



Sources of lanthanides in soils and estimation of their hazards

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Abstract: In recent years, an increase in the input of lanthanides (Ln), an important group of heavy metals (HMs), has been observed in the environment throughout the world (in particular soil). Anthropogenic Ln sources are mainly connected to phosphogypsum, Ln-bearing microfertilizer and micronutrients, and mine waste. Therefore, assessment of the hazards associated with the presence of Ln in the soil is an urgent issue. Hazardousness, depending strongly upon the buffer capacity of a specified soil, is determined by the organic matter content, the pH value and other soil properties. For a standard soil (containing 10% organic matter and 25% clay), the discrepancy of the hazardousness of HMs can be estimated based on the maximum permissible concentration (MPC) values proposed by ecologists in the Netherlands. Low levels of Ln contamination in the soil are found, making it possible to classify them in the group of moderately and weakly acting pollutants. The MPC value proposed for cerium (Ce) in the standard soil is 44 mg kg⁻¹. Approximate MPC values for several other lanthanides (Ln, Pr, Nd, Sm, Gd and Dy) in soils were proposed based on MPC values for freshwater sediments in the Netherlands. According to the MPC value, Ln falls into the group of moderate- and low-hazardous heavy metals.

Keywords: anthropogenic lanthanides; phosphogypsum; Ln-microfertilizers; Ln-micronutrients; maximum permissible concentration; mine waste

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Lanthanides (Ln) are a chemically uniform group of substances belonging to Group IIIB in the Periodic Table possessing similar physical and chemical properties. Formally, Ln has an atomic mass of 138.9–175.0 and includes 14 elements belonging to the group of heavy metals (HMs), with atomic mass exceeding that of V (50.9). Based on the electron configuration of the elements, the lanthanides are divided into two groups: light elements (La–Eu) and heavy elements (Gd–Lu) (Gonzalez *et al.* 2014). Light Ln are generally more soluble than heavy ones (Adeel *et al.* 2019). In some articles, the term rare earth elements (REEs) is used instead of lanthanides (Adeel *et al.* 2019). In recent decades, the interest in Ln has been increasing due to the use of Ln in the industrial production of high technological devices, agricultural fertilizers and animal feed additives (Schwabe *et al.* 2012). The use of Ln in various industrial products has resulted in the release of anthropogenic Ln to the environment, particularly in soil. Lanthanides are soft, malleable, ductile and good electrical conductors, making them suitable for applications in a variety of essential technologies, such as automotive, nuclear, petroleum, electronics (batteries, headphones, TVs), renewable energy, phosphor, ceramics, fertilizers and even as stains for biological cells (Haque *et al.* 2014; Ramos *et al.* 2016). Besides these, light-emitting diodes, batteries and the glass industry are also among Ln utilizations (Zhang *et al.* 2015). According to the United States Geological Survey (USGS), Ln global production increased from 124.000 tons in 2010 to 167.000 tons in 2018 – an increase of 35% in the last eight years. Reserves worldwide amount to 99 million tons (USEPA 2012). The release of REEs to the environment and their subsequent assimilation by organisms can be intensified by mining activities (Galhardi *et al.* 2017).

As demand rises, pressure over mining increases, since Ln recycling is appallingly low. However, one of the main issues with REE mine production is the so-called balance problem, where the vast majority of REE production is dominated by La and Ce but the majority of REE demand is for Nd or Dy (Binnemans and Jones

2015). Jowitt *et al.* (2018) estimated that merely 2.8% of all disposed REEs (10.683 out of 375.000 tons) is recycled.

Anthropogenic Ln strongly pollutes the environment (Adeel *et al.* 2019) when using microfertilizers (Pang *et al.* 2002; Xu *et al.* 2002). Particular attention is paid to the potential risk of intense industrialization and modern agronomy concerning the impact of Ln upon living organisms (Wu *et al.* 2018; Adeel *et al.* 2019). An exponential growth of scientific articles devoted to the impact of Ln began in the year 2000. The number of publications concerning this issue in peer-reviewed journals has increased from 27 in 2000 to 171 in 2018 (Kang and Kang 2020). At the same time, the average share of scientific articles about Ln in the environment accounts for 25% of the total publications in 2009 to 45% in 2018 (Kang and Kang 2020). Despite the availability of data on the toxic impact of high Ln doses on the soil biota and plants (Redling 2006; Pagano *et al.* 2015; Adeel *et al.* 2019; Kotelnikova *et al.* 2021), the permissible content of each Ln in soils remains debatable.

