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ENVIRONMENTAL EPIDEMIC CHARACTERISTICS OF COAL-BURNING ENDEMIC FLUOROSIS AND THE SAFETY THRESHOLD OF COAL FLUORIDE IN CHINA

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SUMMARY: Data on coal-burning endemic fluorosis throughout China and on the exposure-response relationship between concentrations of fluoride determined in coal samples and the prevalence of dental fluorosis reported from 17 representative surveillance stations in Southwest China were used to estimate the safety threshold for coal fluoride. Coal-burning endemic fluorosis occurs mainly in the mountainous areas of this part of China, where the prevalence of the disease is closely linked to geochemical parameters of the local environment. In these regions the incidence of dental fluorosis has a significant positive correlation with the concentration of fluoride in coal. The safety threshold of coal fluoride is estimated to be 190 mg/kg by the criterion of 0% incidence of dental fluorosis.

Keywords: China; Coal fluoride; Endemic fluorosis; Safety threshold.

INTRODUCTION

Fluorine (F), the most electronegative and reactive of the halogens, is a common chemical element in the earth's crust in combined form. F concentrations in rocks and soils are well documented, but data on the F concentration in coal are relatively limited.[1-4] Swaine reported the total F concentration in coal ranges from 20 to 500 mg/kg.[5] Statistical data indicate that the mean concentration of F in coal worldwide is 80 mg/kg, but in China it is 200 mg/kg.[6] In the mountainous areas of Southwest China, it is even higher — up to 3106 mg/kg in local coal.[7]

Fluoride in coal can be released into the ambient environment as atmospheric F, waterborne F, and residue F during mining, handling, and combustion.[6-8] In Southwest China, F from coal combustion has polluted the atmosphere and has impacted negatively on the temporal and geographical distribution of terrestrial vegetation.[9] People living in these areas are affected by chronic endemic fluorosis due to the excessive uptake of F from air or food dried by coal-burning.[6,10,11]

Fluorosis is a prevalent endemic disease in many parts of the world, caused mainly by high F in drinking water, whereas coal-burning endemic fluorosis appears to be known only in China.[11] In the year 2000, over 18.13 million people in China were identified as having fluorosis caused by F from coal burning, including 16.67 million with dental fluorosis and 1.46 million also afflicted with skeletal fluorosis from this source.[12] Coal-burning fluorosis has received widespread attention from the Chinese government, and in recent years a number of investigations have focused on the environmental

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pathogenesis of the disease.[13-15] Many of these studies showed that the affected areas were always concomitant with heavy coal-burning regions and that the incidence of fluorosis increased with coal F concentration.[13,14] Because of the dispersed nature of the primary data, a comprehensive examination of the dose-response relationship between coal F concentration and the incidence of coal-burning fluorosis has not previously been undertaken.

Our objective was to investigate the environmental epidemic characteristics of coal-burning endemic fluorosis and to determine the relationship between the incidence of fluorosis and coal F concentration in coal-burning endemic fluorosis regions in China using existing data plus our coal F analyses. This work may thus provide a guideline for identifying such regions as well as providing a scientific basis for an environmental risk assessment.

MATERIALS AND METHODS

Survey data on coal-burning endemic fluorosis were obtained mainly from two sources. The first source is the year 2000 annual report of endemic diseases in China compiled by the Ministry of Health, People's Republic of China.[12] From this report we extracted data on the occurrence of fluorosis throughout the country. The other source is reports from the endemic disease prevention and control management system. From them we collected survey data on the incidence of dental fluorosis and the corresponding coal F concentration from studies conducted at seventeen key surveillance stations located in Hubei, Guizhou, and Shanxi Province in Southwest China. The natural geographical characteristics of each station have been described.[13,14,16,17]

Dental fluorosis in these studies was diagnosed according to the criteria developed by the Chinese Ministry of Health.[18] In each surveillance station area, dental fluorosis was recorded in children 8 to 15 years old, who were native-born or were immigrants of more than three years and were never exposed to industrial fluoride. Overall, 77,238 children were examined in the 17 surveillance station areas.

