# ENDEMIC FLUOROSIS IN DEVELOPING COUNTRIES

## CAUSES, EFFECTS AND POSSIBLE SOLUTIONS

J.E. Frencken
(Editor)

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Report of a symposium held in Delft, The Netherlands, April 27th, 1990

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# FOREWORD

Fluoride is well known as a substance effective in the struggle to prevent dental caries. However, the intake of excessive amounts of fluoride causes pathological changes in the teeth, and also in the skeleton eventually causing permanent disability. Usually the cause of the fluorosis lies in the use of drinking water that has a fluoride content in excess of 1.5 mg/l. Especially in the rural areas of the developing countries fluorosis may be rampant.

The Netherlands Committee on Endemic Fluorosis was formed in 1985 at the initiative of some persons who had come across different aspects of fluorosis in Africa and India. The members of the committee come from widely different scientific disciplines, as wide apart as hydrogeology and dental care, nutrition and geochemistry This approach has led to very stimulating discussions in the committee's meetings. Therefore it was decided to organize a mini symposium on 'Endemic Fluorosis in Developing Countries' to discuss with a larger group the results of studies and research carried out by individual members of the committee.

The objective of this Symposium is to draw attention to the fluorosis problem in relation to water supply and nutritional habits and its effects on the state of health and the economy in the affected areas. The discussion will be focused on practical -technical and financial- solutions that can be applied in and outside the framework of the international development cooperation

The following aspects will be highlighted in the papers: the primary sources of fluoride, the occurrence of fluoride in drinking water, the kinetics of fluoride in relation to dental and skeletal fluorosis, the epidemiology of dental and skeletal fluorosis, fluorosis as an indicator of the success of improved rural water supply systems (a case study from Andhra Pradesh, India), past efforts and future expectancies in the field of defluoridation of drinking water, and the social implications of fluorosis.

The committee hopes that the Symposium may contribute to an awareness about the perpetual suffering of millions of people who are affected by this disease, because their drinking water supply contains fluoride in "poisonous" amounts.

G.P. Kruseman, *Chairman Committee.*

# PREFACE

The Symposium was organised to draw attention to the problem of fluorosis, not only in general sense but, in particular, in relation to its presence in developing countries. It is for this reason that the authors, in most cases, have relied on literature and examples from these countries in describing the causes, effects and extent of the problem and in presenting possible solutions.

The problem of fluorosis is not restricted to some individuals but it affects thousands of people in large areas in a great number of developing countries. I hope that this proceeding will lead to action aiming at improving the health and wellbeing of these people.

I wish to express my gratitude for the assistance I have received from the contributors and the departmental secretary in the preparation of the book.

J.E. Frencken.

# 1

# PRIMARY SOURCES OF FLUORIDE

L. Vasak, TNO-Institute of Applied Geoscience, Delft, The Netherlands

*In this Chapter, only the primary sources of fluoride will be considered. Firstly, the general description of these sources will be given, followed by a brief description of the factors governing the ultimate concentration of fluoride in water. The Chapter is concluded with examples of fluoride occurrence in groundwater from different geological environments in Kenya.*

## INTRODUCTION

Fluoride is an ion of the chemical element fluorine which belongs to the halogen group. In nature, fluorine never occurs in an elemental form because of its electronegativity and high chemical reactivity. The fluoride-ions can form complexes with metal ions, if the pH of water is below 5. In higher pH ranges, the single fluoride ion ($F^-$) prevails. The geochemical behaviour of fluoride is similar to that of hydroxyl ion ($OH^-$).

The sources of fluorine in human environment can be divided into two categories: 1) primary sources; 2) secondary sources. The primary sources include the "natural" sources, such as fluoride bearing minerals and volcanic gases, which are related to the geological and geochemical processes in a region. The secondary sources include the "pollution" sources which are related to the industrial and agricultural activities in a region. These activities include, for example, the use of phosphatic fertilizers, processing of phosphatic raw materials, use of clays in ceramic industries or burning of coal.

## PRIMARY SOURCES OF FLUORIDE

### *Fluorine bearing minerals*

According to Strunz (1970), about 150 fluorine bearing minerals are distinguished. Table 1.1 lists the number of fluorine minerals in different chemical groups and shows the most important examples for each of these groups.

Fluorite ($CaF_2$) is the most important mineral containing fluorine in chemical bound. In amphiboles, micas and apatite, fluoride can replace the hydroxyl ion.

Table 1.1 Fluorine bearing minerals

| Group | Number | Examples |
|---|---|---|
| Silicates | 63 | Amphiboles, Micas |
| Halides | 34 | Fluorite, Villiaumite |
| Phosphates | 22 | Apatite |
| Others | 30 | Aragonite |

Source: Strunz, 1974.

### *Volcanic gases*

Volcanic gases, produced during the degassing of a magma usually contain fluorine such as HF, $SiF_4$ or $H_2SiF_6$. Though the total volume of HF in volcanic gases amounts only to 1 to 2 %, the fluorine concentration may reach to several thousands of ppm.

## FLUORINE IN DIFFERENT GEOLOGICAL ENVIRONMENTS

According to the origin of the rocks, geological environments can be divided in: igneous, sedimentary and metamorphic. The average concentrations of fluorine in different rock types are given in Table 1.2.

In igneous rocks (including volcanic rocks), the fluorine is mostly bound in micas and amphiboles (up to 80 %) and to a lesser extent in apatite (up to 20 %). However, in basalt all fluorine can be contained in apatite (Allmann & Koritnig, 1974). A great part of the fluorine in volcanic rocks is not in the mineral structure, but bound at the mineral surface as a result of the impossibility for the gases to escape completely (Griffioen, 1986) In general, alkalic rocks contain more fluorine than ultramafic rocks.

In carbonate sedimentary rocks, fluorine is present as fluorite. The concentrations are low. Clastic sediments have higher fluorine concentrations as the fluorine is concentrated in micas and illites in the clay fractions (Allmann &

Koritnig, 1974). High fluorine concentrations may also be found in sedimentary phosphate beds (shark teeth) or volcanic ash layers.

In metamorphic rocks, the highest concentrations of fluorine are found in rocks that were formed by contact metamorphism. In these rocks the original minerals are enriched with fluorine by metasomatic processes.

Table 1.2 Concentration ranges of fluorine in major rock groups

| Type of rock | Fluorine concentration |
|---|---|
| Igneous | 100 ppm (ultramafic) - >1000 ppm (alkalic) |
| Sedimentary | 200 ppm (limestones) - 1000 ppm (shales) |
| Metamorphic | 100 ppm (regional) - >5000 ppm (contact) |

## ROCK-WATER INTERACTION

The presence of fluorine bearing minerals and gases is essential for the occurrence of fluoride in water. The ultimate concentration of fluoride, however, depends also on climatological and geochemical conditions in the region The formation of fluoride rich waters was discussed in detail by Griffioen (1986) and this process is schematized in Figure 1.1.

Figure 1.1 Formation of fluoride rich water. (Source: Griffioen, 1986)

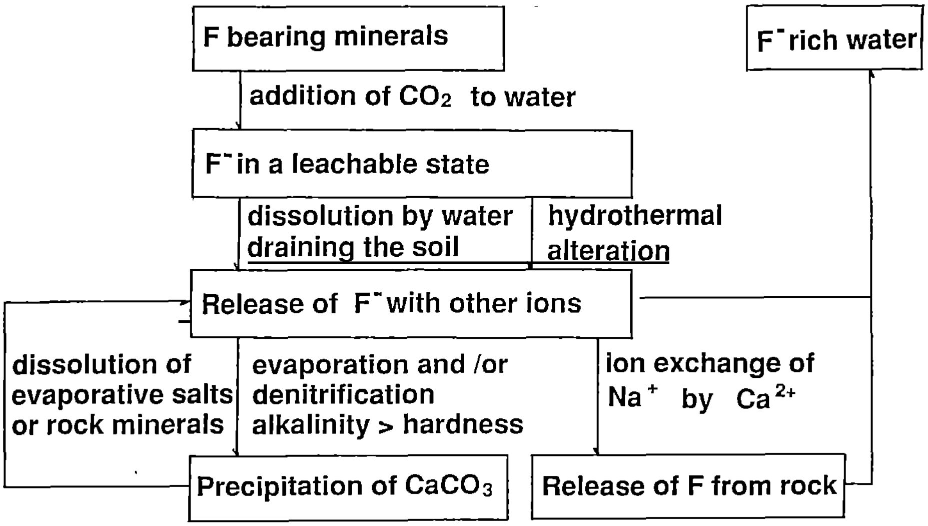
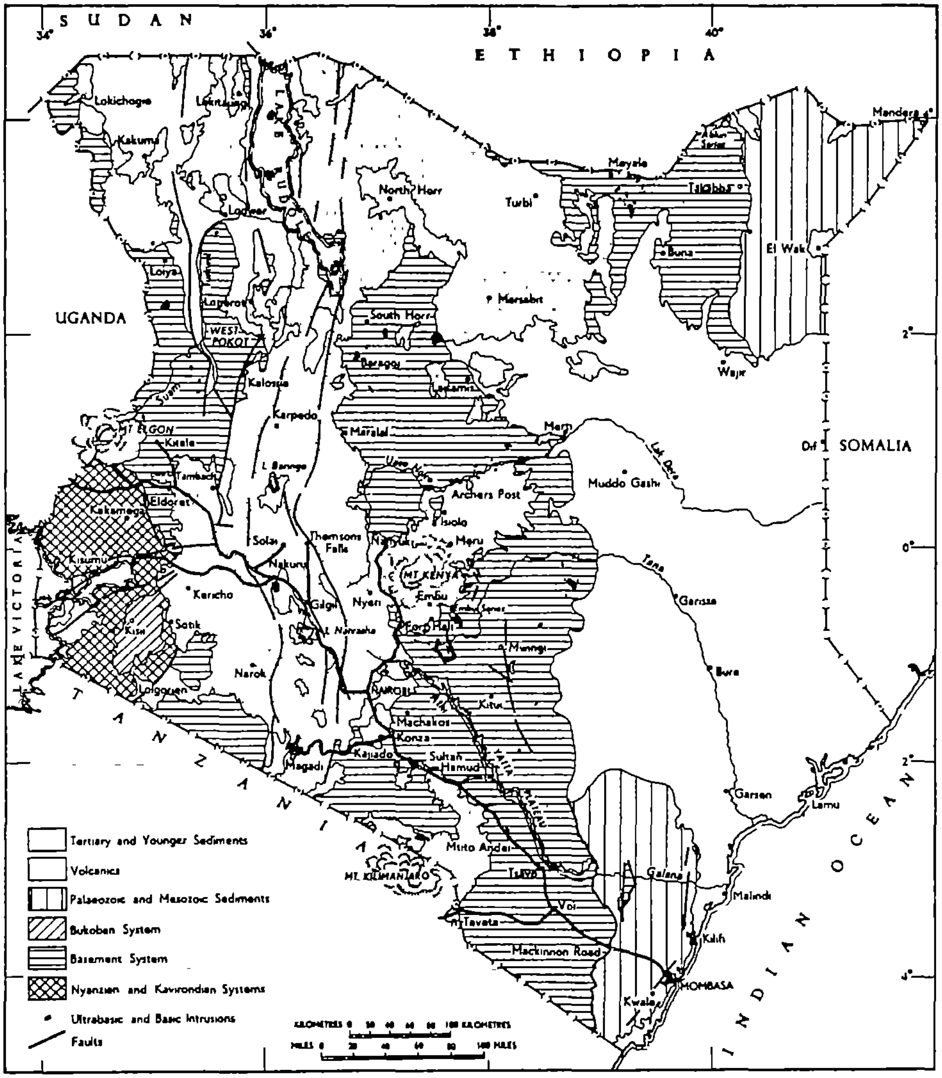


Figure 1.2 Simplified geological map of Kenya. (Source: Pulffry and Walsh, 1969)



The addition of $CO_2$ to water will lower its pH and enhance the weathering of the fluorine bearing minerals. In the absence of $Ca^{2+}$ in leachable state, the fluoride concentration might be high as the fluoride concentration is mainly controlled by the solubility of fluorite. The lowering of the $Ca^{2+}$ concentration may be due to precipitation of calcite or ion-exchange of $Na^+$ by $Ca^{2+}$. During evaporation the $F^-$ contents of the fluid increase if both the following conditions are met: the solution remains in equilibrium with calcite and alkalinity is greater than hardness.

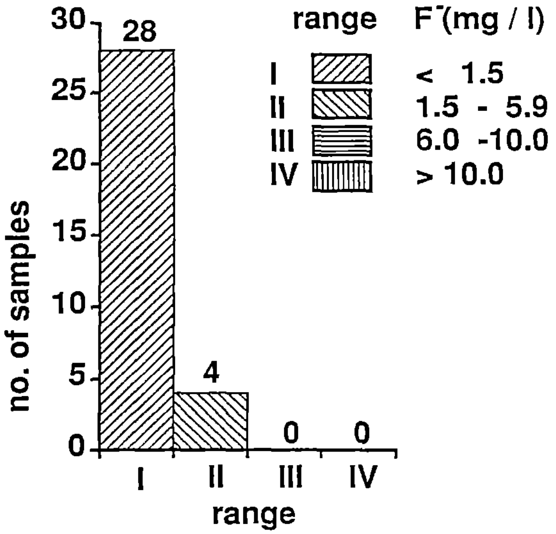
During hydrothermal alteration the solubility of fluorite increases with increasing temperature and fluoride may also be added by dissolution of HF gas. Dissolution of evaporative salts on the surface may be an important source of fluoride.

## EXAMPLES FROM KENYA

### *Physiographic backgrounds*

In Kenya, all the major rock groups, defined in the section on geological environments, occur at or near the surface. A simplified geological map of Kenya is shown in Figure 1.2 on the facing page.

Figure 1.3 Distribution of fluoride concentration in groundwater from Basement Complex, Machakos and West Pokot Districts



The East African Rift Valley, the main geomorphological feature in Kenya, is still an active volcanic region. Large fault systems in the Valley create conditions that allow very deep percolation of infiltrating surface water. During deep percolation, water might come into contact with magmatic gases. The floor of the Rift Valley is characterized by a high hydrothermal activity. The larger part of Kenya belongs to the semi-arid to arid climatological zone. This means that evaporation might strongly influence the water quality.

*Fluoride in groundwater from metamorphic Basement Complex*

The Precambrian rocks of the Basement Complex comprise various types of sediments which were transformed by regional metamorphism into gneisses, schists, quartzites and marbles. Figure 1.3 shows the distribution of fluoride concentration in 32 groundwater samples collected in the Basement areas of West Pokot and Machakos Districts (WRAP, 1984 & 1986).

The concentration ranges I - IV, shown in Figure 1.3, refer to the WHO limit, danger for the mottling of teeth, skeletal fluorosis and danger of crippling fluorosis, respectively. The fluoride concentration ranged from 0.1 - 2.5 ppm. The chemical analyses of the groundwater showed that the fluoride concentrations were directly related to the total content of dissolved solids (represented by the EC value). The chemical analyses of four groundwater samples from Basement areas are given in Table 1.3.

Table 1.3 Chemical analyses of groundwater from Basement Complex (Machakos and West Pokot Districts)

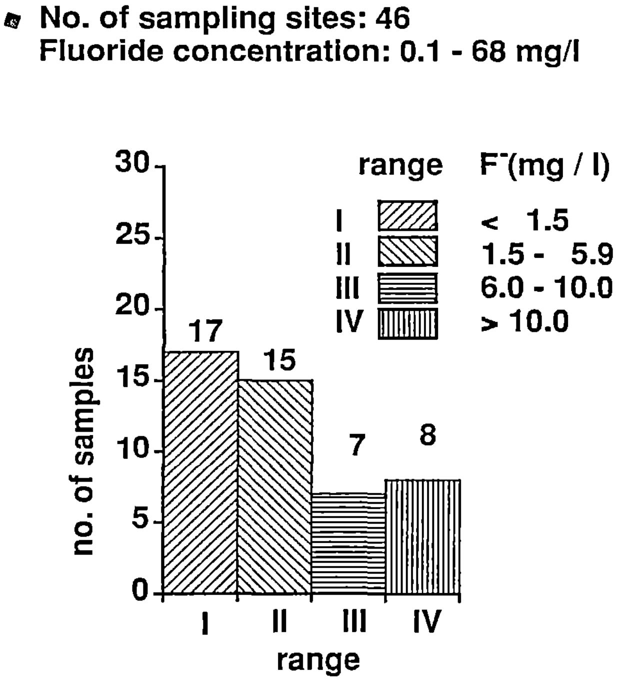
| No. | Source | T | EC | Na | Ca | Mg | Cl | $HCO_3$ | $SO_4$ | F |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | | °C | mS/cm | - | | | mg/l | - |
| 1 | BH 5428 | 24 | 0.30 | 5 | 29 | 9 | 5 | 150 | 5 | 0.2 |
| 2 | BH 5540 | 23 | 0.49 | 8 | 37 | 26 | 9 | 180 | 20 | 0.5 |
| 3 | BH 1578 | 27 | 1.22 | 95 | 125 | 42 | 93 | 375 | 259 | 1.0 |
| 4 | BH 2406 | 26 | 4.65 | 526 | 352 | 253 | 770 | 572 | 1110 | 1.7 |

According to Griffioen & Kohnen (1987), the fluoride concentration in the lower EC ranges (sample 1 & 2) is primarily controlled by the weathering of fluorine bearing minerals. The increase of fluoride concentration with the increasing magnesium concentration in sample no. 2 suggests that the weathering of biotite is the most important weathering reaction with respect to fluoride concentration. In the higher EC ranges (samples 3 & 4), the evapo(transpi)ration becomes the dominant process. Sodium and especially magnesium increases and sulfate and chloride are the dominant anions. Calcium and bicarbonate concentrations are limited by calcite solubility equilibrium. In general, fluoride will precipitate as fluorite together with calcite during evaporation, but a part of fluoride precipitates in easily dissoluble salts. These salts will be dissolved by rain. However, even in the high mineralized groundwater (sample 4), the fluoride concentration remains relatively low.

*Fluoride in groundwater from volcanic areas*

The volcanic areas are composed of basic and intermediate lavas, often intercalated with tuffs. In figure 1.4 the distribution of fluoride concentration in 46 groundwater samples collected in the volcanic areas of Baringo District is presented (WRAP, 1987). It shows that from the 46 samples analysed, about 63 per cent had a fluoride concentration higher than 1.5 mg/l and about 15 per cent was in excess of 10 mg/l $F^-$. The chemical analyses of four groundwater samples from volcanic areas are shown in Table 1.4.

Figure 1.4 Distribution of fluoride concentration in groundwater from volcanic areas, Baringo District



In addition to the processes described above for the metamorphic Basement rocks, the fluoride concentration in the volcanic areas is affected by the hydrothermal activity. According to Table 1.4, the fluoride concentration increases not only with the EC, but also with higher temperatures. In absence of hydrothermal activity (sample 5 & 6), the fluoride concentration of low mineralized water is primarily determined by the weathering of amphiboles or volcanic glass which are important constituents of phonolite, the major lava in Baringo. The relatively high fluoride content in sample 6 may be attributed to the tuff layers, which on the average contain a higher percentage of easily soluble volcanic glass than phonolite. In the samples containing warm (hot) groundwater (samples 7 & 8), the high fluoride concentration must be attributed to the magmatic gas flux of $CO_2$ and HF. Due to removal of calcium ion by calcite precipitation, the fluoride is not equilibrated by fluorite solubility and can reach very high concentration as in sample 8 (68 mg/l).

Table 1.4 Chemical analyses of groundwater from volcanic areas (Baringo District)

| No. | Source | T<br>°C | EC<br>mS/cm | Na | Ca | Mg | Cl<br>mg/l | $HCO_3$ | $SO_4$ | F |
|---|---|---|---|---|---|---|---|---|---|---|
| 5 | BH 5806 | 20 | 0.32 | 64 | 3 | 3 | 12 | 180 | 2 | 0.9 |
| 6 | BH 120 | 24 | 0.50 | 116 | 6 | 1 | 13 | 248 | 39 | 6.2 |
| 7 | BH 3868 | 35 | 2.80 | 880 | 2 | 1 | 108 | 1990 | 94 | 20.0 |
| 8 | SP KAP | 47 | 3.80 | 1020 | 1 | 1 | 225 | 2030 | 98 | 68.0 |

## SUMMARY AND DISCUSSION

As shown in the previous sections, the fluoride concentration in water is mainly dependent on the following factors:

- presence of fluorine bearing minerals and gases;
- weathering processes ($CO_2$ pressure, hydrothermal activity);
- evaporation; and
- calcium concentration.

Although the factors causing the high fluoride concentrations are, theoretically, well defined, more research is necessary to recognize the sources of fluoride in the field.

For proper management of the water supplies with regard to fluoride, a regional hydrogeochemical evaluation is essential. Through such an evaluation, the fluoride sources in the existing supplies may be recognized and categorized in the context of regional movement of water. With this knowledge, potential areas with high fluoride can be excluded in the planning of new supplies.

