

Research Article

Lanthanum Uptake and Distribution in Agricultural and Wild Plants: Insights from Greenhouse and Field Studies in Uncontaminated Soils

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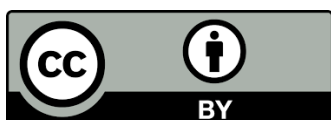
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Lanthanum (La) is widely used in various fields. As a consequence, it can accumulate in soil and thus pose a potential hazard to the environment, including a decrease of plant biomass and chlorophyll content. To date, most experimental studies on the biogeochemistry of La have focused on toxic effects resulting from its accumulation in plants growing in La-contaminated areas, while the behavior of La in plants growing in uncontaminated soils has been the subject of much less research. The primary objectives of this work were to investigate the impact of various factors on La uptake by plants growing in uncontaminated soils and to examine the relationships between La and other lanthanides (Ce, Sm, Eu, Tb, Yb, Lu) in the rhizosphere soil and across different plant species. Greenhouse and field experiments were conducted to investigate the patterns of La uptake and its redistribution between different parts of some agricultural plants (wheat and rye) and weeds (couch grass and plantain) growing in clean soils. The correlation between La in the rhizosphere soil, roots, and leaves and between La and other lanthanides in the soil and plants was studied. Although the natural concentration of La in the soils was usually higher than in plants, plant growth in La-uncontaminated soil did not lead to its bioaccumulation. The correlation between La in the rhizosphere soil and roots, as well as between La in roots and leaves, was statistically insignificant. This may be due to a genetically determined need of a particular plant for La. Different plant species growing under the same conditions were capable of accumulating different amounts of La. Despite the assumption that rare earth elements can substitute for



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Ca, the correlation between La and Ca was often statistically insignificant. On the other hand, in most cases, La had a positive relationship with Ce, Sm, Sc, Fe, and Co.

Keywords

Lanthanum; rare earth elements; rhizosphere soil; plant uptake; agricultural plants; weeds

1. Introduction

Rare earth elements (REEs) are usually defined as lanthanum (La) and 14 elements that make up the lanthanide series [1]. Scandium (Sc) is also commonly included in the REE group. Despite the use of modern analytical methods for the reliable determination of low concentrations of REEs and the increasing information on reference values of these elements in soils and plants, the biogeochemistry of REEs is still poorly understood [2, 3].

Because La is widely used in many fields, there is concern about its potential accumulation in the environment [4]. The bulk of the published literature on La in soils and plants is devoted to the toxic effects of this trace element resulting from its accumulation in plants. On the other hand, the behavior of La in plants growing in clean soils has been the subject of relatively few studies. Among others, the following articles should be mentioned [5-7]. Based on the available information, the following preliminary conclusions can be drawn. In plants growing on soils with high available REE concentrations, the process of REE accumulation occurs, apparently, mainly due to passive transport from the soil to the plant. In cases where REE concentrations in the soil are low and do not exceed a certain level, the process of REE absorption is more complex. Interestingly, the opinions on the effects of La on plants can often be directly opposite. In addition to toxicity, beneficial effects of low doses of La on plants have been noted not only previously but also recently [8-12].

Among the lanthanides, La is an exception because it has 4f electrons in one gas-phase atom [13]. As a result, it is only very weakly paramagnetic, unlike the strongly paramagnetic other lanthanides (except for the latter two, Yb and Lu, which have a filled 4f shell). In this regard, specific differences in the behavior of La and other lanthanides in the environment can be expected. In unpolluted soils, the concentrations of La can vary considerably due to numerous factors. The range of La concentrations in such soils is usually 18-56 mg kg⁻¹, while in plants growing in these soils, it is 0.0032-0.15 mg kg⁻¹.

Lanthanides have similar ionic radius to Ca [14]. It has been suggested that REEs can replace Ca in specific physiological processes [14-16]. However, there is an opposite opinion about the relationships between Ca and lanthanides in biological systems. It has been shown that Ca²⁺ proteins are not inhibited by Ln³⁺ [17, 18]. This is likely because, although the ionic radius of Ca²⁺ and Ln³⁺ are similar, the radius of Ln³⁺ is slightly larger than radius of Ca²⁺. This is especially true for La, since it has the largest ionic radius compared to other lanthanides [19]. It was also reported that the transfer of REEs to chloroplasts is commonly achieved through other specialized processes [20].

Uptake of La by plants is affected by many factors that can have a significant impact on the availability and bioaccumulation of this trace element. In this paper, based on published data [2, 6, 12] and our experimental results, a hypothesis is put forward that, under normal conditions (plant

growth in La-uncontaminated soil), the main factors influencing plant growth are plant species, soil characteristics, and growing conditions.

