Some Medical Considerations Regarding Atmospheric Fluorides

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IN assessing margins of safety and possible toxic hazards from fluorides, it is necessary to consider the body's total intake of fluoride from all sources. This consideration is important when determining the optimum level of fluoride for dental protection before embarking on a scheme for the fluoridation of a water supply, and it is necessary also when considering possible hazards from fluorides to the community in the vicinity of industries emitting them. The total intake will be derived in part from the diet, in part from drinking water, and in part from the atmosphere.

McClure (*1*) made a comprehensive analysis of the fluoride content of individual foods and estimated the adult intake from the diet, excluding that derived from drinking water, was 0.2–0.3 mg. per day. Cholak (*2*), as a result of the analyses of actual dietaries, calculated an intake of 0.34–0.80 mg. F per day exclusive of the fluoride in drinking water. This intake was in Cincinnati, Ohio, which had a fluoride level of 0.1 p.p.m. in the water supply.

Tea is rich in fluorides, and in countries where it is the staple beverage the total fluoride intake is higher. Thus Longwell (*3*) estimated the average fluoride intake of the diet of men in Great Britain as 1.8 mg. exclusive of that in drinking water. Other foods high in fluoride may contribute significantly to the total intake of the population of some countries.

In India, Sita and Venkateswarlu (*4*) found samples of common salt containing 10–20 p.p.m. F while rock salt, still used in some parts of India, contained 170 p.p.m. F, or 40 p.p.m. F after recrystallization. Venkateswarlu (*5*) later found that the sea salt, also used extensively in India, contained 14–20 p.p.m.

Seafoods are rich in fluorides though some may be present in a form less easily absorbed by the body. In addition, variations in average temperature affect the total fluoride intake, and the existence of severe malnutrition in a population will reduce the margin of safety.

Hazards to Man of Industrial Fluoride

Chronic industrial fluorosis was first described in man by Mφller and Gudjonsson (*6*) after an investigation of the workers in the Danish cryolite industry. In 1937, Roholm (*7*) published his

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classic report of the disease, based on a detailed investigation of the cryolite workers. Many of the workers had been exposed to cryolite dust (Na_3A1F_6) for long periods. Typical atmospheric dust concentrations in the works were 30–40 mg. per cubic meter, and in two enclosed areas concentrations up to more than 9,000 mg. per cubic meter were found. Although many workers complained of acute gastric symptoms or shortness of breath, it was difficult to say whether their symptoms could be attributed to the cryolite because they could have been the physical effect of the very heavy dust concentrations.

The most typical symptoms of fluorosis were complaints related to the skeletal and muscular systems, and 35 percent of the workers complained of pains, stiffness, or rheumatic attacks. Eighty-four percent of the exposed workers had radiological evidence of sclerosis, the severity being related to both the duration of exposure and concentration of dust inhaled. Slight or moderate pulmonary fibrosis was found in about half the workers.

Former workers in the industry who were known to have been exposed to concentrations of dust at least as heavy or heavier than the workers currently employed were also examined, but the number and severity of the cases of osteosclerosis was much less. This finding was considered a clear indication of the reversibility of the condition, an observation which was subsequently confirmed by Likins and co-workers (*8*), who also observed that the urinary fluoride excretion remained at a high level for a considerable time after the fluoride intake had been greatly reduced. A higher incidence of spondylitis deformans was found in cryolite workers who had left the industry, but the patients with pulmonary fibrosis did not show any evidence of deterioration since leaving.

Roholm estimated that the workers probably absorbed between 0.2 and 1 mg. per kilogram body weight per day, the former figure being the more likely. The urinary excretion of two workmen, both with osteosclerosis and exposed to heavy concentrations of dust, was 2.45 and 2.09 mg. of fluorine per day as compared with two normal persons whose daily excretions were 0.22 and 0.1 mg., respectively.

