Health Effects of Indoor Fluoride Pollution from Coal Burning in China

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The combustion of high fluoride-content coal as an energy resource for heating, cooking, and food drying is a major exhaust emission source of suspended particulate matter and fluoride. High concentrations of these pollutants have been observed in indoor air of coal-burning families in some rural areas in China. Because airborne fluoride has serious toxicological properties, fluoride pollution in indoor air and the prevalence of fluorosis have been analyzed in a fluorosis area and a healthy nonfluorosis area in China and in a rural area in Japan. For human health, fluoride in indoor air has not only been directly inhaled by residents but also has been absorbed in stored food such as corn, chilies, and potatoes. In the fluorosis area in China, concentrations of urinary fluoride in the residents have been much higher than in the nonfluorosis area in China and in the rural area in Japan. In the fluorosis area, almost all elementary and junior high school students 10–15 years of age had dental fluorosis. Osteosclerosis in the skeletal fluorosis patients was very serious. Urinary deoxypyridinoline in rural residents in China was much higher than in rural residents in Japan. Data suggest that bone resorption was extremely stimulated in the residents in China and that fluoride may stimulate both bone resorption and bone formation. Because indoor fluoride from combustion of coal is easily absorbed in stored food and because food consumption is a main source of fluoride exposure, it is necessary to reduce airborne fluoride and food contamination to prevent serious fluorosis in China. *Key words*: air pollution, bone formation, bone resorption, coal burning, dental fluorosis, fluoride, indoor pollution, skeletal fluorosis. *Environ Health Perspect* 106:239–244 (1998). [Online 27 March 1998]
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Many combustion processes, automobile exhaust, and industrial exhaust are major sources of airborne pollutants (1–4). The combustion of coal is one of the major sources of indoor and outdoor air pollutants in the world (5,6).

Combustion exhaust contains gaseous pollutants and airborne particles, which consist of numerous chemical components (7–9). Many of these chemicals have potential toxicological significance (10–12). Therefore, it is necessary to quantify the human health risks associated with exposure to these pollutants such as suspended particulate matter (SPM) and airborne chemicals.

In China, coal, coal gas, and natural gas are used as domestic fuel energy sources. Combustion emissions, such as SPM and gaseous pollutants, are significantly different for the three kinds of fuels. The highest concentrations of SPM and gaseous pollutants have been observed in indoor air of coal-burning households (13).

In China, there are some environmentally caused diseases such as endemic fluorosis, Keshan disease (selenium deficiency), arsenic poisoning, and endemic goiter (iodine deficiency). Among them, airborne fluorosis and waterborne fluorosis are the most serious pollution-caused diseases. Both coal and the surrounding soil layer have high fluoride contents in some rural areas in China. Because high fluoride-content coal and soil mixture is used as a main fuel energy source, fluoride concentrations are significantly higher in fuel combustion exhaust emissions. As a result, high concentrations of fluoride have been detected in indoor air of coal burning families (14).

In some areas of the world, including the United States, water fluoridation has been used as an effective and inexpensive method for preventing dental caries (15). On the other hand, exposure to excess fluoride has caused dental and skeletal fluorosis in some areas of the world (16). In China, 43 million dental fluorosis cases and 2,370,000 skeletal fluorosis cases may have occurred because of exposure to fluoride emissions from high fluoride-content coal and from drinking contaminated water (17). Therefore, it is necessary to evaluate the risk of environmental fluoride exposure on human health in a well-designed and well-executed study.

Fluoride in indoor air is directly inhaled by residents, and airborne fluoride can easily be absorbed in stored food. Therefore, total exposure such as inhalation and ingestion of fluoride must be determined. Because high concentrations of airborne fluoride and a high prevalence of fluorosis have been observed in some rural areas in China, the potential exposure to fluoride has been a matter of great concern in China. In the present study, changes in the concentrations of SPM and fluoride in indoor/outdoor air and fluoride contamination in stored food were measured in rural areas in China.

Many reports have confirmed that fluoride stimulates bone formation (18–20). On the other hand, almost no confirmed or reliable relationships have been observed between fluoride exposure and the prevalence of bone fractures (21–25). It is generally accepted that the hydroxyapatite structure is a prototype for the structure of bone mineral, and fluoride ions substitute for the hydroxy ions giving rise to partially fluoridated hydroxyapatite or fluoroapatite. Bone also contains calcium ions and phosphate ions in the apatite lattice of mineral crystals (26). Therefore, it is necessary to survey bone formation and bone resorption as a result of absorption of fluoride, as well as urinary contents of fluoride, calcium, and phosphate.

