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Dataset of rare earth elements distribution in soils and vegetables

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ABSTRACT

This paper presents a comprehensive data set on rare earth elements (REE) and their distribution in soils and vegetables in an area where the soil background is enriched with these elements. The study examined REE distribution in six vegetables commonly grown in local residents' gardens, including fennel, garlic, lettuce, parsley, onion, and raddichio, and each plant was analyzed for REE composition in its root, stem, and leaf or bulb. In addition to total REE concentrations in soils and plants, the data set includes transfer factors and fractionation indices for REE in soils, as well as normalized curves. This dataset may be useful in the field of soil geochemistry and research on the uptake of lanthanides into plants and the food chain, as well as their potential effects on human health.

1. Introduction

The increasing use of rare earth elements in many new technological applications related to engineering, medicine, agriculture, and electronics (Kegl et al., 2020; Migaszewski and Gałuszka, 2015; Yan et al., 2022), is leading to their increasing release into the environment (Atinkpahoun et al., 2020; Arienzo et al., 2022). It is, therefore, crucial to recognize the potential risks posed by REEs to both human health and the environment (Pagano et al., 2019; Zhao et al., 2019; Yan et al., 2019; Yin et al., 2021).

A critical first step towards mitigating these risks is monitoring the content of REEs in soil and various plant species, especially those used for human consumption. However, there is currently limited to no available data on rare earth elements in vegetables and their uptake from soil. There is even less data if distribution between different plant parts is taken into account (roots, bulbs, stems or leaves).

Therefore, this dataset aims to provide an insight into the uptake and distribution of rare earth elements in vegetables grown in private gardens and in a substrate naturally enriched with these elements (Salminen et al., 2005). Additionally, the dataset provides information on the distribution of REE among different plant parts, which is crucial to understand as they may be used differently in food preparation. The presented data set can also be used to estimate the intake of rare earth elements via vegetables.

2. Materials and methods

2.1. Sampling and sample preparation

Sampling was performed during November 2019 in Istrian penninsula (Croatia, Fig. 1). Soil and vegetable samples were collected in the Raša region (at two locations in the municipality of Raša (R1, R2) and five locations in the village of Krapan (K1-K5)) and Opatija region (O1-O5). A total of 20 plants and 12 soil samples were collected and analyzed for rare earth element content. Plant samples include six vegetable species, including fennel (2), garlic (2) lettuce (7), parsley (3), onion (3), and raddichio (3). Plant parts were separated for further analysis, including roots (20), stems (3), leaves (20) and bulbs (5), resulting in a total of 48 samples. All sampling sites except one in Krapan (site K5) were private gardens where residents grew vegetables for their own consumption. Map location of sampling area is presented in Fig. 1.

The soil samples were air-dried, passed through a 2 mm sieve to remove gravel, and stored for later analysis. The plant samples were washed with tap and Milli-Q water, separated into different parts (roots, stems, leaves, and/or bulbs), air-dried, homogenized using an agate mill, and stored for further analysis.

To perform total element analysis, plant sub-samples of about 0.07 g were digested in a one-step procedure in a microwave oven (Multiwave 3000, Anton Paar, Graz, Austria) using a mixture of 6 mL nitric acid (HNO_3) and 0.1 mL hydrofluoric acid (HF). Soil sub-samples of about