Murray and Wilson (*9*) in 1946 described an incident involving a family of nine who had been living on a farm in Britain where they were exposed to dust and fumes from an ironstone works in which there was calcining. Some of the nine complained of muscular and joint pains, others of loss of appetite and catarrhal infection. There was no radiological evidence of osteosclerosis, but examinations of urine revealed from 2 to 4 p.p.m. fluorine.

The most notable incident of fluoride exposure in Great Britain was investigated as a result of detecting animal fluorosis in the vicinity of an aluminum factory near Fort William in Scotland (*10*). The report, published in 1949, described detailed clinical, laboratory, and radiological investigations of the workers. None had complained of any symptoms, although there was a suggestion that workers in the furnace room had more frequent digestive disorders and coughs, and indeed the investigators themselves noted the irritating nature of the furnace room fumes. The incidence of aches and pains in the furnace room workers, however, was no greater than in other workers in the factory or in local residents. No dyspnea after exertion was observed, and there was no increased liability to have fractures. There were no physical signs of skeletal fluorosis; one furnace room worker had ankylosing spondylitis which was thought to be unconnected with fluorides. There was also one case of chronic pulmonary fibrosis, and one patient had emphysema and a rigid, barrel-shaped chest.

Radiological examination, however, revealed signs of osteosclerosis in a number of the furnace room workers, the proportion of cases increasing with the duration of the workers' exposure. The signs included lipping of the dorsal and lumbar vertebrae with beaklike exostoses and a pelvis that appeared granular, amorphous, and dense, often with short, bony exostoses. Plaques of dense bone were noted on the tibia and fibula.

Workers in the factory showed an increased urinary excretion of fluoride, the amount being closely related to the severity of exposure. The average excretion of 65 heavily exposed workers in the furnace room was 9.03 mg. per day. Outside the factory no clinical signs or symptoms among local residents were found, and the incidence of mottled teeth in children in the vicinity did not differ appreciably from that in unaffected areas. Measurements of atmospheric fluorides in the factory atmosphere at Fort William ranged from 0.14–3.13 mg. per cubic meter in the furnace room, while outside the factory they varied from 0.22 mg. 200 yards away to 0.04 mg. in the center of Fort William, a distance of 1 mile.

A group of cases of industrial osteosclerosis

acquired in a German chemical works has also been described (*11–13*). As with the Danish patients, their medical condition improved when the workers were removed from the contaminated environment.

A group of 74 workers exposed to relatively high fluoride concentrations in a phosphate fertilizer factory in the United States was compared with a matched control group of unexposed workers (*14*). The fluoride exposure of the individual workers was estimated by repeated examinations of urine samples taken at the end of the night shift, and the percentage of specimens containing or exceeding 10 mg. per liter was calculated as an index of exposure for each person. No disability attributable to fluoride was found in any of the workers, and there was no increase in the number of abnormalities in the gastrointestinal, cardiovascular, metabolic, or hematological systems. Respiratory illnesses were more frequent; however, these conditions might have been caused by the irritating properties of the acid gases. Minimal or questionable degrees of increased bone density were found radiologically in 23 percent of the exposed employees, but in no case would the bone changes have been sufficient for them to have been recognized as having increased radio-opacity in routine radiological practice. An apparent increase in vascular and skeletal conditions and albuminuria in the exposed group was significant only at the 10 percent level.

A number of investigations have been reported from the U.S.S.R. and Czechoslovakia in which dental fluorosis or diminished prevalence of dental caries has been noted in children living in the vicinity of factories emitting fluorides (*15–18*). In both Balazova's and Macuch's series of cases the raised intake of fluorides was confirmed by finding raised urinary fluoride levels, and in Lindberg's investigation atmospheric fluoride levels in the vicinity of the works ranged from 0.098–0.485 mg. per cubic meter.

Air Pollution by Fluorides

Air pollution by fluorides has recently been reviewed by Martin (*19*). Although fluorides are common constituents of the earth's crust and, therefore, are inevitably present in natural waters, they cannot be regarded as natural constituents of the atmosphere because their only natural source is volcanic emissions. They are, however, present in coal smoke and are, therefore, a pollutant of urban atmospheres and are also found in specific localities as a result of the emissions from certain industries.

