The Concentrations and Health Risk Assessment of Rare Earth Elements in Tea in China

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Research

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Abstract

Background

Rare earth element (REE) have been considered to have potential effects on human health. However, little is known about the health risk of REEs exposure among tea drinkers in China.

Objectives

To characterize the concentration of REEs in different categories of tea and assess the health risk of REEs of tea consumers in China.

Methods

4326 tea samples (including oolong tea, dark tea, black tea, white tea, green tea and flowering tea from markets of 24 provinces, autonomous regions and municipalities in China, were analyzed for sixteen kinds of rare earth elements. Combining the REE concentration data with tea consumption data, derived from the China National Food Consumption Survey 2013–2014, the REE exposure level through tea for adults was estimated. Calculate the hazard quotient, and judge the health risk of the population based on the result value.

Results

The average concentrations of rare earth elements in Oolong tea and black tea were the highest (1611.8µg/kg and 1435.2µg/kg), and in green tea and flower tea were the lowest (735.1µg/kg and 564.1µg/kg). The content values of light rare earth elements (LREEs) were dominant in 94.53% of tea samples, cerium (431.5µg/kg) was the most abundant, followed by lanthanum (267.3µg/kg). The average daily dose of REEs in tea intake for Chinese adults was 0.0328µg/kg BW. There are statistical differences in REEs exposure among different genders of tea drinkers (p < 0.001). The average daily intake of REEs in tea was highest in men aged 40–60 (mean: 0.0383µg/kg BW, P95: 0.1421µg/kg BW), but did not exceed the acceptable daily intake (51.5µg/kg BW).

Conclusions

Our study suggested the health risk of REEs exposure from tea is low in tea consumers in China.

1. Introduction

Rare earth elements (REEs), a group of 17 elements of the periodic table including lanthanide elements, scandium (Sc) and yttrium (Y), have similar chemical and physical properties [1]. REEs are widely used in industry, medicine, agriculture, and machinery technology due to their specific properties. Simultaneously, growing evidence shows that REEs in the environment can accumulate in the human body through the food chain [2–3]. REEs are not essential biological elements. Most REEs can accumulate in human bones, blood and brain, and are very difficult to excrete, having detrimental effects on human health [4–6]. Animal studies have shown that REEs have potential neurotoxicity, pulmonary toxicity, nephrotoxicity and cytotoxicity to organisms. Exposure to high doses of REEs can cause adverse
reactions such as destroying the balance of oxidative stress and damaging fibrotic tissue\[^7\text{–}^9\]. Researches for children also showed that REEs might be related to decreased IQ and memory loss\[^10\].

Drinking tea is one of the REE’s dietary intake ways. Tea is a popular drink around the world, particularly in Asia. REEs can help develop plant roots and promote the growth of seedlings\[^11\text{–}^13\]. Therefore, rare-earth fertilizers have been widely used in China since 1990 to improve tea production and quality\[^14\]. REEs were mainly accumulated in tea leaves when applying fertilizer by spraying method\[^15\], and tea leaves have higher REEs concentrations because of longer growth periods and higher maturity\[^16\text{–}^17\]. Studies have shown that the average concentration of REEs in cereals, vegetables, aquatic products, meat, and eggs is between 0.052mg/kg and 0.337mg/kg, while the average concentration of REEs in tea reaches 2mg/kg\[^18\text{–}^19\].

However, Rare earth element exposure and health risk of tea consumers in China remains unknown. Little is known about the REE contents of all categories of tea (green tea, oolong tea, black tea, dark tea, white tea, yellow tea, and flowering tea) in China. Furthermore, the characteristics of drinking tea in Chinese populations are not well established. Considering the increasing worldwide use of REEs and the scant data on the dietary REE intake of humans, there is an urgent need to describe the concentration and the distribution of REEs in the current commercial tea, evaluate the REE exposure and the health risk of tea drinkers.

In this study, the concentrations and distribution patterns of REEs in tea collected from 2012 to 2015 in China were investigated. The risk assessment of tea REE exposure in tea consumers, including the general population and the different age-sex groups, was conducted.