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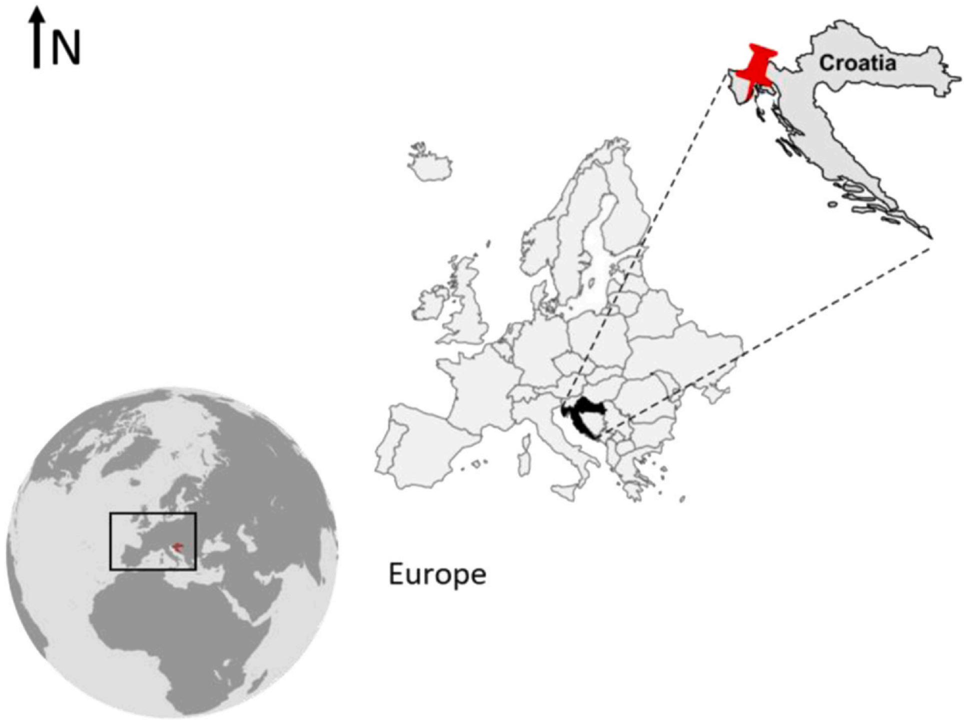

Fig. 1. Map location of sampling area.

0.05 g were digested in a two-step procedure in the microwave oven, using a mixture of 4 mL nitric acid (HNO_3, 65%, pro analysi, Kemika, Zagreb, Croatia), 1 mL hydrochloric acid (HCl, 36.5%, pro analysi, Kemika, Zagreb, Croatia), and 1 mL hydrofluoric acid (HF, 48%, *traceSELECT*, Fluka), followed by the addition of 6 mL of boric acid (H_3BO_3, Fluka, Steinheim, Switzerland).

Before analysis, the soil digests were diluted by a factor of 10 and treated with 2% (v/v) HNO_3 (65%, supra pur, Fluka, Steinheim, Switzerland) and 1 μg L^{-1} of In as an internal standard. On the other hand, the plant digests were treated only with 2% (v/v) HNO_3 (65%, supra pur, Fluka, Steinheim, Switzerland) without any dilution and 1 μg L^{-1} of In as an internal standard was added.

2.2. Analytical procedure

Multielemental analysis of prepared samples was performed by an Agilent 8900 triple quadrupole ICP-MS (ICP-QQQ). External calibration was used for the quantification. Standards for multielement analysis were prepared by appropriate dilution of a multielement reference standard (Analytika, Prague, Czech Republic) containing Ce, La, Nd and Pr (100 ± 0.2 mg L^{-1}), and Dy, Er, Eu, Gd, Ho, Lu, Sm, Tb, Tm, and Yb (20 ± 0.4 mg L^{-1}).

All samples were analysed for total concentration of 14 elements (Ce, Dy, Er, Eu, Gd, Ho, La, Lu, Nd, Pr, Sm, Tb, Tm, and Yb). Limits of detection (LOD) were calculated as three times the standard deviation of three consecutive measurements of REE concentrations in the procedural blanks for the soil and plant digestion protocol. The LODs obtained ranged from 2 to 7 μg kg^{-1} for the soil and from 0.8 to 2.1 μg kg^{-1} for the plant protocol. Accordingly, limits of quantification (LOQ), calculated as ten times the standard deviation of three consecutive measurements of REE concentrations in the procedural blanks for the soil and plant digestion protocol ranged from 6 to 21 μg kg^{-1} for the soil protocol and from 2.5 and 7 μg kg^{-1} for the plant protocol.