Good examples are fractured aquifers in the volcanic areas of the Rift Valley in Kenya. From a quantitative point of view, large faults are attractive sites for well drilling, because of high yields. From a qualitative point of view, however, the composition of water is likely to be affected by high temperature and by dissolution of volcanic gases conducted by faults. Moreover, rain water, enriched with salts which accumulated on the surface due to evaporation, may rapidly infiltrate along the fractures. The chance to strike water with a high fluoride concentration in faulted volcanic areas is therefore high. During the design of a new water supply, the faulted zones should be avoided, if possible. Shallow alluvial aquifers in these areas are, in general, considered to be safe with regard to their fluoride concentration.

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## 2

# FLUORIDE IN DRINKING WATER

J. Smet, IRC International Water and Sanitation Centre, The Haque.

*This chapter starts with a description of sources of fluoride other than primary ones. In particular, attention is paid to the role of drinking water as a source of fluoride, and to the fluoride concentration in the drinking water as a water quality parameter. This Chapter is concluded with a summary of geographical areas in developing countries having high fluoride concentration in water sources.*

## SOURCES OF FLUORIDE

### *Introduction*

In chapter 1, different soil formations were indicated as sources of fluoride and the interrelation between minerals, groundwater and surface water was set out. If the total living environment of man is considered, we see that fluorine and fluoride are present in many substances and organisms, such as rock and soil, water, air, animals and plants. The fluoride concentrations within these sources vary greatly depending on many environmental factors, affecting the importance of these sources for the interaction with man.

Fluoride can enter the human body through ingestion, inhalation and absorption by the skin. Of the total amount of fluoride entering the body, a part is excreted mainly in the urine while the remaining part is absorbed in the tissues. Once retained, only a small percentage of fluoride can be slowly released. Repeated or continuous exposure to fluoride sources will therefore cause accumulation of fluoride in the body.

### *Fluoride in rocks and soil*

Fluoride occurs in natural rock and soil formations as described in Chapter 1. But human activity also contributes, and sometimes significantly, to the concentration of fluoride in soils. This particularly refers to the transfer to the soil of fluorides present in fluoride-containing phosphate fertilizers, pesticides, irrigation water, and the deposits from industrial emissions.

The direct importance of these sources for man is very small, while the indirect importance, as high rock/soil fluoride contents influence surface and groundwater quality, is very significant.

### *Fluoride in the atmosphere*

Traces of airborne fluoride originate from both natural and human activities. Dust containing fluoride is found in volcanic areas, and aerosols of seawater also contain some fluoride. Human activities are the more significant contributors to airborne fluoride which, particularly in urban industrialized areas, may reach serious levels. The main industrial sources are steel and aluminium production plants, superphosphate plants, ceramic factories, brickworks, glassworks, oil refineries and coal-burning power plants. The relative importance for man much depends on the distance between living areas and the polluting industrial plants. In rural areas without such industries, these effects are minimal. Increased levels of air pollution makes the relative importance of airborne fluoride greater. In the neighbourhood of, for instance, aluminium factories - responsible for some 10% of the fluoride emission in the USA (155,000 tonnes/year) (Smith & Hodge, 1979) - the importance may be high and fluoride levels up to 4 $\mu g/m^3$ can be found. Occupational exposures in many of these industries may reach 1 $mg/m^3$.

### *Fluoride in food*

Under normal conditions the fluoride concentration in food products remains low, i.e. in the order of 0.2-0.3 mg/kg. Relatively high values of fluoride, however, can be found in particular foods: up to 7 mg/kg in fresh vegetables (due to industrial and/or pesticide pollution), up to 10.7 mg/kg in polished rice, up to 8.0 mg/kg in fresh or canned fish and up to 3.3 mg/kg in fresh and canned meat. The presence of calcium-containing parts (bones) contributes most to these high values (WHO, 1984b). The use of water with high fluoride concentration in cooking also affects the fluoride content in prepared food. The relative importance of this fluoride source very much varies with diet.

*Fluoride in drinks*
The fluoride content of processed drinks depends mainly on the fluoride content of the water used. The same applies to syrups, juices, coffee, tea etc. prepared at home. In general, breast milk contains less fluoride than its substitutes. A relatively high fluoride content is present in tea leaves: 3.2 - 400 mg/kg dry weight. Most of the fluoride is dissolved in water during brewing. Depending on the fluoride content of the tea leaves and the amount of tea consumed, this source may contribute up to about 2.5-3.0 mg to the daily fluoride intake (WHO, 1984b). The relative importance of tea leaves as a source of fluoride therefore, depends very much on the fluoride contents of the water and concentrates used.

## THE ROLE OF DRINKING WATER

In areas with fluoride-containing geological formations, the groundwater, through its direct contact with the fluoride minerals, usually has a higher fluoride content than the nearby surface water sources. Groundwater from boreholes, wells, and springs may have varying and/or fluctuating fluoride contents, ranging between 0.1 and > 100 mg/l, depending on several influences such as:

- shallow groundwater usually has a lower fluoride content during the rainy season than during the dry season, because of dilution by infiltrating rainwater;
- deep groundwater has a more or less constant fluoride content;
- groundwater may show variation in fluoride content depending on the presence of fluoride-containing formations at different depths.

Surface waters, including rivers, streams, lakes etc., usually have a low fluoride content except when fluoride-containing waste products are discharged into these waters. Lakes in volcanic areas may contain extremely high fluoride values, e.g. a lake in Kenya with more than 2,800 mg/l (Akpabio, 1966).

As the geographical focus of this proceeding is on developing countries, the relative importance of drinking water as a fluoride source is obvious. In these mostly tropical countries drinking water consumption is much higher than in countries with a temperate climate; the ambient temperature is higher and the physical workload is usually greater. Adults consume on average 2 to 5 litres per capita per day (lcd), sometimes even up to 10 lcd of drinking water. If this water has a high to very high fluoride content - and particularly where treatment methods are usually not available - drinking water will form the greatest single fluoride source.

To show the relative importance of drinking water for fluoride intake, the daily fluoride intake and sources of a person from the northern part of Tanzania is compared with that of a person from a temperate climate (Table 2.1).

Table 2.1 Comparison of daily human fluoride intake in the northern part of Tanzania and in a country with temperate climate

| Fluoride source | Intake fluoride north Tanzania | Intake fluoride temperate climate mg/day/personmg/day/person |
|---|---|---|
| Drinking water | 3 litres with 8 mg/l = 24 mg | 2 litres with 0.2 mg/l = 0.4 mg |
| Tea | 10 grams tea dry weight, with 100-200 mg $F^-$/kg, so 1-2 mg $F^-$ through ingestion | Approx. 1.0 mg $F^-$ through ingestion |
| Sodium bicarbonate | (Locally this is heavily contaminated with fluoride) 5 grams $NaHCO_3$ with 1000 mg/kg, so 5 mg $F^-$ through ingestion | nil |
| Other food sources | Average 1 mg | Average 0.6 mg |
| Total intake | 32 mg/day/person | 2 mg/day/person |

Source: Aswathanarayana et al., 1986.

## FLUORIDE CONTENT AS A WATER QUALITY PARAMETER

WHO (1984a) has published 'Guidelines for Drinking-Water Quality' and in Table 2.2 the fluoride values and possible effects are given.

In the setting of the guideline limit of 1.5 mg/l and in defining possible effects for higher concentrations, WHO assumed that people consume a daily average of 2 litres of water. Clearly, drinking water consumption in tropical regions is often higher than 2 litres per day for individuals. Apart from this fact, the urinary excretion of absorbed fluoride is lower and the transpiration is higher than in modest countries while the fluoride content of sweat is less than that of urine. These facts give rise to reconsideration of guidelines.

Table 2.2 Fluoride contents in drinking water and possible effects

| Concentration of fluoride | Possible effects |
|---|---|
| 0.5 - 1.5 mg/l | Fluoride in water has no adverse effects, incidence of caries decreases |
| Above 1.5 mg/l | Mottling of teeth may occur to an objectionable degree = dental fluorosis incidence of caries decreases |
| 3 - 6 mg/l | Association with skeletal fluorosis |
| Above 10 mg/l | Crippling skeletal fluorosis |

Source: WHO, 1984a.

Results of a study carried out by Brouwer et al. (1988) suggest that other guiding values would be more appropriate in tropical regions. These are:

- above 0.6 mg/l: mottling of teeth may occur to an objectionable degree, i.e. dental fluorosis;
- above 7.0 mg/l: crippling skeletal fluorosis.

The new regulations for drinking water quality in the United States of America (Pontius, 1990; Federal Register, 1985 and 1986) give the following values:

- 2.0 mg/l : Secondary Maximum Contaminant Level: to protect against objectionable dental fluorosis, which is not considered to be an adverse health effect,
- 4.0 mg/l : Maximum Contaminant Level: to protect against crippling fluorosis.

The above mentioned Maximum Contaminant Level of 4 0 mg/l is based on an extensive review of research on health effects of fluoride. However, not all countries can adhere tot this standard. In Tanzania, for example, the government has set the drinking water quality standard for fluoride at 8.0 mg/l (MAJI, 1974). Practical, technical and economic arguments have led to the formulation of such a high standard. Another example is Argentina. This country has two different fluoride standards, depending on the location of the water supply (Botteri & Dameri, 1968). These standards are·

- for urban areas: fluoride content should be less than 1.5 mg/l;
- for rural areas (systems supplying water to less than 3,000 people): fluoride content should be less than 2 2 mg/l.

In Table 2.3 the guidelines and standards for fluoride in drinking water for several countries are summarized.

Table 2.3 Summary of guidelines and values for fluoride in drinking water in several countries

| Effects | WHO (1984) | US EPA | Tanzania/Argentina | Suggested (Brouwer et al., 1988) |
|---|---|---|---|---|
| Strong mottling of teeth (dental fluorosis) | > 1.5 | > 2.0 | > 1.5 urban<br>> 2.2 rural | > 0.6 (tropics) |
| Skeletal fluorosis | 3.0 - 6.0 | | > 8.0 | |
| Crippling skeletal | > 10 | > 4.0 | | > 7.0 (tropics) |

## AREAS WITH HIGH FLUORIDE CONCENTRATION IN DRINKING WATER

According to WHO (1984b) more than 260 million people all over the world consume drinking water with a fluoride content of more than 1.0 mg/l. A large part of this group lives in tropical countries where the, by Brouwer et al. (1988), suggested guideline value for dental fluorosis is more than 0.6 mg/l. Countries with areas facing the problem of a fluoride content above 1.5 mg/l in drinking water are listed below. The list is not complete.

Africa: Ethiopia, Sudan, Kenya, Tanzania, South Africa, Nigeria, Senegal, Algeria, Egypt, Zimbabwe, Malawi, Morocco, Uganda, Somalia.
(In Kenya and Tanzania values far above 100 mg/l are reported).

Asia: India, China, Korea, Thailand, Sri Lanka, Indonesia, Yemen, Pakistan.
(In India approximately 25 million people in 8,700 villages drink water with a fluoride content of more than 1.5 mg/l.).

Latin America: Mexico, Peru, Ecuador, Chile, Argentina.

USA: As in the USA more than 300 community drinking water supply systems supply water with a fluoride content of more than 4 mg/l and fluoridation of drinking water is rather common, the government and research institutions give the fluoride problem considerable attention to study adverse effects.

The presence of areas with high fluoride concentrations in groundwater in Tanzania is well-documented by Bardecki (1974). He mapped the areas indicating the percentage of samples of groundwater with a fluoride content above 4 mg/l (Figure 2.1). Some areas with high fluoride content values (in mg/l) are given in Table 2.4.

As stated before, the Tanzanian standard for fluoride is 8 mg/l. This standard is probably based on the old USA standard for skeletal fluorosis. The WHO guideline (1.5 mg/l) and even more the suggested guideline for dental fluorosis for the tropics of 0.6 mg/l (Brouwer et al., 1988) would be an unrealistic standard in Tanzania. The financial and technological problems are too big to meet this standard. On the other hand, the extremely high value of the Tanzanian standard cannot be justified and there is an urgent need to look for appropriate water treatment or other technical solutions.

## TECHNICAL SOLUTIONS

Several water treatment technologies for the removal of excess fluoride in water, both for household and central level, are being used in rural settings. In chapter 6, more specific information on these technologies is given. However, these technologies are often not really appropriate and sustainable considering the specific local technical and economic conditions.

Table 2.4 Areas in Tanzania with different fluoride ranges (above 2.1 mg/l), and the percentage of samples in each range

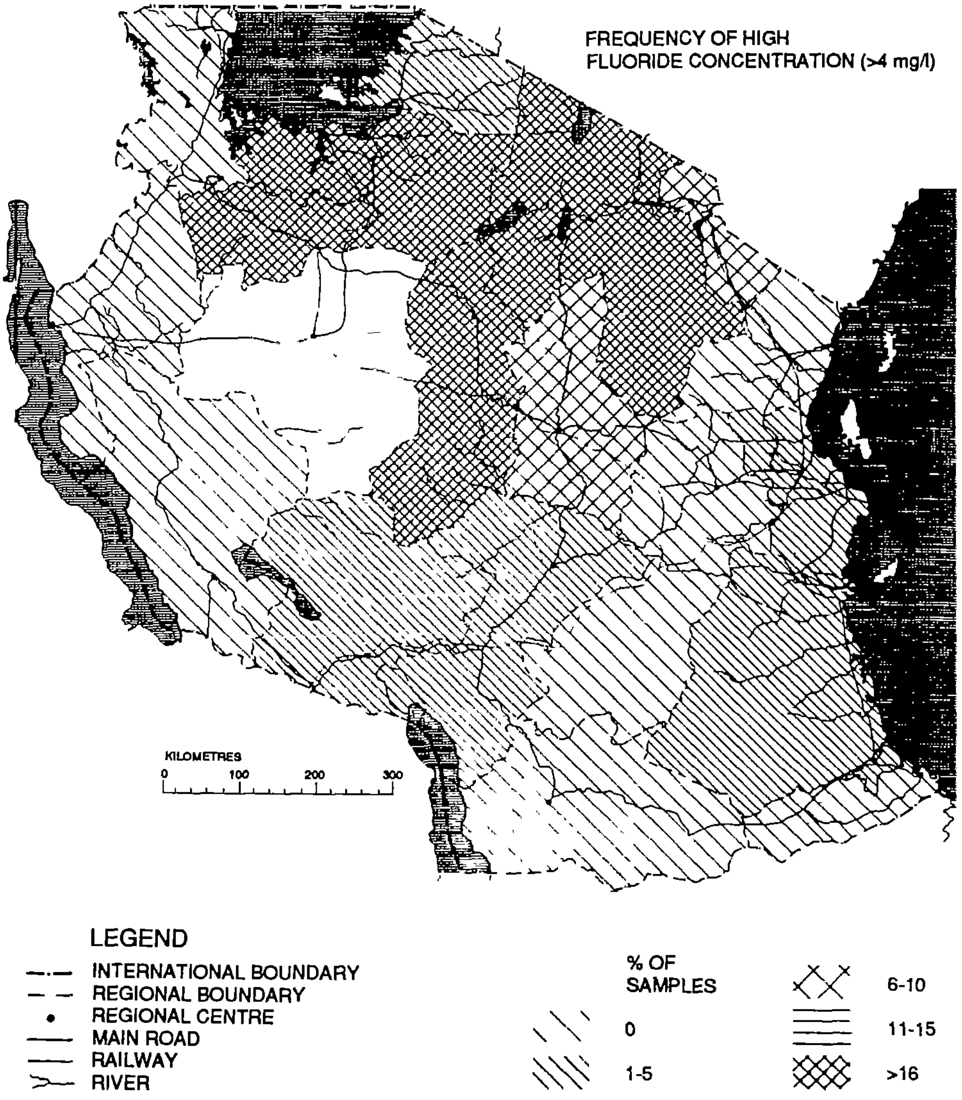
| | Percentage of total samples | | | | |
|---|---|---|---|---|---|
| Region (mg/l F) | 2.1-4.0 | 4.1-8.0 | 8.1-16.0 | > 16.1 | Total > 2.1 |
| Arusha | 18 | 10 | 9 | 14 | 51 |
| Mwanza | 13 | 15 | 10 | 2 | 40 |
| Shinyanga | 11 | 17 | 1 | 2 | 31 |
| Singida | 14 | 9 | 13 | 5 | 41 |
| Kilimanjaro | 8 | 5 | 3 | 1 | 17 |

Source: United Republic of Tanzania, Rural Water Quality Programme in Tanzania, 1979.

The supply of water from low fluoride sources through a piped supply system may be considered as an option Although the capital investment may be high, maintenance cost may remain within acceptable limits, particularly when gravity fed systems could be used. If the people experience and appreciate the benefits of the improved water quality on their health, a reduction in the prevalence of dental and skeletal fluorosis, they may be willing to contribute to the management of the system, particularly in terms of a financial contribution for opertion and maintenance of the piped water supply system. This may be even more so if economic activities, such as cattle-ownership, should benefit from improved water quality.

As the fluoride content of groundwater very much depends on the presence of fluoride containing formations, a proper hydro-geological survey indicating the sites with low, high and very high fluoride content (also in relation to different depths), may help to find the optimum location of sites for groundwater exploration. Obviously, next to the technical criteria, socio-cultural criteria influence the siting of groundwater supply points.

Figure 2.1 Distribution of areas in Tanzania with a fluoride concentration above 4 ppm. (Source: Bardecki, 1974)



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## CONCLUSIONS

Before defining actions to be taken to control fluorosis in a certain area, surveys have to be carried out to identify the main sources of the human fluoride intake. If water appears to be an important source, hydro-geological surveys and identification of low-fluoride surface water sources have to be made. These will then form the basis for discussions with the population on actions to be taken: new sources or defluoridation. These discussions should include all technical, managerial and financial implications of the possible options.

As present international guidelines and standards on fluoride are too high for tropical countries, an adjustment towards more realistic fluoride guidelines and standards is recommended.

Further research on defluoridation technologies is recommended. The need for development and demonstration projects on community-based approaches in reducing the fluorosis problem is identified and needs urgent attention from External Support Agencies active in developing countries.

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## 3

# BIOKINETICS OF FLUORIDE IN RELATION TO DENTAL AND SKELETAL FLUOROSIS

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*Regular ingestion of high quantities of fluoride leads after a number of years to dental and/or skeletal fluorosis. Besides being a health hazard, ingestion of high quantities of fluoride has an effect on the economic potential of a society, particularly, if that society is agriculturally oriented. In this Chapter the physical processes in human bodies related to the development of dental and skeletal fluorosis are described.*

## INTRODUCTION

> "All substances are poisons; there is none which is not a poison. The right dose differentiates a poison and a remedy."
>
> Paracelsus.

Fluoride is an excellent example of Paracelsus famous statement. Both the beneficial and the pathologic effects of fluoride find their basis in an important characteristic of the fluoride ion i.e. the formation in the human body of strong bonds with calcium and phosphate.

In minute quantities (± 0.2 mg/day) fluoride is an essential trace element starting the nucleation and formation of hydroxy-apatite crystals for the building of both bone and teeth.

In slightly higher quantities (0.5-2.0 mg/day) it is a medicine, stimulating in growing teeth (thus before the eruption) the formation of a stronger enamel and after eruption stimulating and guiding the maturation of the enamel and the repair of initial caries lesions resulting in a still stronger enamel with a slightly increased fluoride content.

In still higher quantities, leading to fluoride plasma levels of > 0.15 ppm F*, it starts to interfere with the normal formation of teeth (fluorosis of enamel) and bone (skeletal fluorosis), ending in older people in a non-functional continuing bone formation leading after 10-20 years of high fluoride exposure e.g. to a serious immobilisation of joints and spine. In cases of simultaneous malnutrition, a juvenile skeletal fluorosis (Kenhardt bone disease) may occur with deformation of the legs (genu valgum, "knock knees").

In very high quantities, one to two gram a day, it may be lethal.

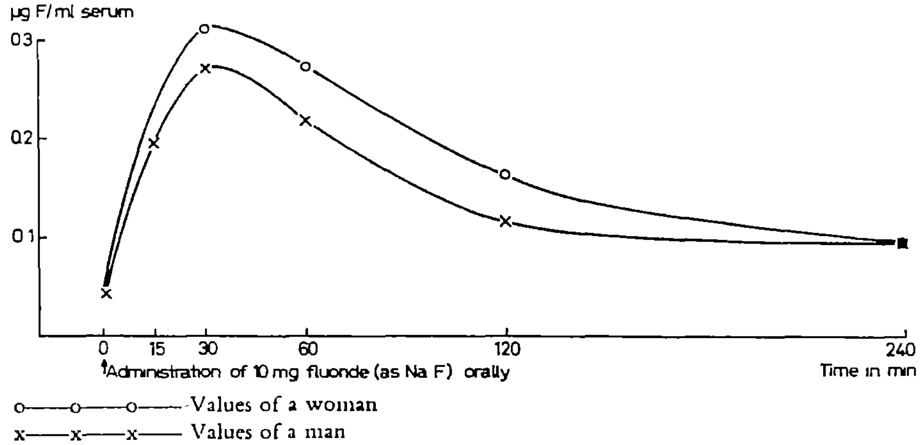
## THE BIOKINETICS OF FLUORIDE

### *Absorption, excretion and retention*

Fluoride in the drinking water is by far the most important source of fluoride absorbed in the human body. Other sources of fluoride (generally less soluble) may be soil and dust, contamination of food, sediment of water etcetera. Tea as beverage (0.5-2.0 ppm F) is probably the only important other source of well soluble fluoride.

