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Geochemical signatures and human health risk evaluation of rare earth elements in soils and plants of the northeastern Qinghai-Tibet Plateau, China

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Abstract: Information on rare earth elements (REEs) in soils and plants of the Qinghai-Tibet Plateau is very limited. Therefore, in this study, we performed field sampling to explore the geochemical signatures and human health risk of REEs in soils and plants of the northeastern Qinghai-Tibet Plateau, China. A total of 127 soil samples and 127 plant samples were collected from the northeastern Qinghai-Tibet Plateau to acquire the geochemical signatures and related human health risks of REEs. The mean total concentrations of REEs in soils and plants of the study area reached 178.55 and 10.06 mg/kg, respectively. The light REEs in soils and plants accounted for 76% and 77% of the total REEs, respectively. REEs showed significantly homogenous distribution in soils but inhomogeneous distribution in plants of the study area. Characteristic parameters indicated that light REEs were enriched and fractionated significantly, while heavy REEs were moderately fractionated in soils and plants. REEs in soils and plants showed significantly negative Europium anomaly. Cerium showed slightly positive anomaly in plants and slight anomaly in soils. The normalized distribution patterns of REEs were generally similar in the analyzed soils and the corresponding plants of the study area. The average bio-concentration factor of REEs ranged from 0.0478 (Scandium) to 0.0604 (Europium), confirming a small accumulation of REEs by plants. Health risks caused by REEs in soils and plants were negligible, while risks for adults were lower than those for children. This study provides important information on REEs in soils and plants of the northeastern Qinghai-Tibet Plateau.

Keywords: rare earth elements; geochemical signatures; human health risk; carcinogenic risk; bio-concentration factor; Qinghai-Tibet Plateau

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1 Introduction

Rare earth elements (REEs) encompass the yttrium (Y), the elements of the lanthanide series (atomic number from 57 to 71), and scandium (Sc), which exhibit similar environmental

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behaviors (Ferreira et al., 2021). Promethium (Pm) does not exist in the Earth's crust since the radioactive decay of Pm is fast (Migaszewski and Gałuszka, 2015). REEs generally exist together in natural environment due to similar physical and chemical properties (Groenenberg et al., 2010; Khan et al., 2017). Many studies reported that pedogenic processes, soil characteristics, and parent material affected REEs background contents in soils (Yao et al., 2010; Wu et al., 2019). REEs are not rare from a geochemical perspective because the abundances of REEs in soils are similar to those of zinc (Zn), lead (Pb), and copper (Cu) and higher than those of mercury (Hg), Stannum (Sn), cobalt (Co), and silver (Ag) (Wang et al., 1998).

Heavy REEs (ΣHREEs; the sum Y, Sc, and from gadolinium (Gd) to lutetium (Lu)) and light REEs (ΣLREEs; the sum from lanthanum (La) to europium (Eu)) compose REEs based on the mass and atomic numbers of REEs. The concentrations of ΣHREEs are generally lower than the concentrations of ΣLREEs (Henderson, 1984; Durn et al., 2021). The ΣLREEs are more mobile and available for plant absorption than ΣHREEs (Brioschi et al., 2013). The concentrations of ΣLREEs in leachate are higher than those of ΣHREEs so that the ΣLREEs are more available for root adsorption than ΣHREEs (Aide and Aide, 2012). The bioavailability of REEs in soil is a considerable factor for the accumulation of REEs by plants (Li et al., 2013; Liu et al., 2018). The concept of bioavailability also was defined the amount of human adsorption in recent years (Li et al., 2018; Ferreira et al., 2022; Wang et al., 2022).

The global annual demands of REEs have increased significantly from 3.00×10^4 to 1.35×10^5 t during 1980–2015 (Barta, 2007; Huang et al., 2016). China is the largest producer of REEs in the world, accounting for approximately 97% of the global supply. The largest REEs ore mine is located in the northern China (Tang et al., 2020). REEs have been widely used in medical technology, industry, and agriculture (Hu et al., 2006; Pagano et al., 2015; Wang and Liang, 2016; Ou et al., 2022). Various field activities have released lots of REEs into the environment (Zhang et al., 2014; Pagano, 2016). Distribution, environmental behaviors, and removal of different contaminants, such as trace elements (Zheng et al. 2010a, b; Liang et al., 2013; Rodríguez-Barranco et al., 2014; Lu et al., 2021; Pirarath et al., 2021; Cui et al., 2022; He et al., 2022), antibiotics and corresponding resistance genes, perfluorooctane sulfonate, polycyclic aromatic hydrocarbons, and microplastics, have attracted attention widely (Liu et al., 2019; Fu et al., 2021; Qiao et al., 2021; Lu et al., 2022a, b). However, studies regarding the negative effect of REEs on natural ecosystems and humans are limited. REEs accumulate in various environmental media (including soil, atmosphere, and water body) after entering into the environment, and finally influence ecosystem via human activities or human health via REEs accumulation in the human bodies (Mihajlovic et al., 2015; Pagano, 2016; Wang et al., 2017; Wang et al., 2019; Godwyn-Paulson et al., 2022). The behavior of REEs in natural environment is strongly affected by soil redox conditions, organic matter content, pH, weathering processes, climate conditions, and parent rock (Zhang et al., 2014; Pagano, 2016; Allajbeu et al., 2016; Huang et al., 2019). There are three ways including ingestion, dermal adsorption, and inhalation for entering the human body (Meryem et al., 2016; Guo et al., 2019). Previous studies have proven that REEs accumulated in the human body can cause numerous diseases (Zhang et al., 2000; Zhao et al., 2017; Ferreira et al., 2022).

Soil is an important medium for receiving pollutants and nutrients to play an important role in ecosystems. In recent decades, the geochemical characteristics and health hazards of REEs have attracted much attention (Kan et al., 2017; Huang et al., 2019; Guo et al., 2019; Wu et al., 2019; Malhotra et al., 2020). However, information about distribution patterns, accumulation in soils and plants, and health risk evaluation of REEs is quite scarce in the study area. The main objectives of this study were to (1) investigate the concentration and distribution patterns of REEs in soils and plants of the northeastern Qinghai-Tibet Plateau; (2) calculate bioavailability and mobility characteristics of REEs; and (3) evaluate the potential human health risk of REEs.

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2 Materials and methods

2.1 Study area and soil and plant samples

The study area is situated in the northeastern Qinghai-Tibet Plateau of China, which spans 4.13×10^5 km^2 (90°37′–101°05′E, 35°01′–38°43′N). The study area has an average annual precipitation of 50–450 mm and an annual average temperature of –5.7–8.5°C. A total of 127 soil samples (soil depth: 0–20 cm) and 127 plant samples were collected from June 14 to June 29 of 2017 by following standard sampling method (State Environmental Protection Administration, 2004) (Fig. 1). All samples were placed in plastic bags, transported back to the laboratory, and stored at –80°C. Soil samples were prepared by following the study conducted by Li et al. (2018).

