A human health risk assessment of rare earth elements in soil and vegetables from a mining area in Fujian Province, Southeast China

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**Abstract**

Contaminated food through dietary intake has become the main potential risk impacts on human health. This study investigated concentrations of rare earth elements (REEs) in soil, vegetables, human hair and blood, and assessed human health risk through vegetables consumption in the vicinity of a large-scale mining area located in Hetian Town of Changting County, Fujian Province, Southeast China. The results of the study included the following mean concentrations for total and bio-available REEs of 242.92 ± 68.98 \( \mu \text{g g}^{-1} \) and 118.59 ± 38.49 (57.89–158.96) \( \mu \text{g g}^{-1} \) dry weight (dw) in agricultural soil, respectively, and total REEs of 3.58 ± 5.28 (0.07–64.42) \( \mu \text{g g}^{-1} \) dw in vegetable samples. Concentrations of total REEs in blood and hair collected from the local residents ranged from 424.76 to 1274.80 \( \mu \text{g L}^{-1} \) with an average of 689.74 ± 254.25 \( \mu \text{g L}^{-1} \) and from 0.06 to 1.59 \( \mu \text{g g}^{-1} \) with an average of 0.48 ± 0.59 \( \mu \text{g g}^{-1} \) of the study, respectively. In addition, a significant correlation was observed between REEs in blood and corresponding soil samples \((R^2 = 0.6556, p < 0.05)\), however there was no correlation between REEs in hair and corresponding soils \((p > 0.05)\). Mean concentrations of REEs of 2.85 (0.59–10.24) \( \mu \text{g L}^{-1} \) in well water from the local households was 53-fold than that in the drinking water of Fuzhou city (0.054 \( \mu \text{g L}^{-1} \)). The health risk assessment indicated that vegetable consumption would not result in exceeding the safe values of estimate daily intake (EDI) REEs (100–110 \( \mu \text{g kg}^{-1} \text{d}^{-1} \)) for adults and children, but attention should be paid to monitoring human beings health in such rare earth mining areas due to long-term exposure to high dose REEs from food consumptions.

**Keywords:** Rare earth elements, Health risk assessment, Exposure, Vegetable consumption, Southeast China

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**Highlights**

- REEs in cultivated soil and vegetables in vicinity of mining site has been studied.
- Farmlands are contaminated seriously due to REEs mining production.
- Accumulation levels of REEs differ significantly among vegetable species.
- Elevated concentration of REEs in human hair and blood is associated with soil.
- Local residents around mining site suffer from higher exposure level of REEs.

**1. Introduction**

Rare earth elements (REEs), including lanthanides from lanthanum to lutetium and scandium and yttrium, share similar chemical and physical properties and tend to exist together naturally rather than in isolation (Henderson, 1984; Hu et al., 2006; Aquino et al., 2009), therefore they are of great concern to many scientists and have been used in the study of many geological and geochemical processes (Lee et al., 2003; Henderson, 2006; Laveuf et al., 2008; Zhou et al., 2012). Generally, low concentrations of REEs is present in soil, plant, water, and atmosphere, but REEs can accumulate in such environments following anthropogenic inputs because of the low mobility of these elements (Cao et al., 2000; Zhang and Shan, 2001; Aquino et al., 2009). In recent years, REEs have caused widespread concern because of their persistence in the environment (Lu et al., 2003; Liang et al., 2005; Tang and Johannesson, 2006; Laveuf et al., 2012), bioaccumulation in biota (Aquino et al., 2009; Šmuc et al., 2012; Dołęgowska and Migaszewski, 2013); and chronic toxicity (Ichihashi et al., 1992; Feng et al., 2006).
It is estimated that the REEs enriched fertilizers as growth promoters released into the cultivated soil are 5200 tons in China in 2002 (Anonymous, 2003). In addition, large-scale and rapid increases of the exploitation activities of REE resources have resulted in substantial increases the contamination levels of soil and water around the mining area (Liang et al., 2005; Olías et al., 2005; Wen et al., 2006; Fang et al., 2007; Miao et al., 2011). Therefore, the presence of excessive REEs contents in soils may have serious consequences for surrounding ecosystems, groundwater, agricultural productivity and human health. Under these circumstances, REEs in soil and water are released and partly enter the human body through multiple exposure pathways, especially food ingestion. To assess the potential risk of REEs to human health, it is therefore necessary to investigate their concentrations levels in daily food of vegetable, grain and meat.