Soluble fluoride is rapidly and almost completely absorbed in the stomach and the intestinal tract into the blood. In case the stomach is relatively empty, the fluoride concentration in plasma will reach a peak in about 30 minutes after the fluoride ingestion, returning to its original level 2-3 hours later (Fig 3.1).

Figure 3.1 The course of the serum fluoride concentration in two young adults after a single oral dose of 10 mg fluoride as sodium fluoride.



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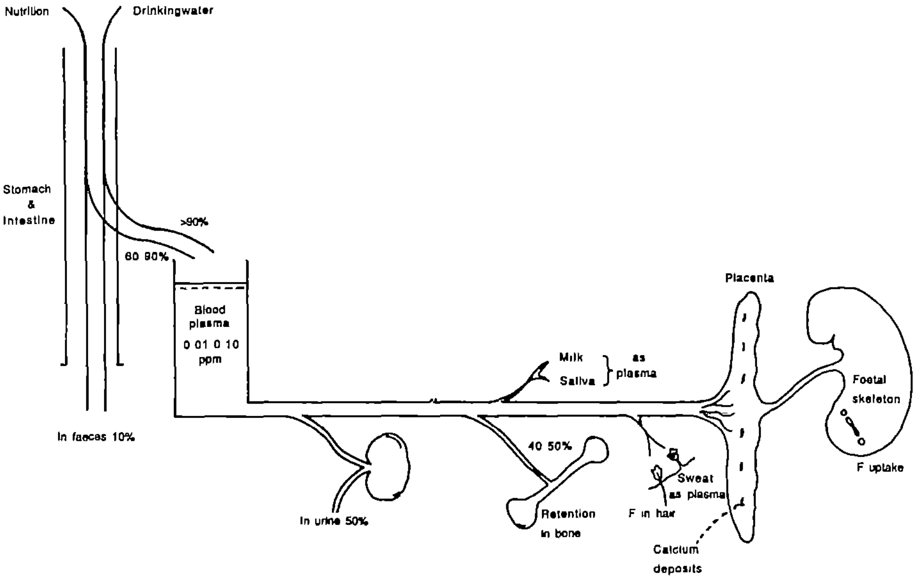
* 1 ppm (parts per million) = 1 mg/L = 0.0001%.

Forty to fifty per cent of the absorbed fluoride is, compared to chloride, rapidly excreted via the urine, a small part (5-10%) is excreted through the faeces. In sweat, mother milk and saliva the fluoride has about the same level as in plasma (Fig. 3.2). Even under extreme conditions (hot and high humidity) the fluoride excretion through these routes will be relatively small.

Figure 3.2 The absorption, excretion and retention of fluoride.



At any level of uptake, 40-50% of the absorbed fluoride is retained in the body, mainly in the bone. Further a small fraction is present in all places with high levels of calcium e.g. in teeth, especially in dentine, in renal stones and in sclerotic blood vessels.

Fluoride is retained in and on the bone in two ways. First it is absorbed in the hydration shell around the apatite crystals, and in a second phase exchanged with the hydroxylgroups in the surface of these crystals. Very slowly the fluoride ions will migrate to deeper layers of the bone. The absorbed part of the fluoride can still be released. However, the fluoride bound in the crystals is released only in case of osteoclastic resorption of bone. The release of the absorbed fluoride is a very rapid process, for the bound fluoride it takes in adults at least 8 years - the half life-time of bone.

*Fluoride levels in plasma*

The fasting fluoride level of plasma is in equilibrium with the fluoride absorbed at the bone surface. At a low level of fluoride absorption (< 5 mg F/day, for children < 2 mg/day) the fluoride plasma level will be less than 0.05 ppm in adults and less than 0.01 ppm in children. Because at each level of intake about 40% is retained, the fluoride concentration at the bone surface will increase and consequently, the equilibrium with plasma will gradually arrive at a higher level. On the other hand, the constant remodelling of the bone will counteract the accumulation of fluoride on the bone surface. However, when the rate of remodelling decreases with age, the fluoride content of bone may increase, leading to a higher fluoride level in plasma as well.

Limited studies in humans have shown that, if the absorption of fluoride in adults is higher than about 5 mg per day, the fasting fluoride plasma level may rise considerably. In the temperate zone a fluoride absorption of 6 or more mg/day may result in a fasting plasma level of 0.10-0.15 ppm fluoride. At a fluoride absorption of about 20 mg/day the plasma level may exceed 0.20 ppm fluoride.

## DENTAL FLUOROSIS

The cause of dental fluorosis is a too high plasma fluoride level during the pre-eruptive mineralization of the enamel. Slight fluorosis, visible as fine white lines across the enamel surface, is probably caused by daily peaks of more than 0.10 ppm fluoride in the plasma fluoride level. The more severe forms of fluorosis are caused by a (continuous) high fasting fluoride plasma level (0.10-0.30 ppm). In these cases the porous hypomineralized enamel takes up, after eruption into the mouth, foreign material from drinking water and foodstuffs such as manganese and iron, leading to a brownish discolouration. Local loss of the poorly formed enamel is caused by surface damage during function.

Daily peaks of fluoride are caused by the daily uptake of too much fluoride in one (e.g. fluoride tablets and fluoride toothpaste at the same time). Severe fluorosis is caused by a high overall fluoride consumption from drinking water. In subtropical areas, with a much higher water consumption, a fluoride concentration of 2-3 ppm will result in high fasting fluoride plasma levels and, consequently, in severe dental fluorosis in a large section of the population.

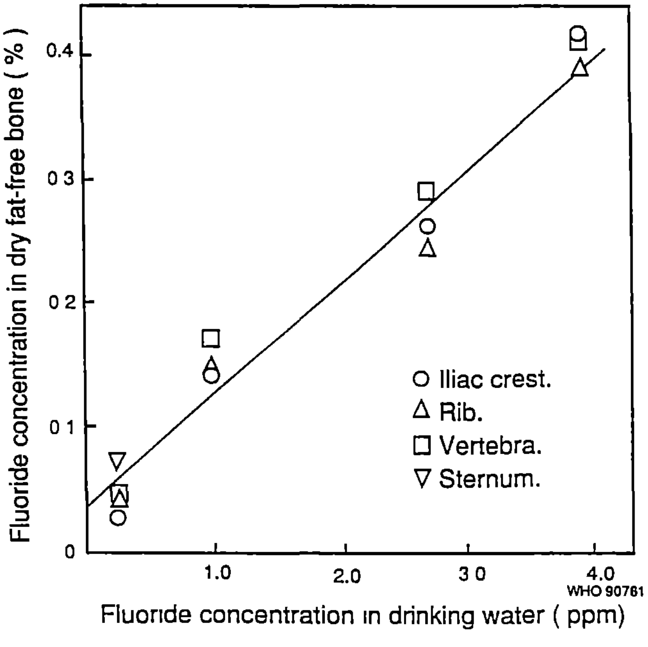
Fluorosis of the primary teeth is far less observed than in the permanent dentition. The reason for this is that the visible part of the enamel of the primary incisors and first primary molar is formed during the first six months of life, the period of the milkdiet which is low in fluoride. In the second primary molar, completed at eleven month, more fluorosis is seen. (At very high fluoride levels during pregnancy the foetal bone, surrounding the forming enamel, may have taken up so much fluoride that under osteoclastic activity so much fluoride is liberated that fluorosis of the primary incisors is possible.)

Dental fluorosis formed in early childhood (in incisors up to 5 years of age, molars and premolars up to 12 years of age) is a life long handicap. Apart from the cosmetic handicap, the severe forms of fluorosis may lead, in young adults, to a functional problem: a severe abrasion of the fluorotic biting surfaces of molars.

## SKELETAL FLUOROSIS OR BONE FLUOROSIS

Skeletal fluorosis in humans was first extensively described in 1937 by Roholm in Denmark as occupational disease. Workers in aluminium factories melting cryolite ($Na_2AlF_6$) from Greenland were the victims. High concentrations of fluoric acid and dust led to a daily fluoride absorption of 20-80 mg. After four years, fluoride concentrations in the bone of 5000-6000 ppm (0.5-0.6% F) were observed. Fluorosis with clinical symptoms was diagnosed in labourers who had been working in the factory for 10 or more years.

Figure 3.3 Relation of fluoride concentration in human bones to fluoride concentration in drinking water (After Zipkin et al., 1958; In: World Health Organization, 1970)

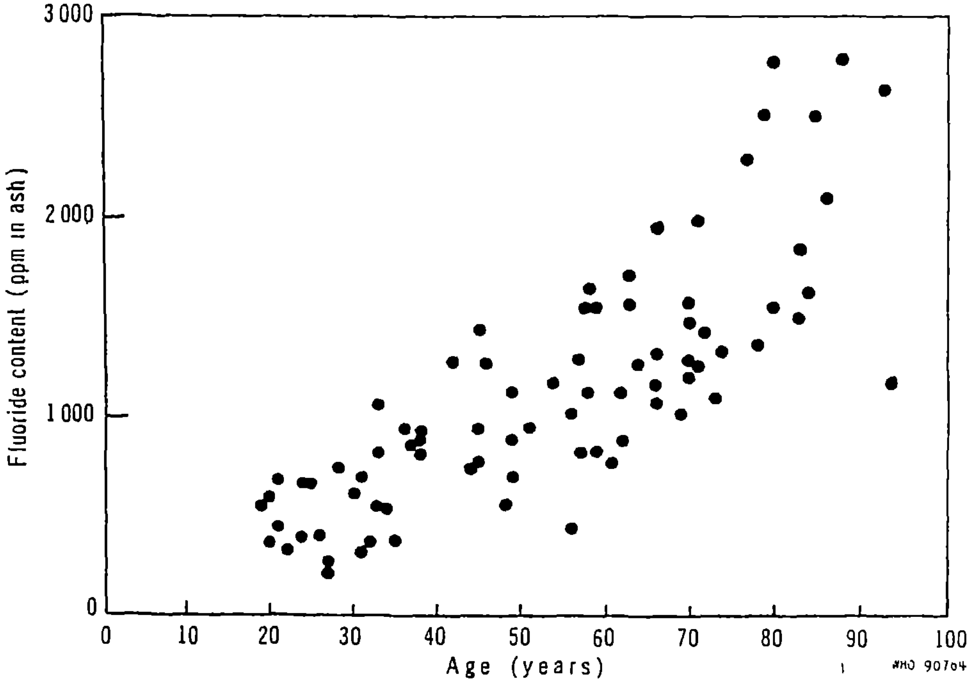


*Endemic fluorosis*

Studies in the U.S.A. demonstrated in adults a linear correlation between the fluoride concentration in the drinking water and in the human bone (Fig. 3.3). At a fluoride concentration in the water of 1.0 ppm, bone will slowly increase in fluoride and contains at 40 years of age 0.1% or a total bone fluoride content of ± 7.5 g. After the age of 50 years, when the rate of remodelling decreases, the fluoride concentration increases more steeply (fig. 3.4). However, the problem is not the relatively high fluoride concentration of bone, but the simulation of non-functional calcifications at atypical places.

Endemic fluorosis is caused by a high fluoride content of drinking water. Two levels of skeletal fluorosis can be differentiated: asymptomatic osteosclerosis and crippling skeletal fluorosis.

Figure 3.4 Fluoride content of femoral compacta from humans of different ages living in districts supplied with drinking water containing < 0.5 ppm F. (After Weatherall, 1966; In: World Health Organization, 1970)



### *Asymptomatic osteosclerosis*

At 6-8 ppm fluoride in the drinking water in the U.S.A., 10-15% of the population shows on X-ray examination a denser bone (osteosclerosis) without any clinical symptoms. However, the fasting plasma fluoride level is raised, but is still below 0.20 ppm. In areas with 4 or less ppm fluoride in the drinking water, no osteosclerosis caused by fluoride was found.

In the tropics and sub-tropics, however, at a much lower level of fluoride in the drinking water, osteosclerosis will be present. The causative factor is the high plasma fluoride level. This level is relatively higher because:

- the water consumption and consequently the fluoride absorption and retention is higher;
- an important fraction of the water is lost without fluoride: evaporation through the skin and the lungs;
- much water is excreted with little fluoride (0.1-0.2 ppm): sweat,
- hard manual labour will aggravate the above points, all leading to higher retention of fluoride and higher plasma levels.

In heat-workers (glass industry) in The Netherlands (1 ppm fluoride in drinking water) these effects could be demonstrated: high water-intake, low urine fraction of excretion and increased level of serum fluoride, compared with office employees of the same factory (Cox & Backer Dirks, 1968).

### *Crippling skeletal fluorosis*

The underlying differences between the fluoride induced asymptomatic osteosclerosis and crippling fluorosis, are the nonfunctional calcifications at atypical places ultimately leading to serious invalidity. There is certainly not an overall critical level of fluoride absorption leading to crippling fluorosis.

Probably, if the fasting plasma fluoride level exceeds 0.20-0.25 ppm, a fraction of the population older than 35-45 years of age will be confronted with the first symptoms of skeletal fluorosis. During the next decade, the number of involved persons will rise and their symptoms will aggravate.

In the tropics, fluoride concentrations in the drinking water of 4-6 ppm (or a daily fluoride absorption of > 10 mg) may cause, under the conditions as outlined in the preceding section, fasting plasma levels high enough to cause a skeletal fluorosis with serious complaints in a substantial part of the population over the age of 45 years. The first subjective symptoms are mostly pain in various joints: hands, feet, knees and spine (polyarthralgia), soon leading to delayed reactions and uncertainty in movements. The mobility becomes more and more limited. Neurological sequelae (loss of function) are observed in a percentage (10%) of the diseased persons. This is caused by mechanical compression of the spinal cord by the formation of osteophytes. Radiographic examination and postmortem studies revealed as most important pathological signs:

- increased density of bone, especially the vertebrae and the pelvis;
- bone contours becoming uneven;
- irregular periosteal growth (exostoses and osteophytes);
- calcification of ligaments;
- bony spurs in joints;
- fusions of the vertebrae (poker back!).

Our knowledge about skeletal fluorosis is still limited. We know relatively little about the secondary factors that play an additional role in the onset of bone fluorosis. There are indications that protein and calcium deficiency are aggravating risk indicators, as is hard manual labour (see Chapter 4). More information about actual absorption and excretion of fluoride, fluoride sources, and fasting plasma levels would be important tools for the longterm prognosis on a population scale Questions such as: "How many persons will become invalid within 10 or 20 years?" should be answered. Epidemiological surveys are needed to assess the magnitude of the present and future problems.

Because after the age of 50 years the rate of bone remodelling is slow, the treatment of advanced crippling fluorosis has a poor prognosis. Prevention of high fluoride absorption is probably the only effective way of master the disease.

*Kenhardt bone disease (Juvenile osteosclerosis)*

A special kind of skeletal fluorosis is described in children below 16 years of age, which was first observed in Kenhardt (South Africa) and later also in India and Senegal. Although the clinical signs (bone deformations and severe genu valgum or "knock knees") and the radiographic findings are serious, the disease does not lead to the adult skeletal fluorosis. The high level of the remodelling of bone in children makes a repair possible.* Apart from high fluoride absorption, other factors must have played an important role such as protein and calcium deficiency.

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* As an example: a girl 13 years of age, since the age of 2 years and 7 month resident of an endemic fluoride area in Ethiopia (10-15 ppm F in the drinking water) migrates to The Netherlands (0.08 ppm F), where she lost by urinary excretion more than 6000 mg fluoride over a period of one year and 8 months (Backer Dirks, 1970).

## BIOKINETICS OF FLUORIDE DURING PREGNANCY

Studies have shown that also for fluoride, the placenta acts as the organ which tries to stabilize the composition of the foetal blood supply. For fluoride this means that the fluoride level of the foetal plasma is stable at the fasting plasma fluoride level of the maternal blood. If the mother absorbs an extra doses of fluoride, leading to a peak in the plasma fluoride level, this peak is cut off by the placenta (Ericsson & Malmnäs, 1962). Table 3.1 gives the serum fluoride values at birth for maternal and foetal blood in three groups of mothers with different fluoride levels in their drinking water (Backer Dirks et al., 1976). The fluoride values for mother and foetus are similar. The fluoride concentration of the milk is low and only for the highest fluoride group somewhat elevated. The high fluoride concentration in the amnion fluid is caused by renal fluoride excretion of the foetus.

Table 3.1 Fluoride concentrations (ppm F) at birth (milk one week after birth) in three areas with different fluoride concentrations in drinking water

| Drinking water (ppm F) | N | Serum Mother | Serum Foetus | Amniotic fluid | Mother milk |
|---|---|---|---|---|---|
| 1 | 7 | 0.01 | 0.01 | 0.02 | 0.01 |
| 2-3 | 10 | 0.10 | 0.07 | 0.17 | 0.01 |
| 4-20 | 10 | 0.21 | 0.22 | 0.40 | 0.06 |

In the group with the highest fluoride absorption, the blood fluoride levels of mother and foetus are extremely high, probably unsuited for a normal bone growth. However, in this, well nourished, group no abnormalities were found that could be attributed to fluoride, also not up to the age of 20 years, except for dental fluorosis. The high remodelling rate of the skeleton and the diet low in fluoride during the first year (milk) gives a good basis for eventual repair.

*Economic effects of high fluoride levels in drinking water*

Because the fluoride absorption per day, thus the total water intake, is the key factor in the development of crippling fluorosis, it is not possible to give a fluoride concentration for water which is definitely safe. Under tropical conditions at 4-6 ppm fluoride, the safe limit for crippling fluorosis is surpassed. In risk areas probably 5-10% of the people in this fourth decade of live or older will observe symptoms of skeletal fluorosis, whereas 1% will be totally unfit for normal labour. The economic burden will be very serious in an agricultural society. In domestic cattle, the use of high fluoride drinking water leads to the same effects as in man. Especially in cows for milkproduction the effect is serious because their fluid intake is high (± 50 l per day). Cows become unable to walk and graze at their knees.

## SUMMARY

In absorption of fluoride (mainly from drinking water), the excretion (mainly with urine) and retention (mainly in bone) is described. The fluoride plasma level of blood, the result of absorption, excretion and retention, is the kermal of the ill-effects of fluoride: dental fluorosis and skeletal fluorosis. Crippling fluorosis is the most serious outcome of skeletal fluorosis in the fourth decade of life when the speed of bone-remodelling is retarded, and generally after a long exposure to high fluoride levels in drinking water.

High fluoride levels in drinking water during pregnancy may endanger the health of the foetus. Because healing of crippling fluorosis, if possible, would last decades, prevention of high fluoride absorption is probably the only effective way to avoid the disease.

Dental fluorosis, which is a live long handicap for millions of persons requires a reduction of the fluoride absorption by children.

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## 4

# EPIDEMIOLOGY OF DENTAL AND SKELETAL FLUOROSIS

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*In this Chapter the prevalence and severity of dental and skeletal fluorosis in endemic areas in developing countries are described. Furthermore, the influence of risk indicators such as drinking water, nutrition, climate and occupation are discussed, and the Chapter is concluded with some suggestions regarding the reduction of the problem of fluorosis through research activities.*

## PREVALENCE AND SEVERITY OF DENTAL FLUOROSIS

### *Indices used for measuring dental fluorosis*

Most probably, the oldest and widely used index is the one recommended by Dean (Dean Index). This index contains six categories; normal, questionable, very mild, mild, moderate and severe (WHO, 1987). An index which has been used more frequently in fluoride studies in recent years, is the one developed by Thylstrup and Fejerskov (TF-index). This index consists of nine categories and is developed on the basis of the histological features in different stages of dental fluorosis observed in areas in Tanzania with a fluoride concentration up to 21 ppm in drinking waters (Thylstrup & Fejerskov, 1978). Dean's category 'severe' (= score 4) is comparable to TF score 5-9 (Fejerskov et al., 1988).

### *Prevalence and severity of dental fluorosis in a number of developing countries*

The review of the available literature on studies reporting on dental fluorosis in developing countries has revealed that little attention has been paid to study this disorder in these countries, and that some of these studies, carried out decades ago, have some methodological flaws if judged according to current knowledge (Royal College of Physicians, 1976). This means that the results of these studies should be treated with caution.

In order to give an impression of the extent of the problem, prevalence and severity values of dental fluorosis in a number of African and Asian countries are compiled and presented in Table 4.1. The table is divided in three parts, reflecting values for children and adults, and for deciduous and permanent dentitions. It shows that dental fluorosis is prevalent in a large number of developing countries in varying degrees of severity, in the deciduous dentitions, in young and in old people. These values suggest that a large number of people in these countries daily consume water with an high level of fluoride. Such high levels of fluoride may lead to the development of skeletal and crippling fluorosis (see page 22-23).