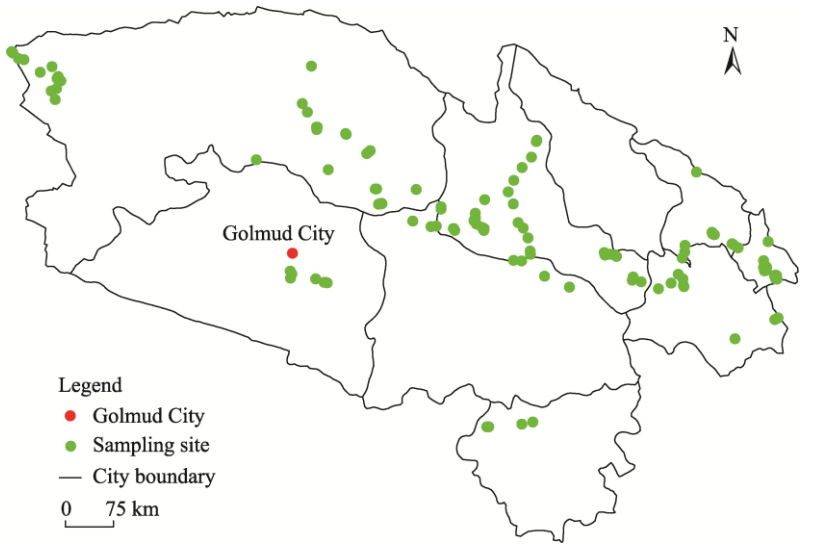

Fig. 1 Study area and soil and plant sampling sites

2.2 Analysis of rare earth elements (REEs)

The soil samples were air-dried, crushed, and then sieved using 0.074-mm sieves. The soil samples were calcined at 700°C for 2 h to eliminate the organic matter. According to related standard (State Environmental Protection Administration, 2004), 100 mg of each soil power was added along with 3 mL HNO_3 with high purity, 2 mL HCl with high purity, and 2 mL HF with high purity in a 100-ml Teflon digestion tube (Wu et al., 2019). These tubes were placed in a microwave dissolving system for 1 h and cooled. The cooled solution was dried at 180°C with 2 mL of high quality $HClO_4$ and 15 mL of high purity HNO_3.

The plant (*Potentilla anserina* L.) samples were pre-treated by following the previously-reported procedures (Shen et al., 2018). All parts of plant samples were washed thoroughly to remove soil particles, washed five times with ultra-pure water, blotted extra water with tissue paper, and dried. Then the samples were grinded to fine powder for digestion. Approximately 100 mg of dry plant powder was digested with 3 mL HCl (37%), 5 mL HNO_3 (65%), and 1 mL HF (65%) (Ayrault et al., 2001; Li et al., 2018). The other digestion procedures for plant samples were same as soils. Using testing instrument (Agilent7900, Agilent Inc, Santa Clara, USA), we measured the concentration levels of La, cerium (Ce), praseodymium (Pr), neodymium (Nd), samarium (Sm), Eu, Gd, terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm) ytterbium (Yb), Lu, Y and Sc in samples.

2.3 Distribution characteristics of REEs

North American shale composite (NASC) was used to normalize REEs concentrations to eliminate the "Oddo-Harkin effect" of REEs (Taylor and Mclennan, 1985). Normalization values

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were calculated by the ratio of the REEs concentrations to the corresponding element concentrations of NASC in this study. The parity effects of REEs were eliminated after the normalized REEs by NASC (Chen and Yang, 2010; Wu et al., 2019).

Characteristic parameters which indicated the distribution of REEs in soils and plants included fractionation between ΣLREEs and ΣHREEs ($(La/Yb)_N$), fractionation between ΣLREEs ($(La/Sm)_N$), fractionation between ΣHREEs ($(Gd/Yb)_N$), Ce anomaly (δ_{Ce}), and Eu anomaly (δ_{Eu}); we calculated these parameters based on normalized values of NASC (Taylor and Mclennan,1985; Mclennan, 1989; Chen and Yang, 2010; Wu et al., 2019; Yu et al., 2019), the calculation process is as follows:

$$(La/Sm)_N = \frac{(La_{sample}/La_{NASC})}{(Sm_{sample}/Sm_{NASC})}, \quad (1)$$

$$(Gd/Yb)_N = \frac{(Gd_{sample}/Gd_{NASC})}{(Yb_{sample}/Yb_{NASC})}, \quad (2)$$

$$(La/Yb)_N = \frac{(La_{sample}/La_{NASC})}{(Yb_{sample}/Yb_{NASC})}, \quad (3)$$

$$\delta_{Ce} = \frac{(Ce_{sample}/Ce_{NASC})}{\sqrt{(La_{sample}/La_{NASC}) \times (Pr_{sample}/Pr_{NASC})}}, \quad (4)$$

$$\delta_{Eu} = \frac{(La_{sample}/La_{NASC})}{\sqrt{(Sm_{sample}/Sm_{NASC}) / (Gd_{sample}/Gd_{NASC})}}, \quad (5)$$

where La_{sample} (mg/kg), Sm_{sample} (mg/kg), Gd_{sample} (mg/kg), Yb_{sample} (mg/kg), Ce_{sample} (mg/kg), and Pr_{sample} (mg/kg) are the contents of La, Sm, Gd, Yb, Ce, and Pr in soil or plant samples, respectively; and La_{NASC} (mg/kg), Sm_{NASC} (mg/kg), Gd_{NASC} (mg/kg), Yb_{NASC} (mg/kg), Ce_{NASC} (mg/kg), and Pr_{NASC} (mg/kg) are the contents of La, Sm, Gd, Yb, Ce, and Pr in reference material, respectively. The content of REEs in reference materials can be found in Taylor and Mclennan (1985).

2.4 Bio-concentration factor (BCF)

Bio-concentration factor (BCF) is usually adopted to calculate the potential transfer of various contaminants from soils to plants (Liu et al., 2015; Ingrid et al., 2022; Nicolas et al., 2022). This approach was also employed to examine REEs. The BCF is calculated as follows:

$$BCF_i = \frac{X_{plant}}{X_{soil}}, \quad (6)$$

where BCF_i is the BCF value of the i^{th} target REEs; and X_{plant} (mg/kg) and X_{soil} (mg/kg) are the concentrations of the target REEs in soil and plant samples, respectively.

2.5 Human health risk evaluation

Three exposure pathways including ingestion, inhalation, and dermal adsorption of REEs in soils and plants might exert threat to human health (Zheng et al., 2020). This study used formulas from the Environmental Protection Agency of USA and other articles (USEPA, 1996; Chen et al., 2022; Li et al. 2022). The hazard quotients posed through three ways can be calculated using the following equations:

$$HQ_{ing} = C \times \frac{IngR \times EF \times ED}{BW \times AT \times CFD} \times 10^{-6}, \quad (7)$$

$$HQ_{inh} = C \times \frac{InhR \times EF \times ED}{PEF \times BW \times AT \times CFD}, \quad (8)$$

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$$HQ_{der} = C \times \frac{SAF \times SAE \times DAF \times EF \times ED}{BW \times AT \times CFD} \times 10^{-6}, \quad (9)$$

$$HI = HQ_{ing} + HQ_{inh} + HQ_{der}, \quad (10)$$

where HQ_{ing}, HQ_{inh}, and HQ_{der} are the hazard quotient of ingestion, inhalation, and dermal adsorption, respectively; C is element concentration (mg/kg); BW is body weight (kg); EF is exposure frequency (d/a); ED is exposure duration (a); AT is averaging time (d); CFD is corresponding reference dose (mg/(kg·d)); IngR (mg/d) and InhR (m^3/d) are ingestion rate and inhalation rate, respectively; PEF is particle emission factor (m^3/mg); SAF is skin adherence factor (mg/(cm^2·d)); SAE is skin area exposed (cm^2); DAF is dermal absorption factor; and *HI* is the sum of HQ_{ing}, HQ_{inh}, and HQ_{der}.