The primary objectives of the research were the following: (1) to assess the uptake of La and some other macro-nutrients (Na, K, Ca, Fe) and trace elements (Sc, Co, Ce, Sm, Eu, Tb, Yb, Lu) by plants growing in uncontaminated soils; (2) to study the influence of a number of factors such as soil pH, texture, plant species, growth media on La uptake by plants; (3) to examine the accumulation of La and its redistribution between different parts of wheat seedlings growing under the same conditions in soil and in different liquid media; (4) to evaluate the relationships between La in the rhizosphere soil and different plant species and also between La and other lanthanides in soils and plants.

2. Materials and Methods

2.1 Test with Wheat Seedlings Grown in Different Media

Five-day-old uniform germinated seedlings of wheat *Triticum vulgare* (vill) Horst were divided into four parts. One part of the seedlings was transferred to pots filled with soil (2 kg of soil in a pot). Other seedlings were transferred to jars filled with either distilled water, or water taken from a spring, or a modified nutrient solution of Hoagland [21] (3 L of water in each jar). The spring was located in a park, in the suburbs of St. Petersburg, Russia (59°83'N, 30°38'E), away from sources of pollution. Soil was collected near the spring from the upper (0-10 cm) soil horizon. The soil was classified as a Ferric Podzol with a loamy texture (sand 38%, silt 37%, clay 25%). The concentrations of La, some other REEs, and essential nutrients (K, Ca, and Fe) in the growth media are shown in Table 1. The soil pH was 6.5 ± 0.4 . First series of soil and water samples was taken before the beginning of the experiment. The water in the jars was aerated throughout the experiment. The soil was watered daily. The wheat seedlings were harvested within ten days after planting.

Table 1 Mean concentrations $\pm$ SD of elements in liquid media (mg L^{-1}) and soil (mg kg^{-1}). *Concentrations of the elements are present in percentages.

Element	Distilled water	Nutrient solution	Spring water	Soil
K	0.41 ± 0.05	34.2 ± 4.0	2.02 ± 0.65	$3.07 \pm 0.65^*$
Ca	0.32 ± 0.04	12.1 ± 2.1	77.5 ± 5.4	$2.94 \pm 0.73^*$
Fe	0.45 ± 0.05	3.53 ± 0.70	0.98 ± 0.12	$1.64 \pm 0.16^*$
La	0.0092 ± 0.0052	1.02 ± 0.05	1.44 ± 0.14	29.6 ± 4.2
Ce	0.015 ± 0.004	1.37 ± 0.15	1.70 ± 0.15	34.6 ± 1.1
Sm	0.002 ± 0.001	0.63 ± 0.06	0.84 ± 0.07	4.08 ± 0.75
Eu	<0.002	0.08 ± 0.03	0.072 ± 0.030	5.34 ± 0.20

2.2 Greenhouse Pot Experiment

Five-day-old germinated seedlings of wheat *Triticum aestivum* L. and rye *Secale cereale* L. were transferred to pots with a volume of 5 kg filled with soil. The soil was classified as an urban podzol with a loamy sand texture (sand 74%, silt 24%, clay 2%). Soil pH was 6.3 ± 0.2 . The temperature in a naturally illuminated greenhouse was usually 25°C during daytime and 20°C at night. Plants and soil

from the surface of the plant roots were collected within ten days after transfer of the seedlings to the soil.

2.3 Field Trial

Two weeds were chosen for the experiment: couch grass *Elytrigia repens* L. Nevski and plantain *Plantago major* L. The site was located in an uncontaminated area (a park in St. Petersburg, Russia). The soil at the site was classified as urbostratozem with a clay loam texture (sand 24%, silt 43%, clay 33%). Soil pH was 6.1 ± 0.3 . Plants and soil from the surface of the plant roots were collected.

2.4 Sampling and Preparation of Plant and Soil Samples for Analysis

All the experiments were performed in triplicate. To provide the reproducibility of elemental analysis, several plants of each species were collected from all the sites. A completely randomized design was used for sampling. The soil was taken from the plant roots. To collect rhizosphere soil samples, the soil was gently shaken from the plant roots. The soil residues adhering to the roots were then collected. After sampling, plants were separated into roots and leaves, washed carefully with deionized water in order to remove from the plant surface dust and soil particles. Then the plant and soil samples were air-dried up to a constant weight. The plant samples were ground. The soil samples were sieved through a 2 mm mesh to remove non-soil materials including plant fragments and then ground to fine powder. The pH (1:2.5 H₂O) of each soil sample was determined. The granulometric composition of the soil samples was determined using the small-angle laser diffraction method on a laser granulometer Shimadzu SALD-2201 (Japan). The data were processed using the WingSALD software.

