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Research paper

The effect of elemental content on the risk of dental fluorosis and the exposure of the environment and population to fluoride produced by coal-burning

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ABSTRACT

Endemic fluorosis is a geochemical disease that affects thousands of people. Growing evidence from domestic and foreign studies indicate that fluorosis is associated with an abnormal level of the elements (such as F, Ca, Fe, Mg, Cu, Zn, P) in the environment and a population exposed to fluoride. To study the effect of the elemental content on the risk of dental fluorosis, the content of 25 elements in the environment produced by coal-burning and a population exposed to fluoride was determined. The results show that an abnormal level of various elements (including F, Al, Se, Zn, Cu, Fe, Mo, Mn, B, V, Ca, Mg, and P) in the population exposed to fluoride, which is related to the increasing or decreasing of the corresponding elements in the environment. Subsequent univariate and multivariate regression analyses show that high levels of F, Al, As, Pb and Cr were a risk factor for dental fluorosis, but not Se, Zn, Cu, B, Ca and P which were a protective factor for dental fluorosis. This study can provide a scientific basis for a further understanding of the causes of health damage caused by fluoride and the improvement of targeted prevention strategies.

1. Introduction

Endemic fluorosis is a worldwide disease that causes a wide range of human health issues. It is also a key prevention and control disease of the “11th Five-Year Plan”, the “12th Five-Year Plan”, the “13th Five-Year Plan”, the “Healthy China 2020 Action Plan” and the “Healthy China 2030 Action Plan” in China. It is characterized by skeletal and dental fluorosis (Sun et al., 2017). Depending on the source of fluoride, endemic fluorosis can be divided into three types originating from drinking water (Mandinic et al., 2010), tea drinking (Fan et al., 2016) and coal burning (Ando et al., 1998), and the last type is unique to China. Guizhou Province is the most prevalent region in China were fluorosis produced due to the burning of coal exists, and it has the largest and most concentrated population. Past data (Li et al., 2005) show that 19 million people were exposed to fluoride, of which approximately 1.2 million had skeletal fluorosis, and approximately 10 million people had dental fluorosis. The etiology of the disease is currently clear, but the pathogenesis is unknown. Therefore, in-depth research on the pathogenesis of endemic fluorosis must be carried out to achieve the goal of the " eliminating of the coal-burning type of fluoride poisoning” in China.

An increasing number of researchers have recently begun to focus on the impact of the elements on human health. The contents of various elements in the human body is very different, and can generally be divided into constant elements and trace elements. A constant element is a main component of the human body or a structural element, including Ca, K, Mg, Na, P, S, etc. Opposed to constant elements, the content of the other elements in the body is very low, and these are known as trace elements whose contents are less than 0.01% of body weight. The World Health Organization (WHO) (Organization and FAO, 1996) classifies trace elements into three categories, including essential trace elements, such as Zn, Se, Cu, Fe, Mo, Cr and Co, probably essential trace elements, such as Mn, Si, B, Ni and V, and potentially toxic elements, such as F, Al, As, Cd, Hg, Li and Pb, which also may have some essential functions when present at low levels. A retrospective study (Zofkova et al., 2017) found that integrity of the skeleton is positively affected by certain trace elements (e.g., Zn, Cu, Mn, Mg, Fe, Se, B and F) and negatively by others (Pb, Cd, Co). In China, a study on drinking

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water fluorosis (Chen et al., 2002) found that the content of Ca, Mg, Cu in the environment samples (i.e., food and vegetables) and the biological sample (i.e., the serum of patients with skeletal fluorosis) was low, which suggested that the low levels of Ca, Mg and Cu in the body may be related to a lack of those elements in the environment. Endemic fluorosis is a typical bio-geochemical disease; therefore, it is closely related to the geological environment in a geographical area. Therefore, it is important to understand the levels of the elements in the environment, to evaluate the environmental effects on the health of a fluoride-exposed population.

In addition to the distribution of the elements in the environment, the content of elements in the body of a fluorosis patient may also be related to the metabolism disorder of the elements. Metabolic disorders of Ca, Fe, Zn were observed in children with dental fluorosis that arose from coal burning (He et al., 2008). Another study (Shi et al., 2011) found that there were differences in the distribution of Ca, Fe, Mg, P, Zn in children exposed to varying amounts of fluoride. The study of the elemental content in a fluorosis area and an exposed population is currently focused on a single or a few elements. Moreover, very little is known about the link between the elemental content and the risk of fluorosis.

In this study, we measured the content of 25 elements [fluorine (F), aluminum (Al), arsenic (As), cadmium (Cd), mercury (Hg), lithium (Li), lead (Pb), selenium (Se), zinc (Zn), copper (Cu), iron (Fe), molybdenum (Mo), chromium (Cr), cobalt (Co), manganese (Mn), silicon (Si), boron (B), nickel (Ni), vanadium (V), calcium (Ca), potassium (K), magnesium (Mg), sodium (Na), phosphorus (P), sulfur (S)] in an environment and a population exposed to fluoride via coal burning, and analyzed the relationship between the elemental content and dental fluorosis, with the aim of studying the effect of the elemental content on the risk of dental fluorosis and the exposure of the environment and population to fluoride via coal burning. The study can provide a scientific basis for a further understanding of the causes of the health damage caused by fluoride and the improvement of targeted prevention strategies.