### 2. Materials And Methods

The commercial tea from different regions of China was sampled to detect different REEs' concentrations in various teas and analyze their distribution patterns. According to the 2013-2014 China National Food Consumption Survey, tea consumption data were obtained. Combined with the REE content and leaching ratio in tea, the REE exposure risk assessment of tea drinkers was carried out.

#### 2.1 Sampling and determination

From 2012 to 2015, 4326 tea samples were collected from supermarkets and local markets in 24 provinces, autonomous regions and municipalities of China, including green tea, oolong tea, black tea, dark tea, white tea and flowering tea. Since only three samples, the yellow tea was not included in the study.

The concentrations of REEs in tea samples were analyzed in local laboratories in 24 provinces, autonomous regions and municipalities. All laboratories used the same analysis procedure and training was provided before the analysis. The sixteen REEs except for Pm in teas were analyzed following a protocol for inductive coupled plasma-mass spectrometry (ICP-MS) elemental analysis in China National Food Safety Standard GB5009.94-2012. Chemical data were independently verified to ensure data quality and accuracy. The limit of detection (LOD) for the method ranged from 0.003 to 0.6 μg/kg for different tea categories. All the samples with results below the LOD were calculated as 1/2LOD depending on the recommended method by the GEMS/Food (WHO 1995)\[^20\]. The percentage of samples in different tea categories to each REE below LOD was over 60%, so the LOD value (UB, upper bound) was assigned to those non-detectable samples. Otherwise, a half of LOD was assigned to the non-detectable (MB, middle bound).

#### 2.2 Tea consumption
Population intake rates for tea were derived from the China National Food Consumption Survey during 2013-2014. In the survey, 43,386 subjects were selected through stratified multi-stage cluster sampling from 16 provinces, autonomous regions, and municipalities in China. Since there are only 21 individuals in the tea-drinking population under 18, the tea-drinking population in this study only considers adults. Food consumption data were collected with a 24-hour dietary recall method on three discrete days, and body weights were obtained in each corresponding individual. Averages of each individual's food type consumption were calculated on three discrete days to represent chronic intake. Because the concentration of REEs in flavored tea beverages and tea infusion was hard to achieve, the consumption rate of tea consumers was derived from surveyed individuals reporting processed tea consumption. Consumption within the same category of processed tea was summed up for every investigated individual to match each tea category's REE concentration data. Tea high consumer group is defined as the 95th percentile of consumers.

2.3 REE exposure assessment of tea consumers

To estimate the risk of REE exposure attributable to tea consumption in adults, used the following formula to calculate the average daily dose (ADD) value:

\[
ADD = \sum ([REEs] \times IR \times LR) / \text{BW} / 1000
\]

Where ADD= average daily dose (in \(\mu\)g/kg BW/d), [REEs] = mean total REEs concentration (in \(\mu\)g/kg) in the specific category of tea, IR = specific category tea intake rate (g/d), LR=specific category tea leaching rate, and BW = each individual body weight (in kg).

Leaching rate was considered in the respect that people only drink tea infusion. Based on food consumption data, white tea was not consumed by the surveyed individuals. Leaching rates of the five tea categories have to be obtained from the available literature. Because the conversion factor of REEs to REOs (rare earth oxides) is in the range of 1.137 to 1.27 (Ce-Ce\(\text{O}_3\)), 1.148 (Dy-Dy\(\text{O}_3\)), 1.143 (Er-Er\(\text{O}_3\)), 1.158 (Eu-Eu\(\text{O}_3\)), 1.153 (Gd-Gd\(\text{O}_3\)), 1.146 (Ho-Ho\(\text{O}_3\)), 1.173 (La-La\(\text{O}_3\)), 1.137 (Lu-Lu\(\text{O}_3\)), 1.166 (Nd-Nd\(\text{O}_3\)), 1.208 (Pr-Pr\(\text{O}_3\)), 1.534 (Sc-Sc\(\text{O}_3\)), 1.160 (Sm-Sm\(\text{O}_3\)), 1.176 (Tb-Tb\(\text{O}_3\)), 1.142 (Tm-Tm\(\text{O}_3\)), 1.270 (Y-Y\(\text{O}_3\)) and 1.139 (Yb-Yb\(\text{O}_3\))), we assume that leaching rated designated as REOs can be considered as REEs. Leaching rates from the literature were calculated at the maximum values, not considering tea shape and tea origin. Leaching ratios of dark tea, oolong tea, green tea, flowering tea and black tea were 26.01%, 65.7%, 35%, 33.3% and 7.04%, respectively\(^{[19,21-23]}\). (See Additional file 1)