Reference values used in the equations are from previous studies (USEPA, 2002; Duan, 2012; Faiz et al., 2012; Chen et al., 2022; Li et al., 2022). We used a unique reference dose value (0.02 mg/(kg·d)) for all REEs in this study (Li et al., 2013; Sun et al., 2017). The values of hazard quotient above 1.0 are considered non-carcinogenic risk to human health and the values of hazard quotient below 1.0 are considered safe for the human body (Wu et al., 2018; Li et al., 2022). Carcinogenic risk was calculated by lifetime mean daily dose (LMDD, mg/(kg·d)) (Sun et al., 2017; USEPA, 2004) and used the following formulas:

$$LMDD = \frac{C \times EF}{AT} \times \left(\frac{CR_{child} \times ED_{child}}{BW_{child}} + \frac{CR_{adult} \times ED_{adult}}{BW_{adult}} \right), \quad (11)$$

$$\text{Carcinogenic risk} = LMDD \times SF, \quad (12)$$

where CR_{child} and CR_{adult} are contact rate for child and adult, respectively; ED_{child} and ED_{adult} are exposure duration for children and adults (a), respectively; BW_{child} and BW_{adult} are body weight of children and adults (kg), respectively; and SF is slope factor, which is 3.2×10^{-12} in the study (Sun et al., 2017). The unacceptable risk for regulatory purposes is higher than 1.0×10^{-4} (Faiz et al., 2012; Chen et al., 2022).

2.6 Statistical analysis

The statistical data of REEs were obtained by Excel while the correlation of REEs with soils or plants was studied by Pearson correlation analysis by using SPSS 22.0. Digital terrain model was established by applying inverse distance weight interpolation (Li et al., 2013; Xu et al., 2016; Wang et al., 2019; Wu et al., 2021). ArcGIS 10.2 processed corresponding distribution for data on human health risk values.

3 Results and discussion

3.1 REEs of soils

Detailed statistical results on the concentrations of total REEs and individual REEs in 127 soil samples were showed in Table 1. The concentrations of REEs normalized NASC, background values in Qinghai Province of China, and background values in China were presented (Taylor and Mclennan, 1985; CNEMC, 1989; MEPC, 1990). The concentrations of REEs in soils ranged from 55.29 to 306.80 mg/kg with an average of 178.55 mg/kg (Table 1). The concentration of REEs in soils from most of sampling sites were generally similar in the study area. The REEs exhibited significantly good homogeneity of distribution in soils. The total concentrations of REEs in 127 soil samples exceeded 100.00 mg/kg. Two soil samples with the highest concentrations of REEs were located in an industrial area (more than 300.00 mg/kg). The results indicated that anthropogenic activities, particularly mining or industrial activities, may contribute slightly to the accumulation of REEs in some sites of study area.

The average concentrations of REEs of NASC, background values in Qinghai Province of China, and background values in China were lower than the concentrations in soils of the study area (Taylor and Mclennan, 1985; CNEMC, 1990; MEPC, 1990). Likewise, the average concentrations

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of ΣLREEs in soils (135.26 mg/kg) were lower than the background values in China (147.92 mg/kg) and higher than that of NASC (73.50 mg/kg) and background values in Qinghai Province of China (126.37 mg/kg). Mean concentrations of ΣHREEs (43.29 mg/kg) were higher than the ΣHREEs of NASC (33.40 mg/kg), background values in Qinghai Province of China (35.46 mg/kg), and background values in China (38.84 mg/kg) (Table1). ΣLREEs in soils accounted for 76% of the total REEs, which was consistent with Tyler's results (Tyler, 2004). The results indicated that concentrations of ΣLREEs were higher than those of ΣHREEs, similar to the results of NASC. The correlation analysis illustrated that there was an obvious positive correlation among REEs of soils (Fig. S1), while the relationship among Eu, Er, Sc, and other REEs was poor. These results indicated that most of REEs might have similar sources in the soils.

Table 1 Statistic parameters of rare earth elements (REEs) in soils and plants of the northeastern Qinghai-Tibet Plateau, China

Rare earth element and parameter	Soils (mg/kg)				Plants (mg/kg)				NASC[a] (mg/kg)	Background values in Qinghai Province of China (soils)[b] (mg/kg)	Background values in China (soils)[c] (mg/kg)
	Min	Mean	Max	SD	Min	Mean	Max	SD			
La	8.36	31.04	54.26	6.6	0.10	1.76	8.66	1.7	16.00	32.80	39.70
Ce	17.09	63.66	112.43	13.2	0.44	3.78	16.70	3.4	33.00	58.30	68.40
Pr	1.82	7.32	12.50	1.6	0.02	0.39	1.93	0.4	3.90	5.87	7.17
Nd	6.95	26.95	46.38	5.8	0.08	1.43	7.19	1.4	16.0	23.70	26.40
Sm	1.33	5.18	8.94	1.1	0.02	0.28	1.45	0.3	3.50	4.77	5.22
Eu	0.33	1.11	3.26	0.3	0.00	0.07	0.32	0.1	1.10	0.93	1.03
Gd	1.31	5.10	8.81	1.0	0.02	0.28	1.41	0.3	3.30	4.15	4.60
Tb	0.18	0.68	1.24	0.1	0.00	0.04	0.19	0.0	0.60	0.60	0.63
Dy	0.99	3.56	6.68	0.7	0.01	0.20	1.02	0.2	3.70	3.80	4.13
Ho	0.20	0.69	1.29	0.1	0.00	0.04	0.20	0.0	0.80	0.77	0.87
Er	0.64	2.19	6.17	0.7	0.01	0.11	0.58	0.1	2.20	2.32	2.54
Tm	0.08	0.28	0.51	0.1	0.00	0.02	0.08	0.0	0.30	0.33	0.37
Yb	0.53	1.82	3.32	0.4	0.01	0.11	0.52	0.1	2.20	2.07	2.44
Lu	0.08	0.27	0.47	0.1	0.00	0.02	0.08	0.0	0.30	0.32	0.36
Y	5.13	18.20	34.64	3.9	0.07	1.07	5.43	1.0	20.00	21.10	22.90
Sc	4.86	10.51	21.93	3.2	0.03	0.47	2.48	0.4	30.00	10.42	/
REEs	55.29	178.55	306.80	35.8	1.03	10.06	48.25	9.3	136.90	172.25	186.76
ΣLREEs	35.88	135.26	234.42	28.2	0.84	7.71	36.27	7.2	73.50	126.37	147.92
ΣHREEs	18.34	43.29	72.39	8.8	0.18	2.35	11.98	2.2	33.40	35.50	38.84
ΣLREEs/ΣHREEs	2.75	4.14	6.09	0.4	2.95	4.21	6.74	0.6	2.20	3.56	3.81
$(La/Yb)_N$	1.71	2.36	3.61	0.3	1.52	2.22	3.01	0.3	/	2.18	2.24
$(La/Sm)_N$	1.16	1.31	1.54	0.1	1.08	1.36	1.67	0.1	/	1.50	1.66
$(Gd/Yb)_N$	1.50	1.87	2.51	0.2	1.29	1.78	3.06	0.2	/	1.34	1.26
δ_{Ce}	0.95	1.01	1.31	0.1	0.93	1.23	3.06	0.4	/	1.01	0.97
δ_{Eu}	0.49	0.67	2.02	0.1	0.51	0.81	3.14	0.3	/	0.65	0.65

Note: La, lanthanum; Ce, cerium; Pr, praseodymium; Nd, neodymium; Sm, samarium; Eu, europium; Gd, gadolinium; Tb, terbium; Dy, dysprosium; Ho, holmium; Er, erbium; Tm, thulium; Yb, ytterbium; Lu, lutetium; Y, yttrium; Sc, scandium; REEs, rare earth elements; ΣLREEs, light REEs (the sum from La to Eu); ΣHREEs, Heavy REEs (the sum Y, Sc, and from Gd to Lu); $(La/Yb)_N$, fractionation between ΣLREEs and ΣHREEs; $(La/Sm)_N$, fractionation between ΣLREEs; $(Gd/Yb)_N$, fractionation between ΣHREEs; δ_{Ce}, Ce anomaly; δ_{Eu}, Eu anomaly; Min, minimum; Max, maximum; SD, standard deviation; NASC, North American shale composite. [a] refers to Taylor and Mclennan (1985), [b] refers to MEPC (1990), [c] refers to CNEMC (1989), and / refers to no data.