2. Materials and methods

2.1. Selection of investigation site

In this study, the selected investigation site was located in Liuchang village, Qinzhen City, Guizhou Province, China, which is a proven to be an endemic area for fluorosis that arises from coal-burning. A fluoride-free area in the suburbs of Qingzhen City was chosen as a control site. The eating habits, economic status and nutritional status of the control area were similar to the fluoride exposure area.

2.2. Selection and grouping of objects

The proposal for this population-based study was reviewed and approved by the Ethical Committee of Guiyang Medical University. Written, informed consent was obtained from all participants. We worked with the Guiyang Centers for Disease Control and Prevention to recruit volunteers. A total of 191 fluoride-exposed villagers were selected by a cluster sampling method (89 males and 102 females, with an average age 51.73 ± 15.29). All participants were required to be permanent residents of the local area(Liuchang village)and were matched for age and sex. The exclusion criteria included smoking, drinking, pregnancy, a fluoride-related occupational history and a recent history of consuming food or drugs that could affect bone metabolism, or the urinary excretion of fluoride. According to the standard “Diagnosis of dental fluorosis”(WS/T 208, 2011, China), the subjects are divided into normal or dental fluorosis. Because dental fluorosis is manifested early in cases of fluoride poisoning, dental fluorosis can be used as a biomarker of fluoride poisoning.

The American Conference of Governmental Industrial Hygienists (ACGIH) biological exposure index is 2 mg/L for fluoride in the urine. In the survey area, the average creatinine in the urine was 1.7 mg/L in the normal population. After being normalized to the average of creatinine (1.7 mg/L), the biological exposure index for fluoride in the urine is approximately 1.2 mg/g creatinine (mg/g Cr). According to the actual detection range in the exposed population, intervals of 0.5 times the biological exposure index were used to define four groups according to fluoride urine content, including a control (< 1.2 mg/g Cr), low (1.2 ~ mg/g Cr), medium (1.8 ~ mg/g Cr) and high (> 2.4 mg/g Cr) fluoride exposure group.

2.3. Collection of samples

2.3.1. Collection of environmental samples

The plum blossom method was used according to the location, terrain characteristics and the incidence of fluorosis, 20 fluorine exposed sampling points and 5 control sampling points were selected. The samples included coal, clay, rice, corn and chilis. The collection of samples followed the principles of representativeness, consistency, sufficient quantity and no pollution. Clay and coal were sampled from multiple locations and then mixed evenly, finally, a sample of approximately 1 kg was formed by the quartation method. Clay was collected 5 cm below the surface, and coal was collected below the surface weathering layer. Grain (rice, corn and chilis) was sampled using a stratified sampling method, and then mixed evenly, finally, a sample of approximately 1 kg was formed by the quartation method.

2.3.2. Collection of hair

The hairs of the subjects were collected, close to the hair root within 3 cm, dry preservation until analysis.

2.3.3. Collection of urine

Polyethylene plastic bottles are soaked with 10% nitric acid overnight, washed with deionized water and dried. After obtaining informed consent, 10 0 ml morning urine samples were collected in the processed polyethylene plastic bottles. All participants were instructed in a method that would avoid contamination. Each urine sample is divided into two aliquots, an untreated sample for the determination of urinary fluoride and urinary creatinine, (urinary fluoride was normalized to the urine creatinine content) and 0. 5 ml of concentrated hydrochloric acid with a pH value less than 2 was added to 50 ml of the other urine sample for the elemental determination. Urine samples were transported at 4 °C, and stored at −80 °C until analysis.

2.3.4. Collection of blood

After obtaining informed consent, 2 ml of non-anticoagulated blood was collected. The serum was separated for the measurement of the elemental content except for fluorine. Serum samples were transported at 4 °C, and stored at −80 °C until analysis.

2.4. Research content and detection methods

2.4.1. Chemical reagents and standards

Nitric acid (GR), hydrogen peroxide (GR), hydrofluoric acid (AR), acetone (AR), ether (AR) and ethanol (AR) were purchased from Wuhan Boster Biological Technology Co., Ltd, China. All standard stock solutions [GBW(E)080549 F, GBW(E)080215 Se, GBW(E)080216 V, GBW(E)080217 B, GBW(E)0802183 Mo, GBW(E)080219 Al, GBW(E)080272 Si, GBW(E)080547 Li, GBW08611 As, GBW08612 Cd, GBW08613 Co, GBW08614 Cr, GBW08615 Cu, GBW08616 Fe, GBW08617 Hg, GBW08618 Ni, GBW08619 Pb, GBW08620 Zn, GBW(E)080118 Ca, GBW(E)080125 K, GBW(E)080126 Mg, GBW(E)080127 Na, GBW(E)080157 Mn, GBW3180 S and GBW(E)080431 P] are provided by the National Center for Standard Material Research in China.

2.4.2. Pre-treatment of the samples

All environmental samples are ground and dried and passed through