

High-fluoride Drinking Water. A Health Problem in the Ethiopian Rift Valley

1. Assessment of Lateritic Soils as Defluoridating Agents

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Purpose: High-fluoride drinking water represents a health hazard to millions of people, not least in the East African Rift Valley. The aim of the present project was to establish a simple method for removing excessive fluoride from water.

Material and Methods: Based on geological maps and previous experience, 22 soil samples were selected in mountainous areas in central Ethiopia. Two experiments were performed:

1. After sieving and drying, two portions of 50 g were prepared from each soil and subsequently mixed with solutions of NaF (500 mL). Aliquots (5 mL) of the solutions were taken at pre-set intervals of 1 hour to 30 days for fluoride analysis – using an F-selective electrode.

2. After the termination of the 30-days test, liquids were decanted and the two soil samples that had most effectively removed fluoride from the NaF solutions were dried, and subsequently exposed to 500 mL *aqua destillata*. The possible F-release into the distilled water was assessed regularly.

Results: Great variations in fluoride binding patterns were observed in the different soils. The percent change in F-concentration in the solutions, as compared to the original [F-], varied: at 1 hour from a decrease of 58% to an actual increase of 7.7%, while – at 30 days – all soil samples had caused a decrease in the F-concentration, varying from 0.5% to 98.5%. Only minute amounts of fluoride would leach from the fluoride-enriched soils.

Conclusion: Lateritic soils may remove excessive fluoride from drinking water. Methods for practical application of this principle should be tested at household level.

Key words: fluoride, defluoridation, drinking water, health

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Due to scarcity and contamination of surface waters, ground water reservoirs have increasingly become the provider of potable water around the world. This is the case not least in arid and semi-arid areas such as the East African Rift Valley.

Ground water is normally considered clean as to anthropogenic contamination (Banks et al, 1998). In a chemical sense, however, these waters are far from pure H₂O. When subjected to sufficiently sophisticated analyses, traces of all naturally occurring elements will be found in the waters.

Table 1 Adapted from Hendrickson & Vik, 1984 – Methods for fluoride removal in water

| |
|-------------------------------------|
| Precipitation methods |
| Alum |
| Lime softening |
| Alum and lime |
| Calcium chloride |
| Adsorption and ion-exchange methods |
| Activated alumina |
| Fluoridized activated alumina |
| Activated bauxite |
| Bone char |
| Tricalcium phosphate |
| Superphosphate |
| Zeolite |
| Charcoal |
| Plant carbon |
| Defluoron |

In most cases, the detailed chemical composition of the water is unknown when new wells are opened. This is unfortunate, as recent studies have indicated that many existing ground water sources may deliver water with one or more components above the maximum acceptable concentration limits (MAC) for drinking water (Reimann et al, 2003; Bjorvatn et al, 1992; Bjorvatn et al, 1994; Edmunds, 1996; Edmunds and Smedley, 1996; Frengstad et al, 2000; Midtgård et al, 1998; Misund et al, 1999; Morland et al, 1997; Morland et al, 1998; Reimann et al, 1996; Smedly et al, 1996; Sæther et al, 1995; Varsanyi et al, 1991; Williams et al, 1996).

The present project is part of a greater, multinational and multidisciplinary program, 'Fluoride in food and water, a health problem in East Africa', initiated in 1996 by the Addis Ababa University, Ethiopia, and the University of Bergen, Norway. As part of the study, water samples from 138 sources were collected in the Ethiopian Rift Valley, and analyzed at the Federal Institute for Geosciences and Natural Resources, Hanover, Germany. More than 70 elements and other parameters were assessed. According to the analyses, 78% of the wells contained one or more ingredients above the MAC values set for drinking water. If the standard for uranium (MAC 2 µg/L), recently suggested by WHO, was included, 86% of the waters would fail to pass the quality tests (Reimann et al, 2003).

