



# 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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## 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.

## ARTICLE INFO

### Article history:

Received 16 January 2013

Received in revised form 17 June 2013

Accepted 29 June 2013

Available online 25 July 2013

### Keywords:

Rare earth elements  
Health risk assessment  
Exposure  
Vegetable consumption  
Southeast China

## 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  $242.92 \pm 68.98$  ( $135.85$ – $327.56$ )  $\mu\text{g g}^{-1}$  and  $118.59 \pm 38.49$  ( $57.89$ – $158.96$ )  $\mu\text{g g}^{-1}$  dry weight (dw) in agricultural soil, respectively, and total REEs of  $3.58 \pm 5.28$  ( $0.07$ – $64.42$ )  $\mu\text{g g}^{-1}$  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  $689.74 \pm 254.25$   $\mu\text{g L}^{-1}$  and from  $0.06$  to  $1.59$   $\mu\text{g g}^{-1}$  with an average of  $0.48 \pm 0.59$   $\mu\text{g g}^{-1}$  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.

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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

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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.,

2000; Zhang et al., 2000a; Yuan et al., 2003; Chen and Zhu, 2008; Aquino et al., 2009).

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, 2005; 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 km<sup>2</sup> 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 production (Statistics Bureau of Changting County, 2011). Therefore, REEs contamination derived from mining can have a potential impact on the growth and safety of such produces.

This is the first study dealing with REEs contamination in soil and vegetables of local households in close proximity to a large-scale rare earth mining site in Southeast China. The main objectives of the present study were: (1) to investigate the concentrations of REEs in soil and vegetables collected from farms in the vicinity of mining site in Hetian Town; (2) to investigate concentrations of REEs in human blood and hair collected from farmers living close to mining site; (3) to evaluate the health risk of dietary REEs exposure to vegetable consumption.

## 2. Materials and methods

### 2.1. Sampling and pretreatment

The samples were collected, during May and June in 2012, from 6 sampling sites of farms located in the vicinity of a large-scale rare earth mining site (Fig. 1). Sample No. 5 was chosen from the site

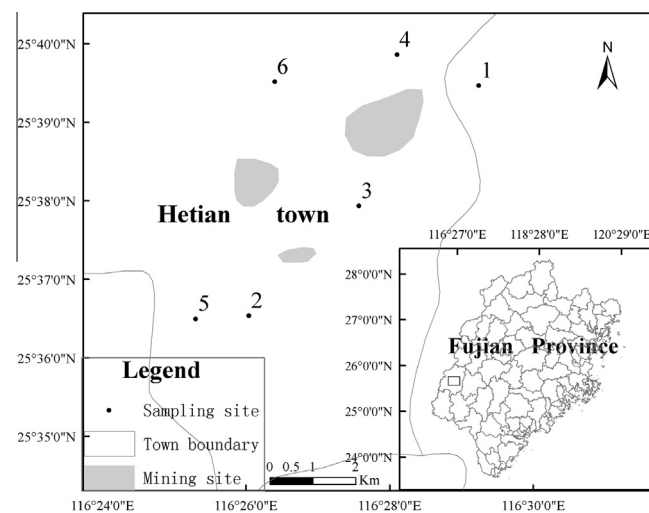


Fig. 1. Locations of the sampling sites in Hetian Town.

located 5 km away from the rare earth mining site as control sample. The other 5 soil samples and vegetable samples including Chinese white cabbage (*Brassica pekinensis* L.), taro (*Alocasia macrorrhiza* L.), Chinese radish (*R. sativus* var. *longipinnatus*), water spinach (*Ipomoea aquatica*), lettuce (*Lactuca sativa* var.), long bean (*Vigna unguiculata* Linn.), pakchoi (*Brassica chinensis* L. var.) and eggplant (*Solanum melongena* L.) were collected from farmlands in close proximity to rare earth mining site. Agriculture soils were collected from a depth of 0–20 cm using a soil sampler and 3–5 mixed plants of each vegetable were randomly cut with ceramic scissors corresponding to soil samples. Meanwhile, blood and hair samples of 12 farmers (one adult man and one adult woman of each household) were collected with the help of Hetian Hospital. After collection, all samples were immediately stored in a portable cryostat and transported to the laboratory.

