasing order: Tanzania (65 µg/kg) taly (11 µg/kg) outh America (9.8 µg/kg) he Balkans (9.3 µg/kg) nd Kazakhstan (9.1 µg/kg). The LREE/HREE ratio ranged between 0.28 (Italy) and 2.5 (Tanzania). The same research group (Squadrone et al., 2020b) measured the concentrations of 24 trace elements and 16 REE (La, Ce, Pr, Nd, Sm, Gd, Eu, Tb, Dy, Tm, Yb, Ho, Er, Lu, Y and Sc) in monofloral and multifloral honeys from Piedmont (Northwestern Italy). The safety of the consumption of the analyzed honeys was also assessed. Ninety-one samples of acacia (n = 26), chestnut honey (n = 18), linden honey (n = 15), rhododendron honey (n = 14) and multifloral honey(n = 18) were collected and analyzed for the 16 REE. For these honeys, the ΣREE were 6.6, 12, 11, 14 and 6.0 µg/kg, respectively. The highest concentrations of REE were found in chestnut honey, while the lowest levels corresponded to acacia and rhododendron honeys. Based on the results (not only for REE, but also for trace elements) of that survey, and taking also into account the nutritional value of the analyzed honeys, their consumptions are recommended and should not mean any potential health risk. In Jordan, Tahboub et al. (2022) conducted a survey aimed

at determining the concentrations of 46 elements in 18 imported samples and 12 local samples of honeys (monofloral and multifloral). Among the analyzed elements, these 15 REE were measured: Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu. The highest levels (in decreasing order) corresponded to Ce > Nd > La > Pr > Sm. The total mean of Σ REE was $145 \pm 157 \mu\text{g/kg}$ (range: 41–489 $\mu\text{g/kg}$), being the mean levels 203 ± 165 and $98 \pm 113 \mu\text{g/kg}$, for local and imported honey samples, respectively. For all analyzed elements, including those potentially toxic and the 15 REE, the concentrations in honeys were found to be much lower than their guideline limits, indicating the safety for human consumption. In China, Wang et al. (2020) measured the levels of 16 REE in 3011 tea samples of four types (black tea, green tea, oolong tea and dark tea), samples that were obtained in the main tea producing areas in different provinces of the country. The highest mean levels of Σ REE (mg/kg) were 2.960, 2.500, 3.870 and 2.955 in black tea (Henan province), green tea (Shanxi province), oolong tea (Guizhou province), and dark tea (Hunan province). The tea leaves accumulated mainly light REE rather than heavy REE. In general, the highest content of REE in teas corresponded to Ce, regardless of the province of collection. The most abundant REE followed this order: Ce > a > c > d in black and green teas, being the trend Ce > c > d > a in oolong tea, and Ce > a > d > c in dark tea. Recently, Kowalczyk et al. (2022) assessed the levels in teas of 16 REE (La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Y and Sc), together with Sb, Ba, B, Li, Te, Tl and V. The potential health risks of the consumption of teas were next assessed. Due to the lack of substantial information on the toxicity of most REE, the related uncertainty was considered as high. La, Ce and Y were found to constitute 60% of total REE contamination in teas, being the contribution from the rest of REE considerably lower. Based on their findings, Kowalczyk et al. (2022) concluded that exposure to REE through tea consumption should pose a negligible risk to the consumers. Even no adverse effects could be expected for high tea consumers. Recently, Imran et al. (2023) determined the levels of 16 REE (Sc, Y, La, Ce, Pr, Nd, Sm, Gd, Eu, Tb, Dy, Tm, Yb, Ho, Er and Lu) in 64 samples of rice grown in Australia and imported rice from various countries (India, Italy, Pakistan, Sri Lanka, Thailand, Vietnam and USA). The average levels of REE were 0.013–2.974, 0.012–3.113 and 0.009–0.919 $\mu\text{g/kg}$ in Australian, Thailand and Vietnamese rice samples, respectively. The highest average levels of REE corresponded to rice samples from Pakistan (0.299–128.2 $\mu\text{g/kg}$), India (0.063–20.574 $\mu\text{g/kg}$) and Sri Lanka (0.022–11.522 $\mu\text{g/kg}$). Individually, the concentrations of light REE (Sc, La, Ce, Nd) were higher than those of heavy REE. According to the authors, the differences detected among samples of the selected countries could be due to different agricultural practices, geological and topographical variation, as well as the use of fertilizers with different REE composition. The health risks for the Australian population due to rice consumption were not assessed in that study. On the other hand, the bioaccumulation of the REE Sc, Ce and Eu (plus Hf and Ta, as well as other trace elements) was evaluated in oats in barley grown in soils of Russia, soils with different characteristics and level of contamination (Shtangeeva, 2022). Soil samples for that survey were collected at 3 different sites (S1, S2 and S3) in St. Petersburg. The plants were grown in these soils, which differed in texture, pH, and concentrations of exchangeable cations, and important, also in the levels of contamination. In the roots, the levels of Sc were in the ranges 0.11–0.37 mg/kg and 0.15–0.23 mg/kg in oats and barley, respectively, being the levels of Ce 1.9–3.8 mg/kg and 1.7–1.8 mg/kg in samples of oats and barley, respectively. In turn, the levels of Eu showed these ranges: 0.04–0.08 mg/kg in oats and 0.009–0.017 mg/kg in barley. The values found in roots were significantly higher than those detected in the leaves of the oats and barley. In a previous study conducted by Squadrone et al. (2019) in Italy, the levels of La, Ce, Nd, Sm, Eu, Gd, Dy, Er, Yb and Lu were measured in samples of plant feed, fruits, honey and wildlife livers. Specifically, 30 samples of