The most problematic element was fluorine: 33% of all samples contained fluoride above WHO's MAC value of 1.5 mg F/L (Range 0.05-11.6 mg F/L; median 0.9 mg/L). Due to the local climatic condition daily water consumption is increased in tropical areas, and in order to avoid excessive intake, the fluoride content of drinking water should be reduced. According to a formula proposed by Galagan and Vermillion (1957) a more relevant value for the Rift Valley area should be 0.7 mg F/L. If this value was used, approximately 60% of the groundwater wells were found to contain excessive fluoride concentrations (Reimann et al, 2003).

An easily discernible consequence of excessive intake of fluoride during childhood, dental fluorosis, is endemic in the East African Rift Valley. In Tanzania, dental fluorosis ranks among the five most common nutritional-based disorders (Mosha et al, 1996). Also in other Rift Valley countries dental fluorosis is an esthetical and practical problem of growing concern to the individual and to society (Walvekar and Qureshi, 1982; Kloos and Tekle-Haimanot, 1993; Fantaye et al, 2003). Skeletal fluorosis, which is a more serious fluoride induced health problem, is frequently seen among middle-aged people with a history of long-term ingestion of harmful amounts of fluoride (Kloos and Tekle-Haimanot, 1993; Tekle-Haimanot et al, 1996).

Manifestations of dental fluorosis, once established, cannot be reversed. However, prevention can be accomplished by avoiding excessive intake of fluoride during childhood. In areas with high-fluoride drinking water, this calls for a change of water supply, or – if alternative sources are unavailable – defluoridation of the water. As demonstrated by Table 1, several methods have been proposed for reducing the fluoride content of water for household use.

The ideal method for defluoridation of high-F drinking water in developing countries should meet the following criteria:

1. Low-cost
2. Low-tech
3. Sustainable
4. Rely on locally available components.

Furthermore, defluoridation should not reduce the quality of the drinking water, e.g. by removing essential elements or introducing unwanted elements.

Previous studies have indicated that clay and clay products may have fluoride binding capacity (Murray, 1983; Moges et al, 1996; Hauge et al, 1994; Agarwal et al, 2001). The present study was made in order to test the possibility of removing ex-

Table 2 Soil and rock samples from Ethiopia

| Sample No. | Location | Coordinates (Easting/Northing) | Nearest town |
|------------|---------------|--------------------------------|------------------|
| 1 | Arbere Kete | 40° 55.64' / 09° 03.27' | Asebe Tefery |
| 2 | Buse | 40° 48.00' / 09° 10.91' | Asebe Tefery |
| 3 | Segeda | 40° 41.45' / 09° 13.09' | Meiso |
| 4 | Metehara Town | 39° 55.27' / 08° 54.19' | Metehara |
| 5 | Bulbula Shoa | 37° 07.64' / 07° 44.45' | Jima |
| 6 | Bada Bura | 37° 03.27' / 07° 46.91' | Jima |
| 7 | Busase | 36° 32.73' / 07° 31.64' | Jima |
| 8 | Chala Guma | 36° 32.73' / 07° 36.00' | Jima |
| 9 | Gibe Shoa | 37° 34.00' / 08° 14.00' | Southern Wolkite |
| 10 | Gibe farm | 37° 32.00' / 08° 14.50' | Southern Wolkite |
| 11 | Jati | 38° 53.45' / 09° 31.63' | Chancho |
| 12 | Chanco | 38° 45.82' / 09° 14.18' | Addis Ababa |
| 13 | Chager | 38° 43.64' / 09° 49.10' | Chancho |
| 14 | Wartu Abeye | 38° 49.09' / 09° 43.64' | Feche |
| 15 | Sebeta | 38° 36.00' / 08° 53.45' | Sebeta |
| 16 | Tefki | 38° 24.00' / 08° 50.18' | Sebeta |
| 17 | Dolota | 39° 21.00' / 09° 25.00' | Sheno |
| 18 | Feto | 39° 10.00' / 09° 10.00' | Sheno |
| 19 | Dukem | 39° 53.50' / 08° 47.75' | Addis Ababa |
| 20 | Dukem | 39° 53.50' / 08° 47.75' | Addis Ababa |
| 21 | Mureja | 39° 25.64' / 08° 39.92' | Wilnchiti |
| 22 | Sendefa | 39° 02.00' / 09° 09.50' | Addis Ababa |

cessive fluoride in drinking water by the use of selected Ethiopian soils.