In this study, the prevalence of dental fluorosis and skeletal fluorosis were examined in a fluorosis area and a nonfluorosis area in China. To determine the total exposure of fluoride, urinary fluoride in residents was determined in the fluorosis and nonfluorosis areas in China and in a rural area in Japan. Urinary deoxypyridinoline, a significant biochemical marker of bone resorption (27–29), urinary calcium, and phosphate were analyzed in the residents of the studied areas.

Materials and Methods

Sampling methods. To study changes in the concentrations of suspended particles in indoor/outdoor air, portable samplers were

used. The sampler was placed at a 1-m height in a kitchen–dining room near the indoor stove. The sampler could separately collect particles that had aerodynamic diameters of >10 µm, 2–10 µm, and <2 µm (4). Low volume samplers were also used to collect gaseous and particulate fluoride in indoor/outdoor air (14). To measure the instantaneous variation of suspended particles, portable digital dust monitors (PCD-1; Shibata Scientific Technology Ltd., Tokyo, Japan) were also used.

Study site. The studied areas were in Pengsui County, Sichuan Province (fluorosis area) and Zhaoxian County, Jiangxie Province (nonfluorosis area) in the southern part of China and in a rural area in Nagano Prefecture, Japan. The population included 2,984 males and 3,516 females for Pengsui County and 1,027 males and 1,055 females for Zhaoxian County. The main occupation of the residents was agricultural work, and their fuel energy source was coal. Measurements of SPM and fluoride pollution were carried out in farmhouses during the winter season of 1995 and 1996. Total intake of fluoride was calculated in farm families in the fluorosis and nonfluorosis areas in China.

Health surveillance. In Pengsui County, 213 students 10–15 years of age were examined for dental fluorosis and dental caries. In Zhaoxian County, 210 control students 10–15 years of age were also examined. Dental fluorosis is characterized by the macroscopic degree of dental fluorosis in relation to the histological abnormalities. The diagnostic classification categories are 0 (normal), 1 (very mild), 2 (mild), 3 (moderate), and 4 (severe) (15,16).

The general population health surveys of the residents were carried out in the clinics.

The prevalence of skeletal fluorosis was 42.10% for Pengsui County (fluorosis area) and 0% for Zhaoxian County (nonfluorosis area). In Pengsui County, 49 skeletal fluorosis patients 40–59 years of age were examined radiologically using an X-ray analyzer. The diagnostic categories of osteosclerosis are classified by an increase in the density of various bones, especially the vertebrae, the pelvis, the forearm, and the lower limb. In Zhaoxian County, 55 normal control residents 40–59 years of age were examined radiologically on the density of forearm bone and lower limb bone.

In Pengsui County, urine was collected between 900 and 1000 hr from 213 students, 49 skeletal fluorosis patients, and 30 residents 30–59 years of age. Urine was also collected in Zhaoxian County from 210 control students and 76 residents 30–59 years of age. To determine the total exposure of fluoride, urinary fluoride of the residents was determined in the fluorosis and nonfluorosis areas in China and in the rural area in Japan. Urinary deoxypyridinoline, calcium, and inorganic phosphate were analyzed in residents in the studied areas and in 74 healthy 30–59-year-old rural residents in Japan. Kidney function and respiratory function of students and residents were checked clinically.

Analytical methods. The filter (19 and 35 mm T60A20; Pallflex Products Corporation,

Putnam, CT) was washed completely with distilled water and the purest grade acetone to decrease background contamination and dried for 24 hr at 20°C and 50% relative humidity before use. At the end of each measurement, the filter of the sampler was removed, dried for 24 hr at 20°C and 50% relative humidity, weighed, and analyzed as follows.

Standard reference material [SRM-2671a; National Institute of Standards and Technology (NIST), Gaithersburg, MD] was used as an analytical standard for urinary fluoride of residents. Extracted fluoride in air and urinary fluoride were analyzed using a fluoride electrode (9609 BN, Model 720A; Orion Research Inc., Boston, MA). Fluoride in food was extracted by 0.25 M hydrochloric acid using an ultrasonic generator, trapped by microdiffusion, and analyzed using a fluoride electrode.