plant feed (bran, oat, forage, wheat and barley), 30 samples of fruits (apples, strawberries, peaches and apricots) and 30 samples of honeys were collected in the Piedmont Region (Northwestern Italy). Additionally, 15 liver samples of wild species (boar, roe deer, deer, bearded sparrow hawk, tawny owl and crow) were collected in that region, from animals that were found dead by local veterinarian services. The highest means of Σ REE were detected in plant feed (mean: 1.8 mg/kg), followed by wildlife livers (0.043 mg/kg), fruits (0.0088 mg/kg), and honeys (0.0078 mg/kg). Among all the analyzed samples, fruits and honeys showed very low REE levels, while plant feed, -mainly represented by raw cereals- reached the highest REE levels, which might mean a major source of exposure. Recently, Kollander et al. (2023) reported the concentrations of 74 elements in traditional and new food varieties in the Swedish market. Fourteen REE (Ce, Dy, Er, Eu, Gd, Ho, La, Lu, Nd, Pr, Sm, Tb, Tm and Yb) were analyzed in that survey. The food groups included cereal products, seeds, potato products, vegetables, algae and algae products, as well as “new” food items (vegetarian protein products, quinoa, teff flour, chia and psyllium seeds). That study provided levels of elements that are not routinely analyzed in foods, and not requested within the EFSA call for data. Because of the enormous quantity of data contained in that survey, we have not here reported the specific concentrations for all the analyzed elements including the 14 REE. For a detailed and extensive information, we refer to Kollander et al. (2023). Recently, Henriquez-Hernández et al. (2023) determined the content of 38 elements in 159 samples of ready-to-eat baby food sold in Spain. These elements included 17 REE (La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Sc and Y). The median concentrations (ng/g fresh product) of Σ REE in ready-to-eat puree for babies were 7.3 (fruit purees), 6.2 (chicken purees), 9.6 (fish purees) and 10.1 (beef purees) in major name brands. In store brands, the median levels were 5.9 (fruit purees), 12.1 (chicken purees), 8.0 (fish purees) and 16.1 (beef purees). For risk assessment, the analysis was carried out for two different exposure scenarios: a) children, who were at the average consumption of the foods considered, and b) children, who were at the 97.5th percentile of consumption. In the first scenario, no risks associated with the intake of REE were found, while in the second scenario, the maximum tolerable intake (according to data reported by other authors) was reached.