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3.2 REEs of plants and BCF

Table 1 also showed the concentrations of total and individual REEs in plants. The order of concentrations of REEs in plants was basically the same as that in soils. The distribution of REEs in plants was extremely uneven in the study area. The average concentrations of REEs in plants were variable in different sites. The average concentration of total REEs was 10.06 mg/kg, and while the concentrations of total REEs ranged from 1.03 to 48.25 mg/kg in plants. The average concentration of ΣLREEs was 7.71 mg/kg in plants, and the concentration range of ΣLREEs was 0.84 to 36.27 mg/kg. The concentrations of ΣHREEs ranged from 0.18 to 11.98 mg/kg, with an average value of 2.35 mg/kg. ΣLREEs covered 77% of total REEs in plants of the study area. La, Ce, Nd, and Y in plants accounted for more than 80% of total REEs. The characteristics of REEs in soils of the study area were well correlated with the high fraction of the above REEs and the distribution order of REEs concentrations. The correlation analysis showed that there was a significantly positive correlation among the REEs in plants to prove that all REEs in plants came from the same sources (Fig. S1b).

BCF can often reflect a plant's capacity to accumulate certain components. The statistical information on BCF values of REEs at various sites was shown in Figure 2. At the sampling sites, BCF values ranged from 0.004 to 0.289 for all target REEs. The average BCF values for REEs followed the order of Eu>Lu>Y>Ce>Yb>Ho>Tm>Dy>La>Tb>Er>Gd>Sm>Nd>Pr>Sc. The average BCF value of Eu in the study area was high compared to other REEs, indicating a relatively high mobility of transplants in soils compared to other REEs. In the same study location, the mean BCF values of REEs were significantly lower than those of heavy metals (Li et al., 2018). However, the average BCF of REEs was lower than that of heavy metals, indicating that REEs were more difficult to transfer from soils to plants (Hu et al., 2013; Jeelani et al., 2017; Liu et al., 2017; Li et al., 2018). BCF values of REEs in all sites were less than 1.000, indicating relatively low bioaccumulation. The results were similar to the recent studies (Ingrid et al., 2022; Nicolas et al., 2022; Tao et al., 2022).

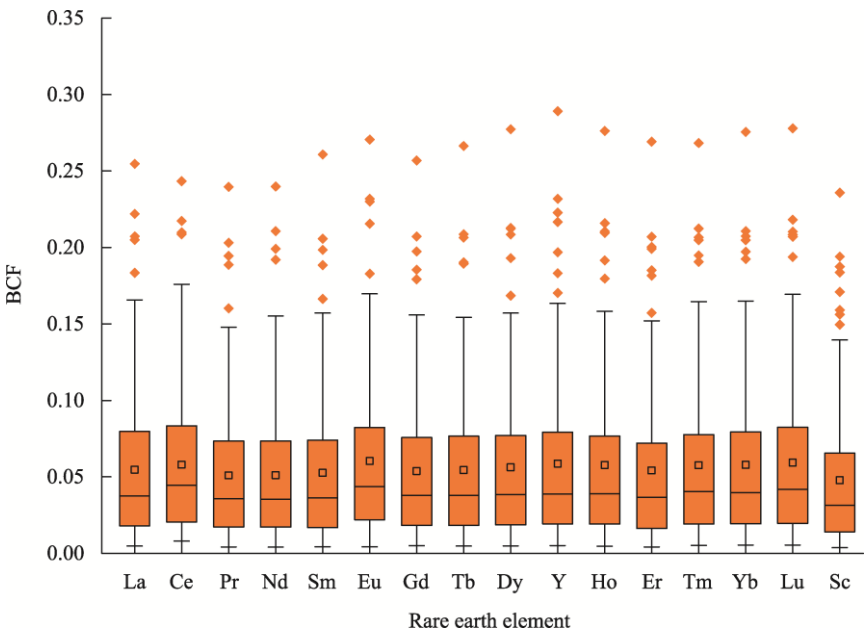

Fig. 2 Bio-concentration factor (BCF) values of rare earth elements (REEs) in the study area. La, lanthanum; Ce, cerium; Pr, praseodymium; Nd, neodymium; Sm, samarium; Eu, europium; Gd, gadolinium; Tb, terbium; Dy, dysprosium; Y, yttrium; Ho, holmium; Er, erbium; Tm, thulium; Yb, ytterbium; Lu, lutetium; Sc, scandium. The boxes represent the range from the lower quantile (Q25) to the upper quantile (Q75). The dots and horizontal lines inside the boxes represent the means and medians, respectively. The dots outside the boxes represent outliers. The upper and lower whiskers show the range within 1.5IQR (interquartile range).

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3.3 REEs Characteristic parameter in soils and plants

Table 1 and Figure 3 showed the characteristic parameters of REEs in soils and plants, including δ_{Eu}, δ_{Ce}, ΣLREEs/ΣHREEs, $(La/Sm)_N$, $(Gd/Yb)_N$, and $(La/Yb)_N$. The average value of ΣLREEs/ΣHREEs in soils was 4.14, and the range of ΣLREEs/ΣHREEs in soils was 2.75–6.09. The results were similar to previous report (Wu et al., 2018). The range of ΣLREEs/ΣHERRs in plants was 2.95–6.74 with an average value of 4.21, indicating that the values of ΣLREEs were more abundant in plants than soils. The characteristic parameter of $(La/Sm)_N$ reflected the fractionation degree of ΣLREEs. The average value of $(La/Sm)_N$ in soils was 1.31, while the range of $(La/Sm)_N$ values was 1.16–1.54 in soils. The average value of $(La/Sm)_N$ in plants was 1.36, while the range of $(La/Sm)_N$ values was 1.08–1.67. These results indicated that ΣLREEs in soils and plants were significantly fractionated in the study area.