Table 4.1 Prevalence of dental fluorosis in deciduous (A), and in permanent dentitions in children (B) and adults (C) by fluoride concentration (ppm) in the drinking water in a number of developing countries. N = number of people examined

| Area | N | Age group | ppm $F^-$ in drinking water | Affected (%) | Dean Score 4 (%) |
|---|---|---|---|---|---|
| (A) | | | | | |
| South India[a] | 387 | 0-5 | 0.6 - 5.0 | 36 | 0 |
| North India[b] | 48 | 0-5 | 9.1 - 10.7 | 82 | 15 |
| (B) | | | | | |
| Senegal[c] | 265 | 7- 9 | 1.1 - 4.6 | 69-100 | 0-60 |
| India[a] | 1115 | 5-15 | 0.6 - 5.0 | 74 | 31 |
| India[d] | 1046 | 5-15 | 1.4 - 9.7 | 23- 81 | - |
| Uganda[e] | 1002 | 5-19 | 0.1 - 3.0 | 3- 91 | 0-0.1 |
| Ethiopia[f] | 1414 | 5-19 | 1.2 - 36.0 | 71- 89 | 7-48 |
| Tanzania[g] | 119 | 9-13 | 18.6 | 100 | 87 |
| (C) | | | | | |
| India[d] | 1186 | 21+ | 1.4 - 9.7 | 10- 70 | - |
| Tanzania[h] | 530 | 40+ | 6.0 - 18.6 | 95-100 | 30-89 |

Sources: a) Pandit et al., 1940; b) Singh et al., 1963; c) Brouwer et al., 1988; d) Jolly et al., 1968; e) Møller et al., 1970; f) Haimanot et al., 1987; g) Grech, 1966; h) Grech & Latham, 1964.

In view of the results of the studies carried out in Tanzania (Table 4.2) and in Senegal (Figure 4.1), a 100 per cent prevalence of dental fluorosis in the permanent dentition in these child populations can already be expected if the water consumed contains a fluoride level between 3.5 and 4.0 ppm. In this range of fluoride concentrations, 17 per cent of the Tanzanian and 60 per cent of the Senegalese children exhibited dental fluorosis in its most severe form. Manji et al. (1986a), using the TF-index, reported a prevalence of 100 per cent in 10-15-year-old Kenyan children who had drunk water from birth which contained an

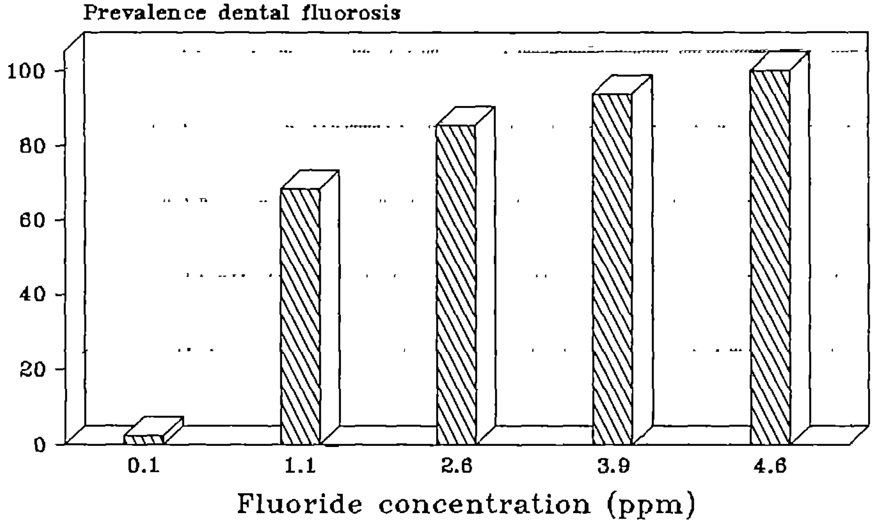
Table 4.2 Percentage distribution of children, aged 7-14 years, according to severity of dental fluorosis in the permanent dentition in three areas in Tanzania. N = number of children examined with two or more permanent teeth

| | ppm $F^-$ in water supplies | Dean Index 0.5 | 1 | 2 | 3 | 4 |
|---|---|---|---|---|---|---|
| Arusha (N = 30) | 3.5 | 0 | 3 | 10 | 70 | 17 |
| Kisongo (N = 29) | 6.0 | 0 | 0 | 0 | 38 | 62 |
| Maji ya Chai (N = 56) | 21.0 | 0 | 0 | 0 | 9 | 91 |

Source: Thylstrup, 1978.

even lower fluoride concentration, namely 2.1 ppm. The percentage of children with severe dental fluorosis (TF-score $\geq$ 5) was 50 per cent. Surely, other risk indicators then drinking water, such as climate, temperature, nutrition and occupation may have played a role in the development of dental fluorosis in the studies referred to. The contribution of these risk indicators will be discussed on page 35.

Figure 4.1 Prevalence of dental fluorosis among children aged 7-9 years in communities in Senegal with different fluoride concentrations in drinking water. (Source: Brouwer et al., 1988).



### *Fluoride concentrations in drinking water and dental fluorosis*

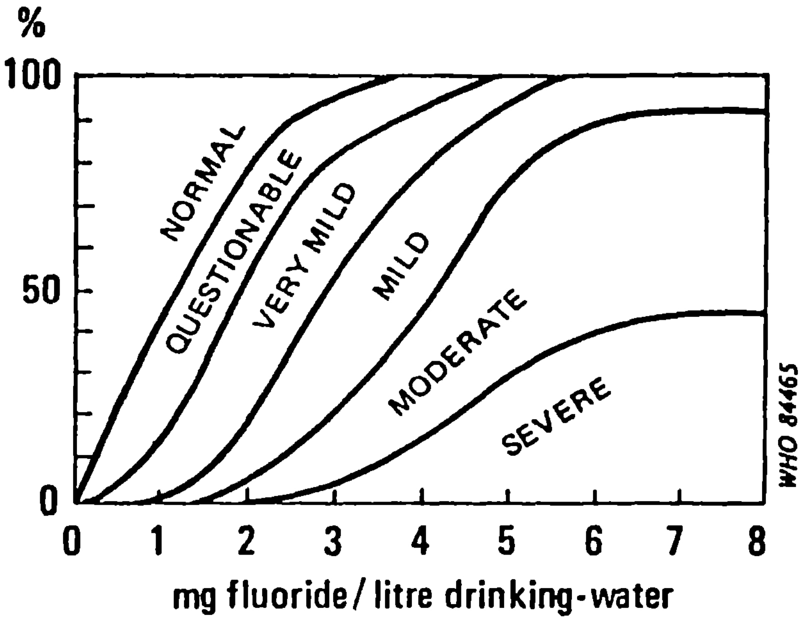
The total amount of fluoride ingested and, in particular, the amount which has become bio-available is important in the development of dental fluorosis. It is known, from the studies done by Dean in the U.S.A. in the thirties, that a direct relationship exists between the prevalence of dental fluorosis and the fluoride content in drinking water. He and his co-workers concluded that a fluoride concentration of 1 ppm in drinking water was the optimum concentration for both the reduction of dental caries and the presence of an acceptable level (10%) of a very mild form of dental fluorosis. The relationship between different levels of fluoride in the drinking water and prevalence and severity of dental fluorosis, based on the data obtained by Dean and his co-workers, is graphically represented in Figure 4.2.

The consumption pattern of drinking water differs from area to area and is dependent on a number of factors, such as temperature, climate, occupation and type of food. It is obvious that in hot climates the amount of water drunk is much higher than in colder climates. Many of the developing countries are in the Tropics where the climate is hot and dry. An assessment of the relationship between the fluoride concentration in drinking water and the prevalence of dental fluorosis in <u>hot</u> climates was therefore performed by a number of investigators. For example, Nanda et al. (1974) carried out a study amongst 16,565 school children in North-Central India. The temperature in the study area ranged from 3° to 45° C, depending on the season. Using the Dean-index they found dental fluorosis (12%) in children who had consumed water with a fluoride concentration of less than 0.4 ppm from birth (Figure 4.3). The figure further shows that the prevalence and severity of dental fluorosis increased in accordance with an increasing fluoride concentration in the drinking water. This trend was also reported for 11-15-year olds in Kenya (Manji et al., 1986b), for a child population in North-Tanzania (Table 4.2) and for 7-9-year olds in Senegal (Figure 4.1).

The findings of the studies referred to above indicate that the prevalence and severity of dental fluorosis is unequivocally directly related to the fluoride concentration in the drinking water and further, and this is very important, that very mild dental fluorosis is already prevalent in areas with a hot climate in developing countries where the fluoride concentration in drinking water has a level of 0.4 ppm and below. This finding calls for analyses of drinking water sources in the developing countries in order to identify the areas with a high level of fluoride.

Results of such an exercise have been reported for Kenya (Nair et al., 1984). Their study showed that the majority (61%) of the water samples of 1286 boreholes and wells from different parts in the country had a fluoride level above 1 ppm, whilst 20% were above 5 ppm. The authors concluded that there is a substantial need for partial defluoridation of many ground water supplies in Kenya.

Figure 4.2 Distribution of dental fluorosis at different levels of fluoride in drinking water according to results published by Dean (1942). (Source: WHO, 1984)



## RISK INDICATORS OTHER THAN DRINKING WATER ASSOCIATED WITH DENTAL AND SKELETAL FLUOROSIS

The contribution of the risk indicators diet and climatological season on the prevalence of dental fluorosis in 5-8-year olds in endemic and non-endemic areas is demonstrated in Figure 4.4. The average fluoride concentration in the drinking water for the non-endemic area was 0.68 (summer), 0.65 (monsoon) and 0.41 (winter), whereas the average fluoride concentration in the drinking water for the endemic area lay between 1.0 and 1.1 ppm for the three seasons. As already described earlier in this chapter, the figure shows that drinking water was the most important source of fluoride intake; ranging from about 66 per cent of the total fluoride ingested in summer to about 46 per cent in winter in the endemic area.

Fluoride concentrations in tea leaves differ from brand to brand. The contribution of tea to the total fluoride intake is further dependent on the concentration of tea in water, the amount of tea consumed and on the time used to brew tea. Looking at figure 4.4, we see that the actual contribution of tea is small, which can only be expected considering the young age of these children, and proves to be independent of the three seasons. The average milligrams of fluoride ingested per child per day was higher in the endemic than in the non-endemic area. This difference is caused by the fluoride in water. In comparison, the ingestion of fluoride by tea drinkers of all ages in the United Kingdom ranged from 0.04 to 2.7 mg per day (Duckworth & Duckworth, 1978).

The contribution of fluoride from the consumption of all other foods and liquids can not be neglected; it varied from about 26 per cent in summer and monsoon to approximately 35 per cent in winter (Figure 4.4). The types of liquid, however, were not specified but we may assume that a large part of these liquids will have consisted of drinking water, the fluoride provider. The same reasoning can be applied to foods cooked in drinking water.

Figure 4.3 Percentage distribution of 1409 Indian children by fluorosis diagnosis; children with full complements of permanent teeth who had consumed water from one source since birth. (Source: Nanda et al., 1974)

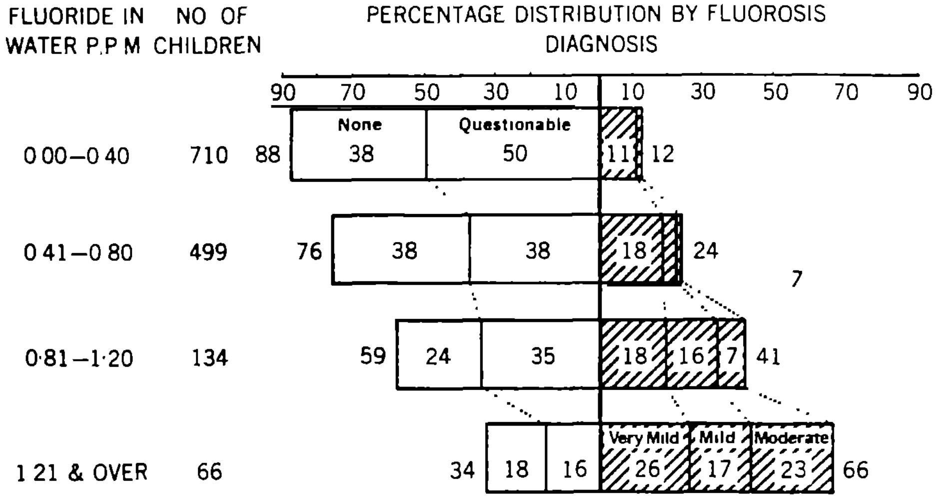
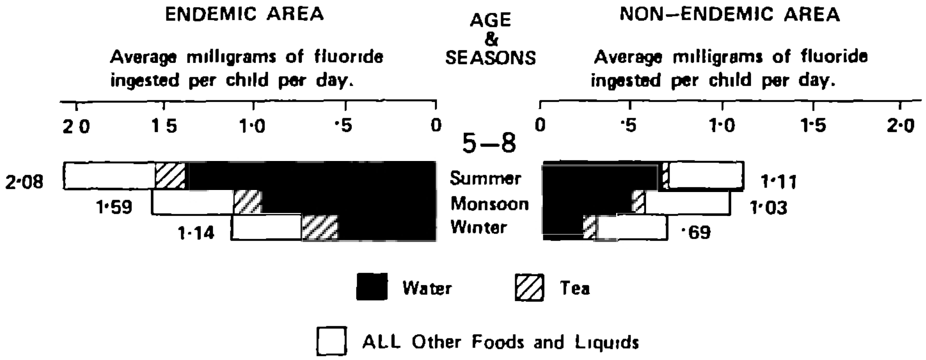


Figure 4.4 Average milligrams of fluoride ingested in 5-8-year-old Indian children per day by area, season and major dietary source. (Source: Nanda et al., 1974)



With different levels in various food items, considerable variations in individual fluoride intake may occur. This means, that subgroups with very low or very high fluoride exposures through the diet may exist (WHO, 1984).

Nutrition, frequency of eating and occupation are reported to be of influence on the total amount of fluoride absorbed (Table 4.3). This was shown in a study by Pandit et al. (1940), who carried out a house-to-house survey on severe bone manifestations in adults in South India. The diagnosis of skeletal fluorosis was based on radiological findings. In area A the fluoride concentration in drinking water ranged from 2.5 to 2.8 ppm. The people belonged to a high socio-economic group and had well-balanced meals three to four times a day. The majority of the people examined in area B were agricultural labourers who only had one meal a day, which was also deficient in nutrients. The fluoride concentration in area B was lower (1.4 - 2.2 ppm) than in area A, but the difference in the prevalence of 'bone trouble' reported was high; 4% in area A and 40% in area B. It was not clearly stated but, considering the nature of the publication, we may assume that what the authors called bone trouble, would nowadays be called skeletal fluorosis. The data were not analysed with the intention to assess associations between risk indicators and disease. It therefore remains unknown to what extent nutritional factors have actually played a role. The authors argued that agricultural labourers, because of their hard work in the field, drink much more than teachers, clerks and merchants (high socio-economic group), and in this way, get a higher fluoride intake than would be expected from the concentrations in the drinking water alone. The same authors carried out a similar study in one

Table 4.3 Possible risk indicators and the prevalence of skeletal fluorosis

| | Village A | Village B |
|---|---|---|
| ppm $F^-$ in drinking water | 2.5-2.8 | 1.4-2.2 |
| Meal | balanced | deficient |
| Frequency of meals | 3-4 | 1 |
| Type of work | light manual | hard manual |
| Prevalence of skeletal fluorosis (%) | 4 | 40 |

Source: Pandit et al., 1940.

and the same town, through which the difference in fluoride concentration in drinking water was eliminated (fluoride level was 1.2 ppm). The agricultural and other labourers doing hard manual work showed a higher percentage of bone lesions and severe manifestations (16%) than light manual labourers (9%). This suggests that occupation, nutrition and frequency of meals should be considered risk indicators.

The influence of nutrition on the development of dental fluorosis was also observed in Morocco, where malnourished children had a higher severity of dental fluorosis than well nourished children (Murray & Wilson, 1948)

Although the exact proportion of nutrition in the development of dental and or skeletal fluorosis is not well established, it is clear that the fluoride content in both water supply and food should be known in order to guarantee the safety of the use of drinking water supplies. Further research in this field is therefore necessary.

Recently, Manji et al. (1986c) have reported about altitude as a possible risk factor. They found the prevalence of dental fluorosis amongst 11-15 year olds living at 2,400 m above sea level (100%) higher than amongst peer groups at 1,500 m above sea level (78%) and at sea level (36.4%). The drinking water in these three zones contained less than 0.5 ppm fluoride. Currently, research is undertaken to confirm these findings and to investigate possible hypothesis.

## PREVALENCE OF SKELETAL AND CRIPPLING FLUOROSIS

### *Diagnostic criteria*

The early symptoms of skeletal fluorosis can only be diagnosed on radiographs. In the less advanced cases the cancellous bones show accentuation of the trabecular structure due to their thickening. Where the sclerosis is completed, the bone appears chalky and structureless. These progressive changes are best observed in the pelvis and the spine (Siddiqui, 1955). In the more advanced cases, which are called 'crippling fluorosis', there is an obvious stiffness of the spine which limits free movement and which may develop into kyphose and flexion deformities of knees. There is difficulty in walking, partly due to stiffness and limitation of movements of various joints, partly to neurologic deficit (Jolly et al., 1969).

### *Occurrence of skeletal and crippling fluorosis in developing countries*

The development of skeletal fluorosis is dependent on the total amount of fluoride ingested per day and the period this ingestion lasts. It is, therefore, important to know with what quantity of fluoride ingested per day and at what length of exposure, the development of skeletal fluorosis can be expected.

In order to study the relationship between various levels of fluoride in drinking water and the onset of bone changes, Wenzel et al. (1982) examined 112 11-15-year olds from three areas in Northern Tanzania dentally and radiographically. By correlating data of skeletal maturity and dental fluorosis, the authors showed that increasing severity of dental fluorosis was associated with a relative delay in skeletal maturity in a region with 3.6 ppm fluoride in the water supplies. They further reported that the relationship was less obvious in regions with ≤ 2.5 ppm fluoride in the drinking water, and concluded that fluoride concentrations in the drinking water above 3 ppm seemed to affect all mineralizing tissues under formation.

The length of exposure to fluoride ingestion is important in the development of skeletal fluorosis as was shown by Siddiqui (1955). In a village in India, where the fluoride concentration in the drinking water was 11.8 ppm, the author observed symptoms of skeletal fluorosis in immigrants into this village between one and four years after their arrival. This is, even taking into account the high concentration of fluoride, a very short exposure period. The author mentioned other risk indicators, such as excessive heat and a poor state of nutrition, as possible reasons for the early development of skeletal fluorosis.

The prevalence of skeletal and crippling fluorosis in adults in three countries are presented in Table 4.4. Crippling fluorosis (4%) was found in Senegalese adults who drunk water with a fluoride concentration of 3.9 ppm. This indicates that skeletal fluorosis is present in individuals living in areas in Senegal with a lower fluoride concentration in the drinking water. High fluoride levels (6.0 - 18.6 ppm) were related to a high prevalence of both skeletal and crippling fluorosis, as can be noted from a study in Tanzania (Grech & Latham, 1964).

Is the development of skeletal fluorosis restricted to adults or can it already occur in children? Grech (1966) examined 119 9-13-year-old school children from a village in Tanzania, known for its high fluoride content in the drinking water (18.6 ppm). Two of these children showed skeletal changes in pelvis and lumbar vertebrae on radiological examination. Similar skeletal changes were observed in six Indian children aged 11-14 years (Teotia et al., 1971). In this latter study the children belonged to a poor socio-economic group and had lived all their life in an endemic area where the fluoride concentration in drinking water sources ranged between 10.4 and 13.5 ppm. In four of the children examined, limited movements of spine, thoracic kyphosis and flexion deformities at the hips and knees were seen, suggesting the presence of crippling fluorosis. Christie (1980) reported data about the physical condition of 251 Tanzanian children below the age of 16 years, who lived in an area where the fluoride level in bore-hole water was 21 ppm. Of the 251 children and adolescents, 23 per cent had knock-knees, 17 per cent had bowlegs and 12 per cent had saber shins. He reported that the deformities were much more prevalent in boys than in girls and usually in a more advanced stage. Except for their deformities, these children and adolescents appeared healthy. Combinations of osteomalacia, osteoporosis and osteosclerosis result in a spectrum of bone changes from an early age. It is not known whether these bone changes will progress into the characteristic symptoms of adult skeletal fluorosis, as the community to which the children belonged moved to this region only eight years previous to the beginning of the study.

It seems that the developing skeleton may be more sensitive to fluoride toxicity than the mature one (WHO, 1984).

Table 4.4 Prevalence of skeletal and crippling fluorosis in adults in a number of developing countries

| Country | N | Age | ppm $F^-$ in drinking water | Skeletal fluorosis (%) | Crippling fluorosis (%) |
|---|---|---|---|---|---|
| Ethiopia[a] | 300 | Adults | 3.7-17.0 | 65 | 10 |
| Senegal[b] | 55 | 40-60 | 3.9 | - | 4 |
| | 42 | " | 7.4 | - | 11 |
| Tanzania[c] | 112 | 40+ | 6.0-18.6 | 87 | 13 |

Sources: a) Haimanot et al., 1987; b) Brouwer et al., 1988; c) Grech & Latham, 1964.