MATERIALS AND METHODS

Based on previous experience (Bjorvatn et al, 1996; Kvalheim et al, 1999) and information from available geology maps, soil and rock samples were collected from 22 selected sites (Table 2). Most sites were localized in the mountainous areas of Oromia, in the central part of Ethiopia (Fig 1). All sites were easily accessible from the Rift Valley. In each site samples of approximately 1 kg rock and soil were collected. With due permission from the appropriate authorities the samples were sent to the University of Bergen, Norway for analysis. Geological and chemical analyses are carried out in the Department of Geology and will be reported separately.

Assessment of the soils' F-binding capacity was made in the Laboratory of Dental Research, Faculty of Dentistry, University of Bergen

The present paper reports on the laboratory tests of fluoride binding potentiality of the soil samples. Two analyses were performed:

1. Approximately 200 g of each soil (n=22) was sieved through meshes ~1x1 mm, and subsequently dried at 50°C for at least 24 h. Two 50 g portions (a and b) were prepared from each of the dried soil samples, and entered into lidded plastic containers (vol. 1 L). 500 mL of NaF solution (8.9, alternatively 10.1 mg F/L) was added to each soil sample. The containers were left unstirred at room temperature. In order to test possible changes in the $|F^-|$ of the solutions, 5 ml aliquots were removed at 1 h, 2 h, 4 h, 24 h, 48 h, 7 days, 16 days and 30 days. Fluoride-analyses were made by the use of F- selective electrodes (Orion 9609BN; Ion plus Fluoride), according to standard procedures.