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 in 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) HNO<sub>3</sub> 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<sub>3</sub> 70% = 1:1:3) and hydrogen peroxide and nitric acid (H<sub>2</sub>O<sub>2</sub> 30%: HNO<sub>3</sub> 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<sub>4</sub>AC, 0.5 M HAC and 0.02 M Na<sub>2</sub>EDTA in 50 mL polypropylene centrifugal tube (Tarvainen and Kallio, 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- $\mu$ m 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<sub>3</sub> 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  $\mu$ g L<sup>-1</sup> 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  $\mu$ g L<sup>-1</sup> concentration. The instrument detection limits of these 15 rare earth elements for this method were 0.003–0.016  $\mu$ g L<sup>-1</sup>, 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 80.1–105.1%, 86.8–114.6%, 90.2–104.7%, 88.2–92.4%, and 97.7–104.7% for soil, vegetable, water, hair, and blood, respectively. The analytical precision and accuracy were accepted only when RSD values were below 5% for the REEs, according to the results of duplicate measurements of all samples and the three standard materials.

### 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):

$$EDI = \frac{C_V * CR}{BW} \quad (1)$$

$$EDI_{bio} = \frac{C_V * CR * BA}{BW} \quad (2)$$

whereby *EDI* and *EDI<sub>bio</sub>* ( $\mu$ g kg<sup>-1</sup> d<sup>-1</sup>) represented the estimated daily intake of REEs and estimated daily intake of bioaccessible REEs, respectively; *BA* (%) was bioaccessibility of REEs in vegetables (mean value of 46%) as cited from literature Shao et al. (2012); *C<sub>V</sub>* 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<sup>-1</sup> and 272 g d<sup>-1</sup> and water of 3 L d<sup>-1</sup> and 2 L d<sup>-1</sup> 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  $\mu$ g kg<sup>-1</sup> d<sup>-1</sup>, 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  $\pm$  68.98  $\mu$ g g<sup>-1</sup> dw, ranging from 135.85 to 327.55  $\mu$ g g<sup>-1</sup> dw. The mean concentration of bio-available REEs was 118.59  $\pm$  38.49  $\mu$ g g<sup>-1</sup> dw, ranging from 57.89 to 158.96  $\mu$ g g<sup>-1</sup> 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 No. 5 (total REEs: 135.85  $\pm$  12.43  $\mu$ g g<sup>-1</sup> dw; bio-available REEs: 57.89  $\pm$  8.45  $\mu$ g g<sup>-1</sup> dw).

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  $\mu$ g g<sup>-1</sup>) (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

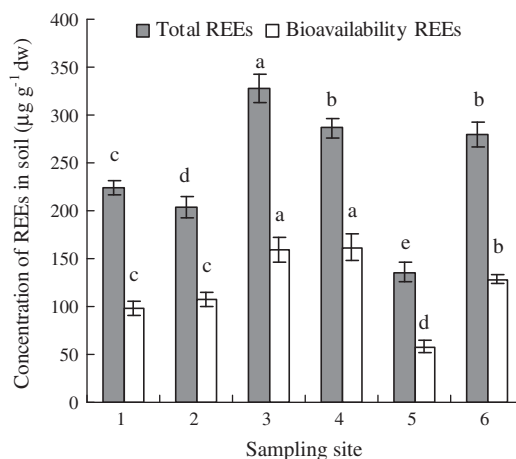


Fig. 2. Concentrations of total and bio-available REEs in soil samples. Values are average (*n* = 3)  $\pm$  standard deviation. Columns with the same letter do not differ significantly at the 5% level according to the Scheffe's multiple comparison test.