2.5 Analysis of Plant and Soil Material

Instrumental neutron activation analysis (INAA) was used to determine the concentrations of Ca, Sc, Fe, Co, La, Sm, Eu, Tb, Yb, and Lu in the plant and soil samples. The elemental analysis by INAA can be performed without additional sample pre-treatment, thereby reducing the level of analytical errors that may arise during sample preparation [22]. The amount of soil and plant material taken for elemental analysis was between 50 and 100 mg. The samples were irradiated in a nuclear reactor for 18 hours (soils) and 24 hours (plants) in a thermal neutron flux of $1 \times 10^{14} \text{ n} \times \text{cm}^{-2} \times \text{s}^{-1}$. The spectra of the irradiated samples were measured twice, once after one week and once after three weeks of irradiation, using high-purity germanium detectors. The k0 method was used to calculate the element concentrations [23]. The certified reference materials (CRMs) NIST 1573 (tomato leaves) from the National Institute of Standards and Technology (NIST, Gaithersburg, MD, USA) and SOIL-7 (International Atomic Energy Agency) were analyzed together with plant and soil samples. The results of the analysis of the CRMs showed a good agreement with certified values. The accuracy of the element determination was 1-2%. The ICP-MS was applied for the analysis of liquid samples. The analysis was performed by a VG Plasma Quad PQ2+ ICP-MS instrument (VG Elemental, Winsford, Cheshire, UK). Single-element standard solutions purchased from Inorganic Ventures (Lakewood, NJ) were utilized to prepare calibration and internal standard solutions. Analysis was performed using an external calibration procedure. Internal standards were included for matrix and instrumental

drift corrections. Procedural blanks were analyzed to determine if there was any contribution from the reagents.

2.6 Statistical Analysis

Data analysis was performed using the STATISTICA for Windows 8.0 work package (StatSoft, Tulsa, OK, USA). Mean concentrations of elements and differences between groups of samples were calculated. For all statistical tests, the significance level was set at $P < 0.05$. The normal distribution of the data set was verified using the Shapiro-Wilk's test. Pearson correlation analysis was used to study the relationships between different elements. In addition, cluster analysis (Ward's method) was applied to better understand the uptake of La and some other REEs by different plant species and in the rhizosphere soil, as well as to assess the contribution of various factors that may influence soil - plant interactions. For cluster analysis, the original data were standardized.

3. Results and Discussion

3.1 Effect of Different Growth Media on Uptake of La by Wheat Seedlings

To simplify the experimental conditions, some researchers often conduct experiments on the absorption patterns of elements using liquid nutrient media [24-26]. The purpose of the test was to compare the ability of plants growing in different media to accumulate La.

Growing wheat seedlings in soil and liquid media had different effects on La accumulation. The concentration of La in roots of the wheat seedlings grown in soil was statistically significantly higher than in roots of the seedlings grown in distilled water, water taken from spring, and in nutrient solution (Figure 1a). However, La concentrations in leaves were similar regardless of where the plants grew (Figure 1b). This may mean that a certain amount of La is required for wheat seedling development. Although roots are able to uptake more of this trace element (e.g. if its concentration in the growth medium is higher than in the plants), the transfer of La to the upper parts of the plants will be limited. A low translocation of La from roots to upper plant parts was also reported by other researchers [27-29].

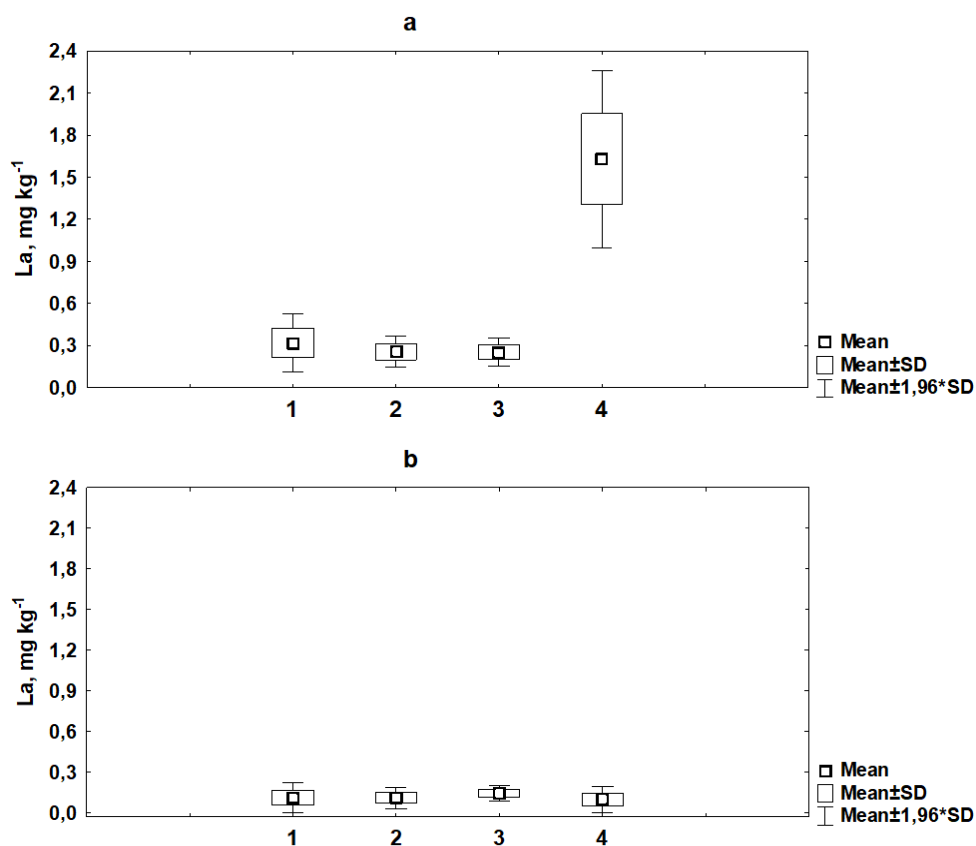


Figure 1 Mean concentration $\pm$ SD of La in roots (a) and leaves (b) of wheat seedlings grown in distilled water (1), spring water (2), nutrient solution of Hoagland (3), and in soil (4).