The hazardousness of Ln in soils is indicated by its maximum permissible concentration (MPC). The development of normative standards values for lanthanides in soils is an urgent issue. The calculation of MPC is hampered by the shortage of data on lanthanides and problems related to harmonizing such data (Sneller *et al.* 2000).

The present work aims to evaluate the sources of soil contamination with lanthanides and proposed MPC values for Ce, Sm, Gd, Dy, Ln, Pr and Hd.

Anthropogenic sources of lanthanides' application to soil

Ln in soils mainly originate from parent rock. In addition to natural processes, Ln content is also governed by anthropogenic factors that can be divided into two groups: ameliorative fertilization and pollution. The first group includes the use of phosphogypsum for ameliorating Solonetz soils and the input of microfertilizers (Liu

et al. 2013; Lyubimova and Salpagarova 2020). The second group consists of the contamination of arable soils with Ln from various sources, i.e. ore mine waste and industrial emissions (Delgado *et al.* 2012; Liang *et al.* 2014; Zhang *et al.* 2019). The recent increase of high-technology industrial applications of Ln in medicine, mobile communication, energy and electronics in many countries has resulted in the contamination of soil worldwide (Levy *et al.* 2017; Squadrone *et al.* 2019). The use of Ln in various industrial products has increased demand for these elements in many countries (Ramos *et al.* 2016). The global demand for these elements was around 30 000 tons for the years 2014–2020 (Weng *et al.* 2015).

Lanthanides in phosphogypsum

Doses of phosphogypsum addition to non-saline soils can vary from 1 to 7 ton ha⁻¹. The increased crop yield in the Krasnodarsk region, Russian Federation, was achieved by adding just 6 ton ha⁻¹ of phosphogypsum during soybean and corn sowing (Efremova *et al.* 2020). High contents of Ca, S, P, Mg, K, Zn, Mn and Cu in the phosphogypsum could enhance soil fertility.

More than 40 ton ha⁻¹ of phosphogypsum is not recommended in order to prevent soil contamination with Sr, F and Cd (Efremova *et al.* 2020). A study of amelioration of Solonetz soils in the Rostov region, Russian Federation, with the compost of manure and phosphogypsum revealed that an optimal compost dose is 40 ton ha⁻¹ of equal proportions of phosphogypsum and manure (Efremova *et al.* 2020). The maximum content of major lanthanides (La and Ce) entering the soil was determined at the phosphogypsum dosage of 40 ton ha⁻¹. Given that the soil layer is 30 cm and its density is 1 ton m⁻³, the soil mass in an area of 1 ha is 3000 ton. Thus, phosphogypsum accounts for 1.3% of the soil mass. The Balakovo acid phosphogypsum obtained from the Khibiny apatite concentrate contains $C_{La} = 1000$ mg kg⁻¹ of phosphogypsum and $C_{Ce} = 1400$ mg kg⁻¹ phosphogypsum (Table 1). In the neutralized phosphogypsum obtained from the Kovdor apatite raw material, contamination with La and Ce in the Murmansk region is an order of magnitude higher (Lyubimova and Salpagarova 2020).

The neutralized phosphogypsum dose accepted in Russia for ameliorating Solonetz soils does not lead to the hazardous Ln saturation in soils. The neutralized phosphogypsum obtained from the Kovdor apatite concentrate contains 0.011% La and 0.027% Ce (Lyubimova and Salpagarova 2020). At a phosphogypsum dose of 40 ton ha⁻¹, the soil gets just La: 1.4 mg kg⁻¹ and Ce: 3.5 mg kg⁻¹. It was also revealed that the amount of La (10 mg kg⁻¹) delivered with phosphogypsum is an order of magnitude lower than its limit value for Chinese soils (Ultisol and paddy soil) (Efremova *et al.* 2020).