**Table 1**

Comparison of concentrations of REEs in soil ( $\mu\text{g g}^{-1}$ ) from different parts of the world.

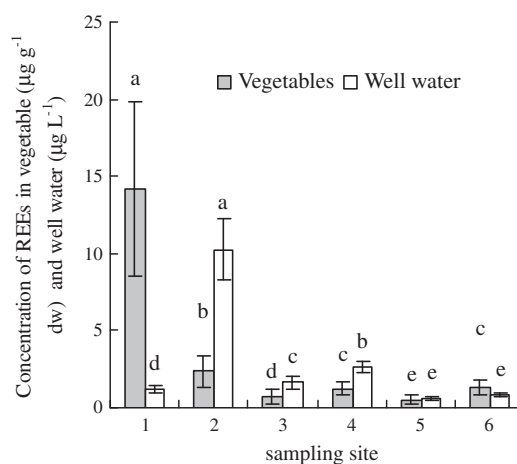
Regions	Range ( $\mu\text{g g}^{-1}$ dw)	Mean ( $\mu\text{g g}^{-1}$ dw)	References
Hetian town, China	135.85–327.55	242.92	Present study
Soil background value, China	–	187.60	Wei et al. (1991)
Yucheng county, China	152.23–338.86	219.87	Mao et al. (2011)
Kočani field, Macedonia	106.40–244.39	173.54	Šmuc 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	40.14–3679.13	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)

deposition (Rühling and Tyler, 2004; Tyler, 2004), sewage irrigation (Wang et al., 2003; Zhu et al., 2012), mining activities (Zhu et al., 1997a,b; Zhang et al., 2000b; Yu et al., 2009), and application of REEs fertilizers (Todorovsky et al., 1997; Zhang and Shan, 2001; Xu et al., 2002; Pan et al., 2002) because of the low mobility of these elements. Soils in Baiyun Obo of Baotou, China, were contaminated seriously by REEs due to the integrated industries involving metallurgy, rare earth production and machinery manufacturing (Li et al., 2010), and the increasing application of REE-salvatable compound fertilizer and REE-phosphate fertilizer on farmland of Baotou (Zhang et al., 2000b). It appears that REEs refined in factory can be attributed as a more serious factor to REEs contamination in soil compared to other field mining activities such as situ leaching.

In this study, the bio-available REEs in soils accounted for 48.53% of total REEs, indicating that it was higher than that of Nidda catchment, in Central Germany, which ranged from 1.5% to 41.2% with an average of 15.9% (EDTA extraction) (Loell et al., 2011). Bioavailability of REEs in soil is primarily controlled by soil pH, clay minerals and organic carbon contents (Diatloff et al., 1996; Cao et al., 2000, 2001; Tyler, 2004; Miao et al., 2007). Harter (1983) reported that low soil pH favors conversion of metals from precipitate fractions into soluble fractions. In addition, REEs releases increase drastically under soil pH 3.5 conditions (Cao et al., 2001). Oxalate and ammonium sulfate widely used in extraction procedures may re-enter the soil environment with discharges of untreated wastewater, resulting in significant decrease in soil pH in mining site of Hetian Town. The climate of subtropical monsoon with warm and humid air in this area contribute to significant weathering of granite, therefore clay minerals containing a large amount of carbonates, silicates and phosphates (Taunton et al., 2000) play an important role in increasing the mobility and bioavailability of REEs in soils (Laveuf and Cornu, 2009; Loell et al., 2011). Consequently, the bioavailability of REEs in soils in the present study are higher than in other regions (Cao et al., 2000; Lu et al., 2003; Wang et al., 2004; Loell et al., 2011; Šmuc et al., 2012).

