Investigation of rare earth elements in urine and drinking water of children in mining area

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Abstract
To compare the contents of rare earth elements in urine and drinking water of children in the mining and control areas and evaluate the health risk of children in the mining area.

Urine and drinking water of 128 children in the mining area and 125 children in the control area were collected from June to July 2015. The contents of rare earth elements were determined using inductively coupled plasma mass spectrometry.

The detection rates of rare earth elements, including yttrium (Y), lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), and samarium (Sm), in the urine of children in the exposed group were all 100%, except for samarium (98%); the rates in the control group were 85.7%, 100%, 100%, 98%, 98%, and 59.2%, respectively, and the remaining elements were not detectable. The concentrations of Y, La, Ce, Pr, Nd, and Sm in the urine of children in the exposed group were significantly higher than that in the control group (P < .01). In addition, the composition ratio of lanthanum was higher than that in the control group. The detection rates of lanthanum and Ce in the urine of children in the exposed group were 1.44% and 0.72%, respectively. The others were not detectable; the rates in the control group were all 0%.

The pollution caused by the presence of Y, La, Ce, Pr, Nd, and Sm in the mining area might affect the health of children in the area, but drinking water might not be the cause.

Abbreviations: Ce = cerium, Dy = dysprosium, Er = erbium, Eu = europium, Gd = gadolinium, Ho = holmium, ICP-MS = inductively coupled plasma mass spectrometry, La = lanthanum, Lu = lutetium, Nd = neodymium, Pm = promethium, Pr = praseodymium, REE = rare earth elements, RF = radiofrequency, Sc = scandium, Sm = samarium, Tb = terbium, Tm = thulium, Y = yttrium, Yb = ytterbium.

Keywords: children, drinking water, rare earth elements, rare earth mining area, urine samples

1. Introduction
Rare earth elements (REEs) have a total of 17 elements, including 15 lanthanide elements in the periodic table group IIIB, with atomic numbers ranging from 57 to 71. These are lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), samarium (Sm), promethium (Pm), europium (Eu), gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), ytterbium (Yb), lutetium (Lu), scandium (Sc), and yttrium (Y), having similar electronic structures and chemical properties. REEs have been widely used recently as superconducting materials, fluorescent materials, and clinical diagnostic tracers in high technology fields such as industry, national defense, and medical science because of their unique physical and chemical properties.[11]

The increase in residual REEs and pollution caused by them in the environment has led to REE enrichment in the human body through the food chain, causing harm.[2] Recent and long-term potential impacts of REEs on human health have attracted much attention. Excessive amounts of REEs could cause acute and chronic toxicity and damage to the nervous and reproductive systems. Hua et al[3] evaluated the acute toxic effects of lanthanum on liver injury and observed blood sinus dilatation, focal congestion, pyknosis, dissolution, vacuolar degeneration, and necrosis of the nucleus in the livers of the exposed group, indicating a damaging effect of REE on liver cells. Zhang et al[4] evaluated the reproductive toxicity of Sm in testicular lesions of male ICR mice given a continuous oral administration of Sm nitrate for 90 days. They found that the testicular tissue and organ coefficients in the treatment group decreased with decreased cytoplasm and decreased testicular spermatogenic cells and testicular sperm, indicating that Sm targeted the testis. Furthermore, electron microscopy confirmed that the lesions of spermatogenic cells and sperms included mainly mitochondrial swelling, mitochondrial vacuolization, and unclear nuclear membrane. The apoptotic rate of spermatogenic cells was found
to increase using the terminal deoxynucleotidyl transferase dUTP nick end labeling (TUNEL) assay and was related to the p53-mediated signaling pathway. Feng et al. showed that lanthanum poisoning could lead to the imbalance of trace elements, enzymes, and neurotransmitter systems in the brain and impair learning ability in rats. Moreover, REEs could affect the normal functioning of neurons by causing intracellular calcium overload, oxidative damage, and abnormalities of AMPA receptor channel functioning of neurons by causing intracellular calcium overload, oxidative damage, and abnormalities of AMPA receptor channel functioning.

The Bayan Obo ore deposit in Baotou in the province of Inner Mongolia has a history of 55 years. It is a world-famous rare earth symbiotic deposit where 16 kinds of rare earth minerals have been found recently. The increase in the status of REEs in the world has led to an increase in the quantity of REEs produced in the Bayan Obo ore deposit area, making it a key pollution source and destruction area of ecological environment. Moreover, its environmental problems have attracted huge attention.