3.2 Effect of Soil Characteristics on Uptake of La by Different Plant Species

The soil textures in the greenhouse and field experiments were different. In the greenhouse test, the soil was a loamy sand, and in the field trial, it was a clay loam. The pH of the rhizosphere soil in the experiments slightly differed. Figure 2 shows the distribution of essential nutrients - Fe, Ca, K, and Na – in the soils.

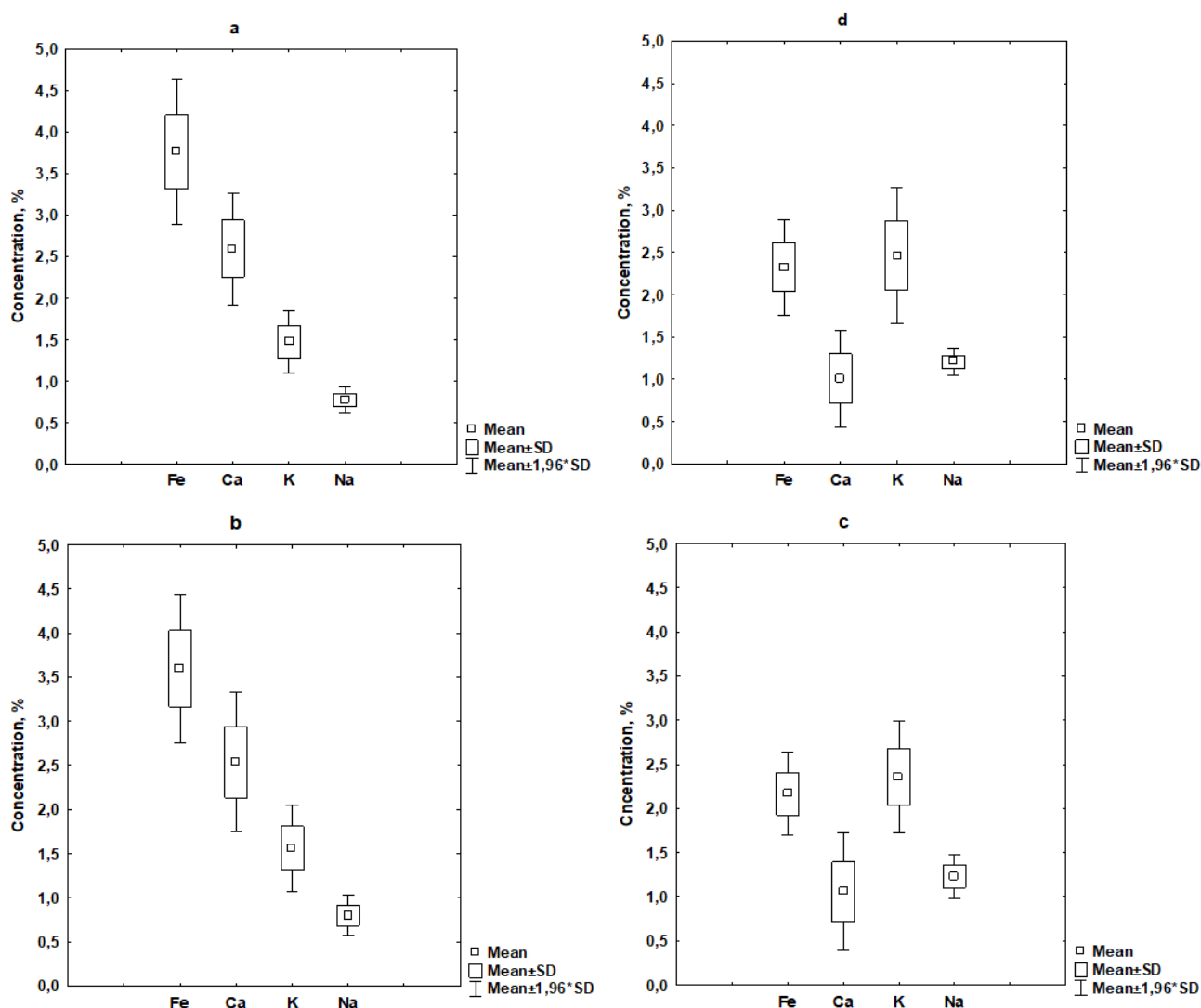


Figure 2 Mean concentrations of Fe, Ca, K, and Na in the rhizosphere soil of wheat (a), rye (b), couch grass (c), and plantain (d).