Although, there is no report on incidents of human poisoning through food chain, potential concerns regarding effects of continuous exposure to low levels of REEs on human health have been arising because they are accumulated in blood, brain and bone after entering into human body (Feng et al., 2000; Yuan et al., 2003; Chen and Zhu, 2008; Aquino et al., 2009). Previous studies have found that high exposure levels of REEs may be related to health problems such as liver function decline (Arvela et al., 1976, 1980; Zhu et al., 2005). In addition, occupational and environmental exposure to REEs can pose health risk to human body (Haley, 1965; Sabbioni et al., 1982), and little information is thus far available about the dose intake and potential health effects of exposure to REEs on human beings living in the rare earth mining areas for a long term. Thus, it is necessary to investigate the accumulation levels of REEs in blood and hair which can hint human health status.

Huang et al. (2007) estimated that REE oxides entering into the soil due to mining activities with low extraction rate of 50% were 119,000 tons in 2005 in China, resulting in potential soil pollution because of their persistence and toxicity (Ding et al., 2004). Hetian Town (25°40′N, 116°20′E) is located in the middle of Changting County in the southwest of Fujian Province, China. It is also widely known for soil erosion because the most serious soil erosion within Fujian Province happened here, affecting a total area of 296 km² with a population of 68435. More than 50 rare earth mining sites scatter throughout the Town. However, this town has also a major industry for agricultural production (e.g. grain, vegetable, and fruit produces), accounting for one sixth of the Country’s total productivity. The demographic information of the study population, including age, gender, stature, weight, occupation, and personal lifestyle was obtained through face to face questionnaire surveys. The blood and hair samples analyzed were approved by every participant in this study.

2.2. Sample analysis

The soil samples were air dried at room temperature for one week and passed through 2-mm polyethylene sieve to remove plant debris and pebbles. Afterwards, the samples were ground in an agate mortar into fine powder and passed through 0.149-mm nylon sieve. Vegetable samples were washed three times with deionized water to remove soil particles and were then oven-dried at 75 °C to a constant weight for 48 h. The dried vegetable samples were ground into fine powder with an agate mortar and passed through 0.149-mm nylon sieve. The powdered samples were then stored in glass containers and kept in a refrigerator at −4 °C until analysis was made. The hair was soaked in acetone solution for 2 h, and then washed three times with deionized water to remove pollutants and was then air dried at room temperature. All glassware was soaked in 50% (v/v) HNO₃ for 24 h and rinsed with deionized water before use.

All samples were analyzed for REEs in the State Key Laboratory Breeding Base of Humid Subtropical Mountain Ecology (Fujian Normal University). Aliquots of the powdered soil and vegetable samples (0.1 g) were mineralized in a Milestone Microwave Laboratory Systems (Multiwave 3000, Anton Paar, Austria), kept under temperature control by a combination solution of hydrofluoric,
muriatic acid and nitric acid (HF 40%:HCl 38%:HNO$_3$ 70% = 1:1:3) and hydrogen peroxide and nitric acid (H$_2$O$_2$ 30%: HNO$_3$ 70% = 1:3), respectively. After digestion the solutions were diluted by deionized water to a volumetric flask of 100 mL. The bio-available fraction of REEs of soil samples (1 g) was extracted using 10 mL pH 4.65 mixture solutions of 0.5 M NH$_4$AC, 0.5 M HAC and 0.02 M Na$_2$EDTA in 50 mL polypropylene centrifugal tube (Tarvainen and Kalilio, 2002; Zhu and Bi, 2008). The tube was shaken for 1 h, centrifuged at 3500 rpm for 25 min, and filtered through a 0.22-μm Millipore filter and then diluted to 100 mL with deionized water. Blood (1 mL) and hair (0.2 g) samples were digested with 2 mL 70% nitric acid and 1 mL 30% hydrogen peroxide in an oven at 120 °C for 1 h to solution clarify. The digested solution was diluted to 50 mL volumetric flask with 5% HNO$_3$ solution after cooling. Working solutions were freshly prepared daily for analysis.