Urinary deoxypyridinoline, a significant biochemical marker of bone resorption, was analyzed using an enzyme-linked immunosolvent assay (ELISA) according to the manufacturer's instructions (Metra Biosystems Inc., Mountain View, CA). Urinary creatinine, calcium, inorganic phosphate, sodium, potassium, chloride, urea nitrogen, and uric acid were determined using an automatic analyzer system (Hitachi 7250, Hitachi Company Ltd., Hitachi, Japan) according to the manufacturer's instructions.
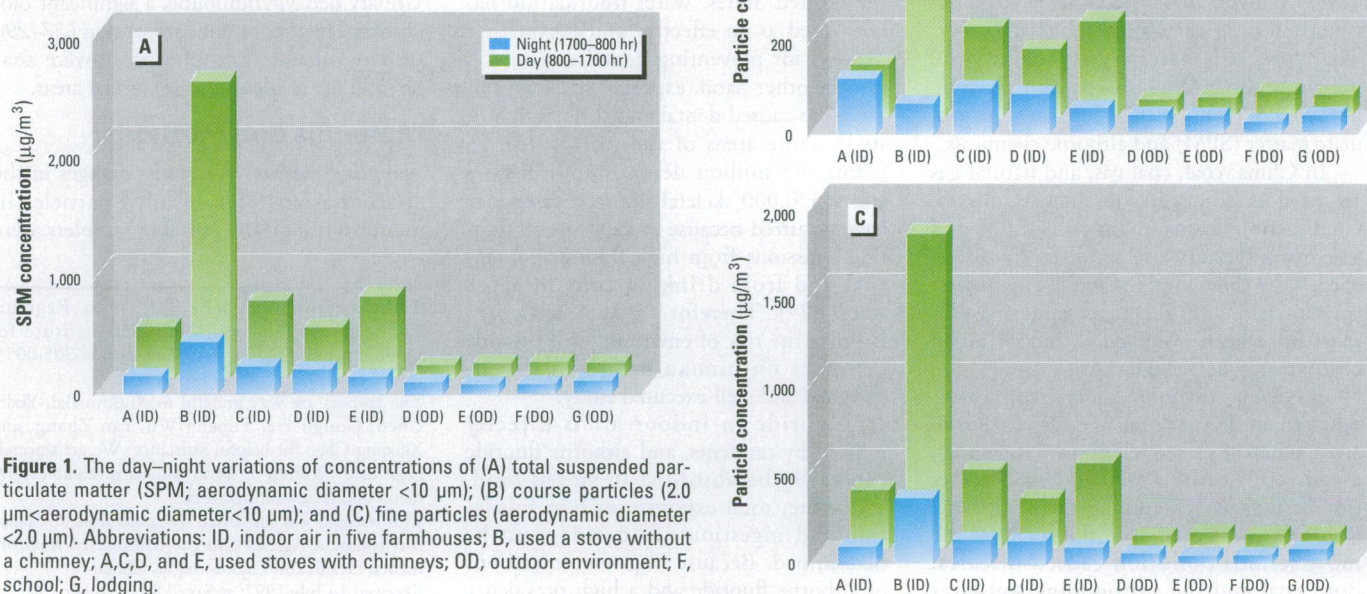

Figure 1. The day–night variations of concentrations of (A) total suspended particulate matter (SPM; aerodynamic diameter <10 µm); (B) course particles (2.0 µm<aerodynamic diameter<10 µm); and (C) fine particles (aerodynamic diameter <2.0 µm). Abbreviations: ID, indoor air in five farmhouses; B, used a stove without a chimney; A,C,D, and E, used stoves with chimneys; OD, outdoor environment; F, school; G, lodging.

Statistical evaluation. Statistical analyses were carried out using the *F*-test and *t*-test as described by Snedecor and Cochran (*30*); $p < 0.05$ was considered significant.

Results

Coal is usually the most important energy source in residential areas, especially in many rural residential areas in China. The residents in rural areas use coal-burning stoves with or without chimneys. Residents use these stoves for cooking, heating, and drying food for storage.

As shown in Figure 1A, SPM concentrations in the farmhouse that used a stove without chimney (B) were substantially higher than SPM concentrations measured in farmhouses that used stoves with chimneys (A,C,D,E). Hourly concentrations of SPM varied considerably. Average concentrations of SPM in indoor air (ID) were significantly higher than SPM concentrations in outdoor (OD) environment samples. Size distributions of airborne particles in indoor and outdoor air were also monitored as shown in Figure 1B and C. High SPM concentrations were observed for fine particles with aerodynamic diameters of <2.0 μm.

From monitoring records of the instantaneous variation of SPM concentrations in the farmhouses, we observed that concentrations of SPM in indoor air varied considerably as a function of time of day. Measured concentrations were highest during periods when cooking and heating occurred. SPM concentrations in outdoor air also varied greatly. High concentrations were usually detected during late morning and early evening at cooking time (data not shown).

In fluorosis areas, domestic coal was usually supplied by local mines in the village or town. In the studied fluorosis area, coal was supplied by two local mines and was naturally contaminated by fluoride in the surrounding soil layer. The fluoride concentrations in coal from local mines were as high as 559 mg/kg and 802 mg/kg. The fluoride concentrations in the surrounding soil layer were also as high as 592 mg/kg and 669 mg/kg.