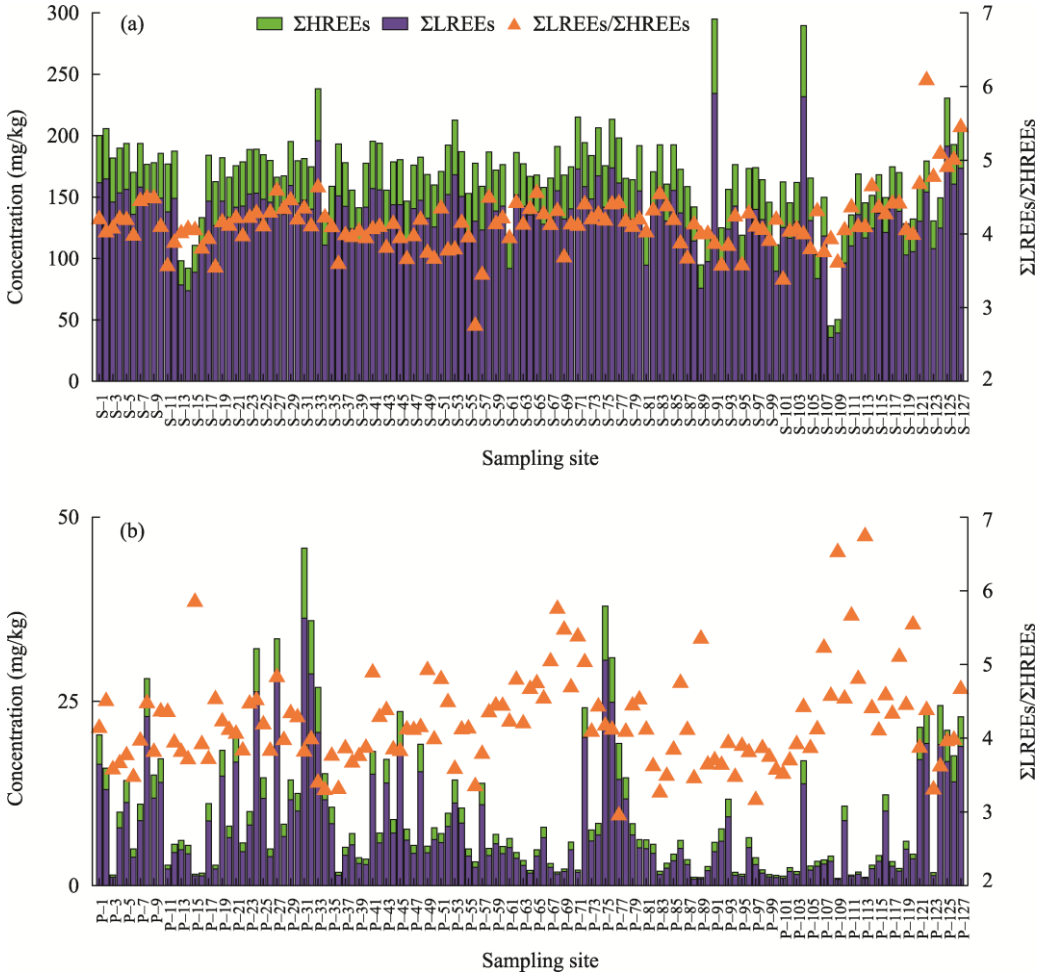

Fig. 3 Concentrations of light REEs (ΣLREEs) and heavy REEs (ΣHREEs) as well as the ratio of ΣLREEs to ΣHREEs (ΣLREEs/ΣHREEs) in soil samples (a) and plant samples (b). P-1–P-127 represent the serial number of sampling sites.

The characteristic parameter of $(Gd/Yb)_N$ illustrated the fractionation degree of ΣHREEs. The range of $(Gd/Yb)_N$ values in soils was 1.50–2.51 with an average value of 1.87, while $(Gd/Yb)_N$ values in plants ranged from 1.29 to 3.06 with an average value of 1.78 in the study area, illustrating that ΣHREEs in soils and plants had moderate fractionation. The characteristic parameter of $(La/Yb)_N$ showed the fractionation degree of ΣLREEs and ΣHREEs. The range of $(La/Yb)_N$ values in soils ranged from 1.71 to 3.61 with an average value of 2.36, while the range

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of $(La/Yb)_N$ values in plants was 1.52–3.01 with an average value of 2.22, illustrating that ΣLREEs-enriched and the curve of the figure was right oblique (Fig. 4). These characteristic parameters in this study were similar to the background values in Qinghai Province of China and the background values in China.

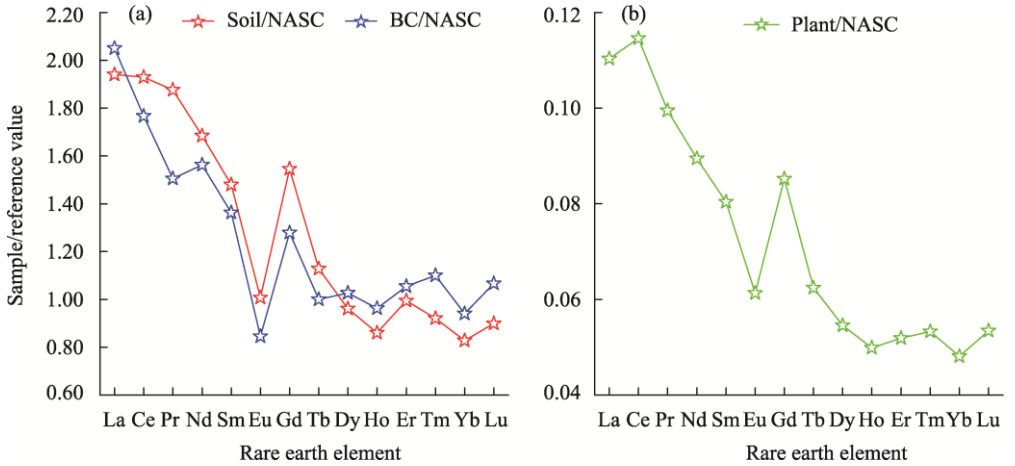

Fig. 4 (a), normalization of average REEs values and background composition in soils; (b), normalization of average REEs values in plants. NASC, North American shale composite; BC, background composition. Soil/NASC represents normalization of average REEs values in soils, BC/NASC denotes normalization of background composition in soils, and Plant/NASC represents normalization of average REEs values in plants.

δ_{Eu} and δ_{Ce} generally indicate anomalies of Eu and Ce (Yao et al., 2010; Gill et al. 2018). The elements are positive anomalies when δ_{Eu} or δ_{Ce} values are greater than 1.05, conversely, they are negative anomalies when δ_{Eu} or δ_{Ce} values are lower than 0.95 (Zhao et al. 2017; Gill et al. 2018; Wu et al., 2019). The range of δ_{Eu} values in soils was 0.49–2.02 with an average value of 0.67 in the study area. The average δ_{Eu} value in plants was 0.81, ranging from 0.51 to 3.14. These results indicated Eu in soils and plants of the study area was significantly negative anomalies. The range of δ_{Ce} values in soils was 0.95–1.31 with an average value of 1.01. The range of δ_{Ce} values in plants was 0.93–3.06 with an average value of 1.23. These results illustrated that Ce did not show significantly anomaly in soils while it showed slightly positive anomaly in plants. The δ_{Eu} and δ_{Ce} of REEs in soils and plants of the study area were very similar to the background values in Qinghai Province of China and the background values in China.

Because REEs with even atomic numbers are always more numerous than those with odd atomic numbers, it is challenging to compare the abundance of REEs directly. The concentrations of REEs were normalized using NASC to remove "Oddo-Harkin effect" in the study area (Fig. 4). The normalization model can be adopted to compare the REEs abundance in various environmental media and to determine whether there is a shortfall or enrichment of REEs (Wu et al., 2019; Ferreira et al., 2021; Lin et al., 2021). The NASC-normalized distribution patterns of REEs were usually similar in soils and plants. The average REEs patterns of the soils and plants matched the average values patterns of the background values in Qinghai Province of China (Fig. 4a and b). The distribution patterns of REEs in soils and plants of the study area were similar to the REEs patterns of the sediments and soils (Wu et al., 2019; Costa et al., 2021). Similar distribution patterns of REEs demonstrated that they primarily came from natural sources.