A survey by the chief alkali inspector in England and Wales in 1961 indicated that some 25,000 tons of fluoride were being emitted annually into the atmosphere from industrial sources. Twelve thousand tons of this fluoride were derived from the industrial use of fluorspar, 10,000 of which were emitted during the manufacture of steel, 5,000 were from the industrial and domestic use of coal, 4,500 from the heavy clay industry, 600 from the treatment of iron ores, 500 from the cement industry, and 150 tons from the pottery industry. Emissions from gas furnaces, the chemical industry, the manufacture of hydrofluoric acid and fertilizers, and phosphorus and zinc smelting were small.

Since 1961 the amount of fluoride emitted from the steel and pottery industries has diminished. Emissions are partly gaseous and partly particulate, and the particulate matter is deposited at varying distances from the point of emission. The most serious problem is the contamination of herbage thus leading to fluorosis in farm animals.

Data collected from various sources by the Department of Health and Social Security showed that deposits of fluoride in a rural area of England (Essex) averaged 0.69 g. of solid fluoride per 100 square meters per month whereas in London values ranging from 0.58 to 2.6 g. were obtained, the excess over rural areas presumably being largely the result of combustion of coal. Measurements by local authorities at sites in Warwickshire varied from 0.5 to 1.08 g. per 100 square meters, the 1.08 g. being recorded downwind from an aluminum works.

In contrast to these readings, a monthly average of 4.5 g. was measured near the center of Rotherham, at a distance of approximately 1½ miles from a steelworks which at that time was using fluorspar in open hearth furnaces. An average of 3.64 g. was similarly obtained at a site near the center of the pottery industry in Stoke-on-Trent.

Deposit gauge readings such as these are of value in assessing risks to agricultural animals, but measurements of the amounts of fluoride in the atmosphere are more important in assessing human risk since they indicate the quantities which might be inhaled. Measurements undertaken for us by the Medical Research Council's Air Pollution Research Unit using a volumetric apparatus in the center of London indicate a normal winter pollution of particulate fluorides of

0.05–0.09μg. per cubic meter expressed as fluorine. Gaseous fluorides are less than 0.05μg. so that the total fluoride content of the normal London air would be of the order of 0.1–0.15μg. During a period of exceptionally heavy pollution associated with severe fog, the amount of particulate fluoride may increase to 0.8μg. per cubic meter.

In contrast, winter measurements of total particulate and gaseous fluoride undertaken by the Warren Spring Laboratory of the Department of Trade and Industry yielded an average of 0.7μg. at Rotherham and 1.9μg. at Stoke-on-Trent. Similar, and in some cases rather higher, atmospheric concentrations have been reported from urban and rural areas of the United States (*20*).

Man's Fluoride Intake from Polluted Air

When measurements of the concentrations of fluorides in the atmosphere are available, it is comparatively easy to determine whether there is any hazard to man from absorption through the respiratory system. The average man engaged in moderately strenuous exercise inhales approximately 20 cubic meters of air per day, and it is therefore possible to obtain an estimate of his daily intake from this source by multiplying the concentration in the atmosphere expressed in micrograms per cubic meter by 20. Such an estimate errs on the side of safety since it assumes that all the fluoride inhaled is absorbed. Thus, while much of the gaseous fluoride would be readily absorbed, some of the suspended matter is insoluble, some because of the particle size would fail to reach the alveoli, and some would be exhaled with the tidal air.

Using the aforementioned method to calculate the amount of fluoride inhaled, workmen in the furnace room described in the Fort William investigation would have an intake of fluoride from the atmosphere of approximately 10–23 mg. per day based on an 8-hour exposure. The clinical picture of the Fort William workmen in fact broadly agrees with that of similar adults in Bartlett, Tex., who would have been obtaining approximately 15 mg. per day from a high fluoride water supply (*21*). A hypothetical person spending 24 hours at a distance of 200 yards from the Fort William factory would have an approximate intake of 4.5 mg., whereas in Fort William itself the 24-hour intake a mile from the factory would only be 0.8 mg.