Lanthanides as microfertilizers

Mixed Ln-fertilizers including a set of Ln are used widely in Chinese agronomy to enhance plant nutrition (Xu *et al.* 2002). The small amount of Ln promotes the production of biomass and the

growth of vascular plants (Liu *et al.* 2013; Kotelnikova *et al.* 2021). Lanthanides have been used in Chinese agronomical practices since 1972 (Tyler 2004). According to Xu *et al.* (2002), the common Ln dose does not exceed 0.23 kg ha⁻¹ per year, corresponding to c. 0.1 mg kg⁻¹ per year. Lanthanides are introduced in the dissolved form as nitrates, chlorides or amino acid mixtures. In 1995, the volume of Ln-fertilizer production corresponded to an area of 16–20 mln ha (Pang *et al.* 2002).

Lanthanides are introduced annually by seed powdering or foliar fertilizing. Direct addition of c. 0.1 mg kg⁻¹ to the soil yields a weak (or zero) result and, therefore, it is not recommended. In heavy soils, the introduced Ln are fixed rapidly by the solid phase and become inaccessible for the plants. In light soils, the indigested Ln are washed out, and can contaminate the groundwater.

Although high crop yield is recorded at a moderate La concentration in the solution under laboratory experiments, such a positive effect is not observed under field conditions. This situation was recorded in China at Ultisol and rise soil with a high sorption capacity that absorbed the main share of the added La (Xu *et al.* 2002).

The optimal dose is different for each specified Ln. The maximum increase in the pea crop (29–35%) is achieved if the dose of La in the soil (35 mg kg⁻¹) is 6 mg kg⁻¹, but the optimal dose of Sm (7 mg kg⁻¹) is two times lower (Kozhevnikova and Ermakova 2008). Therefore, it is necessary to study the influence of each Ln separately, and different MPC values should likely be prescribed for Ln.

The toxic La dose is rather low: the productivity of plants decreased when more than 10 mg kg⁻¹ was added to the highly sorptive Ultisol and paddy field in China (Xie *et al.* 2001). The saturation of the specified soils can govern the low limit dose of La with competing cations (first of all, Ca²⁺).

A high critical Ln concentration was recorded in vegetative experiments with the introduction of variable doses of La, Ce and Nd chlorides in samples of soddy-podzolic soil (Kotelnikova *et al.* 2021). Soils neutralize the Ln toxicity of other HMs due to the buffer capacity (Bauer *et al.* 2018). The main toxic effect is manifested at a very high input of these Ln to the soddy-podzolic soil: $\Delta Ln = 200$ mg kg⁻¹. The buffer capacity can be high, for example, if the soil-absorbing complex (SAC) in the soddy-podzolic soil lacks Ca²⁺ that can drive Ln, such as La³⁺, Ce³⁺, Pr³⁺, Nd³⁺, into the solution. These elements have an ionic radius similar to that of Ca²⁺. Therefore, they are able to competitively replace Ca²⁺ ions in biological systems (Evans 1983). Parameters such as total organic carbon content, particle size distribution, Eh/pH, influence the sorption capacity of soils in relation to lanthanum like other HMs: the higher the parameter's value, the higher the sorption capacity in relation to lanthanum (Pinskii *et al.* 2018).

Because of the hazard of Ln, researchers have studied the possibility of biological remediation of soils by growing plants that can absorb the excess Ln and decontaminate the soil. However, such studies came across difficulties related to the Ln toxicity that