## CONCLUSION

From studies of the literature on the epidemiology of dental and skeletal fluorosis in developing countries it is apparent that it is not possible to predict the prevalence and severity of these disorders from the consumption of fluoridated water alone. The prevalence and severity of dental and skeletal fluorosis is dependent on a number of risk indicators like climatological condition, occupation (hard or light manual work), nutrition, frequency of food consumption, and amount of water, tea and other liquids drunk. From these risk indicators, fluoridated drinking water is the most important one. Depending on the presence of the other risk indicators, it can be concluded that:

- very mild dental fluorosis can occur in children drinking water with a fluoride level as low as 0.4 ppm;
- a 100 per cent prevalence of dental fluorosis in 10-15-year olds can occur if the water they drink contains fluoride in a concentration of 2.1 ppm;
- skeletal changes can already develop in 11-15-year olds drinking water with a fluoride level of 3.6 ppm;
- skeletal deformities may occur in children living in areas where the drinking water contains fluoride in excess of 10 ppm.

These conclusions call for research into the fluoride level of all existing and to be constructed drinking water sources in developing countries. These activities will provide the possibility to map the results of the water analysis. This mapping will assist the health and environmental sanitation authorities in planning appropriate education programmes to deal with the fluoride problem at community level. For example, one should look into the possibility to stop the use of water sources with a high fluoride content and to use water from another source instead. It is very obvious that the availability of water and the distance to the sources are important factors which should be taken into account in the planning stage. Besides this health promotion activity, the problem of dental and skeletal fluorosis can be reduced to a great extent if the test water is analysed for its fluoride content before a drinking water source is constructed; this procedure should become part of the criteria for determining water safe for drinking With the aid of geochemical mapping (aerial survey), soils with expected low levels of fluoride can be identified and this will help the construction engineers to locate the most appropriate place for carrying out exploratory drillings.

Research into simple, but effective methods to defluoridate the drinking water should be intensified (see Chapter 6).

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# 5

# DESIGN FOR A HEALTH IMPACT EVALUATION OF WATER SUPPLY WITH FLUOROSIS AS INDICATOR

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*In order to measure a positive health impact of improved water supplies, indicators such as diarrhoea and nutritial status have been used extensively. However, these indicators turned out to have shortcomings. Because of its specific characteristics, fluorosis has the potential to be a suitable health indicator. In this Chapter a design for a health impact evaluation is presented with fluorosis as indicator.*

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## INTRODUCTION

The Netherlands Assisted Project in Andhra Pradesh is part of the Indo-Dutch bilateral cooperation in the water sector. The project started in 1979. So far 191 villages, covering approximately 480.000 people, are supplied with public standpipes.

Fluorosis was one of the major criteria for inclusion of a village on the list of problem villages to be addressed by the Netherlands Assisted Project. The Indian Technology Mission estimates a total of 1079 villages in 17 districts in Andhra Pradesh with inhabitants suffering from fluorosis. The main causal factor is fluoride ingestion via water. Water samples taken in such endemic villages indicate ranges up to 29 mg/l fluoride (Government of AP/IPM, 1987).

## HEALTH IMPACT EVALUATION

### *Health Impact Evaluation of the Netherlands Assisted Project*

The opportunity to develop the idea of fluorosis as an indicator for a Health Impact Evaluation (HIE) of the project arose when project staff sought assistance to set up a HIE in accordance with project requirements. There was evidence to suggest that fluorosis could offer an interesting indicator of health impact. This impression was supported by comparing fluorosis with other indicators such as diarrhoea and nutritional status.

### *Why do a Health Impact Evaluation?*

The fact that a health impact evaluation on fluorosis could provide an interesting intellectual exercise, is in itself no argument to do such a study. Plausible reasons would be:

- to determine if the project produces the anticipated benefits;
- to establish additional inputs needed to maximize health benefits from the water supply investment.

It is generally accepted that one should only consider to do a health impact evaluation firstly, when the information is needed for project planning, sector planning or inter-sector resource allocation and secondly, when there is reasonable evidence that the project is functioning and being utilized. A third and perhaps obvious condition is that the project or programme to be evaluated is likely to demonstrate a significant impact on the outcome measure (Briscoe et al., 1986). This last condition, however, is proving particularly difficult to fulfil.

### *Why (why not) use fluorosis as an indicator in Andhra Pradesh?*

Most studies dealing with health impact of improved water supplies translate the concept of "health" into an indicator which provides a rather limited measure of absence of health, e.g. diarrhoeal disease which is studied in the most vulnerable age-groups in seasons chosen because of high incidence of diarrhoeal disease. Even so, diarrhoeal disease has been elusive as an indicator to show significant impact of improved water supply.

Fluorosis in affected villages in Andhra Pradesh is a major health problem affecting a large part of the population from school age onwards, with chronic symptoms throughout life and increasing with age. The association between fluoride levels in water used for human consumption and fluorosis is firmly established. Ingestion of high fluoride water may safely be regarded as the main cause of fluorosis in rural villages in Andhra Pradesh (Ramamohana Rao & Rajyalakshmi, 1974). It also is the main reason why an improved water supply was considered worthwhile. In other words: the rationale for improved water supply provides the indicator and the improved water can be expected to have a major effect on the indicator. These are important criteria for a successful indicator.

In addition fluorosis has some peculiar characteristics, which can be made use of in a health impact study. For dental fluorosis these are:

- the finite period of exposure risk for each tooth;
- the permanence and uniformity of symptoms;
- the relative ease with which symptoms can be measured.

These characteristics indicate the possibility to time-locate the fluoride exposure of an individual on the basis of which teeth are and which are not affected. Or, if we study similar teeth in different age groups, to compare past fluoride exposure of different birth cohorts at one point in time.

The reasons why fluorosis has not been used before as an indicator in health impact evaluations of improved water supplies seem to be threefold. Firstly, fluorosis has a limited geographical distribution compared to other water related diseases such as diarrhoea. Secondly, even where fluorosis is regarded as a major problem, typical low cost solutions for small scale water supplies are not yet sufficiently developed. (In Andhra Pradesh, piped water schemes carry low fluoride surface water to affected areas.) Lastly, since fluorosis takes many years to develop, it is probably impossible to detect an impact within the time scale in which agencies are willing to fund a health impact study.

## DESIGN FOR A HEALTH IMPACT EVALUATION OF WATER SUPPLY WITH FLUOROSIS AS INDICATOR

### *Purpose of study*

The study proposal follows the suggestions of project staff, who expressed a wish for such a study to be user-oriented and feasible for execution by regular local staff. The main use of the study would be to provide a demonstration at village level of the beneficial effects of improved (low fluoride) water to recipients. It is felt that, if successful, such a demonstration could be a good entry point for focused health education and for creating popular demand for a functioning, well maintained supply. In that way it would be quite different from other HIE's. A related objective would be to establish additional inputs needed to maximize health benefits from the water supply investment. In addition, the study would aim to determine if the project has produced the anticipated benefits, and to channel this information to decision makers.

### *Which indicator?*

The study design focuses on dental fluorosis because an impact on skeletal fluorosis cannot be measured until many years after the introduction of the improved water system, when skeletal symptoms would otherwise become manifest. Dental symptoms are much more sensitive and quick to show an impact. Moreover, the absence of dental symptoms predicts the absence of skeletal symptoms in future. A defined stage of dental fluorosis in the central incisors is suggested as an indicator. Cohorts of schoolchildren in eligible villages are proposed as the study group.

### *Type of study*

The design is for a cross-sectional study, measuring the prevalence of defined symptoms of fluorosis in different age-groups at the same time. Since working through the schools has some obvious advantages, we could think of a community survey or a school survey.

If the age groups are chosen carefully, a difference in dental fluorosis between the two age groups can be related to improved water supply. It is suggested that in addition the relative risk, or, more precisely, the odds ratio for users and non-users is calculated by distinguishing high and low scores among the postintervention group and comparing their past exposure.

### *Study group*

In order to do such a study the following conditions and limitations seem to apply:

a. Limit the study to project villages with high fluoride levels in unimproved sources and safe fluoride levels in improved sources.
b. Within the above group of villages, limit the study to villages in which the improved supply has been functioning and in use (not necessarily by all households) since commissioning.

(Note that, in adhering to condition a) and b), we forsake the idea of a study representative of all project villages. See below for discussion.)

c. Limit the study to villages with improved water supply completed eight or more years before the study. This long time span refers to the time between the beginning of the at-risk period (around birth) and the time when symptoms become visible (i.e. after eruption of the first permanent teeth, at the age of 7-8 years).
d. Limit the study to villages where one or more schools exist, the pupils of which live predominantly in areas or settlements supplied by the project.

e. Limit both pre- and postintervention study group to children who are born and bred in the selected villages.

Factors such as gender, nutritional status and socio-economic class are not accounted for in the study group. This is, among other reasons, because ingestion of water is by far the most important causal factor of fluorosis.

## DISCUSSION

One could argue that in view of the ease with which health impact can be measured when dental fluorosis is valid as an indicator, some of the limitations that normally apply for health impact evaluations are not applicable to this study. One could go one step further and argue that health impact in this case could be taken as a measure of long time functioning and utilisation of the new water supplies. If we follow this line of thinking and propose health impact as an overall measure of project achievement, it stands to reason to avoid all limitations in the study design that make the study group un-representative. One could then enter all 'fluorosis problem villages' which were in the past selected by the project for improved water supply into the sampling frame. However, some stratification according to pre-project fluoride levels would be required since there is evidence to suggest that there are wide variations in fluoride levels and that some 55% of the villages listed as 'fluorosis problems' actually have alternative water sources with fluoride levels below the Indian norm of 1.6 mg/l. This, together with the knowledge that fluorosis symptoms are related to the fluoride dose ingested, would point to a possible use of the study results for policy purpose. One could for example envisage a change of policy in which priority would be given to those villages that have no alternative for water sources with unacceptable fluoride levels.

The study design as presented here could be useful for health impact evaluations in other areas of the world where fluorosis was a criterion for providing alternative water supplies and where such alternative supplies have been successful in supplying low-fluoride water that was used by the target population for a minimum of 8 years.

The choice of 'indicator teeth' could be made according to the time span after the intervention. In the proposal at hand the (lower) central incisors are chosen simply because they are generally the first front teeth to erupt. Other teeth, however, could provide better indicators because their formation and therefore their exposure takes longer, resulting in clearer symptoms of fluorosis (Fejerskov et al., 1988).

Both dental and skeletal fluorosis would seem exceptional indicators for evaluations long after completion of water supply projects in fluorosis-stricken areas. This type of ex-post evaluation, however, is regrettably rare. This aside, it could be regarded an achievement in itself if the implementation of the study would provide an example of a low-cost, user-oriented health impact evaluation.

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## 6

# FLUORIDE REMOVAL METHODS

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*This Chapter starts with a review of current and proposed defluoridation methods and continues with a detailed description of some selected methods which show the greatest potential for application in developing countries. The Chapter is concluded with a summary of the available fluoride removal methods.*

## INTRODUCTION

Most defluoridation methods are complicated and/or expensive They require a certain level of technological skill and can be applied only in a centralized water distribution system or require expensive chemicals. In this Chapter, current practices and new developments in the field of defluoridation will be discussed in terms of level of applicability (municipal, community or household), experience gained with the method, ability to reduce the fluoride concentration to WHO norms, and suitability for developing countries A comparison between methods based on costs, removal efficiency and expected life span is not made due to insufficient information. Emphasis is laid on inexpensive and effective methods suitable for the removal of fluorides from potable waters at village and household level. Five of such methods are discussed in more detail. These five methods show the greatest potential for application in developing countries. At the end of this Chapter a list with references of the current status of defluoridation methods is given, including information on the removal mechanism of fluoride

## REVIEW OF FLUORIDE REMOVAL METHODS

The methods used for the removal of fluoride from drinking water are precipitation, adsorption and ion-exchange, osmosis, electrochemically stimulated coagulation and electrodialysis. Precipitation methods include the use of lime and alum, aluminium sulfate, gypsum, lime, magnesite, semi-calcined dolomite or calcium chloride. The adsorption or ion exchange media include activated alumina, activated bauxite, bone char, granulated bone media, tricalcium phosphate, super phosphate, zeolites, activated carbon, plant carbon, charcoal, clay pots, coconut shell and several commercially available ion-exchange resins such as Defluoron 1 and 2, Zeocarb 225, Tulsion, Carbion and Agrion O-100. Recently, the defluoridating capacity of kaolinitic clay, china clay, and serpentinite has been investigated. Furthermore, a number of electrochemical methods, reverse osmosis methods and a few new precipitation methods have been tested.

The most extensively tested and used methods include the activated alumina method (Barbier, 1984; Belle, 1984; Hendrickson, 1984; Hepp, 1979; Mazounie, 1984; Mjengera, 1988; Rubel, 1984 and 1979; Schoeman, 1987), the reverse osmosis method (Bellen, 1985; Fox, 1987; Schneiter, 1983), and the electrochemically stimulated coagulation method (Li-Cheng, 1985; Ershov, 1988; Wu, 1987). The activated alumina and electrochemically stimulated coagulation method are applied presently only in municipal plants. The reverse osmosis method is applied presently at household level. These methods are able to reduce the fluoride concentration in the treated water to below the recommended WHO level, but are only suitable for use in technologically advanced areas. Although the electrodialysis method has so far only been tested in the laboratory and at a pilot-plant, the method appears to be promising for the removal of fluoride in technologically advanced areas. Other methods which are able to reduce the fluoride concentration to below the WHO recommended level and which seems to be suitable for application in municipal plants, are methods which use lime and alum, lime alone, the phosphate-calcium mixture used in the Andco process and semi-calcined dolomite. In general, the suitability of these materials for fluoride removal is not studied to the extent as done for activated alumina, reverse osmosis and electrochemically stimulated coagulation.

For application at household and/or community level in developing countries, lime and alum, polyaluminium chloride, gypsum and fluorite, clays, granulated bone medium and bone char are suitable. With the exception of gypsum and fluorite, these materials are capable of reducing the fluoride content to below the WHO recommended level. A great advantage of the gypsum-fluorite method, however, is that the process of removal is well known and very simple in comparison with other methods.

## DEFLUORIDATION METHODS AT HOUSEHOLD OR COMMUNITY LEVEL

In this paragraph, some of the better known methods that are in current use or show greatest potential for application in developing countries are described in short. Reverse osmosis is not described in detail, because it has not yet reached the stage where it can easily be applied under primitive conditions A further development of cheap and robust systems of reverse osmosis may in the near future offer a solution to some fluorosis problems in somewhat technologically advanced areas.

### *Lime and alum method*

In this method, popularly known as the Nalgonda method, lime and aluminium sulphate are added to fluoride containing water. This results in coagulation and flocculation of the water. Thereafter, a sedimentation process takes place and eventually, the water is filtrated and desinfected as illustrated in Figure 6 1. The use of lime , about 1/20 of the alum dose, reduces the amount of alum needed (Bulusu, 1979). Some researchers have reported the use of larger amounts of lime (Mjengera, 1988), or the addition of small amounts of coagulant aids such as calcite, lime, alumina and sodium silicate, followed by a known amount of a saturated solution of filter alum ($Al_2(SO_4)_3 24H_2O$) (Rao, 1988). The method is reported to reduce fluoride to levels lower than 1 mg/l. Alkalinity, pH and the amount of fluoride in the raw water determine the amount of filter alum needed. The amount of alum needed increases with alkalinity and increasing fluoride concentration (Hendrickson, 1984; Mjengera, 1988; Rao, 1988). The optimum removal of fluoride occurs at pH 6 5 (Mjengera, 1988). On the basis of results of numerous experiments, a reagent dosage chart has been prepared (Table 6.1).

The method has been introduced and popularized in several fluorosis affected villages in the State of Andhra Pradesh, India, using a community defluoridation plant (Figure 6.1), and in Kenya, at a pilot level. From experiments and field trials it appears that 260 mg/l of alum is required to reduce the fluoride concentration from 2.05 mg/l to 1.10 mg/l (Gitonga, 1985). No figures have been presented on the amount of lime required and on the pH range at which the optimum fluoride removal was obtained. Furthermore, results from the field trials in Kenya gave a high value of residual aluminium ion in the treated water, related to the amount of alum added. An addition of 1200 mg/l alum gave a residual aluminium ion concentration of 6.0 mg/l (Gitonga, 1985). Jar test experiments with lime and alum showed a removal efficiency of 77%. Efforts were made to reduce the aluminium level in the treated water by increasing the pH. According to these experiments, it is very difficult to achieve high fluoride removal by using lime and alum (Mjengera, 1988).

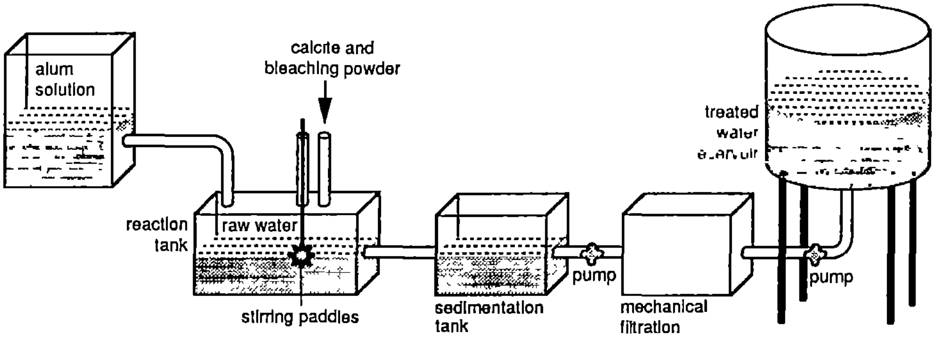
Table 6.1 Reagent dosage chart for domestic defluoridation of drinking water using the Nalgonda technique

| Fl. concentration (mg/l) | Alkalinity | ml reagent required for 10 litres water |
|---|---|---|
| 2 | 300 | 4 |
| 4 | 450 | 8 |
| 6 | 550 | 10 |
| 8 | 600 | 14 |
| 10 | 700 | 16 |
| 12 | 800 | 18 |

Source: Rao, 1988

The method has some disadvantages. For example, it cannot be used in household units, the addition of chemicals requires trained staff, large amount of alum is required to obtain a good removal efficiency, it results in high residual aluminium levels of the drinking water and a sludge disposal device is needed. The advantages of the method are reflected in the experiences and reported success with this method in field trials in India and Kenya, when applied in a community treatment plant, although there are conflicting views with respect to its efficiency.

Figure 6.1 Defluoridation of water with the Nalgonda Technique (Rao, 1988).



*Polyaluminium chloride (PAC)*

Polyaluminium chloride is a new chemical manufactured by Kemira Oy and popularly known as Kempac. The compound is an inorganic polymer with the general formula $Al_n(OH_mCl_{3(n-m)})$ (Kemira Oy, 1987). The properties of PAC are summarized in Table 6.2.

Table 6.2 Properties and composition of Polyaluminium Chloride

| Description of property | | Concentration and water values |
|---|---|---|
| Aluminium oxide | (Al2O3) | 10% |
| Chloride | (Cl) | 9% |
| Sulphate | (SO4) | 2% |
| Density | | 1200 kg/m3 |
| Viscosity | | 15 mPas at -10°C |
| | | 8 mPas at 5°C |
| | | 5 mPas at 20°C |
| Freezing point | | -15°C |
| pH | | 2.7 + 0.3 |
| Colour | | colourless or yellowish |

Source: Kemira OY, 1987

Jar test experiments were performed using raw water with a fluoride content of 19 mg/l and a pH value of 8.5. The results showed that, by increasing the dosage of PAC, the removal percentage of fluoride increased to 96.4% at a dosage of 6 ml PAC/500 ml raw water. This resulted in a fluoride concentration of 0.7 mg/l. The optimum dose of PAC added was approximately 4 ml PAC/500 ml water. At this dosage the fluoride level was reduced to 1.6 mg/l; the removal percentage was 91.6. However, the pH value dropped to 4.3 requiring adjustment to acceptable levels before being able to use as drinking water. The pH value was adjusted by adding sodiumcarbonate and magnesite. The results of these experiments indicated that it was appropriate to add sodiumcarbonate first, and thus increase the pH value of the raw water, followed by a predetermined dosage of PAC. Addition of 3 ml PAC/500 ml water resulted in a fluoride concentration of 1.5 mg/l and a pH value of 7.2. These values are acceptable for drinking water quality as stipulated by the WHO-guidelines (Mjengera, 1988). Combination of the use of PAC and that of magnesite lead to adjustment of the pH and to a reduction in the volume of PAC required for fluoride removal by more than 30%. Some results of the experiments are presented in Table 6.3.

Based on the results of the laboratory experiments, it is concluded that the high removal capacity of the chemical PAC is a big advantage of this method. Proper application of magnesite and PAC allows adjustment of the pH value and reduction of the volume of PAC required.