2.4 Health risk assessment of tea consumers

Hazard quotient (HQ) was applied to judge if the health risk is acceptable. If the value of HQ is less than one, it is believed that there is no significant health risk. If HQ exceeds one, there are possibilities that health risks occur, with a probability increasing as the value of HQ increases\(^{[24]}\). The following equation estimated the HQ:

\[
HQ = \frac{\text{ADD}}{\text{ADI}}
\]

Where ADI is the acceptable daily dose (\(\mu\)g/kg BW/d).

In 2016, the China Scientific Committee on Food Safety Risk Assessment set the temporary ADI (tADI) of three main REE, lanthanum (51.5\(\mu\)g/kg BW), cerium (645.0\(\mu\)g/kg BW), and yttrium (145.5\(\mu\)g/kg BW) based on the NOAEL of the critical effect in the decreased body weight gain from a 90-day feeding study in rats and application of a safety factor of 200 for inter-species and intra-human variability\(^{[25-26]}\). Besides, the China Scientific Committee selected the lowest tADI of lanthanum as the health-based guidance value of the total REEs following the conservation principle of risk
assessments, then derived a group tADI for the total REEs (51.5μg/kg BW). Therefore, we use 51.5μg/kg BW/d as ADI in our study.

2.5 Statistic analysis

REEs concentrations were described by mean, median, interquartile range (IQR), and quantiles. The distributions of total REEs in different categories of tea were positive-skewed, so the median could better reflect the REEs level. Considering the distributions of REEs levels did not follow a normal distribution, Kruskal-Wallis tests were used to assess statistics difference among categories of tea samples and REE exposure of various subgroups, respectively. Moreover, comparisons between multiple groups were carried out by the Wilcoxon test, adjusted by the p.adj function. Statistical significance was two-tailed and set at \( \alpha = 0.05 \). All statistical analyses and figures were performed using R 3.5.1.

3. Results

3.1 Total REE concentrations in different categories of tea

The average concentration of total REE for all 4326 tea samples was 939.1μg/kg, ranged 1.4-62207.8μg/kg. The concentrations of total REEs in the six types of tea are as follows: oolong tea > dark tea > black tea > white tea > green tea > flowering tea. The average total REE concentration in oolong tea and dark tea were 1611.8μg/kg and 1435.2μg/kg, respectively, which were higher than black tea(p < 0.001), white tea (p < 0.001), green tea (p < 0.001) and flowering tea (p < 0.001). (Table 1)