Table 1. Maximum content of lanthanide in soils of different countries, mg kg⁻¹

Country/soil	Source of supply	Content	Reference
Russia/agricultural	Phosphogypsum, Khibiny	$C_{La} = 13$ mg kg ⁻¹ , $C_{Ce} = 18$ mg kg ⁻¹	Recommendations 2006
Russia/agricultural	Phosphogypsum, Kovdor	$C_{La} = 1.4$ mg kg ⁻¹ , $C_{Ce} = 3.5$ mg kg ⁻¹	Recommendations 2006
Russia/podzols	Laboratory experiments	La = 35 mg kg ⁻¹ , $S_m = 7$ mg kg ⁻¹	Kozhevnikova and Ermakova (2008)
Russia/podzols	Laboratory experiments with addition of chlorides La, Ce, Nd	≥ 200 mg kg ⁻¹ of La, Ce, Nd	Kotelnikova <i>et al.</i> (2021)
China/Ultisol and paddy soil	Microfertilizers	0.1 mg kg ⁻¹ of light lanthanides	Xu <i>et al.</i> (2002)
China/Ultisol and paddy soil	Microfertilizers	La = 10 mg kg ⁻¹	Xie <i>et al.</i> (2001)
China/soil in general	Parent rock	$\Sigma Ln = 181$ mg kg ⁻¹	Adeel <i>et al.</i> (2019)
China/upper soil horizons	Near quarries	$\Sigma Ln = 4670$ mg kg ⁻¹	Adeel <i>et al.</i> (2019)
China/agricultural soils	Microfertilizers	$\Sigma Ln = 455$ –502 mg kg ⁻¹	Meryem <i>et al.</i> (2016)

impedes the growth of plant hyperaccumulators of Ln (Rezaee *et al.* 2018).

Lanthanides as micronutrients

Manure of farm animals and sewage sludge have been revealed as additional sources of arable soil contamination (Kawasaki *et al.* 1998; Hu *et al.* 2006; Kaegi *et al.* 2021). Additives in animal fodder change the composition of animal manure used as fertilizers. In order to avoid infection among birds and farm animals, antibiotics were added for many years to the foodstuff. After their prohibition, farmers began to add microelements (Cu and Zn) to replace antibiotics, although the mechanism of their influence is different. Microelements suppress the activity of microbiota that breaks down the food in the animal's stomach, achieving a complete absorption of nutrients (vitamins included) in the organism. This principle occurs in the case of Ln, which appeared more efficient than Cu and Zn and replaced the traditional microelements. Therefore, Ln addition to animal fodder was decreased. For example, only 100–130 mg per 1 kg fodder is added as ascorbates or citrates for pigs (Chen 1997). Higher Ln doses in the fodder provoke a decrease in animal growth and other negative consequences (Panichev 2015).

The technique of Ln addition to fodder initiated in China and Switzerland spread into countries in West Europe (Redling 2006). On the one hand, Ln increase the amount of consumed fodder and decrease the volume of animal faeces, mitigating the environmental impact of cattle farms (Redling 2006). On the other hand, the manure used as organic fertilizer is enriched considerably in Ln, such as Sm, Eu, Gd and Tb (Zhang *et al.* 2019).

Sewage sludge is also contaminated with Ln. In Japan, the Ln content in sewage sludge ash varies from 54 to 130 mg per 1 kg of sediment mass. Contents of Eu and Tb are also increased (Zhang *et al.* 2019). The long-term application of sewage sludge and ash from incinerated domestic wastes can provoke the accumulation of Sc, Sm and Eu in natural Ln-depleted soils.

Thus, arable soil can be contaminated with Ln together with manure and sewage sludge. The properties of these Ln have been studied insufficiently, and normative standards for them are lacking so far.

Soil contamination with lanthanides

Like other HMs, Ln often enter the soil via the atmosphere. However, active and abandoned mines represent the main source of soil contamination with Ln (Humsa and Srivastava 2015; Meryem *et al.* 2016). The Baiyun Mine (Baotou, China) is the largest iron ore and Ln mining site in the world. The greatest amount of Ln (30%) is used in the production of polished glass and ceramics. The second place (28%) belongs to Ln-catalyzers for oil purification. A significant part (19%) of Ln is used as an addition to steel and alloys (Castor and Hedrick 2006). The use of Ln in high-tech productions and medicine is increasing year by year. It has been proved that the aerial contamination of soils with Ln is highest in China (Adeel *et al.* 2019). The soil pollution increases the risk of Ln input to plants and animal and human organisms. Phosphogypsum contaminates the areas located near plants specializing in the production of phosphorus fertilizers. Soils in such areas are enriched in Ln; first of all, elements with a high background value: La, Ce and others (Borges *et al.* 2018). Whereas the total Ln (Σ Ln) content in surficial soil horizons near the mine varies from 156 to 56 500 mg kg⁻¹ (average Σ Ln = 4670 mg kg⁻¹), while the sum of background values of Ln in Chinese soils is 181 mg kg⁻¹ (Adeel *et al.* 2019). In the area of anomaly (up to 56 500 mg kg⁻¹), the buffer capacity of soils decreases, which should be reflected in a decrease in the standards for Ln. Moreover, the contamination of soil and water with Ln in this area increases year by year (Adeel *et al.* 2019). In the

agricultural soil of Xujan, the Σ Ln content is 455–502 mg kg⁻¹ (Meryem *et al.* 2016).