This study aimed to comprehensively evaluate the pollution of REEs in drinking water in the Bayan Obo ore deposit area and its influence on population health by determining the content of REEs in drinking water and urine in children in this area. It provided a scientific basis for revealing the pollution characteristics and pollution control in the Bayan Obo ore deposit area. We have previously reported the reference urinary values of REEs in children of the Bayan Obo ore deposit area and the urinary values of REEs in children of the urban Baotou area. The present study compared the 2 populations (children from mining vs. control area) and examined the drinking water as a potential source of contamination.

2. Methods

2.1. Subject selection

The Bayan Obo mining area is under the jurisdiction of Baotou city of the Inner Mongolia Autonomous Region, located in the north of Yinshan Mountain, and its geographic coordinates are east longitude 109°47′ to 110°04′ and north latitude 41°39′ to 41°53′. It is 149 km north of Baotou, 95 km south of the Sino-Mongolian border, and 42 km west of Bailingmiao, where the Damao County Government is located, and has an area of 328.64 km². Two primary schools in the Bayan Obo mining area in Inner Mongolia and 2 primary schools that were far away from the 2 pollution centers (Baotou tailings dam and Bayan Obo mining area) in Baotou city were selected by simple random sampling method (lottery) from June to July 2015. Children aged 6 to 13 years, with an equal number of boys and girls, were randomly selected from each grade in each school. The inclusion criteria of the subjects were as follows: students had urine samples and complete information, lived as local residents for more than 1 year, were aged 6 to 13 years, had no abnormalities in recent routine physical examination or obvious liver or kidney disease, and had creatinine concentration in the normal range (0.3–3.0 g/L). A total of 128 students based on the aforementioned inclusion criteria were selected as the exposed group, and a total of 125 students from the non-polluted area were selected as the control group. No significant difference was found between the 2 groups regarding cultural level, age composition, gender composition, and living habits.

The study was approved by the ethics committee of Baotou Medical College of Inner Mongolia University of Science and Technology. The researchers explained the purpose of this study to the subjects and obtained guardians’ consent.

2.2. Data collection

The investigators explained the objective of the study to the guardians of the children and obtained their informed consent. Then, the investigators instructed the guardian to complete the survey (face to face) and checked the completeness of the response. For subjects who could not provide key information regarding the general characteristics for any reason, water and urine samples were not collected. In this study, all approached guardians agreed to participate. The effective response rate (i.e., responses usable for statistical analysis) was 92.7%. The response of the survey was recorded into digital format by two independent investigators and the consistency was checked.

2.3. Equipment and reagents

A TCHS-RO/40A pure water machine was purchased from Hangzhou Tianchuang Water Purification Equipment Co., Ltd, and a NexION 300Q inductively coupled plasma mass spectrometer was purchased from Perkin Elmer/Sciex. The inductively coupled plasma mass spectrometry (ICP-MS) operation parameters were set as follows: 1100 W of radiofrequency (RF) power, 171 L/min of plasma gas flow rate, 1.2 L/min of auxiliary gas flow (Ar), 0.80 L/min of atomized gas flow (Ar), 1 × 10⁻⁷ torr of vacuum degree, 1.1 mm of aperture diameter of nickel sampling cone, 0.9 mm of aperture diameter of nickel intercept cone, (0.70 ± 0.1) amu of resolution (10% peak height), peak skip as measuring mode, 1 as the measuring point/peak, 10 times of scan number, 50 ms/time of the residence time, 2 times of the repeat number, and 1.2 mL/min of sample lifting amount. The instrument had a grounding resistance of <4 Ω, an ambient temperature of 25°C ± 5°C, and a relative humidity of 20% to 80% (see Table 1 for details).

**Table 1**

| Parameter                          | Value                     |
|------------------------------------|---------------------------|
| RF power                           | 1100 W                    |
| Wash time                          | 20 s                      |
| Read delay                         | 5 s                       |
| Plasma Ar flow                     | 17 L/min                  |
| Aux. Ar flow                       | 1.2 L/min                 |
| Nebulizer Ar flow                  | 0.8 L/min                 |
| Sampler/skimmer                    | Nickel                    |
| Sample depth                       | 8 mm                      |
| Measuring mode                     | STD                       |
| 140Ce/160O/204Ce                   | <2.0%                     |
| Scan mode                          | Peak hop transient        |
| Points per peak                    | 1                         |
| Dwell time per isotope             | 50 ms                     |
| Readings per replicate             | 2                         |
| Number of replicate                | 2                         |
| Resolution (10% peak height)       | (0.7 ± 0.1) amu            |
| Signal measurement mode            | Dual-mode detector        |
| Sample volume                      | 1.2 mL/min                |
| Vacuum degree                      | 1 × 10⁻⁷ torr              |
| Sampler cone diameter              | 1.1 mm                    |
| Skimmer cone aperture              | 0.9 mm                    |
| Grounding resistance               | <4 Ω                      |
| Ambient temperature                | 25°C ± 5°C                 |
| Relative humidity                  | 20%–80%                   |