15 rare earth elements (excluding scandium and promethium) of all samples were determined using inductively coupled plasma mass spectrometry (ICP-MS, X Series 2, Thermo Scientific, USA). External calibration was performed by measuring standard solutions containing these 15 rare earth elements (Analytical measurement centre of national nonferrous metals and electronic material, China) at 0.5, 1, 5, 10, 20, 50 and 100 μg L$^{-1}$ concentrations. Mixed solutions containing Rh, In and Re (Analytical measurement centre of national nonferrous metals and electronic material, China) were used as internal standard at 5 μg L$^{-1}$ concentration. The instrument detection limits of these 15 rare earth elements for this method were 0.003–0.016 μg L$^{-1}$, which were determined as three times of standard deviation from seven blank solutions.

### 2.3. Quality control

The accuracy and precision of the soil analysis were assessed using National Certified Reference Materials of China such as soil (GBW07406) GSS-5, aspen leaf (GBW07604) GSV-3 and human hair (GBW09101a). Standard solutions were inserted into the sample sequence every 8 samples to verify sensitivity and repeatability. The recoveries of these 15 rare earth elements were significantly higher ($p < 0.05$) than those of the control sample.

### 2.4. Health risk assessment of vegetable consumption

The estimate daily intake of REEs through the vegetable consumption was calculated by the following equations (US EPA, 2000):

\[
EDI = \frac{C_V \times CR}{BW}
\]

\[
EDI_{bio} = \frac{C_V \times CR \times BA}{BW}
\]

whereby $EDI$ and $EDI_{bio}$ represented the estimated daily intake of REEs and estimated daily intake of bioavailable REEs, respectively; $BA$ (%) was bioavailability of REEs in vegetables (mean value of 46%) as cited from literature Shao et al. (2012); $C_V$ for concentration of REEs in vegetable was based on dry weight; $BW$ for body weights of adults and children were 60.1 kg and 38.3 kg, respectively; $CR$ for the consumption rate was 478 g d$^{-1}$ and 272 g d$^{-1}$ and water of 3 L d$^{-1}$ and 2 L d$^{-1}$ for adults and children, respectively, in Fujian Province (Statistics Bureau of Fujian Province, 2011). Vegetable consumption rate for children were estimated as 57% of that of adults (US FDA, 2003). The intake dose of REEs found to be damaging to human health was 100–110 μg kg$^{-1}$ d$^{-1}$, which was certificated from human health survey in REEs mining areas and animal experimental results (Zhu et al., 1997a,b).

### 2.5. Statistical analyses

Statistical analyses for comparing the average results of the different soil, vegetable, and well water samplings were performed using a one-way analysis of variance (ANOVA) followed by Scheffe’s test for multiple comparisons. Stepwise multiple linear regression analysis was used to evaluate the relationships between REEs in soil and human blood, hair, respectively. A level of $p < 0.05$ was considered statistically significant for multiple comparisons and linear regression analysis. Statistical analysis was conducted using SPSS software package 16.0 (SPSS Inc., Chicago, IL, USA) and Excel software package 2003.

### 3. Results and discussion

#### 3.1. Concentration levels of REEs in soil

The concentrations of total and bio-available REEs in soil samples are shown in Fig. 2, with the mean concentration of total REEs being 242.92 ± 68.98 μg g$^{-1}$ dw, ranging from 135.85 to 327.55 μg g$^{-1}$ dw. The mean concentration of bio-available REEs was 118.59 ± 38.49 μg g$^{-1}$ dw, ranging from 57.89 to 158.96 μg g$^{-1}$ dw, with bio-available REEs contributing to 48.53% of total REEs. The highest concentrations of total and bio-available REEs were observed in sample Nos. 3 and 4, respectively. The concentrations of total and bio-available REEs in soil samples collected from farms in the vicinity of mining site were significantly higher ($p < 0.05$) than those of the control sample.