Table 6.3 Results of the experiments with PAC. The raw water contains 19 $mgF^-/l$

| Beaker number | (Na2CO3) added g/500 ml | pH value | PAC added ml/500 | pH value | Fl. concentr. mg/l Residual | Removed | Removal % |
|---|---|---|---|---|---|---|---|
| 1 | 0.5 | 10.0 | 1.0 | 9.0 | 11.0 | 8.0 | 42.1 |
| 2 | 0.5 | 10.0 | 2.0 | 7.3 | 4.6 | 14.4 | 75.8 |
| 3 | 0.5 | 10.0 | 2.5 | 7.2 | 4.1 | 14.9 | 78.4 |
| 4 | 0.5 | 10.0 | 3.0 | 7.1 | 1.9 | 17.1 | 90.0 |
| 5 | 0.5 | 10.0 | 3.5 | 6.8 | 1.0 | 18.0 | 94.7 |
| 6 | 0.5 | 10.0 | 4.0 | 6.6 | 0.9 | 18.1 | 95.2 |

Source: Mjengera, 1988

The disadvantages of the method can be summarized as follows. it cannot be applied to household units at present, the addition of chemicals requires careful training of the user, the method has only been tested in the laboratory, and information on residual aluminium levels in the water is not available. In years to come, the method may be suitable for application at household or community level.

*Gypsum and fluorite filter*

Recently, a new method for the removal of fluoride from water using a gypsum filterbed was tested (Schuiling, 1991; Schuiling, 1988; De Graaff, 1991). In this method, fluoride-rich waters are passed through a bed of sand-sized gypsum. During the passage of the water the calcium content of the water increases, resulting in the precipitation of fluorite. The concept is illustrated by the following chemical equation:

$$CaSO_4 2H_2O + 2F^- \text{ ---> } CaF_2 + SO_4^{2-} + 2H_2O$$

Preliminary experiments indicated that precipitation of fluorite did occur indeed, but that it was not very efficient in the absence of fluorite seed crystals. The effectiveness of the method was improved by using a filter composed of gypsum to which fluorite seed crystals were added. Batch and column experiments were carried out to determine the optimum hydraulic loading rate, its efficiency, expected life span of the filter and the optimum gypsum to fluorite ratio After the batch and column experiments were finished, samples of the fluorite-gypsum mixture were taken from the filter and investigated using scanning electron

Figure 6.2 Laboratory set-up for the defluoridation of drinking water (Schuiling, 1990)

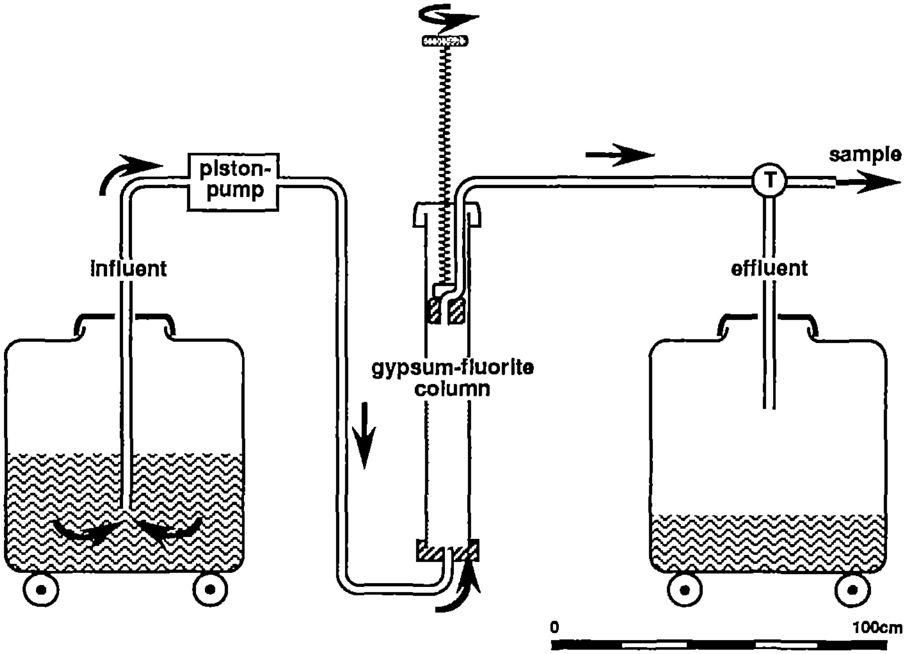
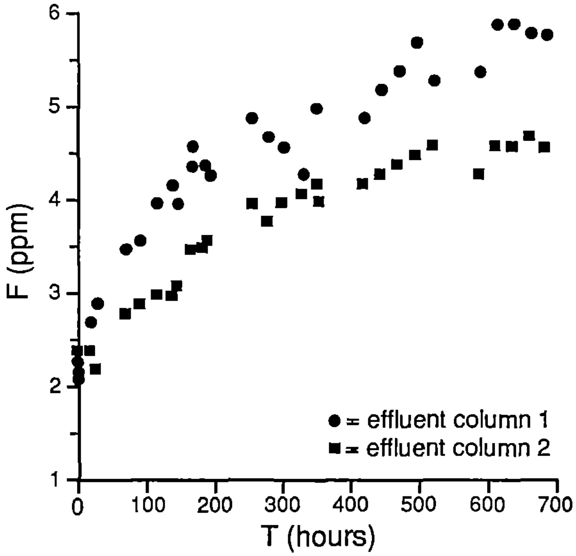


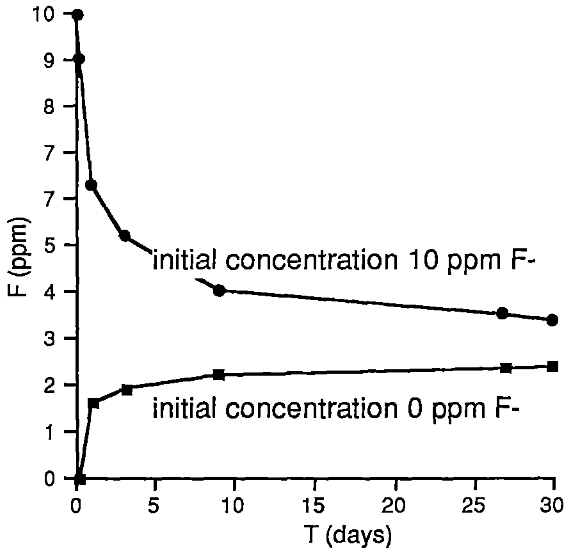
Figure 6.3 Fluoride concentration in column experiment (De Graaff, 1991)



microscopy. Column experiments were carried out using a gypsum-fluorite filter through which raw water containing 10 mgF$^-$/l was passed. In Figure 6.2, the laboratory set-up for the defluoridation of drinking water is illustrated. The concentration of fluoride in the effluent was measured (Figure 6.3). The results showed that, initially, the fluoride concentration dropped to values of approximately 2 mg/l, but, after a certain time, increased again and stabilized eventually at a value of approximately 4 à 4.5 mg/l.

Further experiments were performed with different levels of fluoride in water. A batch experiment with demi-water, containing no fluoride, in contact with a gypsum/fluorite mixture reached a fluoride concentration of 2.3 mg/l. In a parallel batch experiment, starting with demi-water with 10 mg F$^-$/l, the fluoride concentration dropped to 3.3 mg/l. This suggests that the equilibrium concentration lies between 2.3 and 3.3 mg F$^-$/l (Figure 6.4). However, the fluoride concentration of the demi-water containing 10 mgF$^-$/l did not reached the equilibrium fluoride value during the column experiments. There remains a certain oversaturation with respect to fluorite.

Figure 6.4 Fluoride concentration in batch experiment (De Graaff, 1991)



In terms of practical applications, the results mentioned above indicate that, using a gypsum-fluorite filter in which a relatively large amount of water is defluoridated in a relatively short period (e.g. filter diameter: 1 m; hydraulic loading rate: 0.1 $m^3/h$; fluorite/gypsum ratio: 30%/70%; amount of water defluoridated : approximately 2 $m^3/d$), the fluoride concentration will not reach values below 4 mg/l. In addition, the use of a gypsum-fluorite filter will change the composition of the water, increases the calcium concentration to 600 mg/l and the $SO4^{2-}$ concentration to approximately 1500 mg/l.

Considering the results of the experiments it is concluded that the method is capable of defluoridating relatively large amounts of fluoride-rich waters to a level of approximately 4 à 4,5 mg $F^-$/l, but, at the same time, increases the calcium and $SO4^{2-}$ concentrations substantially. The method is relatively new and has not been tested in the field. Nevertheless, laboratory results have shown that, although it does not have the capacity to bring the fluoride concentration down to the WHO recommended level, it has the potential to reduce the occurrence of skeletal fluorosis to a large extent. The advantages can be summarized as follows: it is inexpensive, it uses materials that are often locally available in developing countries and it can be applied at household level and at community level. In situations where it is necessary to defluoridate the water to a concentration below the WHO recommended level, the method can serve as a first step in the purification process. During this first step, the bulk of fluoride is removed, after which the fluoride remaining can be removed by using a second step in the purification process, e.g. a (relatively expensive) adsorption filter.

### *Clays*

Different types of clay are used as natural adsorbents to reduce the fluoride content of water. Several laboratory experiments have been described testing the capacity of clay pot chips (Gitonga, 1985), kaolinite (Jinadasa, 1988), serpentinite (Weerasooriya, 1989) and china clay (Chaturvedi, 1988) for the removal of fluoride from water. For example, tests, carried out at the University of Nairobi, revealed that clay pot chips were able to reduce the fluoride concentration in water. The chips were obtained by breaking the pots bought from the pot makers. No further data on the use of this material have been reported since (Mjengera, 1988). Experiments with kaolinite, serpentinite and china clay have shown that, under laboratory conditions, the concentration of fluoride can be reduced from 10 mg/l to less than 1 mg/l, i.e. below the WHO recommended level in drinking water. Before the experiments started, clay material was powdered and sieved. Different mesh sizes of the tested clay materials were used, resulting in different surface areas ($m^2/g$) available for adsorption. Regarding the efficiency of serpentinite it should be noted that this material was chemically pretreated to enhance uptake of fluoride. Several earlier studies (Kulharni, 1974; Rao, 1975) have shown that the efficiency of serpentinite in removing excess fluoride from fluoride-rich water is limited and that it tends to be deactivated with repeated use.

The above mentioned experiments indicated that the physico-chemical properties, significant for the uptake of fluoride by these clays, include pH, ionic strength, the presence of competing ions such as $OH^-$, $HCO_3^-$ or $SO_4^{2-}$, and, in the case of china clay, also temperature. The time required to attain an apparent equilibrium between fluoride ions in the liquid and solid phases was very rapid. For kaolinitic clay, 95% of fluoride adsorption occurred within the first 30 minutes and 97% within 6 hours. For serpentinite, the adsorption reached a maximum value after 6 hours and for china clay, equilibrium was reached within 120 minutes at 30 °C and a pH value of 5.6. The optimum condition for fluoride uptake by the solid substrate was reached at a pH value of 5.2 for serpentinite and of 5.6 for kaolinite. The adsorption of fluoride on china clay decreased from 93% to 83% by increasing the solution pH from 3.5 to 8.5 at 30 °C. For china clay a higher temperature enhances the uptake of fluoride from the solution. With an increase in temperature of the solution from 30 °C to 50 °C, the adsorption of fluoride (10 mg/l) increased from 84% to 92% at a pH value of 6.5. In the experiments with kaolinite the concentration of salts, present in the solution, affected the concentration of fluoride adsorbed onto clay.

The results of the discussed experiments show that excess fluoride in water can be efficiently removed to levels below the WHO recommended level by the use of natural clays. In order to maximize the adsorption, the pH, temperature and or salt content should be maintained at a level predetermined through laboratory experiments. In the case of serpentinite, the material has to be chemically pretreated. Advantages of the use of natural clays are that the method appears to be relatively cheap and simple, thus making it possible to be applied for defluoridation of water at a household or community level. Disadvantages are that the method has only been tested under laboratory conditions and that application at household level requires training of the user, because the process parameters (pH, temperature and salt content) have to be maintained at a predetermined level.

### *Use of bone media in fluoride removal*

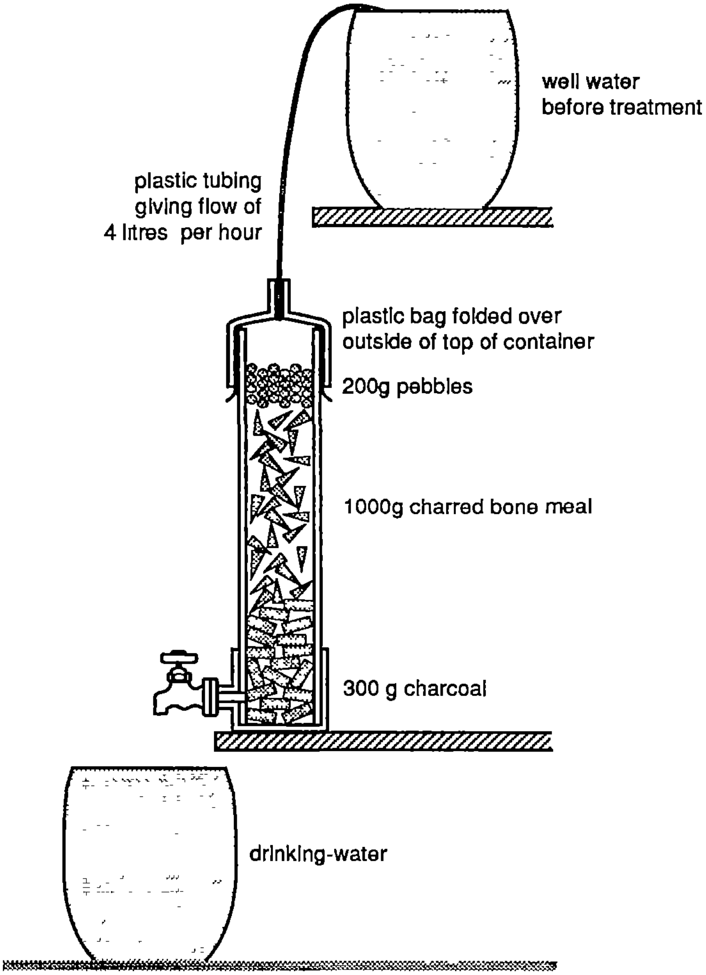
Bone media have been reported to successfully remove fluoride from water. There are two types. These are granulated bone media and bone char. The experiments with granulated bone media and bone char showed that under laboratory conditions (Cardero, 1985; Mosha, 1987; Mjengera, 1988; Phantumvanit, 1985 and 1988) and field conditions (Phantumvanit, 1988), the fluoride concentration can be reduced to less than or equal to 1 mg/l, i.e. below the WHO recommended level in drinking water.

Animal bones, grounded and charred in order to remove the organic materials, can be used as bone char. Bone char was commercially used in Britton, South Dakota USA, from 1953 to 1971 to remove fluoride from drinking water. The capacity of bone char to remove fluoride was reported to be 100 g $F/m^3$. The medium also removes arsenic which is not released during regeneration, and thus poisoning the medium. In the process, influent pH was kept to a level above 7 because bone char is soluble in acid. The medium was regenerated by sodium hydroxide, but the fluoride removal capacity of the bone char decreased during each successive regeneration and was susceptible to attack by low pH. These factors discouraged further development of the commercial use of bone char in the USA (Rubel, 1984).

A laboratory batch experiment with bone char in Norway showed that the material had very little fluoride removing capacity. The authors explained this by the fact that the medium had not been pretreated with sodium hydroxide, the common regenerant (Hendrickson, 1984).

Taking into consideration the objectives of the International Water Supply and Sanitation Decade of improving the people's health by providing them safe drinking water, the Dental Faculty, University of Bangkok, in collaboration with the Intercountry Centre for Oral Health, developed a defluoridator: in short the ICOH unit (Mosha, 1987; Phantumvanit, 1985 and 1988). This unit is based on the filtration and adsorption principle and uses charcoal and charred bone meal. Several experiments were performed to determine the required amounts and proportions of the active ingredients in relation to the amount of water that could be defluoridated before the filter material had to be replaced, the amount of fluoride retained, and the flow rate of the water. The ICOH unit consists of a container and a filter (Figure 6.5). The filter comprises of a bottom layer of 300 g crushed charcoal, mainly for adsorption of colour and odour, a middle layer of 1000 g charred bone meal, and a top layer of approximately 200 g clean pebbles to prevent the intermediate layer from floating (Phantumvanit, 1988). The bone char was prepared using purchased bone meal of 40-60 mesh size as produced for agricultural and industrial purposes. The bone was activated by heating it at a temperature of 600° C for 20 minutes. After cooling, the bone was weighed in 1 kilogram lots for use in the defluoridation unit (Mosha, 1987). During experimentation, 1000 gram bone char was able to defluoridate 480 litres of water with a fluoride concentration of 5 mg/l to less than 1 mg/l with a flow rate of 4 litres/hour. The time over which the filtering material remains active depends on the amount of water to be treated and the initial fluoride concentration (Mosha, 1987; Phantumvanit, 1988).

Figure 6.5 ICOH-defluoridator (Phantumvanit, 1988)



The ICOH defluoridator was tested in a laboratory in Tanzania by Mjengera (1988). The device was ready packed in Thailand and was only repacked in Tanzania to have the ingredients properly placed. The results obtained are illustrated in Table 6.4. The flow rate through the medium ranged between 10 and 11 litres per hour, depending on the arrangement of the unit and the volume of water to be treated. The experiment was also repeated with water from three different wells (Table 6.5). The results confirmed those from Thailand; the material is able to remove fluoride to the lowest possible level Its removal efficiency goes up to 97%.

Table 6.4 Performance of bone char in fluoride reduction

Raw water: Fluoride concentration 19.0 mg/l, pH 8.5
Filter media: 200 gram pebbles, on the surface
1000 grams bone char diameter 3.0 mm, in the middle
300 grams charcoal, at the bottom

| Contact time (min) | pH value | Volume collected (ml) | Fl. content (mg/l) | | % Fl. removed |
|---|---|---|---|---|---|
| | | | Residual | Removed | |
| 15 | 8.6 | 1260 | 1.0 | 18.0 | 94.7 |
| 30 | 8.8 | 1260 | 0.8 | 18.2 | 95.8 |
| 60 | 8.8 | 1260 | 0.5 | 18.5 | 97.4 |
| 90 | 8.7 | 1260 | 0.3 | 18.7 | 98.4 |

Source: Mjengera, 1988.

Besides being tested in the laboratory, the ICOH defluoridator was also tested in the field. A study was performed in 100 households in two districts of Chiang Mai Province in Northern Thailand, where the natural fluoride content ranges between 3 and 7 mg/l. Since this study involved community participation and acceptance, the primary health care approach was applied through the existing local health personnel, village health volunteers, public health workers at the sub-district health station and district health officers. Logistic support was received from Chiang Mai Provincial Chief Medical Office. Results of this field trial confirmed the laboratory results and showed that, with a flow rate of 4 litres an hour, the defluoridator reduced the fluoride content of 480 litres of water from 5 mg/l to less than 1 mg/l. The life span of the filter is dependent on the initial fluoride level and the amount of water consumed. The daily handling and the periodic replacement of the filter caused no problems for villagers. No special persuasion was necessary in order to convince the villagers to use the defluoridator; the provision of cleaner water with a better taste was the best incentive. The next steps in the programme are to provide the villagers with a high temperature furnace so that they can prepare the filter ingredients themselves, thereby reducing running costs, and to design a defluoridator that can provide safe water for a whole community (Phantumvanit, 1988).

Table 6.5 Results when treating water from the 3 water sources using bone char; filtration rate through the media was at an average of 10.5 litres/hour

| Name of water source | pH | Fl. (mg/l) before treatment | Volume treated litres | pH | Fl. (mg/l) Residual | Fl. (mg/l) Removed | % Fl. removed |
|---|---|---|---|---|---|---|---|
| Maij Yard B/H246/78 | 7.7 | 4.0 | 8.2 | 8.4 | 0.4 | 3.6 | 90.0 |
| Sakina B/H92/78 | 8.1 | 6.2 | 9.4 | 8.7 | 0.5 | 5.7 | 91.9 |
| Maij ya Chai (Inatake) | 8.5 | 19.0 | 8.5 | 8.8 | 0.3 | 18.7 | 98.4 |

Source: Mjengera, 1988

Bones can also be used as granulated bone media. Before the bones are used the material has to be pretreated. The preparation for granulated bone media is simple. Bones must be cleaned, all flesh and blood be removed and then degreased. This can be achieved by soaking the material in a strong alkaline solution, e.g. sodium hydroxide solution. Thereafter, the bones are washed, sundried and crushed into smaller particle sizes. This can be accomplished by a crusher or done manually. Care should be taken to ensure that the medium is free of contamination.