Quality control of analytical procedure was performed by simultaneous analysis of the blank and certified reference material for soil (NCS DC 77302, China National Analysis Center for Iron and Steel, Beijing, China) and citrus leaves (NCS Certified Reference Material, ZC73018). For all elements, good agreement was obtained between the analysed and certified concentrations within their analytical uncertainties. The recoveries obtained ranged from 90% to 102% for the soil (NCS DC 77302) reference material and from 88% to 117% for the citrus leaves (ZC73018) reference material.

2.3. Data analysis

For data presentation purposes, the rare earth group (Ce, Dy, Er, Eu, Gd, Ho, La, Lu, Nd, Pr, Sc, Sm, Tb, Tm, and Yb) is denoted REE. The REE group can be further subdivided into light (LREE) and heavy (HREE), i.e., elements from La to Gd and from Tb to Lu, respectively.

The REE concentrations of all soil samples were normalized to North American Shale Composite (Gromet et al., 1984), while plant samples were normalized to average REE content of studied soils.

The decoupling of anomalous behavior of Ce and Eu from the other REE in the NASC-normalized distribution patterns (expressed as Ce/Ce* and Eu/Eu*, respectively) was calculated using the following formulae (Bau et al., 2014; Dai et al., 2016):

$$Eu/Eu^* = Eu_N/(Sm_N \cdot Gd_N)^{0.5} \quad (1)$$

$$Ce/Ce^* = Ce_N/(La_N \cdot Pr_N)^{0.5} \quad (2)$$

Geoaccumulation indices (Müller, 1981) were calculated as follows:

$$I_{geo} = \log_2 \frac{C_n}{1.5 \cdot B_n} \quad (3)$$

Table 1
Descriptive statistics (min – minimum, max – maximum, avg – average, std – standard deviation, all expressed in mg kg^{-1}, and RSD – relative standard deviation, expressed in %) for REE distribution in soil samples.

	min	max	avg	std	RSD
La	9.0	52.3	35.6	12.3	34.5
Ce	17.5	111	71.1	26.0	36.6
Pr	2.16	12.8	8.70	3.05	35.1
Nd	8.09	48.3	32.9	11.6	35.2
Sm	1.55	9.56	6.33	2.26	35.8
Eu	0.37	1.87	1.27	0.43	33.9
Gd	1.41	7.41	5.04	1.67	33.1
Tb	0.20	1.13	0.76	0.26	34.1
Dy	1.27	6.17	4.31	1.40	32.5
Ho	0.25	1.20	0.84	0.27	31.8
Er	0.72	3.38	2.36	0.76	32.1
Tm	0.10	0.49	0.33	0.11	33.8
Yb	0.66	3.08	2.15	0.71	33.1
Lu	0.09	0.46	0.32	0.10	32.6

where C_n is the measured concentration of an element *n* in a soil sample and B_n is the background geochemical concentration of the same element in the soil, given by Salminen et al. (2005).

The accumulation of rare earth elements from soil into vegetables was evaluated using the transfer factor (TF, (Gan et al., 2017)) calculated as follows:

$$TF = (Cn)plant/(Cn)soil \quad (4)$$

where C_n are the concentrations of a studied element *n* in plant (or plant part) and soil at the same site.

3. Results and discussion

The total concentrations of rare earth elements (REE), in the soils and associated vegetables are presented in Tables S1 and S2, respectively, while the calculated transfer factors (TF) are presented in Table S3. In addition to total REE concentrations, data on site description are provided in Table S1. For vegetables, concentrations and transfer factors are given for each part of the plant, i.e. root, stem, leaf or bulb.

Table 3
REE fractionation indices of studied soil samples.