6. Human dietary exposure to REE and risk assessment

In recent decades, due to the widespread use of REE in electronics industry, lasers, advanced materials, superconductors and medicine, among many others, the demand for REE has dramatically increased. Consequently, it has meant an environmental release of these elements, which has been/is being clearly detected in recent years (Gwenzi et al., 2018; Pagano et al., 2019; Pereira et al., 2022; Zapp et al., 2022). Consequently, human exposure to REE, which could have been more or less irrelevant until recent decades, in a few years could be of concern (Oladipo et al., 2023; Yin et al., 2021). For non-occupationally exposed individuals, the diet is the most important route of exposure to metals (Bocio et al., 2005; González et al., 2021). In relation specifically to REE, in recent years, some human dietary exposure to these elements have been conducted in China. The results are now available in the scientific literature. Dai et al. (2022) determined the exposure to REE in the Chinese resident diet through a market-based study. Samples of cereals, beans, potatoes, leafy vegetables, root vegetables, melon vegetables, legumes, edible fungi, pork, beef, mutton, poultry, eggs, pure milk, mixed animal fats, fish, shrimp, shellfish and cephalopods, were collected from 33 cities of China, and the concentrations of 16 REE (La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu and Y) were measured. The

average concentration of Σ REE was 64.95 $\mu\text{g}/\text{kg}$ ww, with a range between 2.12 and 809.68 $\mu\text{g}/\text{kg}$ ww in the different kinds of samples. The highest Σ REE (605.46 $\mu\text{g}/\text{kg}$) corresponded to shellfish samples, while the lowest Σ REE level was detected in milk samples (18.43 $\mu\text{g}/\text{kg}$). The EDIs of REE were estimated for 5 Chinese regions of China, being 0.54 $\mu\text{g}/\text{kg}/\text{day}$ the average intake of REE via food consumption. The maximum EDI was found in Midland (0.90 $\mu\text{g}/\text{kg}/\text{day}$). The highest health risk of dietary exposure to REE corresponded to cereals intake, with a contribution to the total EDI -in the 5 regions- between 45.44% and 69.78%. It was followed by vegetable and aquatic animals' intake. In contrast, the risks due to the consumption of milk, eggs, and beans were relatively lower, contributing approximately with a 10% of the total EDI. In general, the health risks of REE detected in that study were considered as acceptable. A similar survey was also conducted in China by Yang et al. (2022). That study was aimed at investigating the levels and patterns of 16 REE (Pm was not included) in a number of food samples of 11 main categories, obtained in 31 Provinces of China, as well as to assess the dietary intake of REE by the Chinese population. The mean concentrations of Ce, La, Y and Nd (0.11, 0.06, 0.05 and 0.04 mg/kg, respectively) in various food categories were higher than those of the rest of REE. The mean Σ REE in all food samples of the 11 categories ranged between 0.04 and 1.41 mg/kg. In turn, the daily mean dietary exposure of the Σ REE was 1.62 $\mu\text{g}/\text{kg}$ body weight (bw) in the general Chinese population, with a range of 1.61-2.80 $\mu\text{g}/\text{kg}$ bw for the different sex/age groups. Among the 16 analyzed REE, the highest exposure levels corresponded to Ce, La and Y. The highest mean dietary exposure to these 3 REE were 0.54, 0.31 and 0.17 $\mu\text{g}/\text{kg}$ bw, respectively. The sum of the exposures of these 3 REE accounted about 63% of the total exposure to the 16 analyzed REE. On the other hand, the most important contributors to the dietary intake of REE were vegetables and grains, with percentages of 45.3% and 28.6%, respectively. Although the level of REE in tea was the highest among all of the food categories, taking into account that the consumption of this product was comparatively very low, tea resulted in a low contribution (3.6%). Recently, Zhao et al. (2023) reported the results of a research whose main objective was to investigate the REE (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Sc and Y) levels in a number of samples of the most representing foods randomly selected from local wholesale agricultural markets in the Bayan Obo mining area (Inner Mongolia, China). The potential health risks of dietary exposure to these 16 REE was also assessed. The following samples were analyzed for the content of REE: 140 grain samples, 309 meat and aquatic samples, 153 vegetable samples, 15 fruit samples, 39 dairy samples, and 21 egg samples. The order of REE concentrations in all food samples was $\text{La} > \text{e} > \text{d} > \text{r} > \text{c} > \text{Gd} > \text{Eu} > \text{m} > \text{Tb} > \text{Er} > \text{m} > \text{y} > \text{b} > \text{o} > \text{Lu}$. Among all the food analyzed, meat and aquatic samples showed the highest REE levels, followed by grains, fruits, eggs, vegetables and dairy products. La and Ce were the most concentrated REE in the analyzed samples. Regarding human dietary exposure, cereals showed the highest contribution of REE, meaning a major source of exposure. The mean EDI of the total REE was 0.275 g/kg/day (range: 0.116-0.512 $\mu\text{g}/\text{kg}/\text{day}$). This mean value is below the recommended safe daily average intake of REE (70 g/kg/day) (Dai et al., 2022). Also in China, Zhuang et al. (2023) measured the concentrations of 16 REE (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Sc and Y) in various agricultural products collected near a large light rare earth mining area in Shandong Province. Their dietary intakes by local residents were subsequently estimated. The analyzed samples included wheat, maize, dry beans, vegetables, fruits and eggs. Considering all samples, the mean value of total REE was 286.96 $\mu\text{g}/\text{kg}$, being the content of LREE, 270.18 $\mu\text{g}/\text{kg}$. It accounted for more than 94% of total REE. Among the LREE, Ce, La, Nd and Pr were the dominant elements. The content of HREE was 16.77 $\mu\text{g}/\text{kg}$. Wheat, leafy vegetables, and allium vegetables had the