3.2 Concentrations of 16 REEs in different categories of tea

The concentrations of 16 kinds of REEs were measured on the samples of 6 categories of tea. The concentration of Ce (431.5μg/kg) was the most abundant in all tea samples, followed by the concentration of La, Y and Nd (267.3μg/kg, 204.4μg/kg and 175.5μg/kg, respectively). The concentrations of Lu and Tm (4.0μg/kg and 6.6 μg/kg, respectively) were the lowest among these REEs in all tea samples. (Table 2)
| Element | oolong tea | dark tea | black tea | white tea | green tea | flowering tea | all tea |
|---------|------------|----------|-----------|-----------|-----------|--------------|--------|
| Sc      | 101.3(0.2–1390.0) | 138.6(0.3–889.0) | 93.7(0.2–777.7) | 77.3(0.2–549.0) | 69.8(0.2–1483.0) | 47.3(0.3–635.0) | 83.5(0.2–1483.0) |
| Y       | 365.9(0.0–5900.0) | 229.8(23.7–1778.5) | 241.6(0.1–3050.5) | 114.2(1.1–618.0) | 147.9(0.0–3900.0) | 116.7(0.0–520.0) | 204.4(0.0–5900.0) |
| La      | 409.7(0.2–16800.0) | 317.2(9.0–1254.0) | 297.9(0.0–2350.0) | 150.1(1.5–803.0) | 216.2(0.0–2879.5) | 144.6(0.0–1130.0) | 267.3(0.0–16800.0) |
| Ce      | 889.5(0.2–58193.7) | 490.8(9.4–2140.0) | 425.6(0.2–8670.0) | 255.3(1.0–659.0) | 293.2(0.0–6565.0) | 252.9(0.2–1850.0) | 431.5(0.0–58193.7) |
| Pr      | 78.1(0.0–1470.0) | 67.2(2.5–317.9) | 56.7(0.0–579.0) | 31.0(0.1–146.0) | 36.0(0.0–533.5) | 27.4(0.0–198.0) | 48.4(0.0–1470.0) |
| Nd      | 293.3(0.0–6080.0) | 221.6(1.7–1214.0) | 205.4(0.0–2110.0) | 112.6(0.6–420.0) | 130.0(0.0–1907.5) | 110.6(0.1–818.0) | 175.5(0.0–6080.0) |
| Sm      | 59.4(0.0–1120.0) | 54.6(0.3–538.0) | 43.0(0.0–387.0) | 22.3(0.0–67.3) | 26.1(0.0–301.2) | 23.8(0.0–172.0) | 36.4(0.0–1120.0) |
| Eu      | 16.3(0.0–257.0) | 21.3(0.0–230.0) | 17.0(0.0–164.0) | 10.8(0.0–50.0) | 12.7(0.0–817.0) | 18.2(0.0–924.0) | 14.7(0.0–924.0) |
| Gd      | 69.2(0.0–3700.0) | 52.0(0.0–269.9) | 44.5(0.0–393.0) | 24.0(0.0–120.0) | 27.0(0.0–870.0) | 22.9(0.0–121.0) | 38.7(0.0–3700.0) |
| Tb      | 10.2(0.0–152.0) | 15.5(0.0–210.0) | 7.5(0.0–65.5) | 4.9(0.0–32.9) | 5.2(0.0–175.0) | 4.0(0.0–70.0) | 7.2(0.0–70.0) |
| Dy      | 72.3(0.0–4800.0) | 68.0(0.0–4800.0) | 46.1(0.0–3700.0) | 261.1(0.0–5800.0) | 23.4(0.0–4200.0) | 81.0(0.0–3400.0) | 41.2(0.0–5800.0) |
| Ho      | 14.2(0.0–903.0) | 12.6(0.0–190.0) | 8.8(0.0–70.5) | 7.0(0.0–28.7) | 4.5(0.0–504.0) | 10.3(0.0–240.0) | 7.6(0.0–903.0) |
| Er      | 46.5(0.0–2910.0) | 36.8(0.0–1800.0) | 27.6(0.0–1200.0) | 102.6(0.0–2200.0) | 14.6(0.0–1400.0) | 30.4(0.0–1200.0) | 24.5(0.0–2910.0) |
| Tm      | 19.6(0.0–1600.0) | 16.9(0.0–1600.0) | 5.4(0.0–1000.0) | 80.7(0.0–1900.0) | 2.9(0.0–1300.0) | 23.2(0.0–1300.0) | 6.6(0.0–1900.0) |
| Yb      | 44.5(0.0–562.5) | 25.9(0.0–246.6) | 23.8(0.0–325.3) | 9.0(0.0–38.0) | 12.3(0.0–210.9) | 19.7(0.0–739.0) | 20.7(0.0–739.0) |
| Lu      | 7.6(0.0–222.9) | 10.9(0.0–190.0) | 4.0(0.0–49.9) | 1.7(0.0–8.1) | 2.0(0.0–212.0) | 2.1(0.0–20.0) | 4.0(0.0–222.9) |
| LREE    | 1746.3(0.7–60936.8) | 1172.8(40.6–5092.1) | 1045.6(0.6–14224.0) | 582.1(4.6–2006.0) | 714.2(0.1–8579.0) | 577.5(0.7–3311.4) | 973.8(0.1–60936.8) |
| HREE    | 742.4(0.7–13944.0) | 607.1(42.3–8291.0) | 503.1(0.7–6111.5) | 682.5(1.4–10089.5) | 309.5(0.4–7011.0) | 357.6(0.7–6083.5) | 438.3(0.4–13944.0) |
| LREE/HREE | 2.9(0.0–55.1) | 2.2(0.1–5.0) | 2.5(0.1–15.4) | 2.4(0.1–5.9) | 2.8(0.0–38.4) | 4.5(0.1–92.3) | 2.8(0.0–92.3) |
The concentration values of LREE were higher than the values of HREE in 94.53% of tea samples. The ratios of LREE/HREE ranged widely from 0 to 92.35 in all tea samples, but about 85% of tea samples have LREE/HREE ranged from 1 to 4. The ratio of LREE to HREE was the largest in flowering tea and white tea (4.5), the smallest in dark tea (2.2). The proportions of LREEs and HREEs in oolong tea were 70.2% and 29.8%, respectively; they accounted for 67.5% and 32.5% in black tea.