Quotas for the export of Ln were reduced severely by the Chinese government in 2010–2011, affecting the price policy of other countries. Because of a lack of ecological requirements, soils near mine wastes in China are characterized by a high degree of contamination (Folger 2011). The total concentration of all REEs in agricultural soils at the Ln mining sites in northern China varies from 83 to 9840 mg kg⁻¹. The main Ln mineral is represented by bastnaesite – fluorocarbonate of La and Ln (M^{III}CO₃F). The concentration of light rare earth elements (LREEs) in this mineral is higher relative to heavy rare earth elements (HREEs). Among the LREEs, Ce is the most widespread element, while La, Nd and Eu are subordinate. At the same time, the HREE concentration in the agricultural soil in China is as much as 37 mg kg⁻¹. It was established that the Dy content is highest among HREEs in the Chinese soil (Adeel *et al.* 2019). One of the main Fe–REE deposits is located in Bayan Obo, Inner Mongolia, China (Castor and Hedrick 2006). The complex technology of ore dressing produces large amounts of Fe-rich wastes. The carbonate-related deposits are enriched in LREEs (Simandl 2014).

The largest REE-bearing deposits are known in southern Western Australia, where laterites are developed on carbonate rocks. They are marked by a high share of HREEs and Y, including 11.2% of R₂O₃ + Y₂O₃ (Mariano 1989). The abundance of mine wastes containing 0.33% Y₂O₃, Nd and Ta is described in O'Driscoll (1988). In Brazil, laterites containing 13.5% R₂O₃ are confined to phosphate minerals.

Lanthanides occur naturally not as metallic elements but as a constituent in a wide range of mineral types, including halides, carbonates, oxides and phosphates (Table 2). The REEs occur mainly as substitutes for the major ions in rock-forming minerals. High REE concentrations are needed to form minerals of these elements.

At present, great attention is devoted to Ln-bearing ore mining in western countries (Canada and Australia), where the mining of such ores has an industrial significance (Simandl 2014).

Lanthanide deposits can be divided into two types. Type 1 includes deposits mined at present as the main REE sources. These deposits accommodate carbonates and clay minerals. Type 2 includes deposits previously exploited for the mining of apatites, uranium ores and phosphates (Simandl 2010, 2012).

Soils in such mining areas are marked by a higher degree of REE contamination. The urban and agricultural sectors are less contaminated. However, it is also important to examine the concentration of REEs in the urban and agricultural soils because of their bioaccumulation in plants of various anthropogenic media (Hu *et al.* 2004; Tyler 2004; Adeel *et al.* 2019).

Buffer capacity of soil towards lanthanides

Control of the environmental contamination by HMs is based on the notion of MPC that depends strongly on the metal concentration

Table 2. Values of the maximum permissible addition (MPA) for freshwater sediments and standard soil in the Netherlands (Sneller *et al.* 2000)

Lanthanides	Atom no	Sediment (g kg ⁻¹ d.w.)	Soil (mg kg ⁻¹ d.w.)
La	57	4.7	11*
Ce	58	18.7	44
Pr	59	5.8	14*
Nd	60	7.2	17*
Sm	62	2.5	6*
Gd	64	1.8	4*
Dy	66	2.2	5*

Calculated MPA values are marked by asterisk*. d.w., dry weight.

traditionally estimated by the dose-effect correlation. In addition, the MPC value is governed by the buffer capacity of the specified soil. Different buffer capacities of soils rule out the law applied to Ln: prescription of a unified concentration level permissible for the given metal.