Similarly Lindberg's data in the vicinity of a superphosphate plant in the U.S.S.R. points toward maximum possible adult intakes of up to 10 mg. Such an intake would be unacceptable in a population, and the high fluoride intake by children was in fact confirmed by the presence of dental fluorosis.

By contrast, on the basis of the 1963–64 measurements, intakes in the center of London would only be of the order of 0.001–0.004 mg. per day. On a very exceptionally foggy day of high pollution the figure might be increased by a factor of 5–10.

In Stoke-on-Trent during the 1963–64 investigation, the maximum possible intake would be of the order of 0.04 mg. and at Rotherham of 0.01–0.02 mg. These figures are all very insignificant in comparison with the normal fluoride intake from the diet. Figures for American cities quoted by Cholak (*20*) also indicate similar insignificant intakes.

Fluoride Intake from Food

The occurrence of fluorosis in cattle in the vicinity of factories emitting fluorides suggests a possibility of increased fluoride intakes in man arising from the contamination of food grown in these areas. Soil normally contains considerable quantities of fluorine, mostly in a form inaccessible to plant life, and the deposition of fluorides in the vicinity of factories emitting fluorides will add to the soil content.

In general, however, and with the exception of certain plants, such as those of the tea family, the work of MacIntire and others (*22–24*) has shown that comparatively little fluorine is absorbed from the soil. Absorption is greatest with the addition of soluble fluorides to light, sandy soils with little organic matter, but even so, much of the absorbed fluoride remains in the root system (*25–26*). In contaminated areas fluorides are deposited on the foliage, and it has been shown that gaseous fluorides, in particular hydrogen fluoride, are readily absorbed by the leaves (*27*).

To assess the possible human intake from contaminated vegetables, a survey was initiated in Britain by the Ministry of Agriculture Fisheries and Food in 1965 with the assistance of the National Agricultural Advisory Service and the Laboratory of the Government Chemist (unpublished paper by C. M. Jones, J. M. Harries, and A. E. Martin). Leafy vegetables (cabbage, kale, brussels sprouts, and lettuce) were most likely to be affected, and these were collected at different

seasons in the contaminated areas of Stoke-on-Trent, Rotherham, Ampthill in Bedfordshire, and near a brickworks in Oxfordshire. Each sample was halved, and one-half washed and prepared for the pot by stripping off outer leaves where appropriate. The other half of each sample was left intact or with only the obvious outer leaves stripped off, as it would be if prepared for market. The samples were then oven dried and sent to the Laboratory of the Government Chemist for analysis.

The results for all samples are summarized in the tables. All samples were moisture free (table 1) except for those from Oxfordshire which were not dried (table 2). It was noted that in general samples taken nearer the point of emission had a higher fluorine content, though this pattern was not so apparent in Stoke-in-Trent where a number of sources of fluoride emissions are widely scattered over the city (table 1). Samples were difficult to obtain in some areas since vegetables are not grown where there are heavy industrial deposits.

Using data from the tables, it is possible to estimate the sort of intake which might result from consuming vegetables grown in these areas. An average helping of cabbage for an adult would weigh approximately 100–180 g. raw weight or 5–9 g. dry weight. On this basis, a helping of cabbage containing 10 p.p.m. F dry weight grown

Table 1. Fluoride content (p.p.m. dry weight) of leafy vegetables, Stoke-on-Trent, by year and crop