highest content of REE, being the main sources of dietary exposure to REE. In contrast, the lowest contents of REE were found in melons vegetables, root vegetables, fruits, and eggs. The EDI of REE was 4.20 $\mu\text{g}/\text{kg bw}/\text{day}$. This was a low level that did not exceed the acceptable daily intake. There were scarce differences in the intake of REE for the different age/gender groups of population.

7. Conclusions

Due to their expanding usage in a number of high-tech applications, REE are currently being of considerable importance for the modern world economy. A direct consequence of the increased use of these elements also means that REE are being now an emerging pollutant. Since its extensive use in a number of applications it may cause an increasing risk of environmental contamination (Brewer et al., 2022; Gwenzi et al., 2018). Consequently, there is also a potential increased human exposure to REE, elements on whose toxicity the available information are rather scarce (Hirano and Suzuki, 1996; Pagano et al., 2015; Brouziotis et al., 2022). It is more than probable that the use of REE will continue its increase in the next years. Therefore, exhaustive investigations aimed at establishing the human exposure and potential adverse effects of these elements are clearly required. In this paper, the results of the available studies focused mainly on determining the levels of REE in foodstuffs have been reviewed. As above indicated, the diet is the main route of exposure to metals for non-occupationally exposed subjects. Thus, to know the current dietary intake of REE is an important first step. The results of most studies here reviewed do not suggest potential health risks for the consumers of the analyzed freshwater and marine species of higher consumption, or derived from the intake of a number of vegetables, fruits, mushrooms and other various foodstuffs (honey, tea, rice, etc.). However, it should be expected that the environmental concentrations of REE will increase with the increased use of these elements, which could lead to increased levels of contamination of fish, vegetables, fruits, etc. Consequently, a continued monitoring of the levels of REE in food is strongly suggested. This monitoring is being already conducted with well-known toxic elements as arsenic, lead, cadmium or mercury, for example. To assess periodically the potential health risks of the dietary exposure to REE is, logically, another important issue. For this, it is essential to know/update the toxic effects of the 17 REE, for which information is not currently particularly abundant.

Declarations

Credit author statement

- Neus González: Writing – original draft
- José Luis Domingo: Conceptualization; Writing – review & editing

Conflict and/or competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have

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