Fluoride in coal samples from the 17 surveillance station areas (air-dried and passed through a 2-mm sieve) was determined by the fluoride ion-selective electrode method, essentially according to the method of Thomas and Gluskoter.[19] The procedure involves fusing 0.100 g of coal with 2.0 g of NaOH, dissolving the resulting cake in water, adjusting the pH to about 5.2 with HCl, and finally determining the F activity in the presence of TISAB buffer. Calibration was done with standards of known F concentration.

Graphical output and data processing were accomplished by GIS (ArcView GIS Version: 3.2a, ESRI Inc., 1992) and Excel, respectively.

RESULTS

Figure 1 shows the locations of coal-burning endemic fluorosis areas in China. As can be seen, the disease is found mainly in the mountainous regions of Southwest China, such as Hubei, Guizhou, Shanxi, and Sichuan

Province. In these areas, the climate is mainly moderate in temperature and humidity. The geomorphology consists of marine or coastal deposit limestone mountains and fluvial erosion mountains with a widely distributed Permian coal and Cambrian-Ordovician coal strata. Calcium bicarbonate is the dominant chemical constituent in the drinking water, which is characterized by low F concentrations (always below 1.0 mg/L).

Figure 1. Distribution of coal-burning endemic fluorosis in China.

The Table below summarizes data from our seventeen analyses of coal F concentration and the corresponding incidence of dental fluorosis in typical coal-burning endemic fluorosis areas in Southwest China. From the data in this table as plotted in Figure 2, we can see that the incidence of dental fluorosis increases with increasing F concentration in the coal: the higher the concentration, the more prevalent the disease. The correlation between the two variables can be expressed as $Y = 0.075X - 14.28$, where Y is the incidence of dental fluorosis (%), X is the F concentration in coal in mg/kg, and the correlation coefficient is 0.943 ($n = 17$, $P < 0.01$). Linear regression indicates a "safety threshold" for zero percent dental fluorosis at a concentration of 190 mg F/kg of coal. The regression also implies a 50% incidence of dental fluorosis at a concentration of 857 mg F/kg of coal. When the F concentration in coal rises to about 1524 mg/kg, almost every resident in the disease areas is afflicted with dental fluorosis.

Table. Incidence of dental fluorosis and the concentration of coal F in 17 surveillance station areas of Southwest China

Station	Population	Incidence of dental fluorosis %	Coal F^- concentration mg/kg
Libo	171,366	0	58
Luodian	293,994	0	186
Qianxinan	697,075	7.80	212
Pingtang	267,368	4.30	244
Dushan	317,910	16.54	534
Duyun	463,426	22.72	646
Fuquan	292,720	16.54	698
Wengan	390,245	22.00	725
Zhenping	53,719	66.05	867
Ankang	2,613,914	56.59	872
Langao	148,311	53.09	885
Changshun	226,235	92.20	1165
Ziyang	285,018	66.08	1172
Huishui	388,896	93.90	1194
Pingli	201,392	88.03	1282
Guiding	267,809	90.59	1392
Longli	192,436	96.40	1668