As shown in Figure 2, indoor pollution by airborne fluoride is very serious. In the studied fluorosis area, the daily mean concentration of airborne fluoride in indoor air was 74.4 ± 12.9 μg/m³ [mean ± standard deviation (SD)]. Because airborne fluoride is easily absorbed in food that is being dried for storage, contaminated food represents a significant source of exposure to high concentrations of fluoride. Significant fluoride exposure of the residents was caused by the intake of fluoride-contaminated food such

as corn and chilies, as shown in Figure 3. The concentrations of fluoride in contaminated foods were very different from farm to farm. The concentrations of fluoride in corn and chilies were 70.2 ± 26.5 mg/kg dry weight and 367.5 ± 428.5 mg/kg dry weight (mean ± SD), respectively.

To analyze food consumption and fluoride contamination in food, the main sources of fluoride in residents in the fluorosis area were calculated as summarized in Tables 1 and 2. Total daily doses of fluoride were as

high as 49.16 ± 45.49 mg/day/person. Almost 97% of the fluoride dose came from food consumption. Around 2% of the total fluoride dose was from direct inhalation of airborne fluoride.

To evaluate total exposure of fluoride, the concentrations of fluoride in urine were measured in residents of fluorosis and nonfluorosis areas in China and in the rural area in Japan, as shown in Figure 4. The concentrations of urinary fluoride in male residents in the fluorosis area (32.6 ± 18.6 μM
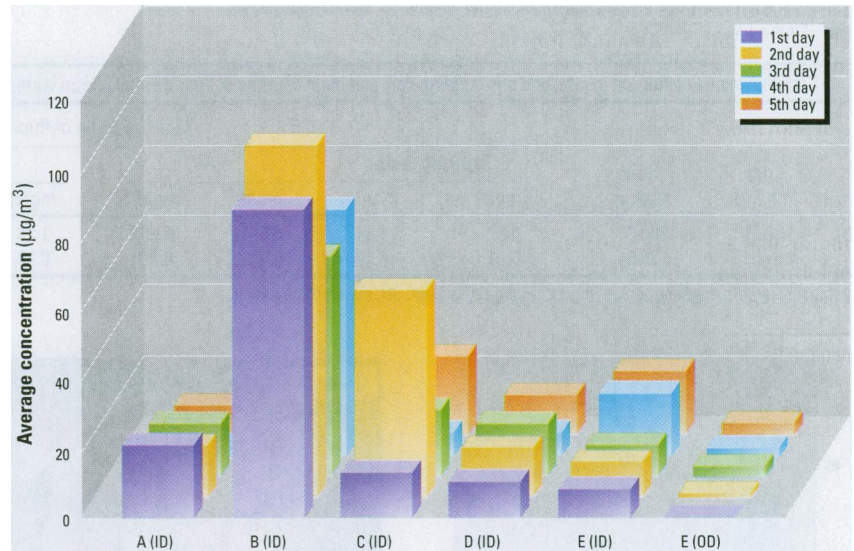

Figure 2. The daily concentration change of airborne fluoride. Abbreviations: ID, indoor air in five farmhouses; B, used a stove without a chimney; A,C,D, and E, used stoves with chimneys; OD, outdoor environment.
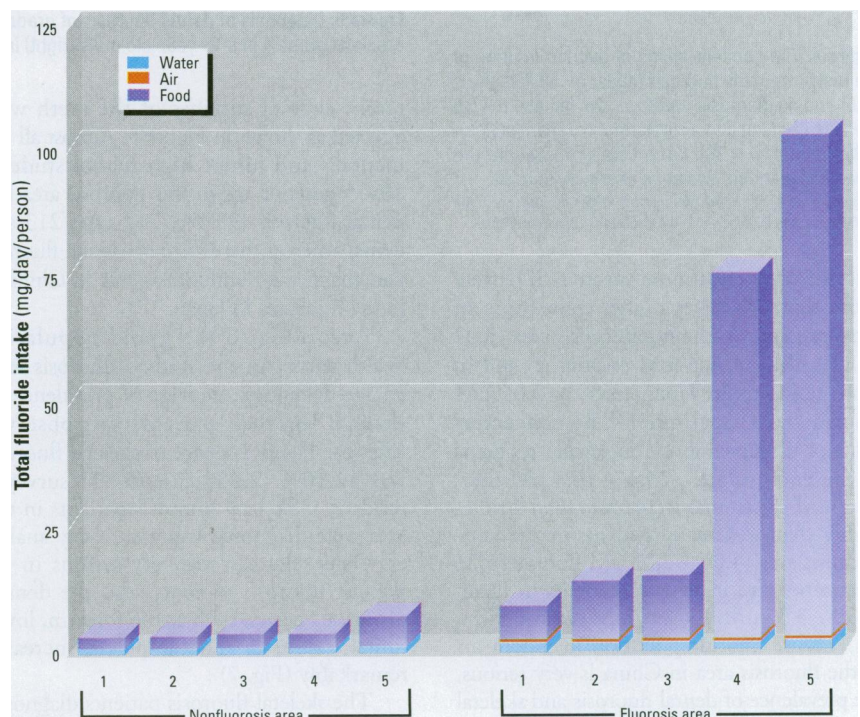

Figure 3. Total daily dose of fluoride in farm families in fluorosis and nonfluorosis areas in China.