ICP-MS = inductively coupled plasma mass spectrometry.
Cesium internal standard reserve solution (1000 μg/mL; National Research Center for Certified Reference Materials) and single standard reserve solutions of Sc, La, Ce, Pr, Nd, Sm, Pm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, and Y (1000 μg/mL; National Research Center for Certified Reference Materials) were used. All reagents were guaranteed reagents (gr.). Mixed standard reserve solution was prepared as follows: 1.00 mL of single standard reserve solutions of Sc, La, Ce, Pr, Nd, Sm, Pm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, and Y were put into a 1000-mL volumetric flask and diluted to the scale with 1% HNO₃. Furthermore, 1 mL of this solution contained 1.00 μg of Sc, La, Ce, Pr, Nd, Sm, Pm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, and Y (1000 μg/mL; National Research Center for Certified Reference Materials) in uniform conditions. The absorbance of mixed standard series solution was measured using ICP-MS, and the standard curve was drawn. The contents of REEs in water, transportation blank, and parallel operation blank were determined under the same determination condition of standard solution.

2.6. Statistical analysis

Statistical analysis was performed using SPSS 19.0 statistics software (IBM). Categorical data were expressed as number and percentage and compared using the chi-square test or Fisher exact test, as appropriate. For continuous data, the Z test was used for the normality test. The total amount of REEs by daily intake of drinking water and the content of REEs in urine in children in the Bayan Obo mining area were expressed as means ± standard deviation. The rank-sum test was used for comparing between different regions and genders (2 independent-samples t test was used for the contents of REEs in urine, which were in logarithmic normal distribution; rank-sum test was used for the contents of REEs in drinking water; the data under the detection limit were uniformly calculated using half of the detection limit). A P value <.05 was considered as the statistically significant difference.

3. Results

3.1. General characteristics of the study subjects

Table 3 presents the general characteristics of the subjects. There were no differences between the 2 groups regarding age, education level, and gender. Compared with the control group, the exposed group presented higher frequencies of exposure to REEs in the residential environment (P < .001), parents working in REE factories (P < .001), drinking tap water (P < .001), parents working in REE factories (P < .001), and mothers exposed to REEs during pregnancy (P = .001).

3.2. Determination of REEs in urine

The detection rates of REEs, including Y, La, Ce, Pr, and Nd, in the urine of children in the exposed group in the mining area were

| Table 2 | Multielementary analytical validation (ng/mL). |
|--------|-----------------------------------------------|
| Element | r     | LOD  | LOQ  | RSD%  |
| 138Ce  | 0.9386 | 0.0787 | 0.2625 | 2.78  |
| 139La  | 0.9998 | 0.0859 | 0.1903 | 4.55  |
| 140Nd  | 0.9999 | 0.0814 | 0.2714 | 0.617 |
| 141Pr  | 0.9999 | 0.0946 | 0.3154 | 3.08  |
| 144Nd  | 0.9999 | 0.0857 | 0.2858 | 0.546 |
| 148Gd  | 0.9999 | 0.0864 | 0.2877 | 7.50  |
| 150Tb  | 0.9999 | 0.0862 | 0.2873 | 3.35  |
| 151Sm  | 0.9999 | 0.0868 | 0.2892 | 1.78  |
| 152Eu  | 0.9998 | 0.0952 | 0.3174 | 4.87  |
| 153Gd  | 0.9998 | 0.0972 | 0.2905 | 5.25  |
| 155 Dy  | 0.9999 | 0.0873 | 0.2910 | 4.98  |
| 156Ho  | 0.9999 | 0.0875 | 0.2915 | 3.37  |
| 157Er  | 0.9998 | 0.0876 | 0.2919 | 6.80  |
| 158Nd  | 0.9997 | 0.0878 | 0.2907 | 7.80  |
| 159Tb  | 0.9999 | 0.0879 | 0.2901 | 5.44  |

| Table 3 | Baseline characteristics of the subjects selected based on the aforementioned inclusion criteria (n = 253). |
|---------|-------------------------------------------------------------|
| Characteristics | All (n = 253) | Exposure group (n = 125) | Control group (n = 128) | P |
| Age, years | 2 | 20 | 2 | 20 | .185 |
| Education level | | | | | .814 |
| Gender | | | | | .748 |