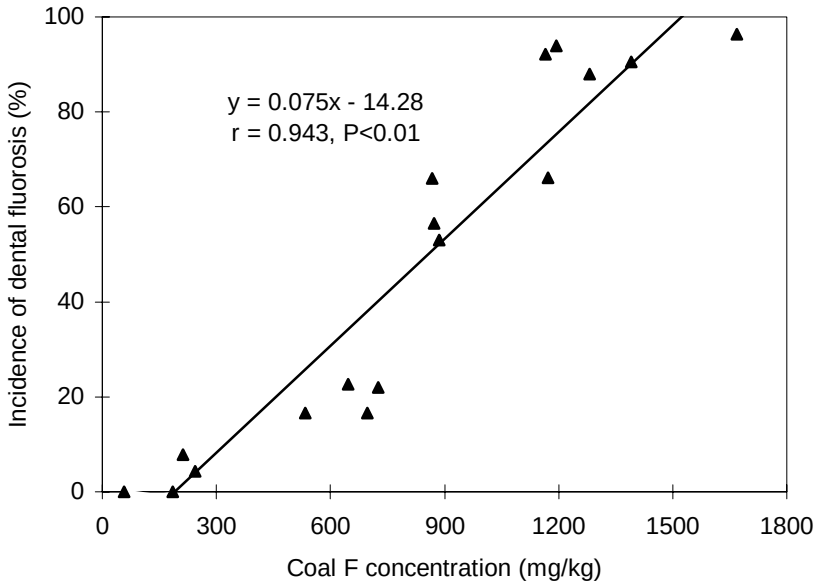

Figure 2. Correlation between the incidence of dental fluorosis and coal F concentration.

DISCUSSION

In the year 2000, the Chinese Ministry of Health reported that China had 16.67 million people in 201 counties who were afflicted with dental fluorosis caused by coal-burning pollution.[12] Among these counties, nearly eighty percent (158) and over ninety percent of the people (15.10 million) were located in Southwest China. The 17 station areas in this study are the key surveillance stations in the coal-burning endemic fluorosis regions of Southwest China. Their reports include the disease prevalence in non-affected areas and slightly affected areas as well as heavily affected areas. Their data reflect the general differences of F concentrations in coal which, in turn, correlate closely with the distribution of the disease in Southwest China, and they also reflect the overall situation of the disease in China.

Although the prevalence of fluorosis is closely linked to local natural environmental parameters, such as climate and geochemical background, the most important factor is the high F concentration in local coal.[8,11] Because of the wet climate in the mountainous areas of Southwest China, the high F containing coal produced locally is used as the major fuel for cooking, heating, and drying grain in chambers without chimneys. The annual mean coal consumption is generally as high as 3000 kg per family. During combustion, most of the F in the coal enters the atmosphere in the form of HF, SiF_4, etc.[7,20] In this way the indoor air and grain hung indoors are polluted by the coal F. Epidemiological investigations show that excessive intake of F from food and air is the primary pathway in coal-burning endemic fluorosis regions.[21-23]

Dental fluorosis is one of the major symptoms of coal-burning endemic fluorosis and has a highly significant correlation ($P < 0.01$) with coal F concentrations, thereby demonstrating that in regions of long-term coal pollution, coal F concentration can be used as a sensitive marker for environmental risk assessment of coal F exposure and thus as an indicator of the level of environmental contamination. Assessment of the prevalence and severity of coal-burning endemic fluorosis by this indicator provides an efficient large-scale monitoring method for environmental pollution by coal F at very low cost. Therefore, the "safety threshold" of 190 mg F/kg coal first determined here, provides a scientific basis for ascertaining coal-burning endemic fluorosis affected areas and potential threaten areas.

This safety threshold is relatively safe because, although the prevalence of coal-burning endemic fluorosis is also affected by other factors, such as coal exposure time and annual coal consumption, coal F concentration is the primary determinant. An investigation conducted in Jianshi County demonstrated that, besides the coal F concentration, the incidence of dental fluorosis was positively correlated with coal exposure time and coal consumption.[24] Moreover, dental fluorosis is a sensitive first-sign indicator of a

chronic endemic disease affecting almost the entire body. Fluoride can injure the cytoplasm, affect enzyme activity, and interfere with normal metabolic processes involving the teeth and the skeleton. Traditionally, emphasis has usually been placed on dental fluorosis and skeletal fluorosis because their clinical symptoms are recognizable. However, even before visible symptoms such as dental fluorosis and skeletal fluorosis appear, some normal metabolic processes associated with the nervous system and bodily organs are already affected.[25-27] Further comprehensive studies are therefore desirable to investigate these issues.

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