Sample	Eu/Eu*	Ce/Ce*	La_N/Lu_N	La_N/Sm_N	$LREE_N/HREE_N$
K1	0.97	0.94	1.54	1.03	1.28
K2	1.01	0.95	1.51	1.02	1.33
K3	0.92	0.92	1.69	1.03	1.33
K4	1.09	0.92	1.47	1.04	1.21
K5	1.01	0.92	1.56	1.06	1.32
R1	1.04	0.98	1.43	1.03	1.29
R2	1.00	0.95	1.55	1.03	1.28
O1	1.01	0.89	1.77	1.02	1.42
O2	0.94	0.84	1.92	1.02	1.43
O3	0.99	0.98	1.72	0.98	1.41
O4	1.04	0.93	1.70	1.00	1.42
O5	0.99	0.99	1.67	0.98	1.41
min	**0.92**	**0.84**	**1.43**	**0.98**	**1.21**
max	**1.09**	**0.98**	**1.92**	**1.06**	**1.43**
avg	**1.00**	**0.93**	**1.62**	**1.02**	**1.34**
std	**0.05**	**0.04**	**0.15**	**0.02**	**0.07**
RSD	**4.9**	**4.3**	**9.1**	**2.0**	**5.6**

Eu/Eu*- europium anomaly (Eu/Eu* = $Eu_N/(Sm_N \times Gd_N)^{0.5}$)
Ce/Ce*- cerium anomaly (Ce/Ce* = $Ce_N/(La_N \times Pr_N)^{0.5}$)
La_N/Lu_N - ratio of normalized concentrations of La and Lu
La_N/Sm_N - ratio of normalized concentrations of La and Sm
$LREE_N/HREE_N$ - ratio of sum of normalized concentrations of light and heavy rare earth elements.

Quality control data including REE concentrations in procedural blanks for soil and plant digestion protocol, and associated limits of detection, as well as the REE distribution in analysed certified reference materials (Soil NCS DC 77302 and Citrus leave ZC73018) are presented in Table S4. Tables S1–S4 can be found in the repository (https://data.mendeley.com/datasets/r4x2vn5w42/3).

The descriptive statistics for REE distribution in the soil samples are shown in Table 1, and the total and sum of light and heavy elements REE are shown in Table 2. Concentrations of the REEs in analysed soil samples ranged over five orders of magnitude, from 0.09 mg kg^{-1} to 111 mg kg^{-1} (Table 1). Of all the measured REE, Ce was found at highest concentrations in all samples, whereas concentrations of Tm and Lu were lowest. Analysed soils displayed variations in terms of REE concentrations as well as distribution patterns, with total REE (ΣREE)

Table 2
REE distribution and pH in studied soil samples.

Sample	ΣREE	LREE	HREE	LREE/HREE	% LREE	% Ce	pH
K1	102	95	7.0	13.5	80	35	8.87
K2	87	81	5.8	14.1	80	36	8.82
K3	213	199	14.0	14.2	81	36	7.51
K4	43	40	3.3	12.2	78	34	8.36
K5	197	184	12.9	14.2	80	35	8.23
R1	108	101	7.3	13.8	80	36	9.04
R2	167	156	11.5	13.5	80	36	8.89
O1	147	137	9.3	14.7	82	35	8.86
O2	161	150	10.2	14.8	81	34	8.51
O3	236	221	14.7	15.0	83	37	8.27
O4	224	210	13.9	15.1	82	36	8.48
O5	259	243	15.9	15.3	83	38	6.89
min	**43.3**	**40.0**	**3.29**	**12.2**	**77.7**	**33.8**	**6.89**
max	**236**	**221**	**14.7**	**15.1**	**83.0**	**37.4**	**9.04**
avg	**153**	**143**	**9.99**	**14.1**	**80.8**	**35.5**	**8.39**
std	**62.2**	**58.4**	**3.79**	**0.85**	**1.45**	**1.05**	**0.63**
RSD	**40.6**	**40.8**	**38.0**	**6.0**	**1.8**	**3.0**	**7.5**

ΣREE - sum of concentrations of all REE (in mg kg^{-1})
LREE - sum of light REE concentrations, from La to Gd (in mg kg^{-1})
HREE - sum of heavy REE concentrations, from Tb to Lu (in mg kg^{-1})
LREE/HREE - ratio of light REEs to heavy REEs
% LREE - share of light REEs in the total REE concentration
% Ce - share of Ce in the total REE concentration (element with the highest concentrations).