There were no differences between concentrations of the elements in the soil taken from roots of wheat and rye seedlings (greenhouse experiment), or between concentrations of Fe, Ca, K and Na in the soil taken from roots of couch grass and plantain (field trial). In the soil used for the greenhouse pot experiment, the concentrations of the elements were distributed as follows: Fe > Ca > K > Na. In the field trial, the concentrations of Fe and K in the rhizosphere soil of couch grass and plantain were similar and exceeded the concentrations of Ca and Na. In soil taken from roots of couch grass and plantain, the concentrations of Fe and Ca were lower, and K and Na were higher than the concentrations of the elements in soil taken from the roots of wheat and rye seedlings (the differences were statistically significant, $P < 0.05$). It may be assumed that the differences in the concentrations of these macro-elements are due to the texture of the soils (loamy sand and clay loam). It was shown that soil C and N concentrations are strongly related to soil texture [30].

As shown in Table 2 and Table 3, the concentration of La in a clay loam soil (field trial) was higher than in a loamy sand soil (greenhouse pot experiment). The concentration of La was usually much higher in the rhizosphere soil than in the plants. The most pronounced differences between the concentration of La in the soil taken from roots and its concentration in roots were observed for

crops: 66 times higher for wheat and 54 times higher for rye. In weeds, these differences were lower: 21 times for plantain and 15 times for couch grass. The concentration of La was also higher in roots than in leaves. Except plantain, the differences between La in roots and leaves were statistically significant ($P < 0.05$).

Table 2 Mean concentration (mg kg^{-1}) $\pm$ SD of La in the rhizosphere soil, roots and leaves of couch grass and plantain. ^a – differences between concentrations of La in roots and leaves are statistically significant ($P < 0.05$). ^b - differences between concentrations of La in leaves of couch grass and plantain are statistically significant ($P < 0.05$).

Couch grass	Plantain
Rhizosphere soil	
30.0 ± 6.0	33.3 ± 2.1
Roots	
2.03 ± 0.73^a	1.58 ± 0.26
Leaves	
0.561 ± 0.147^b	1.09 ± 0.49

Table 3 Mean concentration (mg kg^{-1}) $\pm$ SD of La in the rhizosphere soil, roots, and leaves of wheat and rye seedlings. ^a – differences between concentrations of La in roots and leaves are statistically significant ($P < 0.05$).

Wheat	Rye
Rhizosphere soil	
24.1 ± 3.1	23.8 ± 3.5
Roots	
0.365 ± 0.135^a	0.443 ± 0.260^a
Leaves	
0.076 ± 0.030	0.103 ± 0.073

It should also be noted that the concentration of La in leaves of plantain was statistically significantly ($P < 0.05$) higher than in leaves of couch grass. Both plants were grown under the same conditions, and the La concentrations in the rhizosphere soil of plantain and couch grass were nearly identical. The plants belong to different clades. Couch grass is a monocot, and plantain is a dicot. It can be assumed that plants may have their own genetically determined way of distributing elements, including La, between different parts of the plant. In particular, the study of the effect of elevated La concentration in soil on maize *Zea mays* L. and bean *Phaseolus vulgaris* L. showed that the La concentration in maize roots was lower than in bean roots [31].

3.3 Relationships between La and Other Elements in the Rhizosphere Soil and Plants

Correlation analysis showed that there was no statistically significant correlation between concentrations of La in the rhizosphere soil and roots of wheat and rye seedlings (greenhouse experiment), as well as between La in the soil taken from roots of couch grass and plantain and in

roots of the plants (field trial). In both experiments, there was also no statistically significant correlation between La in roots and leaves of all plants.

Lanthanum, like other metals, can accumulate in plants if the soil where the plants grow is enriched with this trace element [16, 32, 33]. This is probably a result of the passive transport of La. However, plants are usually “not interested” in accumulating La, and its concentration in different plant species growing in clean soil is quite low, especially in the green plant parts [32, 34]. In most cases, La uptake by roots significantly exceeds its transport from underground to the upper parts of plants [35]. Roots act as a kind of barrier, preventing the transfer of many elements to leaves. Therefore, the absence of a positive correlation between La in the roots and leaves of plants growing in La-uncontaminated soil is expected.

In addition, the correlation between La and Ca, other REEs, Sc, Fe and Co in the rhizosphere soil, roots and leaves of the plants grown in greenhouse and field experiments was calculated (Table 4). Considering the publications on the possible replacement of Ca by REEs, the relationship between La and Ca was of particular interest. However, among all samples, a statistically significant ($P < 0.05$) positive correlation between La and Ca was found only in roots of rye seedlings (greenhouse pot experiment). In all other cases, the correlation between La and Ca was statistically insignificant, and usually the correlation coefficients did not exceed 0.32. This suggests that it remains unknown whether La can be a physiological substitute for Ca.