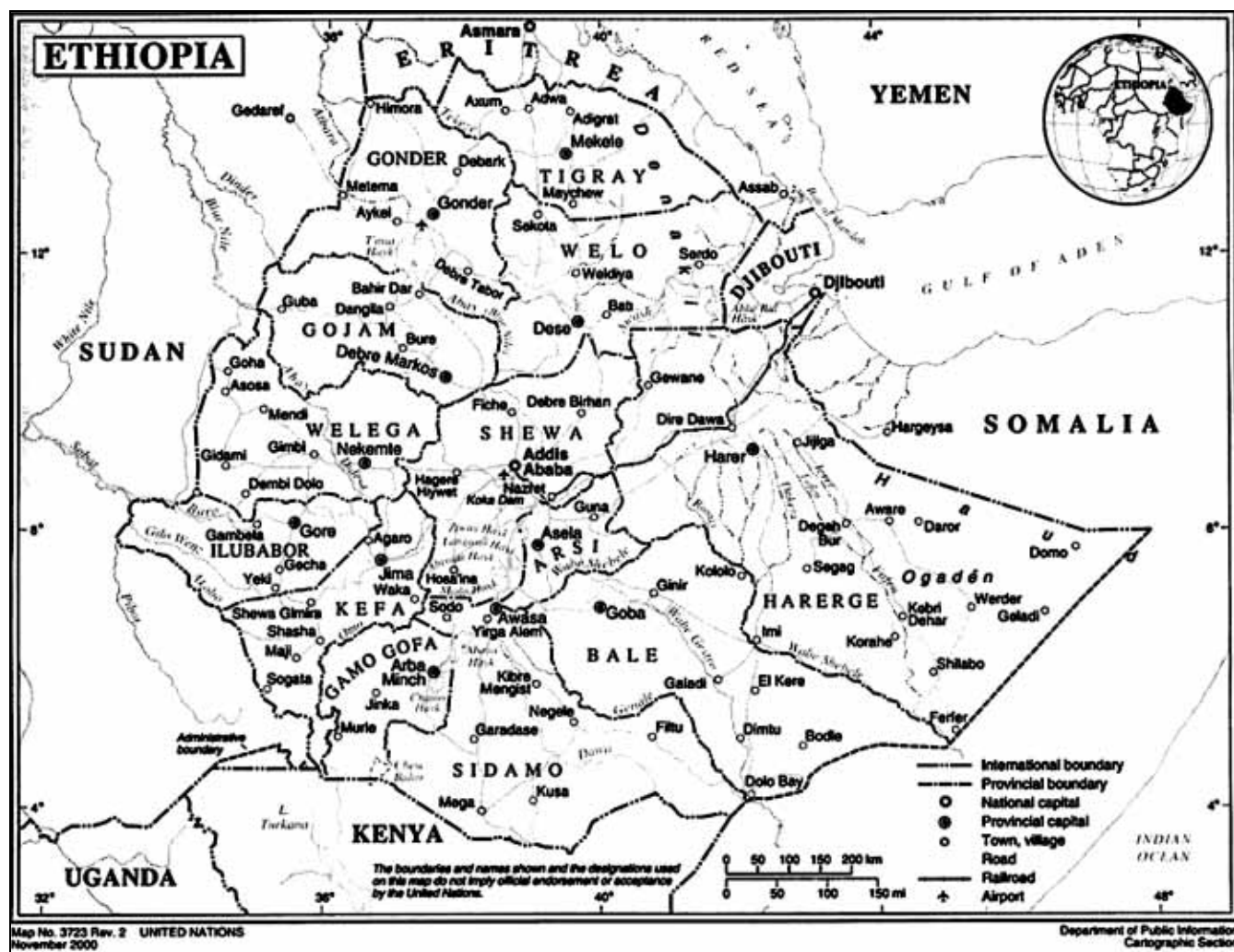


Fig 1 Map of Ethiopia.

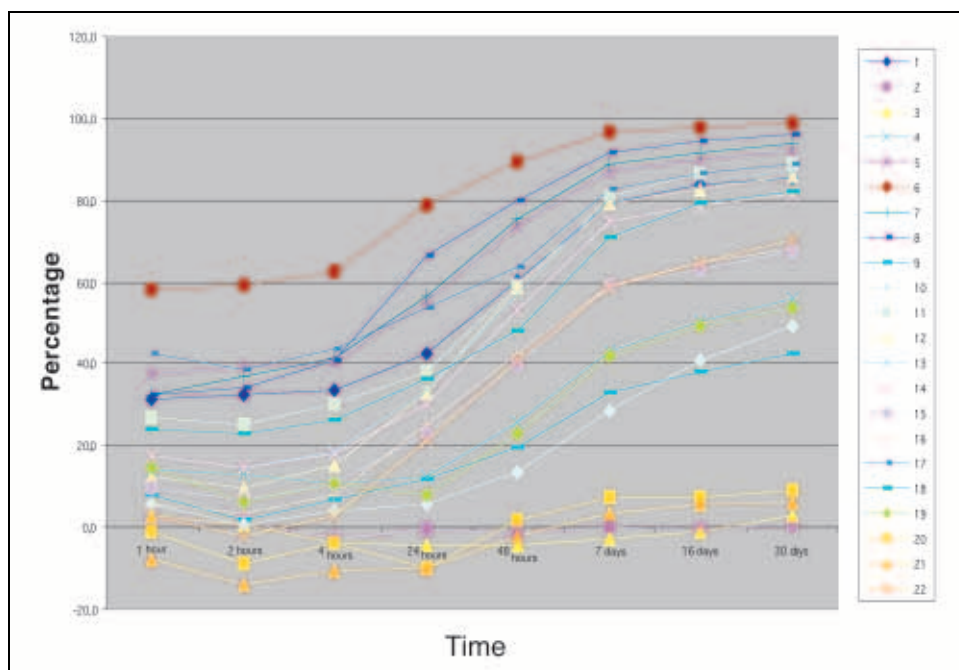
2. In order to test the possible release of F⁻ from fluoride-enriched soil, the soil-samples that had most effectively removed fluoride from the water, #6a and #8b, were removed after termination of the 30-day experiment. After drying at 50°C for 24 hours, each sample (50 g) was exposed to 500 mL distilled water. Aliquots of 5 ml were taken at regular intervals from each container and analyzed for fluoride, as previously described.