It has been demonstrated that granulated bone media is able to successfully remove fluoride from water in experiments in Argentine (Cardero, 1985) and in Tanzania (Mjengera, 1988). In the experiments carried out by Cardero, a contact time of half an hour was enough to allow the fluoride to chemically combine with the granulated bone media. At a production of 20 litres per day and a concentration of 10 mg/l fluoride in the raw water, the medium needed to be replaced every three months (Cardero, 1985). In Tanzania a laboratory experiment with granulated bone media was performed using the ICOH defluoridator. The preparation was done carefully to make sure that the material was free of contamination. The material was sieved, thus getting three types of filtering media of particle size: 1.0 mm, 2.0 mm and 3.0 to 6.0 mm in diameter. In the experiments 1.0 mm and 2.0 mm particle sizes were used. The bone media was filled into the ICOH defluoridation device. About 300 grams of charcoal was placed at the bottom, followed by 500 grams of 2.0 diameter bone media and than 500 grams of 1.0 mm bone media. The material was thoroughly washed with tap water. After the washing was over, the device was well fixed and thus ready for the experiment. A bucket containing 10 litres of raw water with a fluoride concentration of 19 mg/l and a pH of 8.5 was placed on top. Using a small tube, the water was allowed to flow into the defluoridator. When the defluoridator was full, 15 minutes of contact time was allowed. After this period, the water was collected and analysed for fluoride content and pH level. The experiment was

Table 6.6 Performance of granulated bones in fluoride reduction

Raw water: Fluoride concentration 19.0 mg/l, pH 8.5
Filter media: 500 grams diameter 1.0 mm, on the surface
500 grams diameter 2.0 mm, in the middle
300 grams charcoal, at the bottom

| Contact time (min) | pH value | Volume collected (ml) | Fl. content (mg/l) | | % Fl. removed |
|---|---|---|---|---|---|
| | | | Residual | Removed | |
| 15 | 9.7 | 750 | 0.5 | 18.5 | 97.4 |
| 30 | 9.8 | 750 | 0.3 | 18.7 | 98.4 |
| 60 | 9.4 | 1100 | 0.2 | 18.8 | 98.9 |
| 90 | 9.2 | 1100 | 0.1 | 18.9 | 99.5 |
| 120 | 8.3 | 1100 | < 0.1 | 18.9 | 99.5 |
| 150 | 8.0 | 1145 | << 0.1 | 18.9 | 99.5 |

Source: Mjengera, 1988

repeated several times. Each time the contact time of the water and the material was increased. The results are presented in Table 6.6 The experiment was repeated with three different types of well water (Table 6.7). The results show that granulated bone media have a removal capacity of up to 99% when particle sizes of 1 and 2 mm are used. Such a level of efficiency has not been reported earlier. The experiments showed that care should be taken when selecting proper bone Bones have to be chalky white and should exclude joints and other parts that are porous in nature (Mjengera, 1988).

The results obtained in Thailand (charred bone meal) and in Tanzania (charred bone meal and granulated bone media) with the ICOH defluoridator are in line with those obtained in Argentine (granulated bone media), although the bone treatment was different. The expected time for media replacement is also the same in both cases.

Table 6.7 Results when treating water from the 3 water sources using granulated bone; filtration rate through the media was at an average of 10.0 litres/hour

| Name of water source | pH | Fl. (mg/l) before treatment | Volume treated litres | pH | Fl. (mg/l) | | % Fl. removed |
|---|---|---|---|---|---|---|---|
| | | | | | Residual | Removed | |
| Maji Yard B/H226/78 | 7.7 | 4.0 | 10.0 | 7.9 | < 0.1 | 3.9 | 97.5 |
| Sakina B/H 92/78 | 8.1 | 6.2 | 8.3 | 7.3 | < 0.1 | 6.1 | 98.4 |
| Maji ya Chai (Inatake) | 8.5 | 19.0 | 12.6 | 8.3 | < 0.1 | 18.9 | 99.5 |

Source: Mjengera, 1988

It can be concluded that both granulated bone media and bone char can bring rich fluoride water to a level below that recommended by WHO. In order to maximize the performance, the materials have to be pretreated properly. It is recommended to activate bone char at 600° C and for granulated bone media, it is advised to select clean, non-porous bones and use particle sizes of 1 and/or 2 mm. In both cases it is recommended to treat the material with sodium hydroxide before it is used. Another big advantage of the method is that it has performed well at a household level. It may also be applied at community level, but in order to have a successful performance, community participation is essential.

## LOW-COST, LOW-TECHNOLOGY FLUORIDE REMOVAL

If only drinking water, high in fluoride concentration, is available, the water must be defluoridated. The selection of the above mentioned methods was made on the basis of their potential use in developing countries. These methods can be used in decentralized units, either at individual wells, or in households, or at the community level. They make use of cheap, often locally available materials, or of materials which have a very high fluoride removal capacity. A disadvantage of several of these methods is that they do not achieve a fluoride reduction till the WHO recommended level of 1.5 mg $F^-$/l. On the other hand, fluorosis is widespread in areas in dry climates, where the only available water may be groundwater with fluoride levels in excess of 5 mg/l. For example, in Tanzania the threshold value has been set at 8 mg $F^-$/l, because of unavailability of water with acceptable fluoride levels. A simple method which deals with excessive fluoride concentrations may already alleviate the worst problems in such a situation.

A further disadvantage of most of these methods is that they have only been tested in the laboratory using artificial waters. Neither information on the lifespan nor on the bacteriological quality is, unfortunately, available for these technologies. As most of these methods have not yet been tested under field conditions, it is not clear whether they will be acceptable to the population they are suppose to serve.

## CONCLUSIONS AND RECOMMENDATIONS

There are several well-tested defluoridation methods available, although most of them can at present only be used in technologically advanced areas. Efforts should shift from the development of ever more defluoridation methods on the laboratory scale to testing of available methods under field conditions in developing countries. Such tests will provide information on cost, lifespan, and chemical and bacteriological quality of the treated water. Even more important, however, is the social acceptability of the method and the degree of community participation without which no technology will meet with success.

In some cases the introduction of a "quick and dirty" cheap partial defluoridation method, such as the gypsum + fluorite method, which can be applied at the local level, may already eliminate most of the severe health consequences of excessive fluoride intake.

## LIST OF FLUORIDE REMOVAL METHODS

### 1. PRECIPITATION

#### *1. Lime and Alum method*

Principle: Coagulation and flocculation with lime and alum, sedimentation and filtration.

Current status: Tested in the laboratory and in the field; applicable at community level and in central treatment plants. Successful field tests have been done in rural parts of India and Kenya. Conflicting views with respect to its efficiency.

References: Hendrickson, 1984; Mjengera, 1988; Rao, 1988; Nawlakhe, 1978.

#### *2. Lime*

Principle: After lime is added to water, fluoride coprecipitates with magnesium hydroxide if enough magnesium is available, to lower fluoride levels down to less than 1 mg/l. If the magnesium concentration is too low, lime is used to form fluorite ($CaF_2$), and in these cases the fluoride concentration of high fluoride waters can be reduced to 8 mg/l.

Current status: Tested in the laboratory and in the field; is a well known process. Trained operators are needed. Poor removal efficiency at low fluoride concentrations. Applied in central treatment plants.

References: Choi, 1979; Hendrickson, 1984; Mjengera, 1988; Rabosky & Miller, 1974.

#### *3. Andco*

Principle: Addition of phosphate-calcium mixture; precipitation of fluorapatite compound.

Current status: The process is sophisticated, but the consumption of chemicals is low. Reduction to fluoride concentration below 1 mg/l is possible. The facilities for using the

Andco process are supplied by Andco Environmental Process Inc., U.S.A. as package plants.

References: O'Brien, 1983.

*4. Polyaluminium chloride*

Principle: Pretreatment by filtering the water through a magnesite filter, followed by the addition of polyaluminium chloride, reduces the fluoride level to below the WHO recommended level and adjusts pH to acceptable level.

Current status: Tested in laboratory only. High removal capacity of the chemical polyaluminium chloride. Possibility to develop the method for application at community level.

References: Mjengera, 1988.

*5. Aluminium sulphate (Alum)*

Principle: Addition of alum, flocculation and sedimentation, followed by filtration. Reduction of fluoride concentration to below 1 mg/l is possible.

Current status. Tested in both laboratory and under field conditions, well known process. Trained operators are needed. Applied in central treatment plants

References: Hendrickson, 1984.

*6 Calcium chloride*

Principle: Addition of calcium chloride and an inorganic flocculation aid (Al, Fe, Si) followed by precipitation of calcium fluoride. Reduction of fluoride to a level of about 1 mg/l is possible.

Current status: Process is not well known and well trained operators are required.

References: Biver, 1982; Hendrickson, 1984.

### 7. *Gypsum and fluorite*

Principle: Filtration through a gypsum filterbed mixed with fluorite seed crystals, followed by precipitation of fluorite. Reduction to a fluoride level of approximately 4 mg per litre.

Current status: Tested in laboratory only; well known process. Very simple method, application at household and community level is possible. Method can be used as a first step in which the bulk of fluoride is removed, followed by a more expensive adsorption method in which the fluoride remaining is removed.

References: Graaff van de, 1991; Schuiling, 1991 and 1990.

### 8. *Magnesite ($MgCO_3$)*

Principle: Magnesite is treated at a temperature of 800° C - 1000° C, thus releasing carbondioxide and obtaining magnesium oxide (MgO). The magnesium oxide is crushed and water is filtered through the medium. Experiments indicate that fluoride reduction is obtained through adsorption and ion-exchange, followed by precipitation of magnesium hydroxyfluoride.

Current status: Tested in the laboratory. The removal efficiency depends on the contact time allowed. Maximum observed removal efficiency is 81%, but the pH increases substantially, thus making the treated water unfit for domestic use.

References: Mjengera, 1988.

### 9. *Dolomite ( semi-calcined dolomite)*

Principle: In the removal of excess fluoride from water by filtration through semi-calcined dolomite particles, magnesium hydroxyfluoride is formed in the granular bed of semi-calcined dolomite, which consists of magnesium oxide and calciumcarbonate. The material can be regenerated using 1% sodium hydroxide.

Current status: Tested in the laboratory. The experiments indicate that the average residual concentration of fluoride is between 1.0 and 1.5 mg/l, which corresponds to the WHO recommended level. The capacity of the filter medium was 400g $F/m^3$, which is about two times less than the capacity of granules of activated aluminium oxide, used for water defluoridation. In view of the low cost of the granular filter medium of semi-calcined dolomite and the fact that it is a waste product in the production of refractures, its use for water defluoridation seems promising. The method can be developed for application at municipality, community and household level.

Reference: Shablovskaya et al., 1988.

## 2. ADSORPTION AND ION-EXCHANGE

### *10. Defluoron 1*

Principle: This medium is prepared by treating dried sawdust with concentrated sulfuric acid, washed with sodium carbonate or sodium chloride, followed by 1% aluminium sulfate Adsorption and ion-exchange take place in the medium, which can be regenerated with 1% alum.

Current status: The medium is reported to have poor hydraulic properties and high attritional losses. Trained technical operators are needed for media regeneration. Application in central treatment plants only.

References: Hendrickson, 1984.

### *11. Defluoron 2*

Principle: The material consists of sulphonated carbonaceous material which is loaded with aluminium ions. In principle, the method is an ion-exchange process passed through resin. Regeneration is carried out with commercial aluminium sulphate solution.

Current status: Difficult to evaluate due to limited information in literature. There is a need for skilled operators for plant operation and media regeneration.

References: Bulusu, 1979; Hendrickson, 1984; Mjengera, 1988.

### *12. Activated alumina (Compalox, Granulated activated alumina, fluidized activated alumina)*

Principle: Adsorption and filtration of water through a filterbed of activated alumina. Fluoride is removed in the filterbed through ion-exchange. The activated alumina has a high fixation capacity for fluoride and the fluoride concentration can be lowered to less than 1 mg/l.

Current status: The method has been well documented and studied. A number of publications have demonstrated its advantage over other excess fluoride removal methods. The method is quite suitable for municipal plants in developed countries and has been applied in several countries. Experiments for household defluoridation in Sweden have demonstrated that an acceptable reduction of fluoride can be achieved.

References: Barbier, 1984; Belle, 1984; Hepp, 1979; Mazounie, 1984; Mjengera, 1988; Schoeman, 1985 and 1987.

### *13. Activated bauxite*

Principle: Activated bauxite, mainly $Al_2O_3$, may be used instead of activated alumina. Adsorption and filtration of water through a filterbed of activated alumina, where fixation of fluoride occurs.

Current status: Difficult to evaluate due to limited information in literature. It may be economically feasible for some plants to use activated bauxite instead of activated alumina, although the capacity of activated bauxite for fluoride removal is less and the media losses during regeneration are greater Suitable for municipal treatment plants.

References: Hendrickson, 1984.

### *14. Tri-calcium phosphate (synthetic bone char, synthetic hydroxyapatite)*

Principle: Filtration of water through a filter of tri-calcium phosphate where fixation of fluoride occurs.

Current status: Tested in laboratory and under field conditions. Material is able to reduce the fluoride concentration to less than 1 mg/l. Application in municipal plant, community plant and household level is possible. Disadvantages are high media losses (42% per year), it is less effective compared to bone char and requires more media regeneration.

References: Naujoks, 1977; Hendrickson, 1984.

### *15. Superphosphate*

Principle: Granular superphosphate is used in a conventional ion exchange column. Regeneration is accomplished with 1% sodium hydroxide. Powdered superphosphate is used as well. Residual fluoride concentration in treated water is about 1 mg/l.

Current status: Difficult to evaluate due to limited information in literature. Well trained operators are needed A disadvantage is the need for acidification of source water prior to treatment.

References: Hendrickson, 1984.

### *16. Clays as natural adsorbents*

Principle: Different types of clay are used as natural adsorbents to lower the fluoride content of the water. Kaolinite, serpentinite, china clay and clay pot chips have been tested. Depending on the type of clay, pretreatment of material is necessary.

Current status: Tested in laboratory only. Fluoride concentration can be reduced to approximately 1 mg/l. A simple method which can be developed for application at household and community level. Uses locally available material.

References: Chaturvedi, 1988; Gitonga, 1985; Jinadasa, 1988; Mjengera, 1988; Weerasooriya, 1989.

*17. Granulated bone media*

Principle: Granulated bones react with fluoride in a similar manner as to bones and teeth of the human body. Fluoride is immobilized in the filter medium through the process of ion-exchange. The material has to be pretreated with sodium hydroxide and, preferably, particle sizes of 1 and 2 mm should be used. The bones have to be non-porous and clean. The medium has to be regenerated with sodium hydroxide (municipal plant) or replaced after a certain period, depending on the amount of water treated and the initial fluoride concentration in the water (household and community level).

Current status: The method has been tested in the laboratory and in the field. The method is simple and the medium has a very high removal capacity for fluoride. The residual concentration of fluoride can be as low as 1 mg/l. Proper pretreatment of the medium is essential for its success. Application at household and community level is possible.

References: Cardero, 1985; Mjengera, 1988.

*18. Bone char*

Principle: Same principle as granulated bone media. Pretreatment of material is necessary, activation at 600° C is recommended.

Current status: Tested in the laboratory and under field conditions. Has been applied in municipal plants (U.S.A.) and at household level (Thailand). Conflicting views with respect to its efficiency. The most recent experiments indicate that the material has a very high removal capacity for fluoride. Residual concentration of fluoride can be as low as 1 mg/l. Very simple method. Has been applied at household and municipal level and can be applied at community level as well.

References: Mjengera, 1988; Mosha, 1987; Phantumvanit, 1985 and 1988.

*19. Granulated activated carbon*

Principle: Granulated activated charcoal, pretreated with a aluminium sulphate solution, can remove fluoride from water. In the removal process, fluorides form a complex compound with aluminium ions. There is adsorption of both free and bound fluoride ions by the products of hydrolysis of the aluminium sulphate and at the surface of the activated charcoal. The optimal condition for fluoride removal occurs at pH values of 4.8 - 5.5. The adsorption of fluorides by granular activated carbon is intensified in an acidic medium. The medium can be regenerated with a solution of aluminium sulphate.

Current status: This method was tested in the laboratory and in a pilot plant. The method is capable of reducing fluoride to the WHO recommended level, but the medium has to be regenerated after a certain time. During treatment of the water, the concentration of $SO_4^{2-}$ and $Al^{3+}$ in the water increases but remains below 100 mg/l and 0.2 mg/l, respectively. The method is reported to be not inferior to the treatment method using aluminium oxide. The method has the potential for application in a municipal plant.

References: Slipchenko, 1984 and 1987.

### *20. Charcoal*

Principle: Charcoal, prepared through burning wood under natural conditions followed by soaking in alum before being used, has been reported to remove fluoride by adsorption.

Current status: The method has been tested in the laboratory. The most recent experiments indicate that the material has little or no capacity for fluoride removal. An additional disadvantage is the fact that contamination problems may arise if the charcoal is later used as fuel for cooking or heating.

References: Hendrickson, 1984.

*21. Plant carbon*

Principle: Treatment of paddy husks by digestion in 1% potassium hydroxide followed by soaking in 2% alum also produces a medium which will remove fluoride through adsorption. The removal capacity is reported to be 320 mg F/kg of medium at pH 7.0.

Current status: Difficult to evaluate due to limited information in the literature. The method has only been tested in the laboratory.

References: Hendrickson, 1984; Ndegwa, 1980.

*22. Coconut shell*

Principle: Sulphonated carbonaceous materials have been prepared from coconut shell using sulphuric and fuming sulphuric acids. Fluoride is removed by adsorption and ion-exchange.

Current status: The method has been tested in the laboratory. The defluoridation capacity is reported to be 780 mg/kg. The material was able to defluoridate the water to levels between 0.1 and 2.0 mg/l depending on pH conditions. When compared with other ion-exchange resins, the material has a lower fluoride removal capacity and, therefore, does not appear very promising. The material can be regenerated with an aluminium-sulphate solution (2-4%)

Reference: Rao, 1988.

*23. Zeolites*

Principle: Removal occurs by adsorption on the material.

Current status: Zeolites were tested and found to be impractical for fluoride removal. Different zeolites are selective for different ions, and it is possible that a zeolite exists which more effectively removes fluoride, but the investigation does not seem to be promising.

Reference: Hendrickson, 1984.

*24. Zeocarb 225*

Principle: Commercially available type of ion-exchange resin, which is selective for removing fluoride.

Current status: The material was tested in the laboratory. The removal capacity is reported to be 1650 mg/kg, which is relatively high when compared with other ion-exchange resins. The material can be regenerated with an aluminium-sulphate solution (2-4%).

Reference: Rao, 1988.

*25. Tulsion*

Principle: Commercially available type of ion-exchange resin, which is selective for removing fluoride.

Current status. The material was tested in the laboratory. The removal capacity is reported to be 960 mg/kg. The material can be regenerated with an aluminium-sulphate solution (2-4%).

Reference: Rao, 1988.

*26. Carbion*

Principle: Commercially available type of ion-exchange resin which is selective for removing fluoride.

Current status: The material was tested in the laboratory. The removal capacity is reported to be 820 mg/kg. The material can be regenerated with an aluminium-sulphate solution (2-4%).

Reference: Rao, 1988.

*27. Agrion O-100*

Principle: The material is a strongly acidic cation-exchange resin which has successfully been used for the removal of fluoride from water. Regeneration of the resin material can be done with caustic soda and alum, common salt and alum, and alum only.

Current status: The material has been tested in the laboratory and is capable of removing fluoride to concentrations less than 1 mg/l. Advanced technical skill is required.

Reference: Krishnaswamy, 1987.

*28. Filtercarbon, Shell sand, Perlites, Alginates, Filton, Hustad Marmor N2, Hustad Marmor N5.*

Principle: Adsorption and filtration of water through a filter bed of the above listed materials. Fluoride is removed in the filter bed through ion exchange.

Current status: The removal capacity of these materials was tested in the laboratory using a batch process. Some of the materials were pretreated before testing. The results of the experiments indicate that filtercarbon showed some fluoride removal ability but the amount of media needed was relatively high. The anion exchange resins Hustad Marmor N2 and N5 showed little fluoride removal ability as well. The shell sand, perlites, alginates and filton showed virtually no capacity for fluoride removal.

Very little information is available in the literature. From the results described it can be concluded that these materials are not suitable for fluoride removal.

Reference: Hendrickson, 1984.

## 3. ELECTROCOAGULATION

*29. Electrochemically stimulated coagulation*

Principle: The initial process of electric coagulation is an electrolytic one. Generally speaking, the electrode reaction is arranged in series with the following procedures.

- reaction ions or reactive materials move towards the electrode surface;
- reaction ions or reactive materials are adsorbed on the electrode surface;
- electrons are gained or lost on the electrode surface, resulting into new reactive materials;

- the reactive materials are decomposed on the electrode surface or chemically changed in the liquid in the vicinity of the surface and reactive materials are formed into new phases and conveyed from the surface of the electrodes into the solution itself.

Usually aluminium plates are used as electrodes in the fluoride removal process. Fluoraluminium complexes are formed resulting in the removal of fluoride from the treated water.

Current status: The method has been extensively tested in the laboratory in the USSR and Japan and is used in municipal plants as well. The method is capable of reducing the fluoride concentration to the WHO recommended level. The method has been well documented and studied, but most of the literature has been published in the Russian language. From the information available, it appears that the method is complicated and requires well trained operators.

References: Ershov, 1988; Li-Cheng, 1985; Matveevich, 1984.

## 4. OSMOSIS

### *30. Reverse osmosis point-of-use treatment*

Principle: In this process, the principle of membrane filtration is applied. Membrane filtration is the general name for the process that uses a semi-permeable membrane for the separation of water and contaminants. The driving process for the separation process is the water pressure. Point-of-use reverse osmosis (RO) systems are typically composed of a prefilter for sediment removal and, if necessary, of a preactivated carbon filter to remove chlorine. Behind the prefilters, a RO membrane of varying composition and design is placed, followed by a storage tank and a granular activated carbon filter, respectively. Manufacturers have added or subtracted different components, but the components listed are common to most point-of-use RO systems.