Table 4 Pearson correlation coefficients between La and other elements in the rhizosphere soil and plants (statistically significant correlation is shown in bold).

*Concentrations of the elements in the plant samples were below the limit of determination.

Pot experiment						
Element	Wheat			Rye		
	Soil	Roots	Leaves	Soil	Roots	Leaves
Ca	0.13	0.35	0.32	0.17	0.64	0.20
Sc	0.24	0.70	0.59	0.68	0.80	0.14
Fe	0.35	0.70	0.55	0.54	0.89	-0.06
Co	0.37	0.24	0.59	0.51	0.72	0.19
Ce	0.59	0.23	0.09	0.62	0.49	-0.11
Sm	0.61	0.88	0.49	0.70	0.83	0.31
Eu	0.59	0.77	0.51	-0.30	0.02	0.14
Tb	0.59	*	*	0.50	*	*
Yb	0.20	*	*	0.13	*	*
Lu	0.29	*	*	0.05	*	*
Field experiment						
Element	Couch grass			Plantain		
	Soil	Roots	Leaves	Soil	Roots	Leaves
Ca	-0.15	0.25	0.27	-0.33	0.09	0.31
Sc	0.51	0.51	0.52	0.98	0.83	0.68
Fe	0.51	0.40	0.48	0.95	0.48	0.72
Co	-0.19	0.66	0.40	0.88	0.52	0.28

Ce	0.94	0.00	0.46	0.93	0.46	0.47
Sm	0.97	0.61	0.41	0.99	0.76	0.57
Eu	0.08	-0.36	0.47	0.47	-0.18	0.74
Tb	-0.42	*	*	0.36	*	*
Yb	0.48	0.28	0.29	0.40	0.32	0.40
Lu	0.54	0.04	0.28	0.85	0.19	0.47

As shown in Table 4, a significant ($P < 0.05$) positive correlation was observed between La and Sm in most samples. The exceptions were leaves of rye and couch grass. In many cases, La was also correlated with Sc, Fe, and Co. These last three elements are often present in the environment in the trivalent state [36-38]. Therefore, they can have similar properties to REEs. It has been noted that the uptake of REEs by plant roots is associated with the uptake of Fe [39]. An interesting situation is observed with Sc. On the one hand, it was reported that the chemical behavior of Sc can differ from that of other REEs [2]. This author also reported a low correlation of Sc with REEs [40]. But on the other hand, it is known that Sc, Y and La are considered as a triad, since they belong to the 3rd group of the Periodic Table [41]. Thus, it is expected that these elements may behave similarly. It is also interesting that the correlation between La and Eu in roots and leaves of wheat seedlings and in the rhizosphere soil of the plants was statistically significant and positive, while in the rhizosphere soil, roots and leaves of rye that grew under the same conditions and in the same soil the correlation between La and Eu was statistically insignificant. This can be a result of the release of different root exudates into the surrounding soil by these two plant species (wheat is a monocot and rye is a dicot).

3.4 Cluster Analysis

Cluster analysis was employed to examine the behavior of La and several other elements (Ca, Sc, Fe, Co, Ce, Sm, Eu, Yb, Lu) in soil and plants. Figure 3 shows an example of cluster analysis of the rhizosphere soil and roots of couch grass and plantain. Based on the data, the following conclusions can be drawn:

- in the rhizosphere soil of both plant species, light REEs La, Ce, and Sm formed a separate cluster;
- similar close relationships between La, Ce, and Sm were found in plantain roots, but in couch grass roots, Ce moved to the second cluster group;
- the two heaviest REEs, Yb and Lu, were always closely associated;
- in all samples (except plantain roots), Sc, Fe, and Co formed their group, separated from other elements (this could be expected, since these elements are chemically similar);
- except for Eu, Ca was separated from other elements.

Results of cluster analysis were in good agreement with the data of correlation analysis.