3.4 Health risk of soils and plants

Non-carcinogenic effects of REEs in soils and plants of the study area on human beings were calculated (Fig. 5; Tables S1 and S2). The total non-carcinogenic risks of REEs in soils for adults ranged from 0.0081 to 0.0450, while those in plants for adults ranged from 0.0002 to 0.0071. The range of total non-carcinogenic risks from REEs in soils for children was 0.0187 to 0.1038, while

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the total non-carcinogenic risks of REEs in plants ranged from 0.0003 to 0.0163. The higher non-carcinogenic risks occurred in the eastern study area where soils and plants were underlain by human activities (Fig. 5). The lowest non-carcinogenic risk value was acquired from inhalation, followed by dermal adsorption and ingestion pathways for children and adults from soils and plants. Total non-carcinogenic risk values of soils and plants obtained in this study were less than 1.0000, illustrating low non-carcinogenic risk of REEs. The non-carcinogenic risk posed by REEs from soil and plant ingestion were relatively low in the study area by considering the low REEs concentrations in soils and plants.

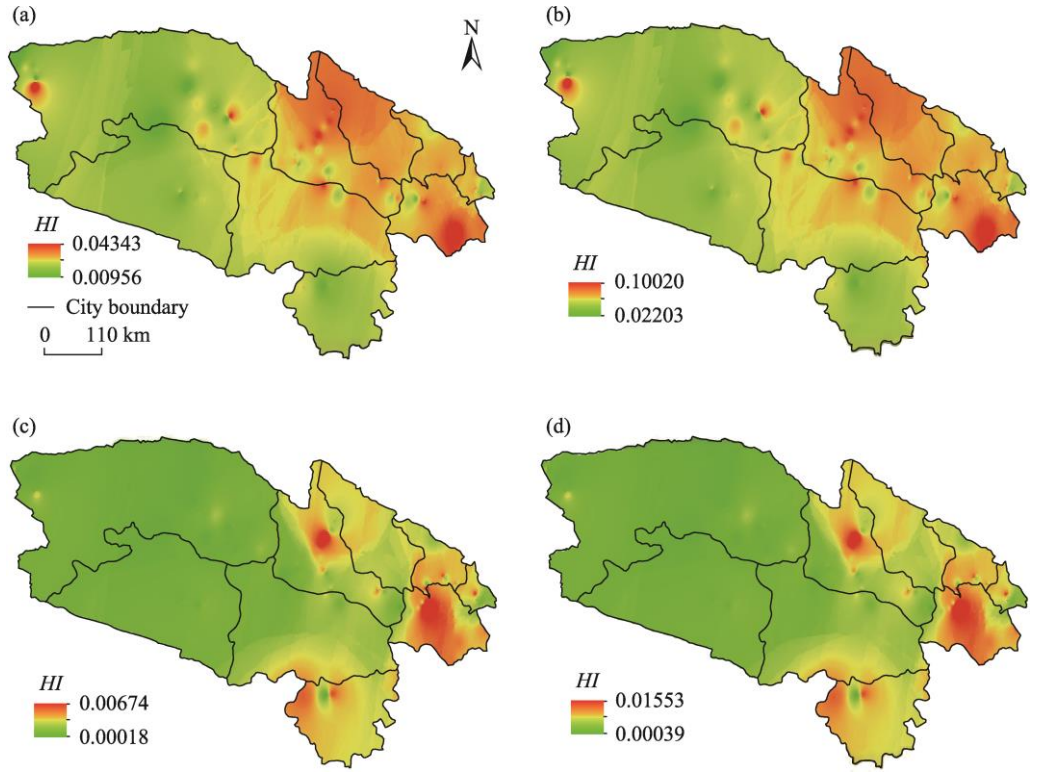

Fig. 5 Non-carcinogenic risk of REEs from soils and plants in the northeastern Qinghai-Tibet Plateau, China. (a and b), non-carcinogenic risk of REEs for adults and children from soils, respectively; (c and d), non-carcinogenic risk of REEs for adults and children from plants, respectively. *HI* is the sum of the hazard quotient of ingestion, inhalation, and dermal adsorption.

Previous studies on the health evaluation of REEs in polluted areas have shown that the non-carcinogenic risk values exceeded more than 3.000×10^{-3} for adults and 1.800×10^{-2} for children (Pagano et al, 2015; Ferreira et al., 2022), significantly higher than those values observed in plants from this study. The average non-carcinogenic risk values of La, Ce, and Nd in soils for adults was higher than 3.000×10^{-3}, while the non-carcinogenic risk value of Ce in soil for children was higher than 1.800×10^{-2}. Normally, children are more susceptible to ingesting REEs due to hand and mouth movements. In addition, children exhibited greater health risks for incomplete formation of physiologic system and low body weight (Sun et al., 2017; Li et al., 2022). Based on the results of hazard quotient in Chinese cities, total non-carcinogenic risk values higher than 0.1000 for children should be more attention (Sun et al., 2017; Guo et al., 2019).

The effect of carcinogenic risk of REEs in soils and plants of the study area on human beings were analyzed (Fig. 6; Tables S1 and S2). The total carcinogenic risk values of REEs in soils ranged from 1.593×10^{-10} to 8.839×10^{-10} (Fig. 6a), while the total carcinogenic risk values of REEs in plants ranged from 2.976×10^{-12} to 1.390×10^{-10} (Fig. 6b). Carcinogenic risk posed by REEs in the study area had similar distribution. Carcinogenic risk through ingestion, dermal

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adsorption, and inhalation were all below safe level (<1.000×10^{-6}) in the study area, illustrating that carcinogenic risk generated through REEs to human body was acceptable in the study area.

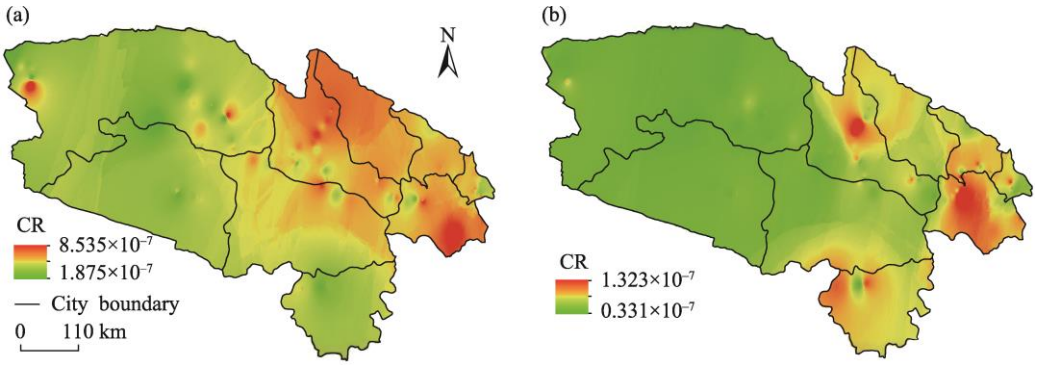

Fig. 6 Carcinogenic risk (CR) of REEs from soils (a) and plants (b) in the northeastern Qinghai-Tibet Plateau, China

4 Conclusions

The distribution of REEs in soils was noticeably homogeneous although that in plants was not homogeneous. ΣLREEs in soils and plants account for 76% and 77% of total REEs, respectively. Most of REEs might have similar sources in soils or plants. The distribution patterns of REEs in soils and plants from the study area were generally similar. ΣLREEs had significant fractionation while ΣHREEs had moderate fractionation in soils and plants. ΣLREEs enriched in soils and plants. Significant negative anomaly of Eu occurred in soils and plants. Ce was not significantly anomaly in soils and had slightly positive anomaly in plants. The average BCF values followed the order of Eu>Lu>Y>Ce>Yb>Ho>Tm>Dy>La>Tb>Er>Gd>Sm>Nd>Pr>Sc. BCF values for REEs indicated that they had relatively low bioaccumulation. Eu had relatively high mobility of transplants of the study area. These characteristic parameters of REEs in soils and plants of the study area were similar to the background values in Qinghai Province of China and the background values in China. Both non-carcinogenic and carcinogenic risks posed by REEs in soils and plants were acceptable. Adults were less sensitive to REEs ingestion compared to children.