### 3.3 REE distribution pattern in six categories of tea

The average concentrations of REEs measured in six types of tea samples were standardized by chondrite. Although the REE abundances in different kinds of tea were different, the REE distribution patterns except white tea were consistent. The distribution curve shows a negative slope, the La-Eu curve is steep, and the Eu-Lu curve is relatively flat. Dy, Er and Tm in white tea, and Tm in black tea and flowering tea all showed apparent positive abnormalities. (Fig. 1).

The LREE/HREE ratio reflects the degree of differentiation between light and heavy REEs in tea samples to a certain extent. The results showed that the L/H ratio of the six categories of tea was more significant than 1, indicating the LREEs were enriched in tea. The $\delta$E value of flowering tea and the $\delta$Ce value of oolong tea are the largest, indicating that flowering tea and oolong tea have a substantial enrichment degree for Eu and Ce, respectively. (La / Sm) N and (Gd / Yb) N reflect the degree of internal fractionation of LREE and HREE, respectively. The enrichment degree of green tea and flowering tea for LREE and oolong tea for HREE were higher than other tea types. (Table 3)

| category         | L/H | $\delta$Eu | $\delta$Ce | (La/Yb)N | (Gd/Yb)N | (La/Sm)N |
|------------------|-----|------------|------------|----------|----------|----------|
| oolong tea       | 2.9 | 0.79       | 1.16       | 6.08     | 1.25     | 4.20     |
| dark tea         | 2.2 | 1.23       | 0.79       | 8.09     | 1.61     | 3.54     |
| black tea        | 2.5 | 1.20       | 0.77       | 8.27     | 1.50     | 4.22     |
| white tea        | 2.4 | 1.44       | 0.88       | 11.01    | 2.14     | 4.10     |
| green tea        | 2.8 | 1.48       | 0.78       | 11.61    | 1.76     | 5.05     |
| flowering tea    | 2.8 | 2.41       | 0.94       | 4.85     | 0.93     | 3.70     |

### 3.4 REEs exposure attributed to drinking tea

Most people are loyal to one certain category of tea, which accounts for 95.01% of all tea consumers. About 53% of tea consumers drink green tea, 18% drinking black tea, similar to oolong tea (18%), and about 8% for both flowering tea and dark tea. No individuals reported drinking white tea or yellow tea. Among 16 REEs exposures, Ce was the highest for tea consumers. Consistent with distributions of REE in tea, LREE exposures play a significant role. Exposures from HREE are in low levels except for Y. (Fig. 2)

The average daily dose of REEs from tea in Chinese adults was 0.0328μg/kg BW/d, 0.0352μg/kg BW/d for male adults and 0.0269μg/kg BW/d for female adults, respectively. In comparing the REE exposures of male and female tea drinkers of different ages, the exposure of men is higher than that of women ($p < 0.05$). Among Chinese tea drinkers,
men between 40 and 60 years old and women over 60 years old have the highest average exposure to REEs, which was 0.0383µg/kg BW/d and 0.0310µg/kg BW/d, respectively. (Table 4)