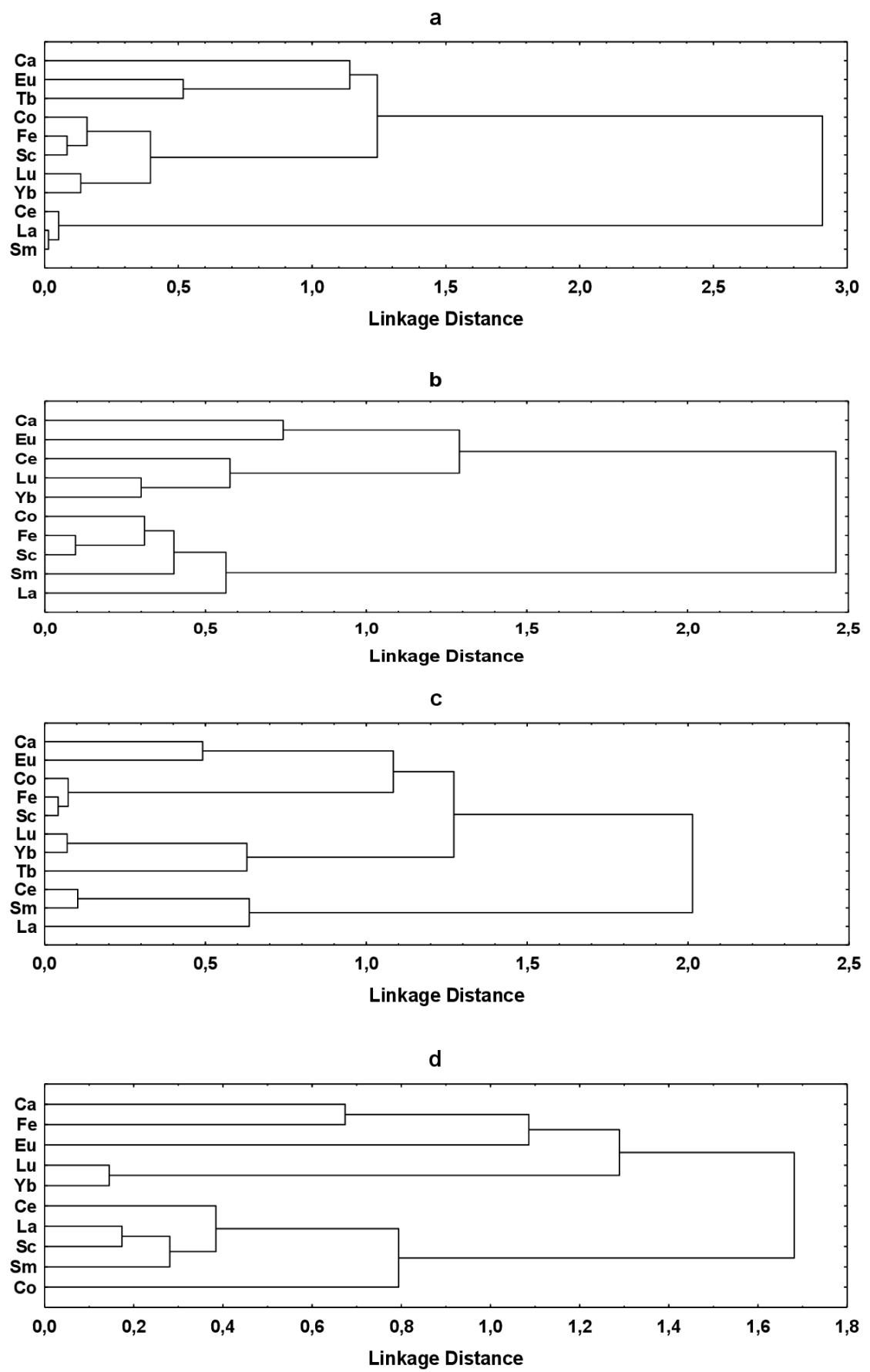


Figure 3 Cluster analysis (Ward’s method) of the rhizosphere soil (a) and roots (b) of couch grass, rhizosphere soil (c) and roots (d) of plantain.

3.5 Europium Anomalies

In plants and soils, normalized REE patterns may help better understand biogeochemical processes in the rhizosphere [42]. In this respect, among other REEs, Eu is one of the most interesting trace elements because it can change its oxidation state when interacting with organic and inorganic phases of soil. As a consequence, the mobility of Eu in soil and its uptake by plants may change.

A standard method for studying the behavior of REEs in different objects involves the use of normalized models, where the concentration of each REE in the sample is divided by its corresponding concentration in the chondrite [43]. Figure 4 shows chondrite-normalized patterns of some REEs in the rhizosphere soil, roots, and leaves of couch grass and plantain. Due to the absence of significant differences in the concentrations of REEs in the soil taken from the roots of couch grass and plantain, both curves were similar, and no Eu anomalies were detected (Figure 4a). An apparent positive Eu anomaly was found in roots (Figure 4b). It has been suggested that the positive anomaly of Eu in plants may result from the reduction of Eu^{3+} to Eu^{2+} in the rhizosphere and thus increasing its mobility in the soil-plant system and leading to preferential uptake of Eu^{2+} [43, 44]. A slight negative Eu anomaly was observed in plantain leaves, while no Eu anomaly was found in leaves of couch grass (Figure 4c). A negative Eu anomaly in plants may be due to depletion of Eu relative to other REEs in plant tissues [39]. Perhaps these results indicate, first of all, a different situation at the rhizosphere-soil-plant root boundary, and also that the situation may be different for other plant species, even if they grow under the same conditions.