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Appendix

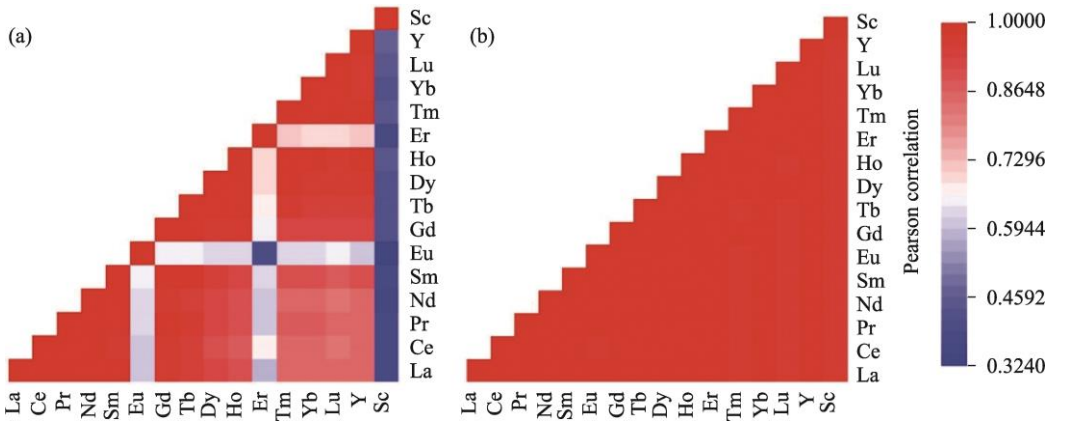

Fig. S1 Pearson correlations among rare earth elements in soils (a) and plants (b) of the northeastern Qinghai-Tibet Plateau, China. La, lanthanum; Ce, cerium; Pr, praseodymium; Nd, neodymium; Sm, samarium; Eu, europium; Gd, gadolinium; Tb, terbium; Dy, dysprosium; Ho, holmium; Er, erbium; Tm, thulium; Yb, ytterbium; Lu, lutetium; Y, yttrium; Sc, scandium.

Table S1 Non-carcinogenic and carcinogenic risk for rare earth elements (REEs) in soils of the northeastern Qinghai-Tibet Plateau, China

REE	Non-carcinogenic risk for children				Non-carcinogenic risk for adults				Carcinogenic risks			
	Ingestion	Inhalation	Dermal adsorption	Total	Ingestion	Inhalation	Dermal adsorption	Total	Ingestion	Inhalation	Dermal adsorption	Total
La	1.02×10^{-2}	7.50×10^{-6}	2.86×10^{-4}	1.05×10^{-2}	4.37×10^{-3}	3.22×10^{-6}	1.75×10^{-4}	4.55×10^{-3}	8.00×10^{-11}	6.93×10^{-12}	2.52×10^{-12}	8.94×10^{-11}
Ce	2.09×10^{-2}	1.54×10^{-5}	5.86×10^{-4}	2.15×10^{-2}	8.97×10^{-3}	6.60×10^{-6}	3.58×10^{-4}	9.33×10^{-3}	1.64×10^{-10}	1.42×10^{-11}	5.18×10^{-12}	1.83×10^{-10}
Pr	2.41×10^{-3}	1.77×10^{-6}	6.74×10^{-5}	2.48×10^{-3}	1.03×10^{-3}	7.58×10^{-7}	4.11×10^{-5}	1.07×10^{-3}	1.89×10^{-11}	1.63×10^{-12}	5.95×10^{-13}	2.11×10^{-11}
Nd	8.86×10^{-3}	6.52×10^{-6}	2.48×10^{-4}	9.12×10^{-3}	3.80×10^{-3}	2.79×10^{-6}	1.52×10^{-4}	3.95×10^{-3}	6.94×10^{-11}	6.01×10^{-12}	2.19×10^{-12}	7.76×10^{-11}
Sm	1.70×10^{-3}	1.25×10^{-6}	4.77×10^{-5}	1.75×10^{-3}	7.30×10^{-4}	5.36×10^{-7}	2.91×10^{-5}	7.59×10^{-4}	1.33×10^{-11}	1.16×10^{-12}	4.21×10^{-13}	1.49×10^{-11}
Eu	3.64×10^{-4}	2.68×10^{-7}	1.02×10^{-5}	3.75×10^{-4}	1.56×10^{-4}	1.15×10^{-7}	6.23×10^{-6}	1.62×10^{-4}	2.85×10^{-12}	2.47×10^{-13}	9.01×10^{-14}	3.19×10^{-12}
Gd	1.68×10^{-3}	1.23×10^{-6}	4.70×10^{-5}	1.73×10^{-3}	7.19×10^{-4}	5.29×10^{-7}	2.87×10^{-5}	7.48×10^{-4}	1.31×10^{-11}	1.14×10^{-12}	4.15×10^{-13}	1.47×10^{-11}
Tb	2.23×10^{-4}	1.64×10^{-7}	6.23×10^{-6}	2.29×10^{-4}	9.54×10^{-5}	7.01×10^{-8}	3.81×10^{-6}	9.93×10^{-5}	1.74×10^{-12}	1.51×10^{-13}	5.51×10^{-14}	1.95×10^{-12}
Dy	1.17×10^{-3}	8.60×10^{-7}	3.27×10^{-5}	1.20×10^{-3}	5.01×10^{-4}	3.69×10^{-7}	2.00×10^{-5}	5.22×10^{-4}	9.17×10^{-12}	7.94×10^{-13}	2.89×10^{-13}	1.02×10^{-11}
Ho	2.26×10^{-4}	1.66×10^{-7}	6.34×10^{-6}	2.33×10^{-4}	9.70×10^{-5}	7.13×10^{-8}	3.87×10^{-6}	1.01×10^{-4}	1.77×10^{-12}	1.54×10^{-13}	5.60×10^{-14}	1.98×10^{-12}
Er	7.19×10^{-4}	5.29×10^{-7}	2.01×10^{-5}	7.40×10^{-4}	3.08×10^{-4}	2.27×10^{-7}	1.23×10^{-5}	3.21×10^{-4}	5.64×10^{-12}	4.88×10^{-13}	1.78×10^{-13}	6.30×10^{-12}
Tm	9.08×10^{-5}	6.68×10^{-8}	2.54×10^{-6}	9.34×10^{-5}	3.89×10^{-5}	2.86×10^{-8}	1.55×10^{-6}	4.05×10^{-5}	7.12×10^{-13}	6.16×10^{-14}	2.25×10^{-14}	7.96×10^{-13}
Yb	5.99×10^{-4}	4.40×10^{-7}	1.68×10^{-5}	6.16×10^{-4}	2.57×10^{-4}	1.89×10^{-7}	1.02×10^{-5}	2.67×10^{-4}	4.69×10^{-12}	4.06×10^{-13}	1.48×10^{-13}	5.25×10^{-12}
Lu	8.87×10^{-5}	6.52×10^{-8}	2.48×10^{-6}	9.12×10^{-5}	3.80×10^{-5}	2.79×10^{-8}	1.52×10^{-6}	3.95×10^{-5}	6.95×10^{-13}	6.02×10^{-14}	2.19×10^{-14}	7.77×10^{-13}
Y	5.99×10^{-3}	4.40×10^{-6}	1.68×10^{-4}	6.16×10^{-3}	2.57×10^{-3}	1.89×10^{-6}	1.02×10^{-4}	2.67×10^{-3}	4.69×10^{-11}	4.06×10^{-12}	1.48×10^{-12}	5.24×10^{-11}
Sc	3.45×10^{-3}	2.54×10^{-6}	9.67×10^{-5}	3.55×10^{-3}	1.48×10^{-3}	1.09×10^{-6}	5.91×10^{-5}	1.54×10^{-3}	2.71×10^{-11}	2.34×10^{-12}	8.55×10^{-13}	3.03×10^{-11}
Total	5.87×10^{-2}	4.32×10^{-5}	1.64×10^{-3}	6.04×10^{-2}	2.52×10^{-2}	1.85×10^{-5}	1.00×10^{-3}	2.62×10^{-2}	4.60×10^{-10}	3.98×10^{-11}	1.45×10^{-11}	5.14×10^{-10}