### 3.2. Concentration levels of REEs in vegetable and water

The concentrations of REEs in vegetables ranged from 0.06 to 64.42  $\mu\text{g g}^{-1}$  dw with an average of 3.58  $\mu\text{g g}^{-1}$  dw. The mean concentrations of REEs in vegetable samples collected from the vicinity of mining site were significantly higher ( $p < 0.05$ ) than those in the control site (Fig. 3). The concentrations of REEs in the vegetable



**Fig. 3.** Concentrations of total REEs in vegetable and well water samples. Values are average  $\pm$  standard deviation. Columns with the same letter do not differ significantly at the 5% level according to the Scheffe's multiple comparison test.

species of taro (75% of water content) and water spinach (85% of water content) were higher than China's National Standard ( $0.7 \mu\text{g g}^{-1}$  fresh weight, GB2762-2005), which indicates that REEs from rare earth mining was a source of REEs pollution. The concentration level of REEs in taro was 77 fold higher than Chinese white cabbage. Generally, concentrations of REE in leafy vegetables were obviously higher than those in non-leafy vegetables (Table 2). The concentration of REEs in well water ranged from 0.59 to 10.26  $\mu\text{g L}^{-1}$  with an average of 2.87  $\mu\text{g L}^{-1}$  (Fig. 3), which was significantly higher than drinking water (mean value of 0.054  $\mu\text{g L}^{-1}$ ) in Fuzhou City, China.

The concentration levels of REEs in vegetables in our study region were higher than local markets' vegetables of China ( $0.28 \mu\text{g g}^{-1}$ , Jiang et al., 2012;  $0.59 \mu\text{g g}^{-1}$  dw, Liu et al., 1997), and also higher than rice grain ( $0.074 \mu\text{g g}^{-1}$  dw) (Šmuc et al., 2012). However, concentration levels of REEs in vegetables in Datian and Anxi, China ( $6.55 \mu\text{g g}^{-1}$ ,  $7.12 \mu\text{g g}^{-1}$  dw, respectively) (Zhang et al., 2000c) were significantly higher than those in our results, because they were seriously affected by the intensive mining activities.

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 area. Therefore, further investigation is needed to explain the impacts of concentration levels of REEs in soil and water on the vegetables.

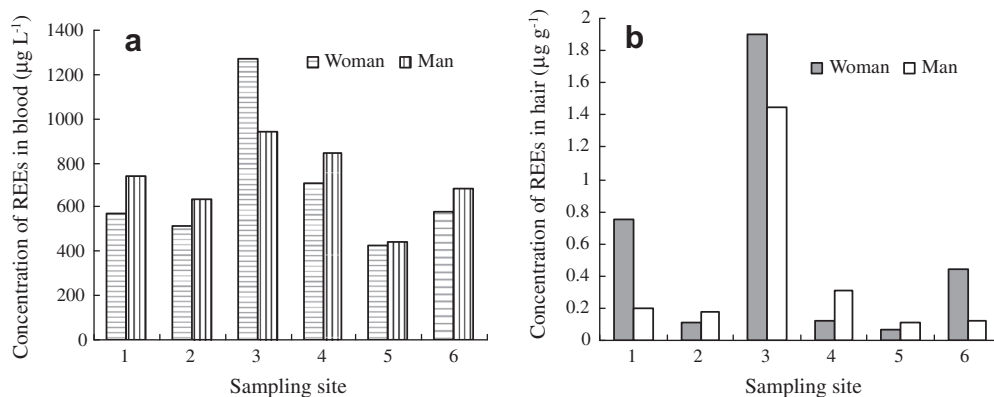
### 3.3. Concentration levels of REEs in human blood and hair

The concentrations of REEs in human blood and hair ranged from 424.76 to 1274.80  $\mu\text{g L}^{-1}$  with mean value of  $689.74 \pm 254.25 \mu\text{g L}^{-1}$  and ranged from 0.06 to 1.89  $\mu\text{g g}^{-1}$  with mean value of  $0.48 \pm 0.59 \mu\text{g g}^{-1}$ , 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  $\mu\text{g L}^{-1}$  and 1.67  $\mu\text{g g}^{-1}$ . This



**Table 2**  
Concentrations of REEs in different vegetable species in present study.

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



**Fig. 4.** Concentrations of total REEs in local human blood (a) and hair (b) 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.