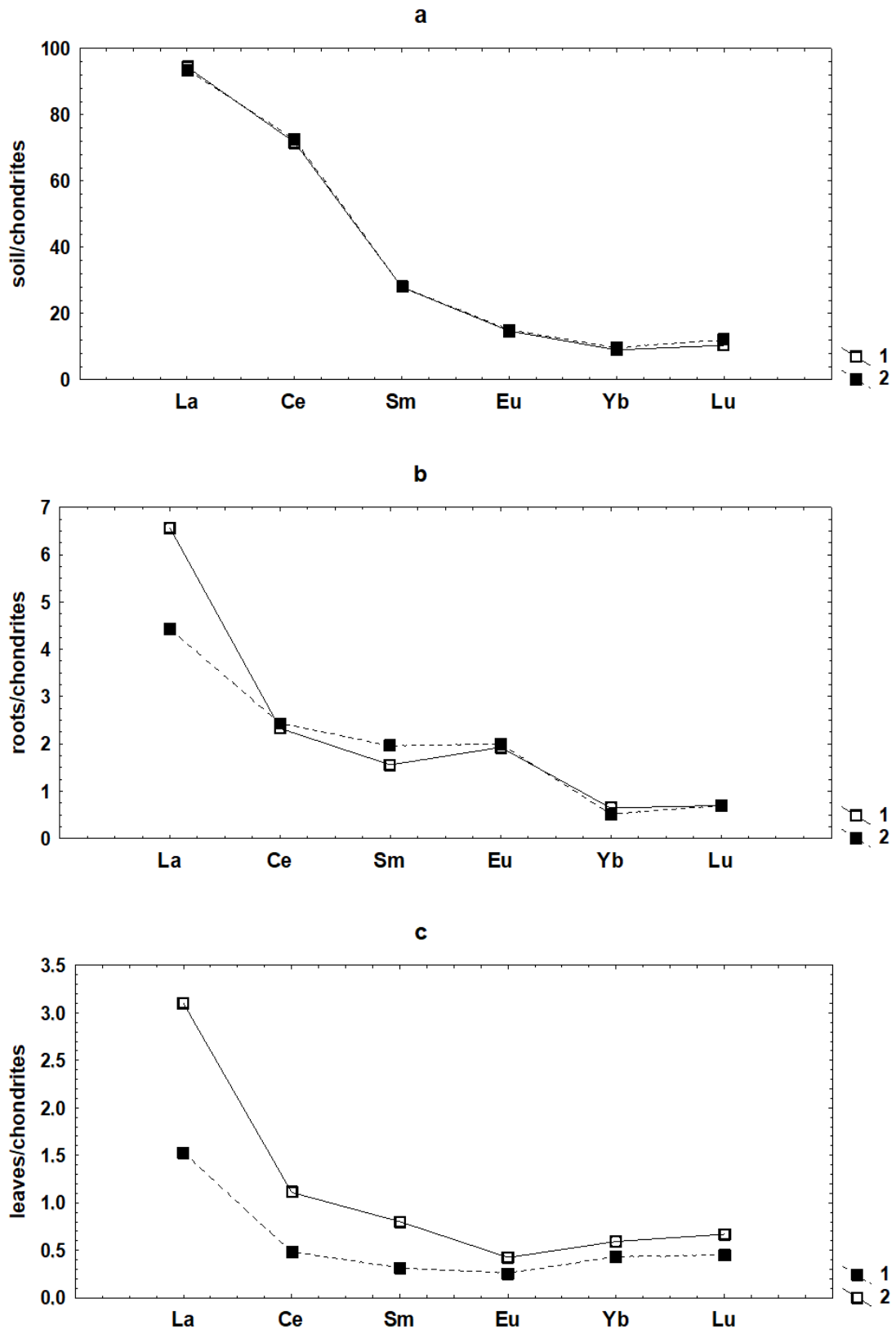


Figure 4 Chondrite normalized concentrations of REEs in the rhizosphere soil (a), roots (b) and leaves (c) of couch grass (1) and plantain (2).

4. Conclusions

Our experimental results confirmed that among the main factors influencing the uptake of La by plants are cultivated species and soil characteristics. The concentration of La even in La-uncontaminated soil is usually higher than its concentration in plants, especially in upper plant parts. However, plants growing in such uncontaminated soil do not accumulate this trace element. Different plant species, such as monocots and dicots, growing under the same conditions, can absorb different amounts of La. Another point to emphasize is that the correlation between some REEs in roots, leaves, and rhizosphere soil of monocots and dicots can also be quite different. In crops (wheat and rye), La uptake by roots and its transfer from roots to leaves was lower than in weeds (couch grass and plantain). It can be assumed that for plants growing in uncontaminated soils, the amount of La (as well as other macro- and trace elements) necessary for the normal functioning of plants is genetically determined for each plant species and depends to a lesser extent on external factors. In most cases, the correlation between La and Ca in the rhizosphere soil and plants is statistically insignificant. Therefore, there is some doubt whether La can serve as a physiological substitute for Ca. On the other hand, in many cases, La is highly correlated with Ce, Sm, Sc, Fe, and Co. Therefore, it can be assumed that the biogeochemical behavior of La is more similar to these trace elements than to Ca.

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Author Contributions

The author did all the research work for this study.

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Competing Interests

The author has declared that no competing interests exist.

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