Note: REE, rare earth element; La, lanthanum; Ce, cerium; Pr, praseodymium; Nd, neodymium; Sm, samarium; Eu, europium; Gd, gadolinium; Tb, terbium; Dy, dysprosium; Ho, holmium; Er, erbium; Tm, thulium; Yb, ytterbium; Lu, lutetium; Y, yttrium; Sc, scandium.

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Table S2 Non-carcinogenic and carcinogenic risk for REEs in plants of the northeastern Qinghai-Tibet Plateau, China

REE	Non-carcinogenic risk for children				Non-carcinogenic risk for adults				Carcinogenic risks			
	Ingestion	Inhalation	Dermal adsorption	Total	Ingestion	Inhalation	Dermal adsorption	Total	Ingestion	Inhalation	Dermal adsorption	Total
La	5.80×10^{-4}	4.27×10^{-7}	1.62×10^{-5}	5.97×10^{-4}	2.49×10^{-4}	1.83×10^{-7}	9.92×10^{-6}	2.59×10^{-4}	4.55×10^{-12}	3.94×10^{-13}	1.44×10^{-13}	5.08×10^{-12}
Ce	1.24×10^{-3}	9.14×10^{-7}	3.48×10^{-5}	1.28×10^{-3}	5.33×10^{-4}	3.92×10^{-7}	2.13×10^{-5}	5.54×10^{-4}	9.74×10^{-12}	8.44×10^{-13}	3.08×10^{-13}	1.09×10^{-11}
Pr	1.28×10^{-4}	9.38×10^{-8}	3.57×10^{-6}	1.31×10^{-4}	5.47×10^{-5}	4.02×10^{-8}	3.57×10^{-6}	5.83×10^{-5}	1.00×10^{-12}	8.66×10^{-14}	3.16×10^{-14}	1.12×10^{-12}
Nd	4.71×10^{-4}	3.46×10^{-7}	1.32×10^{-5}	4.84×10^{-4}	2.02×10^{-4}	1.48×10^{-7}	8.05×10^{-6}	2.10×10^{-4}	3.69×10^{-12}	3.19×10^{-13}	1.16×10^{-13}	4.12×10^{-12}
Sm	9.25×10^{-5}	6.80×10^{-8}	2.59×10^{-6}	9.52×10^{-5}	3.96×10^{-5}	2.91×10^{-8}	1.58×10^{-6}	4.13×10^{-5}	7.25×10^{-13}	6.28×10^{-14}	2.29×10^{-14}	8.11×10^{-13}
Eu	2.22×10^{-5}	1.63×10^{-8}	6.21×10^{-7}	2.28×10^{-5}	9.50×10^{-6}	6.98×10^{-9}	3.79×10^{-7}	9.88×10^{-6}	1.74×10^{-13}	1.50×10^{-14}	5.48×10^{-15}	1.94×10^{-13}
Gd	9.24×10^{-5}	6.80×10^{-8}	2.59×10^{-6}	9.51×10^{-5}	3.96×10^{-5}	2.91×10^{-8}	1.58×10^{-6}	4.12×10^{-5}	7.24×10^{-13}	6.27×10^{-14}	2.29×10^{-14}	8.10×10^{-13}
Tb	1.23×10^{-5}	9.06×10^{-9}	3.45×10^{-7}	1.27×10^{-5}	5.28×10^{-6}	3.88×10^{-9}	2.11×10^{-7}	5.49×10^{-6}	9.65×10^{-14}	8.36×10^{-15}	3.05×10^{-15}	1.08×10^{-13}
Dy	6.64×10^{-5}	4.88×10^{-8}	1.86×10^{-6}	6.83×10^{-5}	2.84×10^{-5}	2.09×10^{-8}	1.13×10^{-6}	2.96×10^{-5}	5.20×10^{-13}	4.50×10^{-14}	1.64×10^{-14}	5.81×10^{-13}
Ho	1.31×10^{-5}	9.65×10^{-9}	3.67×10^{-7}	1.35×10^{-5}	5.62×10^{-6}	4.13×10^{-9}	2.24×10^{-7}	5.85×10^{-6}	1.03×10^{-13}	8.90×10^{-15}	3.25×10^{-15}	1.15×10^{-13}
Er	3.75×10^{-5}	2.76×10^{-8}	1.05×10^{-6}	3.86×10^{-5}	1.61×10^{-5}	1.18×10^{-8}	6.42×10^{-7}	1.67×10^{-5}	2.94×10^{-13}	2.55×10^{-14}	9.28×10^{-15}	3.29×10^{-13}
Tm	5.26×10^{-6}	3.86×10^{-9}	1.47×10^{-7}	5.41×10^{-6}	2.25×10^{-6}	1.66×10^{-9}	8.99×10^{-8}	2.34×10^{-6}	4.12×10^{-14}	3.57×10^{-15}	1.30×10^{-15}	4.61×10^{-14}
Yb	3.48×10^{-5}	2.56×10^{-8}	9.74×10^{-7}	3.58×10^{-5}	1.49×10^{-5}	1.10×10^{-8}	5.95×10^{-7}	1.55×10^{-5}	2.73×10^{-13}	2.36×10^{-14}	8.61×10^{-15}	3.05×10^{-13}
Lu	5.27×10^{-6}	3.88×10^{-9}	1.48×10^{-7}	5.42×10^{-6}	2.26×10^{-6}	1.66×10^{-9}	9.01×10^{-8}	2.35×10^{-6}	4.13×10^{-14}	3.58×10^{-15}	1.30×10^{-15}	4.62×10^{-14}
Y	3.51×10^{-4}	2.58×10^{-7}	9.82×10^{-6}	3.61×10^{-4}	1.50×10^{-4}	1.11×10^{-7}	6.00×10^{-6}	1.56×10^{-4}	2.75×10^{-12}	2.38×10^{-13}	8.68×10^{-14}	3.07×10^{-12}
Sc	1.55×10^{-4}	1.14×10^{-7}	4.34×10^{-6}	1.59×10^{-4}	6.64×10^{-5}	4.88×10^{-8}	2.65×10^{-6}	6.91×10^{-5}	1.21×10^{-12}	1.05×10^{-13}	3.83×10^{-14}	1.36×10^{-12}
Total	3.31×10^{-3}	2.43×10^{-6}	9.26×10^{-5}	3.40×10^{-3}	1.42×10^{-3}	1.04×10^{-6}	5.80×10^{-5}	1.48×10^{-3}	2.59×10^{-11}	2.25×10^{-12}	8.19×10^{-13}	2.90×10^{-11}

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