RESULTS

The consequences of exposing high-F waters to the various soils varied greatly (Fig 2). According to results at 7 days, the soils may roughly be divided into three groups: Those which showed high fluo-

ride reduction (75% or better, 9 samples), those with medium results (25–74%, 9 samples) and those with little or no effect (< 25%, 4 samples). Initially, some soils (# 2, 3, 20, 21 and 22) actually leached fluoride into the water, but in the long run even these soils seemed able to reduce the |F⁻| of the surrounding water. An interesting observation was that after the first, rather quick average reduction (18±16% at 1 h) there was a slight re-bounce at 2 h (16±19% reduction). Thereafter the average fluoride removal increased gradually to 18±19% (4 h), 28±24% at 24 h and 42±29% at 48 h. At one week's exposure to the various soils, the average reduction in fluoride content of the waters was 57±32% with a range from a 3% increase in |F⁻| to a 97% reduction. A further, slow decrease in the F⁻ concentration of the solutions was seen; at 30

Fig 2 Fluoride reduction in percentage, according to time and soil. (Mean of two analyses) Soil/solution (w/v) 1:10, Original F-conc. 8.9 alternatively 10.1 mg/L Legends refer to the soil number (Table 2).



days the average reduction was $64 \pm 32\%$, with a range from 0.5 to 98.5%.

A graphic presentation of the effect of the various soils (Fluoride reduction/increase in percentage over time, is given in Fig 2).

2. As demonstrated by Table 3, the 'invert-test' failed to show a substantial release of F- from soils # 6 and # 8, which had previously removed 98.5% and 97%, respectively, of the fluoride in waters containing 10.1 mg F/L.

DISCUSSION

Already Paracelsus (1493-1541) realized that: "All substances are poisons; there is none that is not a poison. The right dose differentiates a poison and a remedy."

However, because fluoride is a valuable anti-carries agent, many dentists from western, high-carries countries have been reluctant to consider the possible detrimental effects of fluoride. On the other hand, in some parts of the world fluoride ingestion is held responsible for a series of hard- and soft-tissue ailments, and the focus has been primarily on the negative aspects of fluoride (Susheela, 2001). This is unfortunate. Like all natural elements, fluoride may be a blessing or a curse, depending upon fluoride intake and the size and age of the individual.

Table 3

| Exposure time | F- conc. in mg/L (mean of two) | |
|---------------|--------------------------------|------|
| | #6 | #8 |
| 1 h | 0.08 | 0.15 |
| 2 h | 0.08 | 0.13 |
| 4 h | 0.09 | 0.14 |
| 24 h | 0.14 | 0.26 |
| 48 h | 0.16 | 0.30 |
| 7 days | 0.19 | 0.34 |

Millions of people, especially in East Africa, the Indian subcontinent and China, are subjected to high-fluoride drinking water and, thereby, excessive intake of fluoride. In these areas dental and skeletal fluorosis are endemic. The relevant authorities are aware of the problems, and various methods have been proposed to remove fluoride at various levels (Bower and Hatcher, 1967; Fox and Sorg, 1987; Hendrickson and Vik, 1984; Chaturvedi et al, 1988; Hauge et al, 1994; Padmasiri and Disanayake, 1995; Moges and Zevge, 1996; Zevenbergen et al, 1996; Kloos and Tekle-Haimanot, 1999; Ambelu and Faris, 1999).

Our present findings show that selected Ethiopian soils, stored for a week in contact with high-fluoride water, may reduce the water's fluoride content from ~ 10 to less than 0.5 mg F/L. Various mechanisms have been proposed to explain the fluoride binding in soil and clays (Murray, 1984). We do not know in detail processes observed in the present study, but the fluoride sorption isotherm seems to follow the Freundlich's formula (Bårdsen and Bjorvatn, 1996; Mashresha, 2002), which indicates that the fluoride binding capacity of the clay increases with increasing fluoride concentration of the water. Once we have a closer knowledge on the physico-chemical composition of the clays, a more detailed discussion of the possible mechanisms will be undertaken.