| Age group      | Sex     | Number of subjects | REEs exposure(µg/kg BW/d) | HQa     | HQb     |
|----------------|---------|--------------------|--------------------------|---------|---------|
|                |         |                    | Range  Mean  SD  Median  P95     |         |         |
| General population | 860    | 0.0004– 0.6598      | 0.0328  0.0575  0.0126  0.1283  0.3077 | 0.0002  0.0025 |
| 18- Male       | 160    | 0.0005– 0.6598      | 0.0320  0.0667  0.0119  0.1148  0.4738 | 0.0002  0.0022 |
|                | Female  | 78                 | 0.0010– 0.1461       | 0.0237  0.0344  0.0099  0.1075  0.1408 | 0.0002  0.0021 |
| 40- Male       | 326    | 0.0003– 0.5124      | 0.0383  0.0640  0.0135  0.1421  0.3756 | 0.0003  0.0028 |
|                | Female  | 120                | 0.0006– 0.4194       | 0.0272  0.0518  0.0116  0.1036  0.2113 | 0.0002  0.0020 |
| >60 Male       | 127    | 0.0005– 0.3026      | 0.0316  0.0484  0.0118  0.1419  0.2787 | 0.0002  0.0028 |
|                | Female  | 49                 | 0.0007– 0.1412       | 0.0310  0.0360  0.0160  0.1070  0.1400 | 0.0003  0.0021 |

a Calculated with mean of REEs exposure
b Calculated with 95% quantile of REEs exposure

3.5 Risk assessment of REE exposure among tea consumers

To further explore the health risk of the REEs intake in tea, the HQ of all sub-population was estimated. The general population's HQ value was 0.0006, for male in 40–60 years and female over 60 years were 0.0007 and 0.0006, respectively. Considering the ADI is 51.5µg/kg BW/d, the HQ values of different age-sex groups are far less than 1. Even the HQ for the tea high consumer group(calculated with 95% quantile of REEs exposure) does not exceed 1 (HQ = 0.0025).(Table 4)

4. Discussion

According to our research, despite the high concentration of REEs in tea, the health risk for REEs exposure attributed to drinking tea in Chinese tea consumers is acceptable. The mean REEs daily intake through drinking tea among the general Chinese tea consumers was 0.0328µg/kg BW. Compared to exposure of REEs attributed to the other foods, such as seafood and vegetables, the level was lower[27–28]. A study on dietary exposure to REEs in Shanghai residents found that the contribution rate of tea is low, which is consistent with our conclusion[29].

By analyzing six kinds of tea on the markets in China, we found that the concentration of REEs in different types of tea is different. Oolong tea and dark tea had a high level of REEs among these categories of tea, owing to high tea leaf
maturity. The low concentration of total REEs in flowering tea might be related to the distribution of most rare earth elements in plants which were root > stem > leaf > fruit. These types of tea with high Ce (accounting for about 93% of all REEs), spraying cerium fertilizer with Ce and La as the main components in China may be the potential cause. In addition, studies have found that tea trees have a specific selectivity in the absorption of REEs, especially regarding Ce.

The distribution pattern of REEs in tea shows certain regularity. We found that the concentration of LREE in tea China was higher than that of HREE, and the proportion of LREEs and HREEs was about 70% and 30%. Consistent with the study of Guo detected oolong tea, rock tea, dark tea, black tea and green tea and found that the concentration proportion of LREEs in tea was 66.7% ~ 71.9%, and the concentration proportion of HREEs was 28.1% ~ 33.3%. This phenomenon may be related to the differentiation of REEs in the soil-plant system, indicated that the fractionation enrichment of LREEs in tea leaves and fractionation depletion of HREEs, follow the general law of plant REE fractionation. The enrichment of REEs in different teas may be related to many factors, such as the growth cycle of tea, the application of rare earth fertilizer and tea's specific absorption.

Our study results suggest that the exposure risk of REEs in tea for Chinese tea drinkers was acceptable among different age-sex groups. The average daily intake of REEs through tea drinking was higher in men than in women, and the REEs exposure was higher in male tea drinkers in 40–60 years old than in other age-sex groups. Due to living habits and other factors, male' high tea consumption leads to more REEs ingested through tea. According to the survey of tea consumption behavior in China, the proportion of male tea consumers is higher than that of females, and the Middle-aged and old people are the main body of tea consumption. Ni found that the exposure of REEs caused by drinking flowering tea would not produce any health risk. Guo also found that the exposure of REEs caused by drinking green tea with a high content of REEs was negligible. The above research is consistent with our results. HQ values in the different age-sex groups were far less than 1, indicating an acceptable level of health risk.