In contrast to air and water, soils, even within a zonal-genetic series, differ so much in chemical and mineral composition that it is impossible to prescribe single MPC values for them. Therefore, when specifying the MPC value for metal, one should take into account the parameters of buffer capacity defining the Ln fixation in soil. The buffer capacity is governed by the contents of organic matter and silty particles and the pH value (Minkina *et al.* 2018). The factor of organic matter is particularly important because of the tendency of Ln to form Ln-organic complexes (Fedotov *et al.* 2019).

This problem is resolved partly by introducing the approximate concentration (APC) that considers some indicators of the buffer capacity of soil: grain-size composition and the pH value of soil (HN 2009). For example, the APC value of Ni increases from 20 mg kg⁻¹ for light-textured soils to 40 mg kg⁻¹ for acid-heavy-textured soils and 80 mg kg⁻¹ for neutral heavy-textured soils (HN 2009). Thus, the permissible Ni concentration correlates with the buffer capacity of the soil (i.e. increase of the share of clay particles and pH value). The sorption capacity of cationogenic metals (Ni included) correlates directly with the pH value. Since Ln belong to the group of cationogenic metals, their APC values should also correlate positively with the pH value of soil.

Light Ln show a physical and chemical affinity to Ca²⁺ because of the similarity of ionic radii: Ca²⁺ (0.099 nm), La³⁺ (0.110 nm), Ce³⁺ (0.102 nm), Pr³⁺ (0.099 nm) and Nd³⁺ (0.098 nm) (Greenwood and Earnshaw 1997). High Ca²⁺ content in the soil-adsorbing complex (SAC) increases Ln mobility, which is harmful to the development of plants (Brown *et al.* 1990).

Phosphorus has a reverse effect. Ln are fixed in the soil in phosphates. In P₂O₅-rich soils, the bulk La content also certainly increases (Viers and Wasserburg 2004). Thus, we should take into account the share of the SAC-incorporated Ca²⁺ in the soil and the content of active phosphates.

The MPC/APC standard has a common shortcoming associated with soil pollution. It was estimated based on fixed concentrations of heavy metals and metalloids, without differentiation of their natural and anthropogenic components, consequently leading to overestimation of the danger of contamination in the positive geochemical underestimation in the negative natural anomaly area. The fixed normative values ignore the natural climatic and geochemical peculiarities of regions that can be very contrasting.

Flexible normative standards for lanthanides

The concept of flexible MPC makes it possible to dispense with the fixed normative standards. The flexible MPC takes into account both background element content and MPC (Van de Plassche and De Bruijn 1992; Crommentuijn *et al.* 1997). Evidently, the MPC value can be used for characterizing the hazardousness of a specified lanthanide element in soils. The MPC value for a chemical element in the soil is one of the addends in the MPC determination formula:

$$\text{MPC} = \text{background} + \text{MPC}.$$

The buffer capacity of soils is indicated by the grain size composition (silty particles <2 µm) and the content of organic matter (Crommentuijn *et al.* 1997). The standard is represented by soil containing 25% of silt and 10% organic matter. Empirical formulas are proposed for calculating MPC for Ln in the case of a real soil of another composition.

The flexible normalization based on the differentiation of concentration into two Ln groups – natural (background) and anthropogenic MPC – makes it possible to define the local significance of MPC and eliminate the shortcomings of normative standards related to the application of fixed numbers. MPC values were obtained by numerous and diverse ecotoxicological studies. The principle accepted in the Netherlands (Crommentuijn *et al.* 1997) is based on the mathematical harmonization of a great number of experimental works (160 papers) dedicated to the influence of heavy metals/metalloids on the biota and plants.

According to the proposals of the Dutch researchers (Crommentuijn *et al.* 1997), MPC_{Ce} = 44 mg kg⁻¹ soil. This value is 4.5 times lower than MPC = 200 mg kg⁻¹ for most Ln (Ce included) proposed in Kotelnikova *et al.* (2021). Let us compare this value of MPC Ce = 44 mg kg⁻¹ with the data obtained in Russia on soddy-podzolic soil with low C_{org} content. It has been shown that the mitotoxic effect on plants is manifested only at MPC = 200 mg kg⁻¹ for most Ln (including Ce) introduced in the form of mobile salts (Kotelnikova *et al.* 2021). Thus, the prescribed normative value is 4.5 times more rigorous than the permissible Ce addition to the soil. The maximum phosphogypsum addition to the soddy-podzolic soil is 28 mg kg⁻¹ soil, which is 1.7 times lower than MPC_{Ce} = 44 mg kg⁻¹ reported in Van de Plassche and De Bruijn (1992) and Crommentuijn *et al.* (1997). Soil with a high buffering capacity is able to neutralize the toxic effect of Ln introduced in the composition of mobile forms in laboratory experiments (Kotelnikova *et al.* 2021). Therefore, the proposal of Dutch ecologists regarding Ce as a moderately hazardous pollutant can be considered quite reasonable.