Table 1. Concentrations of fluoride in coal and surrounding soil, and main sources of fluoride intake from water, air, and food in farm families in the fluorosis area in China

Family[a]	Coal (mg/kg)	Soil (mg/kg)	Drinking water mg/l	Drinking water mg/d/p	Air mg/m³	Air mg/d/p	Corn mg/kg	Corn mg/d/p	Potatoes mg/kg	Potatoes mg/d/p	Wheat mg/kg	Wheat mg/d/p	Chilies mg/kg	Chilies mg/d/p	Vegetables mg/kg	Vegetables mg/d/p	Total fluoride intake mg/d/p
1	559.0	592.5	0.13	0.26	0.075	0.91	51.45	7.20	24.0	1.10	1.60	0.19	189.2	0.19	1.21	0.37	10.22
2	559.0	592.5	0.13	0.26	0.084	1.02	62.27	17.44	24.0	0.67	1.60	0.22	217.2	1.09	1.21	0.05	20.75
3	802.5	660.0	0.28	0.56	0.053	0.65	109.95	25.29	24.0	2.16	1.60	0.11	203.2	8.17	1.21	0.19	37.13
4	802.5	660.0	0.28	0.56	0.085	1.03	44.64	24.55	24.0	9.84	1.60	0.66	98.4	13.77	1.21	1.99	52.40
5	559.0	592.5	0.13	0.26	0.075	0.91	82.75	113.30	24.0	3.94	1.60	0.43	1129.6	5.65	1.21	0.83	125.32
Mean	656.4	619.5	0.19	0.38	0.074	0.90	70.21	37.56	24.0	3.54	1.60	0.32	367.5	5.77	1.21	0.69	49.16
SD	133.4	37.0	0.08	0.16	0.013	0.15	26.48	42.96	–	3.74	–	0.22	428.5	5.54	–	0.79	45.49

Abbreviations: mg/d/p, mg/day/person; mg/kg, mg/kg dry weight; SD, standard deviation.
[a]Families 1–5 in this area.

Table 2. Concentrations of fluoride in coal and surrounding soil, and main sources of fluoride intake from water, air, and food in farm families in the nonfluorosis area in China

	Coal (mg/kg)	Soil (mg/kg)	Drinking water mg/l	Drinking water mg/d/p	Air mg/m³	Air mg/d/p	Rice mg/kg	Rice mg/d/p	Chili and vegetables mg/kg	Chili and vegetables mg/d/p	Total fluoride intake mg/d/p
Mean	152.01	285.0	0.57	1.14	0.0048	0.057	1.26	0.752	1.68	0.154	2.11
SD	–	35.0	0.01	0.02	0.0013	0.016	0.37	0.361	–	0.017	0.35

Abbreviations: mg/d/p, mg/day/person; mg/kg, mg/kg dry weight; SD, standard deviation.
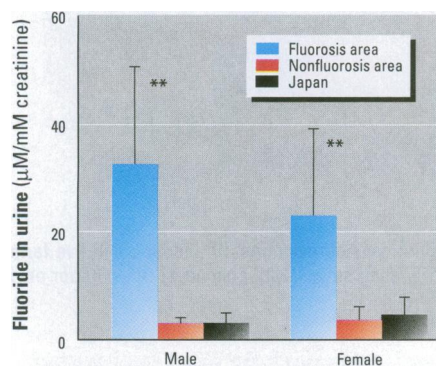

Figure 4. The concentrations of fluoride in urine of the residents in the fluorosis (male, $n = 53$; female, $n = 26$) and nonfluorosis (male, $n = 26$; female, $n = 50$) areas in China and in a rural area in Japan (male, $n = 36$; female, $n = 38$). Each value represents the mean and standard deviation of urinary fluoride.
**Values were significantly different from values of residents in the nonfluorosis area in China and in the rural area in Japan ($p<0.001$).