The concentrations of REEs in all sample sites, except for the control site (No. 5), were higher than the soil background value of China (187.60 μg g$^{-1}$) (Wei et al., 1991). The mean concentration of total REEs in the present study was lower than Baiyun Obo, Baotou and Ganzhou (Li et al., 2010; Xu et al., 2011; Zhang et al., 2000b; Gao et al., 2001), and it was higher than other regions (Table 1). Our results suggest that the agricultural soil environment in mining sites in Hetian Town was moderately polluted by REEs. Previous studies have reported that REEs can continuously accumulate in surface soil following various pathways such as atmospheric
Comparison of concentrations of REEs in soil (µg g⁻¹) from different parts of the world.

| Regions                   | Range (µg g⁻¹ dw) | Mean (µg g⁻¹ dw) | References          |
|---------------------------|-------------------|------------------|--------------------|
| Hetian town, China        | 135.85–327.55     | 242.92           | Present study       |
| Soil background value, China | –                 | 187.60           | Wei et al. (1991)   |
| Yucheng county, China     | 152.23–338.86     | 219.87           | Mao et al. (2011)   |
| Košani field, Macedonia   | 106.40–244.39     | 173.54           | Šmuc et al. (2012)  |
| Ganzhou city, China       | 315.00–1355.00    | 572.00           | Zhang et al. (2000a,b), Gao et al. (2001) |
| Kielce, Poland            | 34.11–93.09       | –                | Dołęgowska and Migaszewski (2013) |
| Baotou, China             | 40.14–3079.13     | 284.07           | Xu et al. (2011)    |
| Baiyun Obo, China         | 475.27–27549.58   | –                | Li et al. (2010)    |
| Nidda catchment, Germany  | 41.50–544.20      | 201.10           | Loell et al. (2011) |

As we have discussed above, vegetables can significantly transfer REEs from soil to edible part, and can accumulate continuously, resulting in REE pollution of vegetables in the study region. High concentration level of REEs in soil can also lead to more absorption and accumulation of REEs by vegetables (Tyler, 2004). Actually, plant uptake of REEs depend on variables of plant species (Tyler and Olsson, 2001; Loell et al., 2011), and mobility and bioavailability of REEs in soil (Loell et al., 2011). The accumulations of REEs in vegetables are likely attributed to the high bioavailability of REEs in soil and high concentration levels in wastewater of the mining activities. Bioavailability of REEs in soils in this study region were higher than those from the control site (No.5). The highest concentrations of REEs in human blood and hair were observed in sample 3, which contained 1108 µg L⁻¹ and 1.67 µg g⁻¹. This

3.3. Concentration levels of REEs in human blood and hair

The concentrations of REEs in human blood and hair ranged from 424.76 to 1274.80 µg L⁻¹ with mean value of 689.74 ± 254.25 µg L⁻¹ and ranged from 0.06 to 1.89 µg g⁻¹ with mean value of 0.48 ± 0.59 µg g⁻¹, respectively (Fig. 4). The mean concentrations of REEs in human blood and hair samples of exposure sites were higher than those from the control site (No. 5). The highest concentrations of REEs in human blood and hair were observed in sample 3, which contained 1108 µg L⁻¹ and 1.67 µg g⁻¹.
Hair Man Man

...was observed between REEs in hair and soil samples (REEs (Arvela et al., 1976, 1980), and high concentration levels of previous studies have demonstrated that liver damage is sensitive to eventually accumulate REEs through food digestion and absorption. Pre-