Hazardousness of lanthanides in soil relative to other heavy metals

Since the MPC of Ln depends strongly on the buffer capacity of the soil, the issue of the Ln toxicity can only be solved if the buffer capacity of soils is taken into consideration. However, equations for recalculating the MPC value of Ln in the standard soil into the MPC value in the given soil are lacking so far. Therefore, flexible normative standards cannot be applied for soils strongly differing from the standard soil in the Netherlands.

However, MPC values can be useful for comparing the hazardousness of different metals under comparable conditions (standard soil in the Netherlands). The MPC values could be applied for determining the Ln hazard in soil relative to other heavy metals.

To solve this issue, MPC values proposed by the Dutch ecologists for freshwater sediments could be applied. The main distinction between the standard soil and sediments under consideration lies in elevated buffer capacity due to the organic matter's high content.

For Ce, the increase of MPC due to soil buffer capacity (at 10% organic matter) relative to the freshwater sediments is 44: 18.7 = 2.35. If this multiplying coefficient is applied for other Ln with a high background value, we obtain approximate MPC values for Ln in soils, taking into account the effect of C_{org} (Table 2).

Although the calculation is conventional, MPC values provide insight into the relative hazardousness of different soils with a similar buffering capacity. In general, the odd-numbered Ln are more hazardous than the even-numbered ones. La and Gd, respectively, are the most hazardous elements among the odd- and even-numbered Ln.

Values of MPC make it possible to rank 17 heavy metals/metalloids (7 Ln included) and distinguish among them the most hazardous metals in soils (Crommentuijn *et al.* 1997; Sneller *et al.* 2000). Values of MPC show a wide variation range: from 0.0061 for the light metal Be (most toxic element) to 253 mg kg⁻¹ for Mo (least toxic element). The wide variation reflects the difference in the hazardousness of elements in the soil.

Table 3. Hazardousness of chemical elements, according to the general toxicological normative standards for soils accepted in Russia (GOST 2008) and the Netherlands (Crommentuijn *et al.* 1997)

Hazard class	Russia	MPA, mg kg ⁻¹	Netherlands
1. High	As, Cd, Hg, Se, Pb, Zn	<1	Be, Se, Tl, Sb, Cd
2. Middle	Co, Ni, Mo, Cu, Sb, Cr	1–10	V, Hg, Ni, Cu, Cr, As, Ba, Sm*, Gd*, Dy*
3. Low	Ba, V, W, Mn, Sr	>10	Zn, Co, Sn, Ce, Pb, Mo, Ce, La*, Pr*, Nd*

Note: (*) Computed MPA values of lanthanides.

The values of MPC of elements could be divided into three hazard classes, as accepted in Russia (GOST 2008). Class 1 includes elements with MPC < 1 mg kg⁻¹; class 2, elements with MPC 1–10 mg kg⁻¹; and class 3, elements with MPC > 10 mg kg⁻¹ (Table 3).

There is a significant distinction in the hazardousness of heavy metals/metalloids based on the MPC value in soils, according to the general toxicological normative standards accepted in Russia and the Netherlands. The list of highly hazardous elements includes only two common elements: Se and Cd. Based on low MPC values, the list of the remaining highly hazardous elements is supplemented with three certainly hazardous elements (Be, Tl and Sb) (Greenwood and Earnshaw 1997), for which MPC/APC values have not been prescribed in Russia.

The number of moderately hazardous elements (with Sm, Gd and Dy added by us) increased from 6 to 10. According to this list of moderately hazardous elements, the hazardousness of three medium-classed Ln is comparable with that of known pollutants, such as Hg and As.