Figure 5. Diagnosis of dental fluorosis of grade 3–4 in a 13-year-old female (left) and of serious dental fluorosis of grade 4 in a 13-year-old male (right) in the studied fluorosis area in China.

fluoride/mM creatinine; mean ± SD) were more than 10 times higher than those in male residents in the nonfluorosis area (3.02 ± 1.19 μM fluoride/mM creatinine) and in male residents in Japan (3.05 ± 2.05 μM fluoride/mM creatinine). The concentrations of urinary fluoride in female residents in the fluorosis area (23.3 + 16.3 μM fluoride/mM creatinine) were more than 4 times higher than in female residents in the non-fluorosis area (4.00 ± 2.33 μM fluoride/mM creatinine) and in female residents in Japan (4.89 ± 3.37 μM fluoride/mM creatinine).

Because fluoride pollution in indoor air in the fluorosis area in China is very serious, the prevalence of dental fluorosis and skeletal fluorosis is extremely high. Dental fluorosis patients showed slight to severe mottling of
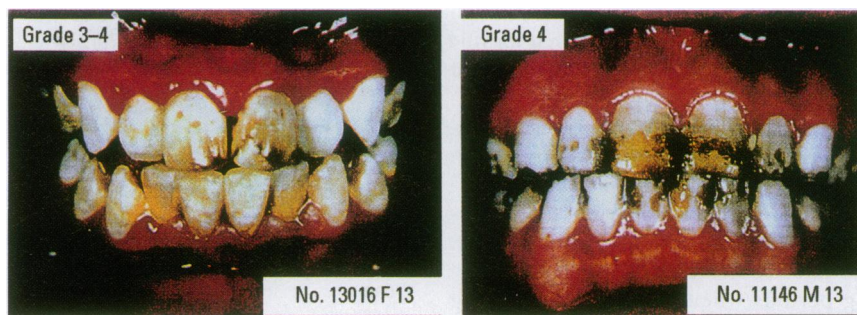
teeth; enamel surfaces of the teeth were affected as shown in Figure 5. Almost all elementary and junior high school students 10–15 years of age in this fluorosis area had dental fluorosis (99.5%; 212 cases/213 students) whereas there were no dental fluorosis patients in the nonfluorosis area in China or in the rural area in Japan.

According to the grand population health survey in the studied fluorosis area, an age-dependent increase of prevalence on skeletal fluorosis patients was observed (Fig. 6). The prevalence of skeletal fluorosis was 42.10% (2,455 cases/5,831 surveyed residents). Skeletal fluorosis patients in this area were diagnosed using an X-ray analyzer. Osteosclerosis was very serious in the skeletal fluorosis patients, and the density of various bones, such as the forearm, lower limb, vertebrae, and the pelvis, increased remarkably (Fig. 7).

The skeletal fluorosis patients diagnosed as stage 1, 2, and 3 using the diagnostic categories of Singh and Jolly (31) were 6%

(3/49), 6% (3/49), and 88% (43/49), respectively. Twenty nine percent of the osteosclerosis patients also showed marble bones syndrome in the pelvis.

The biochemical and nutritional effects on bone metabolism were evaluated by analyzing urinary parameters and the epidemiological data of residents. As shown in Figure 8, urinary deoxypyridinoline, a significant biochemical marker of bone resorption, was much higher in the residents of both studied areas in China than in the rural residents in Japan. The concentrations of urinary inorganic phosphate were also significantly lower in the residents in the fluorosis area of China than in the nonfluorosis area and in the rural area in Japan (data not shown).

Discussion

Combustion of coal as an energy resource is one of the major sources of SPM, gaseous pollutants, and some hazardous toxicants in many countries, including China (5,6,11).

Airborne fluoride from the combustion of high fluoride-content coal is one of the most hazardous air pollutants and has serious impacts on human health (16,17).

In China, coal, coal gas, and natural gas are used as domestic fuel energy sources. The highest concentrations of airborne pollutants were observed in indoor air of coal-burning families (13). In some rural areas in China, high fluoride-contaminated coal, which is produced by local mines, is used as a main fuel energy source. As a result, hazardous air pollutants such as SPM and fluoride are significantly higher in the combustion exhaust emissions from this high fluoride-content coal (14). In this study, the highest concentrations of SPM and fluoride were observed in indoor air during periods of cooking and heating. Daily mean concentrations (± SD) of SPM and fluoride in indoor air were 712 ± 648 $\mu g/m^3$ and 74.4 ± 12.9 $\mu g/m^3$, respectively.