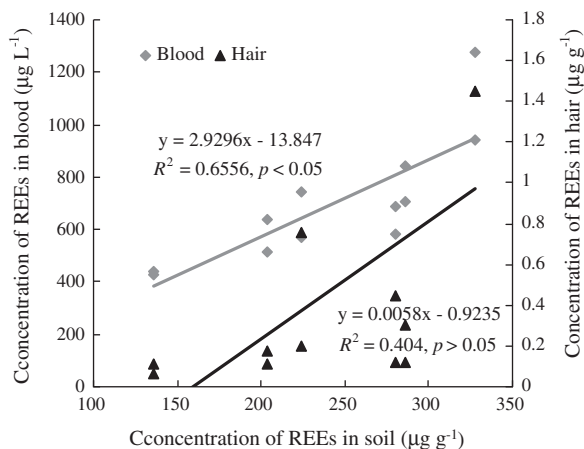
Concentration levels of REEs in local residents' blood samples were markedly higher than those of normal human blood samples

from Changchun Blood Center, northeast China, which ranged from 1.40 to 13.30  $\mu\text{g L}^{-1}$  with an average of 4.07  $\mu\text{g L}^{-1}$  (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  $\mu\text{g g}^{-1}$  with mean value of  $4.73 \pm 2.43 \mu\text{g g}^{-1}$  (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 ( $100\text{--}110 \mu\text{g kg}^{-1} \text{d}^{-1}$ ) (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



**Fig. 5.** The relationships between concentrations of REEs in human blood and hair samples and corresponding soils.

**Table 3**Estimated daily intake (EDI) of total REEs and bioaccessible REEs via vegetables and water consumption for adults and children in Fujian Province ( $\mu\text{g kg}^{-1} \text{d}^{-1}$ ).

Vegetable species	Adult	Adult <sub>bio</sub>	Children	Children <sub>bio</sub>
Chinese white cabbage	(0.0517–0.2418, 0.1567)	(0.0238–0.1112, 0.0721)	(0.0462–0.2159, 0.1399)	(0.0212–0.0993, 0.0644)
Water spinach	(1.0944–14.8928, 4.8683)	(0.5034–6.8507, 2.2394)	(0.9772–13.2982, 4.347)	(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	(0.0636–0.8367, 0.3730)	(0.0293–0.3849, 0.1716)	(0.0568–0.7471, 0.3331)	(0.0261–0.3437, 0.1532)
Eggplant	(0.0541–0.2139, 0.1607)	(0.0249–0.0984, 0.0739)	(0.0483–0.1910, 0.1435)	(0.0222–0.0879, 0.0660)
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.

consumption of any of the sampled vegetable species should not result in an EDI of REEs value exceeding  $100\text{--}110 \mu\text{g kg}^{-1} \text{d}^{-1}$  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 health ( $100\text{--}110 \mu\text{g kg}^{-1} \text{d}^{-1}$ ) for both adults and children, but the long-time impacts of REEs on human beings should be taken into account. This is a preliminary study on REEs exposure in mining site located in Southeast China, and epidemiologic investigations with a large number of exposure participants are warranted to evaluate the subsequent health effect of exposure to REEs.

#### Acknowledgments

We are very grateful to the anonymous reviewers and editors for their constructive comments and suggestions which improve our manuscript greatly. This work was financially supported by the National Natural Science Foundation of China (Nos. 41171232; 41001170; 40871141) and State Key Laboratory Breeding Base of Humid Subtropical Mountain Ecology (Fujian Normal University). Many heartfelt thanks are given to the enthusiastic help of the staff of Soil and Water Conservation Bureau of Changting County, Fujian, China. We also thank Prof. Caisheng Liao (College of Foreign Languages, Fujian Normal University) for helping us with the improvement of the language.

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