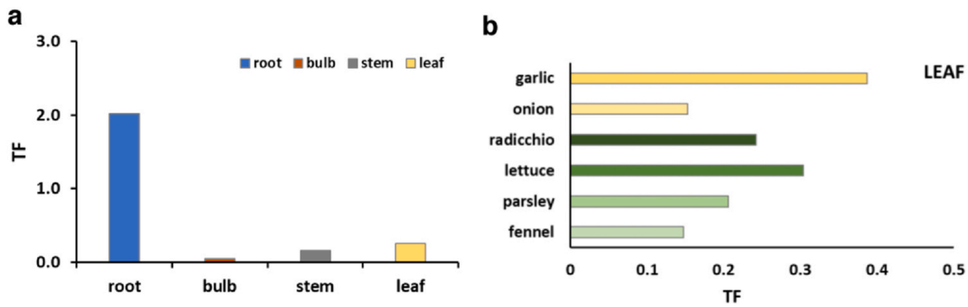

Fig. 2. a) Average TF for ΣREE in different plant parts; b) average TF for ΣREE in leaves of different vegetables.

ranging from 43.3 mg kg^{-1} to 236 mg kg^{-1} (Table 2). Moreover, the range of ΣREE values in the soil samples in the Raša region was wider (43 mg kg^{-1} to 213 mg kg^{-1}) compared to the range of values obtained for the Opatija region soils (147 mg kg^{-1} to 259 mg kg^{-1}).

In general, the soil samples displayed a predominance of light rare earth elements (LREE, La-Gd) over heavy rare earths (HREE, Tb-Lu) with the average proportion of LREE being 81% and LREE/HREE ranging from 12.2 to 15.1 (Table 2).

The pH of the soils ranged from 6.89 to 9.04, being predominantly alkaline with an average value of 8.39.

The calculated REE fractionation indices, including Eu and Ce anomaly, La_N/Lu_N, La_N/Sm_N and $LREE_N/HREE_N$, for studied soil samples are shown in Table 3. Soils exhibited slightly negative to slightly positive europium anomaly (Eu/Eu* =0.92–1.09), while cerium anomaly values displayed slightly negative values (Ce/Ce* = 0.84–0.98). Both anomalies, europium and cerium, were found in a rather narrow range and are comparable with literature values reported for soils from this region (Fiket et al., 2016). In addition, soils are characterized by an overall predominance of LREE, i.e. $La_N/Lu_N > 1$, $LREE_N/HREE_N > 1$ and $La_N/Sm_N \sim 1$ (Table 3), resulting in a convex shape of the normalized curve (Fig. 2a). The predominance of LREE over HREE is typical of soils formed on sedimentary and carbonate rocks (Fiket et al., 2016, 2018), while the observed ratios are consistent with literature data reported for regional soils (Fiket et al., 2016).

Geoaccumulation indices (I_{geo}) were calculated to evaluate soil contamination with REE and are shown in Table 4. As can be seen from the table, all I_{geo} values obtained were negative and ranged from −3.2 to −0.4, which is classified as practically uncontaminated soil (Müller, 1981) and suggests that the concentrations of REE in the studied soils are consistent with natural background levels.

Concentrations of the REEs in analysed vegetables ranged from below detection limits to 25 mg kg^{-1}, with the overall highest concentrations measured in roots (Table 5). Similar as in soils, of all the measured REE, Ce was found at highest concentrations while the concentrations of Tm and Lu were lowest.

While Table S3 lists the calculated transfer factors for all rare earth elements in all vegetables studied, Fig. 2a shows that, on average, the transfer factors for ΣREE are highest in roots, followed by leaves, stems and bulbs. Within the average TF values for ΣREE in leaves, garlic and lettuce have the highest values, followed by radicchio, parsley, onion and fennel (Fig. 2b).