Year and crop	Number of sites	Unwashed			Washed		
		Maximum	Mean	Standard error	Maximum	Mean	Standard error
1965–66							
Winter cabbage	6	28.7	12.7	4.2	24.6	9.1	3.7
Summer cabbage	7	18.5	8.9	2.6	20.9	7.9	3.3
Summer lettuce	7	44.8	21.3	6.0	65.8	19.6	8.5
Autumn lettuce	7	44.1	22.2	5.2	35.2	14.1	4.2
Autumn cabbage	7	19.3	6.2	2.3	8.5	2.8	1.0
Kale	1	[1] 296.6	[1] 296.6	.....	[1] 171.3	[1] 171.3	.....
1968							
Winter cabbage	2	24.3	19.1	.....	10.5	8.5	.....
Summer cabbage	7	42.2	18.2	5.8	30.6	10.6	3.6
Summer lettuce	3	22.3	18.3	.....	16.8	12.0	.....
Autumn lettuce	1	[1] 42.0	[1] 42.0	.....	[1] 16.8	[1] 16.8	.....
Autumn cabbage	8	28.5	12.2	3.2	23.6	10.1	2.9
Winter sprout tops	5	79.7	38.5	15.5	66.2	26.2	10.9
1969							
Winter cabbage	4	49.6	37.4	.....	30.6	21.0	.....

[1] Only 1 sample obtained.

Table 2. Fluoride content (p.p.m. dry weight) of leafy vegetables harvested in three localities, 1965–66

Locality and crop	Number of sites	Unwashed			Washed		
		Maximum	Mean	Standard error	Maximum	Mean	Standard error
Oxfordshire							
Autumn cabbage	1	[1 2] 2.5	[1 2] 2.5	.....			
Kale	1	[1 2] 10.0	[1 2] 10.0	.....			
Bedfordshire							
Winter cabbage	3	55.7	43.4	.....	39.2	32.4	.....
Winter sprout tops	1	[1] 27.7	[1] 27.7	.....	[1] 23.8	[1] 23.8	.....
Rotherham							
Winter cabbage	6	31.4	16.8	3.6	2.7	2.0	0.25

[1] Only 1 sample obtained.
[2] Wet weight.

in Stoke-on-Trent might contribute 0.05–0.09 mg. F to the daily intake. Much of this would in fact have been leached into the water used for cooking, but a few families use this water for making soups. On the same basis a 100–180 g. helping of autumn cabbage from the Oxfordshire locality with 2.5 p.p.m. F wet weight would contribute somewhat less than 0.25–0.45 mg. F. Kale, the vegetable liable to be the worst contaminated, might yield 0.8–1.5 mg. F on the Stoke-on-Trent figures and something under 1.1–1.7 mg. F on the Oxfordshire figures. Assuming an average meal of lettuce to be 50 g. or 2.5 g. dry weight, then that containing 19.6 p.p.m. F from Stoke-on-Trent would yield approximately 0.05 mg. F.

Longwell's figures on fluoride intakes show that an increase of 1 p.p.m. F in a water supply increases the average fluoride intake by approximately 1.4 mg. in an adult male. If the water supply in one of the affected areas was fluoridated at 1 p.p.m. and if a family consumed homegrown vegetables every day, they would still have a lower fluoride intake than the population in some parts of the country with a naturally high fluoride water supply.

Other possible hazards from food are also remote. It has been shown that the increase in fluoride content of milk from cows with fluorosis is negligible. There is no significant accumulation of fluoride in the meat from such cattle, and the prolonged boiling of bones with a high fluoride content has shown there is no hazard in the making of soups and stews (*28*), although it would in fact be unlikely that cattle with marked fluorosis would be slaughtered for human consumption.

Conclusions

Human fluorosis of industrial orgin is now a very rare disease and should not occur in countries where adequate safety precautions are enforced. Experience in the United Kingdom shows that factory emissions are now reduced to such a level that atmospheric pollution by fluorides offers no hazard to human health although in localized areas deposits of fluoride on vegetation still cause fluorosis in cattle.

Man's consumption of vegetables grown in such areas would not present a hazard to health, but in exceptional cases a grossly excessive consumption of heavily contaminated vegetables might lead to a slight increase in dental fluorosis in children. Such fluorosis, however, is unlikely, though experience from certain other countries indicated that where atmospheric pollution from fluorides is higher, signs of dental fluorosis may be found. Where necessary a realistic assessment of the margin of safety or of the existence of a fluoride hazard may be obtained by adequate monitoring of pollution in the vicinity of any factory.

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