Current status: The RO system has been extensively tested in the laboratory and in the field. The experiments conducted in the laboratory confirmed that the RO system is suitable for removing fluoride. A laboratory study done by USEPA with a point-of-use RO unit that was used in the San Ysidro project showed excellent removal of beryllium, mercury, selenium (4+ and 6+), lead, cadmium, chromium (3+ and 6+), and fluoride. In San Ysidro New Mexico (U.S.A.), the point-of-use treatment with a RO system was evaluated. Seventy-three units were installed in homes, restaurants, gas stations and municipal buildings. Maintenance, filter and membrane replacement were provided by a manufacturer contracted by the village, when necessary. The RO units were able to reduce the fluoride concentration from 5.3 to less than 1 0 mg/l. Besides removing fluoride, the units were able to remove many other inorganic contaminants as well, thus improving the taste of the drinking water. A field test in Deseret Utah (U S.A.) showed that fluoride concentrations could be reduced with approximately 60% after defluoridating water which contained 1.7 mgF/l. No figures were given on the amount of water which could be defluoridated before the membrane had to be replaced.

The R.O. unit is suitable for application at household and community level. Primary concerns are the management and operation of the RO units once they have been installed. Routine sampling and monitoring is necessary in order to assure that the units continue to operate correctly.

References: Bellen, 1985; Fox, 1987; Schneiter, 1983.

## 5. ELECTRODIALYSIS

### *31 Electro-dialysis*

Principle. In the electrodialysis process water is separated from contaminants. The driving force for the separation process in the electrodialysis process is an electrical field, whereas in reverse osmosis the driving force is the water pressure. In the electrodialysis process, the water does not pass the membrane but the ions migrate through ion-selective membranes by a direct current electrical field. The permselectivity of the membrane determines the amount of fluoride that can be removed. A method to select the best membrane available has been developed. It is recommended to select a membrane with a permselectivity close to the fluorion and chlorineion.

Current status: The method has been extensively tested in the laboratory and has been applied in a pilot plant. With this method, a residual fluoride concentration lower than 1 mg/l is obtained. The method can be developed for application at a household, community and municipal level in technologically advanced areas.

Reference: Li-Cheng, 1985.

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# 7

# TOWARDS MEASURES AGAINST FLUOROSIS: A STUDY OF NECESSITY AND POSSIBILITIES

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*In the previous Chapters the problem of fluorosis has been elucidated from a physical, medical and technical side. This Chapter considers social, financial and political aspects and it is concluded with a survey of possible action following from the facts and views stated in the present and previous Chapters.*

## INTRODUCTION

These last few years experts have been studying the causes of fluoride intoxication in several regions of the third world. In 1985 Dutch scientists launched a working group on endemic fluorosis with the objective to approach the problem from different disciplines.

An important question confronting the working group was: is further study of causes and solutions of endemic fluoride intoxication only interesting from a mere technical and scientific point of view, or is fluoride intoxication a serious threat to public health, so that further research is fully justified? To answer this question, social, financial and political aspects should be considered as well as scientific data. In addition, two other underlying questions arise:

1. will implementation of measures be effective and does it need a high priority?
2. is it advisable to initiate an international programme of measures from The Netherlands?

To answer these questions discussion of the following issues is vital:

1. severity and extent of the fluorosis problem;
2. possible solutions for the fluorosis problem;
3. accordance to policy priorities established by Dutch national and local government.

## SEVERITY AND EXTENT OF THE FLUOROSIS PROBLEM

Endemic fluorosis occurs almost exclusively in economically underdeveloped areas, i.e. the third world and other countries with insufficient financial means for the supply of unpolluted drinking water. It is a well-known fact that these countries can only cope with their manifold problems when supported financially. In the perspective of other complex third world problems, implementation of measures can only be justified if the severity of the problem is being sufficiently acknowledged.

In the following the problem will be illustrated from different angles.

### *Perception of the extent of the problem*

Exposure to fluorides may cause toxic effects on human beings in the form of dental and skeletal fluorosis (Chapter 3). In general, these effects are irreversible. Policy makers in the stricken areas seem to acknowledge the problem more and more as a problem demanding serious measures. Several governments set national fluoride standards and took action, curatively as well as preventively, though often on an ad hoc basis.

### *Perception of cause and effect*

In examining the effects of fluoride exposure we may distinguish chronic and acute symptoms. Acute symptoms appear immediately after exposure to high concentrations, whereas chronic symptoms do not appear until after long-term exposure to low concentrations. However, as the effects of exposure to existing fluoride concentrations in the affected areas are chronic, in this framework only chronic effects are relevant.

As the interval between exposure and the appearance of symptoms is relatively long and part of the symptoms is not exclusively caused by fluoride intoxication, determination of the specific relationship between dose and effect is extremely difficult and will only be possible through profound research.

### *Number of people exposed*

The number of people risking exposure can only be roughly estimated. It is well-known that the number of exposed people in India and large areas of Africa is huge. Over 80 million people show symptoms of fluorosis. A Tanzanian report has mentioned a percentage of 40 and in the northern province of Tanzania it is said to be as high as 90 (Mosha & Moshi, 1982). As geochemically and climatologically large areas of the African Rift Valley (Kenya, Ethiopia, Mozambique) are most probably similar, the percentage of people exposed will be just as high. Not having the means to reduce the fluoride concentration in drinking water sources, the Tanzanian government decided to set the fluoride standard at a level approximately five times higher than the maximum fluoride level by WHO standards. Should Tanzania adopt the WHO standard, it would be forced to close at least 30 per cent of the existing drinking water sources (Bardecki, 1974). Considering the prognosis for population growth, the problem can only be expected to increase rapidly when no measures are taken The explosive rise in population leads to opening up deeper bore holes, which, as a rule, are more contaminated with fluoride (see Chapter 1).

### *Financial-economic aspects*

The financial-economic consequences of endemic fluorosis are indirect, being a mere consequence of health damage to human beings, or of contamination of crops and animals.

With our current knowledge we cannot estimate the real economic damage due to fluorosis. However, taken into consideration that the majority of people exposed is fully incapacitated according to Western standards, economic damage due to endemic fluorosis must be considerable. Therefore, further research into the economic consequences seems fully justified.

## SOLVABILITY OF THE FLUOROSIS PROBLEM

Efforts to improve living conditions in the third world are only effective, provided that these are aimed at practical solutions and that solutions are certain to be effective. This prerequisite, which is an essential condition stipulated by donor countries when establishing priorities, should be a guideline when approaching fluoride problems. In Chapter 6, various realistic options are summarized.

## ACCORDANCE WITH (INTER)NATIONAL POLICY PLANS FOR DRINKING WATER SUPPLIES

A most essential prerequisite for effective defluoridation projects is a continuous aiming at an optimal accordance with policy directives as formulated internationally in general, and in the Dutch developmental policy in particular. Although national and international efforts to improve the quality and supply of drinking water in developing countries have had impressive results, it is to be regretted that the situation for each individual has not improved at all. Due to the rising foreign debts and poor maintenance of existing supplies, it seems that, for their daily consumption, more and more people rely on water from lakes, rivers, surface waters, wells or other natural sources, which as a rule are seriously contaminated, either biologically or chemically.

In view of the enormous population growth in many developing countries, improving the supply and quality of drinking water needs continuous attention and a high priority. For this very reason the Dutch government has decided to continue its policy of initiating and stimulating supporting activities for the improvement of drinking water supplies.

Doing research and proposing measures in order to prevent and reduce fluorosis, which means to improve the quality of drinking water, would be an interpretation of the following Dutch governmental policy directives:

- sufficient water of high-quality for all individuals in the year 2000;
- improvement of living conditions in rural areas;
- development of methods with the help of locally available means and manpower;
- adjustment to existing initiatives in the sector of drinking water

In the perspective of the problems mentioned here, Dutch developmental policy is being concentrated at "the supply of drinking water". However, it is obvious that an integrated development of defluoridation programmes should be in accordance with the Dutch policy programmes of "food supply" (crop protection) and "health care".

## CONCLUSIONS AND RECOMMENDATIONS

The following conclusions and recommendations are stated by the Dutch working group on endemic fluorosis.

*General*

Because of their very diversity these problems need an overall approach. A sectorial approach to these problems, until now the usual approach, has failed to be sufficiently effective. It is to this end that the Dutch working group on endemic fluorosis has been initiated. The working group proposes:

- to restrict research activities for the time being to India and/or East Africa because of earlier exploratory research and good relationships with institutions in both areas;
- a Dutch-initiated effort to a global approach through inter-national cooperation;
- readjustment of the existing national and international standards and guidelines, those of the WHO included, for the very reason that studies have proved that exposure to concentrations lower than regulated may cause health damage.

*Sources and occurrence*

The working group proposes:

- to obtain factual insight in the occurrence of increased fluoride concentrations in problem areas, through study of existing literature as well as through carrying out water sampling programmes;
- to analyse the relationship between fluoride concentrations in drinking water and the daily intake of fluorides.

*Exposure and effects*

The working group proposes:

- to determine the severity and extent of the fluorosis problem in problem areas through qualitative and quantitative specification of symptoms;
- to describe the dose-effect relationship at different exposure levels (influence of varying daily intake, geological conditions, etc.).

*Development of measures*

The working group proposes:

- to make an inventory of previously tested defluoridation methods and to evaluate these on the basis of clearly defined criteria (costs, social aspects, etc.);
- to make an inventory of alternative ways of reducing fluoride exposure (pipe lines, rainwater reservoirs);
- to develop action programmes in consultation with local authorities.

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# FLUOROSIS

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## SUMMARY AND RECOMMENDATIONS

Certain diseases, like malaria or AIDS, affect the life of many millions of people, and programmes to combat these diseases are extensive and expensive. In contrast with the attention paid to them, only modest attention and effort is given to fluorosis, although fluorosis affects in an often dramatic way the lives and the quality of life of many tens of millions of people, mostly in some of the poorest developing countries. In this symposium on fluorosis an attempt is made for an integrated approach to the fluorosis problem, ranging from a description of the geochemical and climatological causes of fluorosis, through a description of the symptoms of dental and skeletal fluorosis to possible defluoridation schemes or alternative water supplies. It is only in passing that the possible industrial contributions to high levels of fluoride are mentioned, as these are restricted to the immediate surroundings of industries with high fluorine emissions (steel and aluminium plants, phosphate fertilizer plants, brickworks). The symposium focuses on areas with natural high concentrations of fluoride in drinking water, such as found in a number of usually semi-arid tropical countries.

Fluoride is an essential constituent of the human body, where it concentrates mainly in bones and teeth; as with many other natural substances, deficiencies as well as excesses have both negative consequences, and there is an optimal range of concentrations. As is shown in this symposium, the optimum range is not necessarily uniform for all age groups, climates, and occupational groups. For young people living in temperate climates with mainly intellectual occupations, the optimum level is likely to be higher than for poorly nourished adults in tropical countries, doing heavy manual work. From a survey of the possible intake routes of fluoride for humans, drinking water has been identified as the single dominant factor. Even in tropical countries, fortunately, the combination of the geochemical characteristics of the local rock-types and the prevailing climate leads only in a minority of cases to excessive fluoride levels in drinking water.

The major geochemical environments with high fluoride incidence are usually Pre-cambrian basement terranes with high-fluoride granites, or areas with active volcanism, such as found along the East-African Rift Valley. Sometimes 'high fluoride is also found, as in Senegal, in terranes with young marine sediments. Almost always the areas with high fluoride in groundwater have a hot and dry climate. The groundwaters will first take up fluoride from the surrounding rocks or ascending volcanic gases. Due to evaporation, the fluoride content will increase after this initial uptake. In many places, evaporation may lead to the formation of continental salt crusts in the dry season, which are often extremely enriched in fluoride. This fluoride, which is temporarily stored in soluble salt crusts during the dry season, is dissolved again during the monsoon and added to the groundwater reservoir

A careful consideration of the possible source rocks of fluoride in an area may permit the selection of well locations with lower fluoride contents It has been observed that even within one village community, different wells often show widely divergent fluoride contents, apparently as a result of differences in the local hydrogeological conditions.

Continued consumption of drinking water with fluoride levels in excess of the optimum for the local climatological and social conditions, may lead to the onset of fluorosis, which manifests itself in symptoms ranging from mild colorations of the enamel (beginning dental fluorosis) to severe malformations of the bones and stiffness of the joints (skeletal fluorosis) The mildest forms of dental fluorosis are objectionable mainly from an aesthetic (and therefore social) point of view. In the case of skeletal fluorosis, however, life itself, and certainly the quality of life for the suffering individual, is seriously affected. The WHO-norm for drinking water, presently set at 1 5 mg F/litre, is adequate in most cases, but may under adverse conditions already lead to mild forms of dental fluorosis In many arid regions, however, drinking water is such a scarce commodity, that governments have been forced to set the norm at higher levels, in order to have any drinking water at all. A case in point is Tanzania, where the norm has been set at 8 mg/liter, and where fluorosis is widespread in certain districts.

A major issue, of course, is whether there is a course of action to improve the situation Basically, there are two options, alternative water sources or defluoridation In the case of alternative water sources, this may involve bringing piped (surface) waters to the afflicted communities. Another possibility might be the drilling and tapping of other aquifers, after a careful consideration of the geochemistry of the local rocks and the hydrogeological conditions.

Defluoridation of water is technically feasible, and can routinely be carried out in central water distribution systems. Most problem areas have no central water distribution, the village communities depending on local wells. In order to provide defluoridated water under these conditions, a defluoridation method must be devised that can be applied at individual wellsites, and makes use of cheap and preferably locally available chemicals As the technical know-how at the local level is usually not available, the method should be capable to run without technical supervision for months at a time. Several solutions have been offered which seem to fulfil most of these conditions, but so far none of these methods has been routinely applied under real conditions for any length of time. A method that was developed in The Netherlands, with support from the Ministry of Development Cooperation, holds promise for application at the local level, but is unable to bring fluoride levels down to much below 4 mg/litre. Even so, this would already constitute a major improvement over the situation at many places, where only high fluoride waters are available and drinking water with over 10 mg F/litre is consumed. Mixing of partly defluoridated water with scarce surface water, or the application of an absorption method to further reduce the fluoride content of this partly defluoridated water could be envisaged.

As the symptoms of dental fluorosis can be reasonably well quantified, a health impact evaluation using fluorosis as an objective indicator is proposed. Much of the subjectivity and the methodological pitfalls of other health evaluation impact studies can be circumvented. In this symposium such an approach is outlined for close to 200 villages in India suffering from endemic fluorosis, which have in recent years been supplied with low-fluoride drinking water. The results of such a study should find wider application.

Fluorosis as an illness is not amenable to treatment. The cause of it, however, can be lifted, and there is no reason why fluorosis cannot be combated at its source, the local drinking water. As an outcome of our studies, and as an outcome of this symposium, we can formulate the following recommendations:

- to further study the geochemical behaviour of fluoride in a small number of typical geological environments, and the pathways of fluoride from rock to drinking water;
- to make an inventory of defluoridation methods, and evaluate these on the basis of criteria like costs, efficiency and social aspects;
- to include fluoride content consequently as a parameter in the planning of water supply schemes in arid tropical countries;
- to determine the severity and extent of the incidence of fluorosis in problem areas;
- to describe the dose-effect relationships at different exposure levels.

As a further recommendation it should be accepted to reduce the fluoride content of drinking water sources to below 4 mg F/liter as a first step in the strategy to reduce the problem of fluorosis to a realizable level

Although fluorosis does not catch the public eye like a number of other diseases, a fairly modest, directed effort to eliminate its causes will predictably meet with success. The elimination of fluorosis can thus become a partial implementation of the Dutch governmental policy directives, aiming at providing sufficient water of high-quality for all individuals in the year 2000, and improving the living conditions in rural areas.

# NOTES

Notes of the discussion following the symposium on Endemic Fluorosis in Developing Countries. Compiled by J. Frencken and J. Smet.

Six questions were posed, which were grouped in 3 categories:

A. 1. What are the symptoms of fluoride-deficiency.

2. Does the fluoride in toothpaste affect the environment?

B. 1. What is the economic feasible level to control dental and skeletal fluorosis?

2. Which 'human values' ought to be protected: death vs. disease vs. psyche?

3. Are there ways to prevent high fluoride contamination levels in the environment? How is it done?

C. 1. What are the ways in which people in western countries could be made aware of the seriousness of the problem of endemic fluorosis in developing countries?

Regarding A.1. **Backer Dirks**: Fluoride-deficiency does not exist' The minimum daily requirement of fluoride per capita is 0.2-0.4 mg. Most water and food does contain some fluoride, which is accumulated daily to the minimum required amount. One third of the Netherlands has been fluoridated for some time, not to fight deficiency but to reduce caries. After heavy public discussions ("Can one force people to medicate certain drugs?") fluoridation was stopped. Nowadays this discussion is being held in the USA and Australia. Other factors, such as possible detrimental effects to health of increased fluoride

levels, do also play a role in the discussion. In general, the need for measures against caries became, especially after the last war, relevant as the consumption of sweets in-creased tremendously.

Regarding A.2. **Backer Dirks**. Fluoride in toothpaste has no measurable effect on the environment. The average toothpaste contains 0.1% fluoride. For a country like The Netherlands, the yearly intake equals about 500 mg fluoride. The fluoride is partly stored in bones and is partly released into the environment, particularly in surface waters receiving waste water. Compared to the contribution from the application of superphosphate as fertilizer, which leaves yearly about 500,000 kg fluoride on the soil, the fluoride in toothpaste has no practical importance for the environment.

Other important point-sources of fluoride emission are aluminium and glass factories. In the Dutch province of Groningen, fluoride emission from a glass factory has once caused complete defoliation of an adjacent forest.

Regarding B.1 and 2. **Backer Dirks**: Studies assessing the socio-economic effects of endemic fluorosis have not been conducted sofar. There is no doubt that the productivity of a substantial number of people with skeletal fluorosis is much reduced and that the costs (expressed in time and/or money) for additional care are high. In the early stages of fluorosis, patients in rural areas in developing countries will continue to work on their fields as there are no means available to substitute the loss of income due to incapacity. At a later stage these people will be fully unable to work because they are cripple.

The psychological problems caused by dental fluorosis amongst young people but also for people at a higher economic level are very serious and should not be underestimated. These people may be reluctant to open their mouth. Often they cover their mouth when talking or smiling. The question can be posed whether it is better to be dead than crippled as a result of longtime ingestion of large amounts of fluoride?

Regarding B.3. **Smet**: There are some problems concerning the assessment of water quality. Several parameters are in use to control the quality of water and fluoride is usually not the most obvious one to have the attention of the users. Parameter effects of fluoride will only become visible over a long period of time, this in contrast to bacteriological parameters. On the other hand, if consumers see and experience the benefits of improved water supply obtained by lower fluoride levels, they would be more aware of the importance of this asset and most probably be more willing to support the operation and maintenance of water supply systems financially. It would result in an accepted and more sustainable system. This is particularly so, if people's livestock also benefits from the improved water supply.

**Vasak**: Bacteriological and other organic contamination of ground water can be prevented by measures such as protection of the well or borehole. Prevention of anorganic pollution, say fluoride, is not simple. However, if sufficient and reliable data on the geohydrological situation, including water quality data, could be collected, the chances of locating aquifers (e.g. deeper aquifers) with lower and acceptable levels of fluoride would be greater. The present main selection criterion for ground water sources is the discharge of the well (amount of water that can be pumped from the well per hour). Hydrogeological mapping is often not or not properly done in water projects.

**Smet**: The location of aquifers or other water sources with low, acceptable fluoride levels in endemic fluorosis areas is very much prefered to the treatment of (easy accessible) water with high fluoride levels. Presently, most practical

technologies for fluoride removal are still too complicated and financially not sustainable for rural areas in developing countries.
**Deleman and Pelt**: Emphasized the importance of hygiene education in relation to water supply projects.

Regarding C1. Possibilities of solving or reducing the problem of endemic fluorosis compared to other health problems in rural areas of developing countries should be highlighted:

- there are solutions available; for the fluoride in water: either by selecting water sources with acceptable fluoride content or by removing the fluoride in the water using appropriate technologies;
- the solutions (related to water supply) are financially feasible;
  1. alternative water source: probably the investment costs in the implementation phase are higher but the costs in the operation and maintenance phase of the system will be lower as no treatment system is required;
  2. if fluoride removal technologies are applied, these should fit in the specific socio-economic situation and be technically feasible;
- beneficial effects of the absence of fluorosis on man are: higher productivity, less expenditures on care, less psychological problems.

More publicity on the problem and the solutions should be given to the general public and even more to the target group of funders, Governments, NGO's, consultants, researchers etcetera, who work in areas in developing countries facing this problem.
New data on the issue should be regularly published in journals, magazines, newspapers and newsletters.

**Wöltgens**: There is an International Society on Fluoride Research (ISFR) which monitors research on this topic. The ISFR plans to organize an international congress on fluoride in Amsterdam in September 1991.