Airborne fluoride is one of the most hazardous pollutants in fuel combustion exhaust emissions and industrial waste exhaust emissions in the world. Because high concentrations of airborne fluoride are generated by burning high fluoride-content coal, it is estimated that approximately 18 million dental fluorosis cases and 1,080,000 skeletal fluorosis cases may be caused by airborne fluoride in China (17). In these fluorosis areas, domestic coal is usually supplied by local mines and is naturally contaminated by fluoride in soil layers surrounding the coal seams. In the studied fluorosis area, the concentrations of fluoride in coal supplied by two local mines were very high (559 mg/kg and 802 mg/kg); therefore, the probability of fluoride pollution resulting from coal combustion in indoor air and personal exposure to high concentrations of fluoride is also extremely high.

Fluoride in indoor air is directly inhaled by residents. Moreover, airborne fluoride is easily absorbed by foods that are being dried for storage, such as corn, chilies, and potatoes. In the studied area, the concentrations of fluoride in water, which was supplied by two local water sources, were very low (0.13 mg/l and 0.28 mg/l) compared to concentrations of airborne fluoride (74.4 ± 12.9 mg/m^3).

The contamination of fluoride absorbed in corn, wheat, chilies, potatoes, and vegetables was extremely high, and the daily mean intakes of fluoride from these foods were 37.6 mg, 0.32 mg, 5.77 mg, 3.54 mg, and 0.69 mg per person, respectively. The residents ingested these fluoride-contaminated foods every day. In the studied area, 97% of fluoride exposure was due to the ingestion of food and only 2% was from direct inhalation of fluoride.

In some areas of the world, including the United States, water fluoridation has been used as an effective and very cheap method to prevent dental caries (15). In the studied fluorosis area, there were no students with dental caries (0 cases/213 students). However, exposure to excess fluoride is very dangerous for human health and has caused serious dental and skeletal fluorosis.

Because urinary fluoride is the most important indicator of personal exposure, it is possible to evaluate the risk of environmental fluoride on human populations (15,16). The concentrations of urinary fluoride in the residents in the fluorosis area in China were much higher than in residents in the nonfluorosis area in China and in the rural area in Japan.

The personal exposure to fluoride was very serious; thus, the prevalence of fluorosis was extremely high. Almost all elementary and junior high school students 10–15 years of age

in the fluorosis area suffered dental fluorosis (99.5%), whereas there were no dental fluorosis patients in the nonfluorosis area in China and in the rural area in Japan.

Fluoride directly stimulates bone formation *in vivo* and also stimulates fluoroapatite formation (18–20). Because serious fluoride pollution has caused skeletal fluorosis, it is necessary to analyze bone metabolism of the residents in the fluoride-polluted areas. In the studied fluorosis area, the grand population health survey determined that the prevalence of skeletal fluorosis was 42.10% in the residents. Using an X-ray analyzer, 49 skeletal fluorosis patients in this area were diagnosed in detail. Eighty-eight percent of skeletal fluorosis patients had serious osteosclerosis and were diagnosed as having stage 3 of the disease according to the categories of Singh and Jolly (31).