The soil REE normalized patterns of studied vegetables are shown in Fig. 3b. Fig. 3 clearly shows that the root system does not fractionate significantly the REE taken from the soil. It is also interesting to note that both the bulb and the stem are characterized by a positive Eu anomaly and a slight fractionation of HREE. The positive Eu anomaly could be due to an analytical artifact caused by insufficient correction of BaO and BaOH interferences (Pourret et al., 2022), while the slight fractionation of HREE could be related to their higher mobility in soil

Table 4
Geoaccumulation indices (I_{geo}) for studied soils.

	I_{geo}														
	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
K1	-1.9	-1.9	-1.8	-1.8	-1.7	-1.8	-1.8	-2.0	-1.9	-1.8	-1.9	-1.8	-2.0	-2.0	-2.0
K2	-2.2	-2.1	-2.0	-2.0	-2.0	-2.0	-2.0	-2.3	-2.3	-2.1	-2.2	-2.1	-2.3	-2.3	-2.1
K3	-1.0	-0.8	-0.8	-0.7	-0.7	-0.7	-0.8	-1.0	-1.0	-0.8	-0.9	-0.9	-0.9	-1.0	-1.0
K4	-2.9	-3.1	-3.1	-3.0	-3.0	-3.1	-2.8	-3.1	-3.2	-2.9	-3.0	-3.0	-3.0	-3.0	-3.1
K5	-1.0	-0.9	-0.9	-0.8	-0.8	-0.9	-0.8	-1.0	-1.0	-1.0	-1.0	-1.0	-1.0	-1.1	-1.0
R1	-1.8	-1.8	-1.7	-1.7	-1.7	-1.8	-1.6	-1.9	-2.0	-1.8	-1.8	-1.8	-1.8	-2.0	-1.8
R2	-1.2	-1.2	-1.1	-1.1	-1.1	-1.1	-1.1	-1.3	-1.2	-1.1	-1.2	-1.2	-1.4	-1.2	-1.3
O1	-1.6	-1.3	-1.4	-1.2	-1.2	-1.2	-1.2	-1.4	-1.4	-1.4	-1.4	-1.5	-1.6	-1.6	-1.6
O2	-1.4	-1.2	-1.3	-1.1	-1.0	-1.1	-1.1	-1.3	-1.3	-1.3	-1.3	-1.3	-1.5	-1.5	-1.6
O3	-1.0	-0.7	-0.6	-0.6	-0.5	-0.6	-0.6	-0.9	-0.8	-0.8	-0.8	-0.8	-0.9	-0.9	-1.0
O4	-1.0	-0.7	-0.7	-0.6	-0.6	-0.6	-0.6	-0.9	-0.9	-0.8	-0.9	-1.0	-0.9	-1.0	-1.0
O5	-0.9	-0.6	-0.4	-0.5	-0.4	-0.4	-0.4	-0.7	-0.7	-0.7	-0.7	-0.7	-0.7	-0.8	-0.8

Classes of I_{geo} (Müller, 1981):
practically uncontaminated ($I_{geo} \leq 0$),
uncontaminated to moderately contaminated ($0 < I_{geo} < 1$),
moderately contaminated ($1 < I_{geo} < 2$),
moderately to heavily contaminated ($2 < I_{geo} < 3$),
heavily contaminated ($3 < I_{geo} < 4$),
heavily to extremely contaminated ($4 < I_{geo} < 5$),
extremely contaminated ($I_{geo} > 5$).

Table 5
REE distribution (expressed in mg kg^{-1}) in different plant parts of the studied vegetable expressed as minimum (min), maximum (max) and average (avg) values in root, bulb, stem and leaf samples.