...source of REEs pollution in foods, and human body can continually accumulate REEs through food digestion and absorption. Previous studies have demonstrated that liver damage is sensitive to REEs (Arvela et al., 1976, 1980), and high concentration levels of REEs in human body’s blood can cause immunogenic damage to the vascular wall, facilitating the formation of arteriosclerosis (Zhu et al., 1997a,b); therefore the REEs toxicity through long-term intake of small doses REE could not be negligible for human beings. Concentration levels of REEs in local residents’ blood samples were markedly higher than those of normal human blood samples from Changchun Blood Center, northeast China, which ranged from 1.40 to 13.30 μg L⁻¹ with an average of 4.07 μg L⁻¹ (Meng et al., 1999). In comparison with other studies, concentrations of REEs in hair obtained in the present study were slightly lower than those of human exposed to REEs mining site, Shandong Province, east China (Lu et al., 2007), and also distinctively lower than those of children’s hair (0–3 years) from an ion-adsorptive type mining in southern China, which ranged from 1.64 to 9.32 μg g⁻¹ with mean value of 4.73 ± 2.43 μg g⁻¹ (Peng et al., 2002).

### 3.4. Human health risk assessment through vegetables consumption

The EDIs of REEs through vegetable consumption for both adults and children in Fujian Province, indicate that not all vegetable consumption would result in an exceeding value of EDI found to be damaging to human health (100–110 μg kg⁻¹ d⁻¹) (Zhu et al., 1997a,b), while the EDIs of adults and children for the vegetable species including taro and water spinach were much higher than the other species (Table 3). The daily intake dose for both adults and children of the eight vegetables declined in the order of taro > water spinach > lettuce > pakchoi > long bean > eggplant > white radish > Chinese cabbage. Children were more easily exposed to REEs than adults because of high daily intake dose of REEs via vegetables consumption (Table 3), and attention should be paid to avoiding high intake dose of taro. Although the EDI of REEs in the water was lower than that of all the vegetables, water was easily absorbed by gastric and intestinal digestion. In addition, human beings are at the top of the predatory food chain, therefore health damage caused by long-term exposure to REEs from water and vegetables should not be neglected.

It should be noted that the health risk assessment results may be influenced by other factors such as other food (grain, meat, and fruits) ingestion and bioavailability of REEs in foods to humans. Considering the limitations in economic budget and labor force, the bioavailability of REEs in vegetables was cited from literature (Shao et al., 2012) for this study. The results indicate that the

### Table 2

Concentrations of REEs in different vegetable species in present study.

| Vegetable species | Mean (µg g⁻¹ dw) | Range (µg g⁻¹ dw) | Locations | Samples (n) |
|-------------------|------------------|------------------|-----------|------------|
| Chinese cabbage   | 0.197 ± 0.128    | 0.065–0.304      | No. 2, No. 3, No. 4, No. 6 | 4          |
| Water spinach     | 6.121 ± 6.453    | 1.376–18.725     | No. 1, No. 2, No. 3, No. 4, No. 5, No. 6 | 6          |
| Lettuce           | 0.726 ± 0.337    | 0.452–1.103      | No. 1, No. 3, No. 5 | 3          |
| Chinese radish    | 0.199 ± 0.087    | 0.127–0.314      | No. 1, No. 2, No. 4, No. 6 | 4          |
| Long bean         | 0.469 ± 0.514    | 0.080–1.052      | No. 2, No. 5, No. 6 | 3          |
| Eggplant          | 0.202 ± 0.099    | 0.068–0.269      | No. 1, No. 4, No. 5 | 4          |
| Pakchoi           | 0.713 ± 0.245    | 0.522–0.965      | No. 1, No. 3, No. 4, No. 5 | 4          |
| Taro              | 14.719 ± 27.904  | 0.542–64.419     | No. 1, No. 2, No. 3, No. 4, No. 5 | 5          |

**Note:** The concentration of REEs in blood and hair samples may be due to the higher concentrations of REEs in soil at this location compared to the other sampling sites. In addition, there was a significant correlation between REEs in blood and corresponding soil samples ($R^2 = 0.6556$, $p < 0.05$), but no significant correlation was observed between REEs in hair and soil samples ($p > 0.05$) (Fig. 5). These results indicate that soil containing REEs may be the source of REEs pollution in foods, and human body can continually accumulate REEs through food digestion and absorption. Previous studies have demonstrated that liver damage is sensitive to REEs (Arvela et al., 1976, 1980), and high concentration levels of REEs in human body’s blood can cause immunogenic damage to the vascular wall, facilitating the formation of arteriosclerosis (Zhu et al., 1997a,b); therefore the REEs toxicity through long-term intake of small doses REE could not be negligible for human beings.