Furthermore, combined with the levels of REEs from other foods, we can also determine that tea drinkers have an acceptable level of dietary exposure to REEs. The study by Zhou estimated 16 REE intake levels from food, the average of REEs exposure through food for each adult was 4.18µg/kg BW/d. Based on this, together with the REEs exposure data from tea obtained in our study, the risk is also acceptable for tea consumers (HQ = 0.0818). By investigating REE concentration in food in Shaanxi Province and Shanghai, it is found that the consumption of rare earth in residents is at a safe level. According to Sun's research, only 10.29 percent of REES is absorbed by the body through other means, such as breathing and skin. Even considering all approaches, the rare earth element exposure level of tea drinkers in China is much lower than that of tADI.

Due to the use of many national samples and reliable personal consumption data, our research has considerable advantages. We explored the concentration and the distribution of rare earth elements in different categories of tea. Furthermore, we provided convincing evidence that the risk of REEs exposure caused by drinking tea is acceptable based on the consumption data. However, our assessment also has limitations. First, literature values of the leaching ratio of tea differ from each other, even for the same category of tea. Although many factors can affect the leaching ratio, tea infusion extraction procedures may play an essential role. We used the maximum leaching ratio of the same category of tea, given the health risk associated with the ingestion of REEs. The leaching ratio of tea infusion extraction procedures based on the Chinese population's drinking tea habits could be more convincing. Second, we only considered oolong tea, black tea, black tea, green tea, and flowering tea for the tea consumption of the tea drinkers and did not include other tea in the study. Therefore, we may underestimate the REEs intake of the tea drinkers. However, teas such as white tea and yellow tea are not the main types of tea consumed by Chinese residents,
and their production and purchases are low. Finally, we only estimate the health risks of rare earth element exposure based on the tADI obtained from the latest animal experiments as a health guide value. However, to determine the appropriate health guide value, reliable epidemiological research evidence is also needed to determine the harmful effects of rare earth elements on human health.

5. Conclusions

Our study has indicated that the concentrations of the total rare earth elements in six kinds of tea sold in China are oolong tea > dark tea > black tea > white tea > green tea > flowering tea. The distribution pattern of REEs in different categories of tea is similar; Ce, La and other LREEs are more enriched than HREEs. For tea drinkers of different age-sex groups, their exposure to REEs through tea drinking is also inconsistent. The average daily exposure to REEs is higher in the male 40 to 60 years old population. The exposure risk assessment showed that the estimated daily intake of REEs by drinking tea is significantly lower than the acceptable daily intake. The health risk of REEs for tea consumers is acceptable.

List Of Abbreviations

REE
rare earth element
LREE
light rare earth element
Ce
cerium
La
lanthanum
Sc
scandium
Y
yttrium
ICP-MS
inductive coupled plasma-mass spectrometry
LOD
the limit of detection
ADD
average daily dose
IR
intake rate
LR
leaching rate
BW
body weight
REOs
rare earth oxides
HQ
hazard quotient
ADI
acceptable daily dose
tADI
temporary acceptable daily dose

Declarations

Ethics approval and consent to participate
Not applicable

Consent for publication
Not applicable

Availability of data and materials
The datasets during and/or analysed during the current study available from the corresponding author on reasonable request.

Competing interests
The authors declare that they have no competing interests

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Author contributions: conceived and designed the study: YJZ, ZPL, LN, YS and SW. Performed the study: YJZ, LY, WFM, HXS, LZ, DGJ and DJY. Analyzed the data: HBB, YJZ, YS, XX, and WFM. Wrote the paper: HBB, YJZ, MNR, YYG, SW and YS.

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Figures
Figure 1

Chondrite-normalized REE distribution patterns for tea samples.
Figure 2  

Distribution of REE exposures for tea consumers  

Supplementary Files  

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