Data are expressed as number (%).
all 100%. The detection rate of Sm was 98%, while that of Sc, Pm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu were all 0%. The detection rates of REEs, Y, La and Ce, Pr and Nd, and Sm were 85.7%, 100%, 98%, and 59.2%, respectively, in the urine of children in the control group, while that of Sc, Pm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu were all 0%. The concentrations of Y, La, Ce, Pr, Nd, and Sm in the urine of the exposed group were all higher than that in the control group, and the difference was statistically significant ($P < .01$) (Table 4). The constituent ratios of REEs in the urine of children of the exposed and control groups are shown in Figure 1. Figure 1A shows that the contents of REEs in the urine of children of the exposed group were mainly La 44%, Ce 30%, Pr 7%, Nd 14%, Sm 3%, and Y 2%. These components accounted for nearly 100% of the total REEs, and the remaining elements accounted for only 0.42%. Figure 1B shows that the contents of REEs in the urine of children of the control group were La 21%, Ce 33%, Pr 10%, Nd 17%, Sm 2%, and Y 4%, while the proportion of other elements was as high as 13%.

### 3.3. Determination of REEs in drinking water

The detection rates of REEs were 1.44% and 0.72% for La and Ce, respectively, and that of Sc, Pr, Nd, Sm, Pm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, and Y in drinking water of children in the exposed group in the mining area were all 0%. The detection rates of all REEs in the control group were all 0%. The results of REE determination in drinking water showed no statistical difference between the 2 groups, and did not exceed the safe doses (Table 5).

### 4. Discussion

In this study, the contents of 17 REEs in urine and drinking water of children in the Bayan Obo mining area and control area were determined using ICP-MS. The concentrations of Y, La, Ce, Pr, Nd, and Sm in the urine of children of the exposed group were significantly higher than that in the control group. The proportion of REEs in the urine of the exposed and control groups was significantly different. The content of La in the urine of children of the exposed group (44%) was significantly higher than that in the control group (21%). In addition, the content of Sm in the exposed group (3%) was higher compared with that in the control group (2%), whereas the content of Ce, Pr, Nd, and Y

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**Table 4**

| Element | Control group (n=125) | Exposed group (n=128) | $P$ |
|---------|-----------------------|-----------------------|-----|
| $^{44}$Sc | 0.0363±1.8437 | 0.0307±1.8365 | .028 |
| $^{89}$Y | 0.1447±2.1852 | 2.4820±1.9156 | <.001 |
| $^{139}$La | 0.8969±1.9107 | 46.050±1.9391 | <.001 |
| $^{140}$Ce | 1.3725±1.9792 | 30.9813±1.9765 | <.001 |
| $^{141}$Pr | 0.8304±2.3367 | 7.2594±2.2187 | <.001 |
| $^{144}$Nd | 0.8502±2.2406 | 14.5412±2.1014 | <.001 |
| $^{150}$Pm | 0.0491±1.8425 | 0.0404±1.8365 | .024 |
| $^{152}$Sm | 0.0625±1.8941 | 2.6394±2.6804 | <.001 |
| $^{153}$Eu | 0.0480±1.8437 | 0.0406±1.8365 | .028 |
| $^{157}$Gd | 0.0483±1.8437 | 0.0408±1.8365 | .028 |
| $^{159}$Tb | 0.0530±1.8437 | 0.0448±1.8365 | .028 |
| $^{162}$Dy | 0.0486±1.8437 | 0.0410±1.8365 | .028 |
| $^{164}$Ho | 0.0486±1.8437 | 0.0411±1.8365 | .028 |
| $^{165}$Er | 0.0487±1.8437 | 0.0412±1.8365 | .028 |
| $^{167}$Tm | 0.0488±1.8437 | 0.0413±1.8365 | .028 |
| $^{168}$Yb | 0.0489±1.8437 | 0.0414±1.8365 | .028 |
| $^{171}$Lu | 0.0490±1.8437 | 0.0414±1.8365 | .028 |

REEs = rare earth elements.

**Table 5**

| Element | Control group (n=139) | Exposed group (n=130) | $P$ |
|---------|-----------------------|-----------------------|-----|
| $^{145}$Sc | 0.0326 | 0.0326 | |
| $^{89}$Y | 0.0394 | 0.0394 | |
| $^{139}$La | 0.0299 | 1.44 | 0.0299 |
| $^{140}$Ce | 0.0407 | 0.77 | 0.0407 |
| $^{141}$Pr | 0.0473 | 0.0473 | |
| $^{144}$Nd | 0.0429 | 0.0429 | |
| $^{146}$Sm | 0.0429 | 0.0429 | |
| $^{147}$Eu | 0.0431 | 0.0431 | |
| $^{150}$Gd | 0.0434 | 0.0434 | |
| $^{152}$Tb | 0.0476 | 0.0476 | |
| $^{153}$Er | 0.0436 | 0.0436 | |
| $^{154}$Tm | 0.0437 | 0.0437 | |
| $^{157}$Yb | 0.0438 | 0.0438 | |
| $^{159}$Lu | 0.0439 | 0.0439 | |
| $^{159}$Sm | 0.0440 | 0.0440 | |

REEs = rare earth elements.
in the exposed group was lower compared with that in the control group.