Concentration levels of REEs in local residents’ blood samples were markedly higher than those of normal human blood samples.
consumption of any of the sampled vegetable species should not result in an EDI of REEs value exceeding 100–110 μg kg⁻¹ d⁻¹ for both adults and children (Table 3). However, due to toxic pollutants entering into human body mainly through food ingestion, dermal absorption and breath inhalation, therefore a more systematic risk assessment is taken into account for expounding the impacts of REEs to human beings in the future.

4. Conclusions

Soil and vegetable samples collected from farms in the vicinity of mining site showed higher concentrations of REEs than those of the control site, and thus indicating that the REEs derived from mining production processes does actually accumulate in the agriculture soil and vegetables. The mean concentration of REEs in well water was significantly higher than that in drinking water of non-mining area. Concentrations of REEs in blood and hair samples collected from local peasants were higher than those of normal human beings, possibly due to high concentrations of REEs in foods and local farmers’ long-term ingestion via food chain. The risk assessment indicates that the consumption of vegetable should not result in an excessive value of REEs EDI found to be damaging to human beings, possibly due to high concentrations of REEs in foods and local farmers’ long-term ingestion via food chain. The risk assessment indicates that the consumption of vegetable should not result in an excessive value of REEs EDI found to be damaging to human health (100–110 μg kg⁻¹ d⁻¹) for both adults and children (Table 3). However, due to toxic pollutants entering into human body mainly through food ingestion, dermal absorption and breath inhalation, therefore a more systematic risk assessment is taken into account for expounding the impacts of REEs to human beings in the future.

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Table 3

| Vegetable species     | Adult        | Adultᵦᵣᵢᵠ | Children | Childrenᵦᵣᵠ |
|-----------------------|--------------|------------|----------|-------------|
| Chinese white cabbage | (0.0517–0.2418, 0.1567) | (0.0228–0.1112, 0.0721) | (0.0462–0.2159, 0.1399) | (0.0212–0.0993, 0.0644) |
| Water spinach          | (1.0944–14.8928, 4.6863) | (0.5034–6.8507, 2.2394) | (0.9772–12.2982, 4.3477) | (0.4495–6.1172, 1.9996) |
| Lettuce               | (0.3595–0.8773, 0.5774) | (0.1654–0.4035, 0.2656) | (0.3210–0.7833, 0.5156) | (0.1477–0.3603, 0.2372) |
| Chinese radish        | (0.1010–0.2497, 0.1583) | (0.0465–0.1149, 0.0728) | (0.0902–0.2230, 0.1413) | (0.0415–0.1026, 0.0650) |
| Long bean             | (0.0636–0.3867, 0.3730) | (0.0293–0.3849, 0.1716) | (0.0568–0.7471, 0.3331) | (0.0261–0.3437, 0.1532) |
| Eggplant              | (0.0541–0.2130, 0.1607) | (0.0269–0.0984, 0.0739) | (0.0483–0.2110, 0.1439) | (0.0222–0.0879, 0.0690) |
| Pakchoi               | (0.4152–0.7675, 0.5671) | (0.1910–0.3531, 0.2609) | (0.3707–0.6853, 0.5064) | (0.1705–0.3153, 0.2329) |
| Taro                  | (0.4311–51.235, 11.7066) | (0.1983–23.5681, 5.385) | (0.385–45.7493, 10.453) | (0.1771–21.045, 4.8085) |
| Water                 | (0.3000–0.5121, 0.1433) | – | (0.0313–0.5358, 0.1499) | – |

Note: Values in parentheses were range and mean.
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