	root			bulb			stem			leaf		
	min	max	avg	min	max	avg	min	max	avg	min	max	avg
La	0.552	11.8	4.03	0.018	0.201	0.072	0.121	0.573	0.374	0.231	1.141	0.510
Ce	1.12	25.0	7.94	0.039	0.409	0.146	0.240	1.087	0.700	0.458	2.31	0.999
Pr	0.137	2.96	0.972	0.004	0.052	0.018	0.028	0.140	0.091	0.056	0.279	0.122
Nd	0.549	10.9	3.64	0.018	0.199	0.073	0.105	0.552	0.368	0.214	1.070	0.469
Sm	0.105	2.32	0.753	0.005	0.041	0.016	0.017	0.106	0.068	0.033	0.218	0.087
Eu	0.020	0.447	0.144	0.001	0.009	0.004	0.022	0.027	0.024	0.008	0.044	0.019
Gd	0.081	1.68	0.559	0.004	0.029	0.013	0.004	0.069	0.042	0.016	0.151	0.057
Tb	0.012	0.245	0.085	< 0.001	0.005	0.002	0.002	0.011	0.008	0.005	0.025	0.011
Dy	0.072	1.36	0.467	< 0.001	0.027	0.010	0.011	0.069	0.042	0.023	0.130	0.055
Ho	0.014	0.259	0.092	0.001	0.005	0.003	0.007	0.018	0.013	0.010	0.033	0.016
Er	0.038	0.741	0.253	0.002	0.014	0.008	0.017	0.050	0.034	0.024	0.081	0.041
Tm	0.005	0.101	0.035	< 0.001	0.002	0.001				0.001	0.007	0.003
Yb	0.042	0.682	0.231	0.001	0.012	0.006	0.001	0.022	0.013	0.005	0.066	0.022
Lu	0.006	0.090	0.033	< 0.001	0.003	0.001	0.004	0.007	0.005	0.004	0.013	0.007

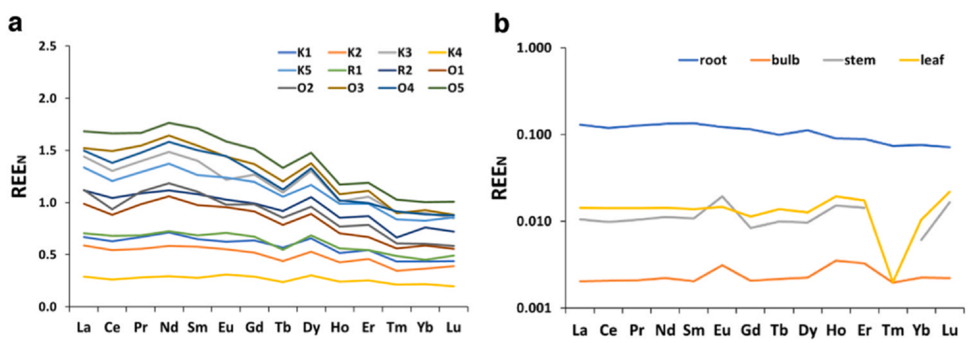

Fig. 3. a) NASC-normalized patterns of REE (REE_N) in studied soils; b) average soil-normalized patterns of REE (REE_N) in different parts of studied vegetables.

solution, i.e. lower affinity for particles (Fiket et al., 2018). The fraction of REE, reaching the leaf, is similar to that in the stem, while the bulbs show the lowest average fraction of REE from the soil.

4. Conclusion

The analysis of the studied vegetables showed that they are able to accumulate rare earth elements (REEs) from the soil. However, the distribution patterns of rare earths varied between different parts of the plants. The roots of the vegetables generally had the highest concentrations of rare earths, while the bulbs, stems, and leaves showed different fractions and anomalies for certain REEs.

These results highlight the complex nature of uptake and translocation of REE in plants, which can be influenced by a number of factors including soil composition, plant species, and specific uptake mechanisms. Understanding these factors is critical to assessing the potential risks associated with the accumulation of REE in vegetables.

Given the increasing use of rare earth elements in various industries and their potential release into the environment, monitoring the content of REE in vegetables and other foods is essential to ensure food safety and minimize potential health risks to consumers.

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Ethics statements

Informed consent was obtained from the private farm owners before collecting soil and plant samples from their place.

CRediT authorship contribution statement

Željka Fiket: Conceptualization, Methodology, Writing – original draft. **Gordana Medunić**: Writing – review & editing.

Data availability

I have shared a link to my data. Rare earth elements in soils and vegetables, Mendeley Data, doi: 10.17632/r4x2vn5w42.3.

Declaration of Competing Interest

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

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.rines.2023.100004.

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