Recently, many reports on the acute and chronic toxicity of La, Ce, and Nd have been published. For example, Wang et al. treated mice with lanthanum nitrate via feeding water for 1 month, and observed accumulation of lanthanum in the body. The content of lanthanum in the liver of poisoned mice increased. In addition, the micronucleus rate of liver cells significantly increased and was proportional to the concentration of lanthanum nitrate solution. Moreover, in vitro tests showed that lanthanum nitrate could cut off the DNA strand and cause genetic damage. Cheng et al. demonstrated that mice continuously exposed to Ce chloride for 90 days showed cell apoptosis, oxidative stress, and space memory impairment in the hippocampus, and altered expression profiles of immune- and inflammatory response-related genes. Kumari et al. found that high-dose Ce oxide nanoparticles could induce DNA damage in peripheral blood leukocytes and micronuclei changes in hepatocytes, blood cells, and bone marrow cells. The activity of lactate dehydrogenase and the content of glutathione in the hepatocytes, blood cells, and bone marrow cells. The activity of lactate dehydrogenase and the content of glutathione in the hepatocytes, blood cells, and bone marrow cells. The activity of lactate dehydrogenase and the content of glutathione in the hepatocytes, blood cells, and bone marrow cells.

Wang et al. found that exposure to excessive amounts of yttrium chloride resulted in bronchogenic lung inflammation in animals through the gastrointestinal tract was significantly higher compared with that by adult animals, suggesting that juvenile animals might be more sensitive to the action of REEs. Moreover, the present study found that the 3 REEs with the relatively high constituent ratio in the urine of the exposed group in mining area were La > Ce > Nd. However, a previous study found it to be Ce > La > Nd. The reasons for the inconsistency might be different sample size and sampling error. Recently, the mode of entry of REEs into the human body in an ion adsorption–type rare earth mining area in the south of China was mainly discussed. The mining area with complex deposit type, such as Bayan Obo, was not a major concern. Because of the different types of deposits, the existence of REEs was different, leading to the absorption of REEs through the environment. In addition, the mode of entry of REEs into the human body and risk factors were different. The experimental data showed that the contents of lanthanum, Ce, Nd, Pr, Sm, and Y in the urine of children in the exposed group in the Bayan Obo mining area were all higher than those in the control area. REEs could enter the human body through soil, atmosphere, water, food, and other routes, and these factors might be the risk factors for rare earth exposure in children.

This study, through the field investigation of Bayan Obo mining area, found that the local soil was not suitable for planting crops. The local population feeds mainly on food supplied from other districts of Baotou, but the drinking water sources are located in the vicinity of the Bayan Obo mining area, and the main drinking water source of the residents is groundwater. Drinking water might be 1 of the rare element exposure pathways in this area. Therefore, this study discussed mainly the health effects of drinking water exposure on the children in the mining area. It provided the theoretical and scientific bases for the health protection of children in the mining area and the safety evaluation of REEs. Nevertheless, no significant difference was found between the 2 groups in the determination of REEs in drinking water. The study could not conclude that exposure to REEs in drinking water was a risk to the health of local children.

This study had some limitations. As the residential area was in the downwind direction of the Bayan Obo mining area, the rare earth element dust in the air of the mining area flowed with the predominant wind to the residential area and fell into the soil of the area. It furthermore entered the daily diet and caused accumulation of REEs in children of that area. Nevertheless, only the content of REEs in drinking water of children in the exposed and control groups was determined and compared in this study. Therefore, the contents of REEs in soil and air should also be measured in future studies to determine the main possible sources of pollution. Moreover, rational planning of residential and mining areas according to the predominant wind direction might greatly reduce the level of rare earth exposure of the residents in the mining area. Selection biases could be present only in children with legal guardians who could not provide complete information were excluded. Of course, rare earth elements are present in very small amounts and any deviation from the technical study guidelines could result in measurement imprecision.

The results showed that the pollution of REEs such as Y, La, Ce, Pr, Nd, and Sm might affect the health of children in the Bayan Obo mining area, but it could not be concluded that drinking water was the cause.

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