The number of low-hazardous elements increased most of all: from 5 to 10. Their list includes four Ln with a high background value: Ce, La, Pr and Nd.

Technologies for remediation of Ln-contaminated soils

Based on the foregoing, the effective recovery of soils contaminated with REE and further extraction of valuable REEs is an urgent task. In general, soil restoration methods can be divided into stabilization methods and extraction methods (Lima and Ottosen 2021). Soil amendments and bioremediation are well-known methods as the common type of stabilization methods. The amendments consist of adding ameliorants to the soil (Dresel *et al.* 2011). The basic principle of application of various additives is based on the pollutants' sorption or their precipitation in a new phase. Several adjustments for REE reduction have been used, including phosphates (Jin *et al.* 2019), biochar (Riedel *et al.* 2015; Pourret and Houben 2018), iron nanoparticles (Su 2017) and coal fly ash (He *et al.* 2019). In addition, calcite, apatite and iron hydroxides (goethite and magnetite) were found to significantly reduce the mobility of elements in studies of the REE sorption by the surface of minerals (Edahbi *et al.* 2018). The role of humic acids in the binding of REEs was shown (Marsac *et al.* 2010).

Bioremediation continues to be one of the most common approaches to soil restoration (Mani and Kumar 2014; Azubuike *et al.* 2016). This situation is largely due to the relatively low cost, flexibility in application, and a wide range of adapted conditions for which these approaches can be used. At the same time, the effectiveness of bioremediation, as a rule, depends on the quality of the work performed and the choice of a specific technology. In addition, bioremediation methods are quite laborious, do not always show high efficiency for highly contaminated soils and are more often used for agricultural land (Paudyn *et al.* 2008).

Although stabilization methods show good effectiveness in soil remediation, they only have a short-term effect – until environmental conditions become prone to re-leaching the contaminant (for example, changes in soil pH, redox conditions, etc.).

The second group of methods has the opposite effect of stabilization methods: removing and isolating contamination from the soil. This group of methods includes three main types: soil leaching, phytoremediation and electrokinetic remediation.

Due to its speed and cost-effectiveness, soil leaching is one of the most effective methods of soil remediation (Li *et al.* 2020). The efficiency of leaching is inextricably linked with the ability of the extractants to dissolve the target pollutants in the soil. In this regard, the aim of leaching techniques is desorbing, dissolving and leaching the target contaminant with biosurfactants saponin and rhamnolipid (Zhou *et al.* 2017) or nitric acid, citric acid and EDTA (Tang *et al.* 2017). Previous studies using shaking and washing of soil columns have confirmed the leaching efficiency of saponins in removing REEs from raw ores (Gao *et al.* 2018).

Special attention should be paid to phytoremediation methods based on the ability of plants to accumulate and promote REE resistance and indirectly influence the biological properties of soils, such as metabolic activity and rhizosphere functioning (Chour *et al.* 2018). The undoubted advantage of this approach is the validity and clarity of the operating mechanisms and good knowledge of the specific technological solutions. For example, in the USA and Canada, private companies in ecology and nature management have been developing and using phytoremediation technologies for a long time (Sharma and Pandey 2014). Phytore can be applied for a long time, and the collected biomass can be reused for energy and biofuel production (Dastyar *et al.* 2019). This method is already widely used around mining in China, reporting high removal yields (up to 96% REEs) (Chour *et al.* 2018).

Thus, the choice of a specific direction in implementing measures for the restoration of the soil contaminated with REE should include a detailed analysis of the studied objects, and the possible risks and delayed effects of a specific strategy require further research.

Conclusion

Lanthanides fall into the moderate low-hazardous heavy metal groups. At the same time a significant increase in Ln input to the soil makes the critical assessment of associated hazardous risk an urgent issue. The hazardousness of Ln relies on both peculiarities of each metal and the buffer capacity of the given soil. The buffer capacity, in turn, depends on the content of organic matter, pH value and other soil properties. Special attention is paid to the necessity of defining the MPC Ln contents in soils with different buffer capacities and methods of soil remediation contaminated with REEs.

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(supporting), project administration (lead), writing – review & editing (equal); **TB**: data curation (supporting), investigation (supporting), project administration (supporting), writing – review & editing (supporting)

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