The concentrations of urinary inorganic phosphate in the residents in China were
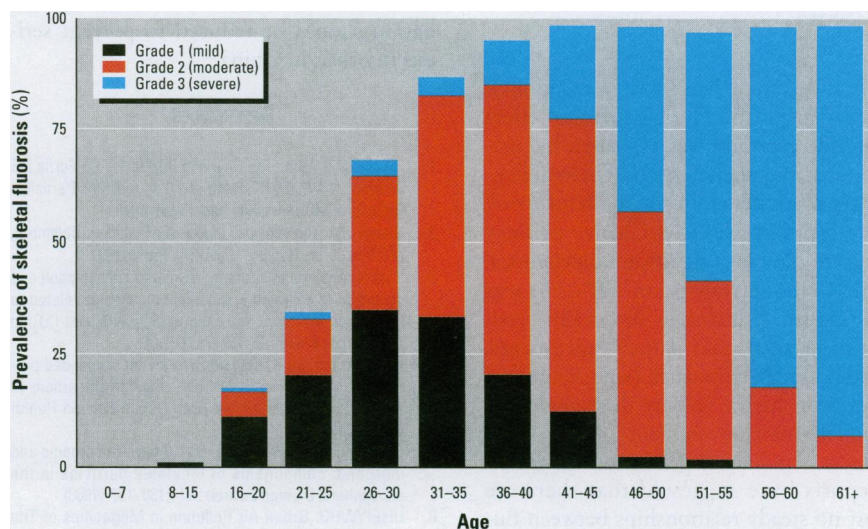
Figure 6. Age-dependent increase in prevalence of skeletal fluorosis patients in the studied fluorosis area in China.
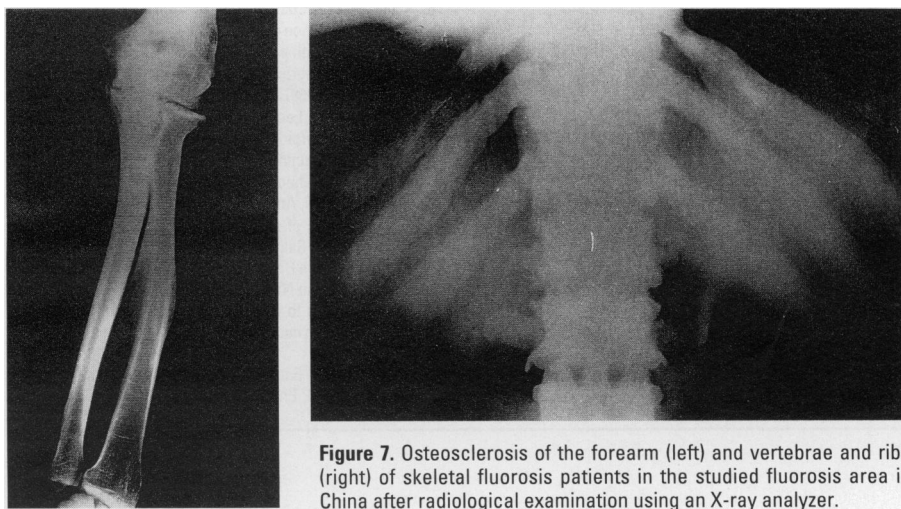
Figure 7. Osteosclerosis of the forearm (left) and vertebrae and ribs (right) of skeletal fluorosis patients in the studied fluorosis area in China after radiological examination using an X-ray analyzer.
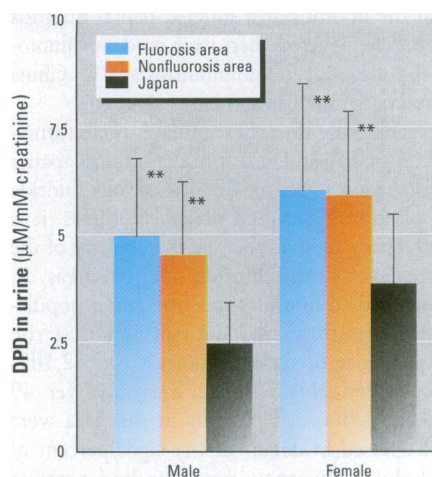
Figure 8. Concentrations of urinary deoxypyridino-line (DPD) of the residents in the fluorosis (male, n = 53; female, n = 26), and nonfluorosis areas in China (male, n = 26; female, n = 50) and in the rural area in Japan (male, n = 36; female, n = 38). Each value represents mean ± standard deviation of urinary fluoride.
**Values were significantly different from values of rural residents in Japan (p<0.001).

significantly lower than those in the healthy residents in Japan. The biochemical effect of fluoride exposure was evaluated by analyzing urinary parameters and epidemiological data of residents. In the residents living in the studied areas in China, urinary deoxypyridinoline, a significant biochemical marker of bone resorption (27–29), was much higher than that in the healthy residents in Japan. In our animal experiments, fluoride directly stimulates bone resorption and excretes higher urinary deoxypyridinoline in vivo (data not shown).

Recent epidemiological and experimental reports have suggested that there are almost no steady relationships between fluoride exposure and bone strength (21–25). Lee and Hanson (32) reported that bone quality was slightly improved by a low dose of fluoride, whereas it was markedly deteriorated by high dose. In this study, bone resorption was extremely stimulated in the residents in China. Grynpas (26) reported that fluoride directly stimulates fluoroapatite formation. In our animal experiment, high concentrations of fluoride stimulated both bone resorption and bone formation in vivo (data not shown). Because serious fluoride pollution caused osteosclerosis (33), fluoride may stimulate both bone resorption and bone formation. Further study is necessary to analyze bone resorption and bone formation in the residents in fluoride-polluted areas.

In the studied area, the skeletal fluorosis patients and residents did not have serious abnormalities of kidney function or respiratory function. As a result of fluoride exposure, the concentrations of urinary inorganic phosphate were significantly lower in the residents in the fluorosis area than in the nonfluorosis area in China and in the rural residents in Japan.

In indoor air of coal-burning families in China, the concentration of fine particles (aerodynamic diameters <2 μm) was markedly high. Because respiratory absorption and aerial diffusion are markedly different according to aerodynamic diameter of particles (34), it is necessary that fine particles and coarse particles (diameter ranges between 2 and 10 μm) should be monitored separately.

Because indoor fluoride exhausted by combustion processes of coal is easily absorbed in stored food and food consumption is a main source of fluoride exposure, airborne fluoride and food contamination should be reduced to prevent serious fluorosis in China.

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