An interesting finding was that a quick fall in the average $[F^-]$ during the first hour of the experiment was followed by a slight increase during the next hour. Similar observations have been made by Hundhammer, working with Ethiopian clays at the Addis Ababa University (personal communication; 2002). These findings indicate that rather complex physicochemical mechanisms are at work, and, as discussed by Masresha (2002), the fluoride removal may e.g. involve a 'site activation process' whereby initially adsorbed fluoride ions through an expansion of the clay create additional space for incoming F^- .

The soils used in the present study were selected more or less empirically. In most cases soils sampled in the same region showed similar fluoride-binding capacity: Thus, all the four samples from the Jima area removed more than 90% of the fluoride during a 30-day period. On the other hand, two soil samples from the neighborhood of Asebe Tefery, as well as two samples taken at the same location in Dukem, varied greatly in fluoride binding capacity. According to the present study, the color of the soil might serve as a useful indicator for the selection of soils to be tested: The red (iron-containing) soils seemed particularly effective fluoride-removers. A better understanding of the soil chemistry would facilitate the selection of optimal soils.

Previous experiments have shown that agitating the water/soil system may speed up the fluoride binding in clay (Agarwal et al, 2001). According to a recent study by Larsen and Pearce (2002), an effective defluoridation of high-fluoride water may also be accomplished by heating water with brushite and calcite. In the Ethiopian Rift Valley heating

may put an extra burden on already overtaxed local forests, but alternative energy sources (e.g. the sun), might possibly be used to facilitate the fluoride removal process. The effect of heating on a fluoride removal system using laterite as the active fluoride-removing agent is now being studied.

Fluorine is prominent among the health related trace elements in the drinking water. It is also the element for which the excessive intake is most easily documented. The general public as well as central and local health authorities are keenly aware of the need for a removal of fluoride from drinking water in high-fluoride areas.

The use of lateritic clay as described above, would seem to offer a simple and inexpensive method to improve the water quality. The fluoride binding capacity of the various soil samples that have been tested, vary greatly, and the 22 samples that have been collected, may not include the 'optimal' clay. It is, however, encouraging to see that untreated clay (sample #6), under the given conditions ($w/v = 1/10$, no pre-treatment, no heating or stirring) is able to remove fluoride from the water at a rate of ~ 30 mg F/kg soil/hour, or ~ 100 mg F/kg soil/week. This is not per se a high fluoride removing capacity, but provided the same effect can be retained in defluoridation at household or village levels 100 kg laterite in one week should be able to reduce the fluoride content of 1.000 liter of water from 10 to 0.5 mg/L or less.

The feasibility of defluoridation methods depends not only on the cost, but also on the possibility of discarding or regenerating spent material. In the present experiments, the fact that very little fluoride leached from the saturated soils indicates that the fluoride/soil binding is fairly strong. The F-saturated soil may possibly be reactivated, but should, for practical and economical reasons be discarded. Soils in the high-fluoride areas of Rift Valley already contain high amounts of 'natural' fluoride (Kvalheim et al, 1998) and it is highly unlikely that laterite used for defluoridation of water will further pollute the local environment. As previously shown, storage in simple clay pots, fired at relatively low temperatures, may also reduce the fluoride content of drinking water (Hauge et al, 1994). A return to this traditional method for water storage should, therefore, be encouraged in high-fluoride areas.

Fluoride reduction does not, per se, guarantee the quality of drinking water. Other requirements, such as the bacteriological and trace element qual-

ities, should also be taken into consideration. Further studies are needed to ascertain acceptable bacteriological standards of the proposed method.

CONCLUSION

Lateritic soils from central Ethiopia may be used for removing excessive fluoride from drinking water.

Methods for practical application of this method should be tested out at the household level. More soils should be collected and tested to identify the best material for fluoride removal.

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