

FLUORIDE IN DRINKING WATER

A CAWST Literature Summary
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CAWST is a Canadian non-profit organization focused on the principle that clean water changes lives. Safe water and basic sanitation are fundamentals necessary to empower the world's poorest people and break the cycle of poverty. CAWST believes that the place to start is to teach people the skills they need to have safe water in their homes. CAWST transfers knowledge and skills to organizations and individuals in developing countries through education, training and consulting services. This ever expanding network can motivate individual households to take action to meet their own water and sanitation needs.

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Photo credit on front cover: An Indian girl displays her teeth affected by dental fluorosis. ©Ruhani Kaur/UNICEF India. Available at: www.drinking-water.org/html/en/ImageView/00000381.html

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Introduction

This literature summary provides short summaries of reports and research papers that have been conducted on fluoride in drinking water, including the following topics:

- Fluoride, occurrence, and its chemical specifications
- Health impacts of fluoride exposure
- Fluoride removal technologies, including field and laboratory studies

All documents are categorized and listed in the following order:

1. Peer reviewed, published papers
2. High quality, unpublished papers
3. Informal reports (e.g. field studies, PowerPoint presentations, press releases, personal communication)

PART 1: FLUORIDE, OCCURRENCE AND CHEMICAL SPECIFICATIONS

CATEGORY 1 – PUBLISHED PAPERS

Fluoride in Drinking-Water

Reference: Fawell, J., Bailey, K., Chilton, J., Dahi, E., Fewtrell, L. and Y. Magara (2006). Fluoride in Drinking-Water: Chapter 2, Environment Occurrence, Geochemistry and Exposure. IWA Publishing, on behalf of the World Health Organization.

Available at:

www.who.int/water_sanitation_health/publications/fluoride_drinking_water/en/index.html

Chapter 2 of this document presents information on fluoride environmental occurrence, geochemistry and exposure. Key information includes the following:

- Fluorine is one of the most reactive of all chemical elements and therefore not found as found as fluorine in the environment
- It is the most electronegative of all the elements which means it has a strong tendency to acquire a negative charge, and in solution form F⁻ ions (fluoride)
- Fluorides are found at significant levels in a wide variety of minerals, such as fluorspar, rock phosphate, cryolite, apatite, mica, hornblende
- Fluoride is found in all natural waters at some concentration
- In groundwater, low or high concentrations can occur depending on the nature of rocks and the occurrence of fluoride-bearing minerals
- The other sources of exposure to fluoride are: air, dental products, food and beverages other than water

Chapter 7 provides information on the country by country occurrence.

CATEGORY 2– HIGH QUALITY UNPUBLISHED PAPERS

GIS-Mapping of Fluoride Contaminated Groundwater in Nakuru and Baringo District, Kenya

Reference: Näslund, J. and I. Snell (2005). GIS-Mapping of Fluoride contaminated groundwater in Nakuru and Baringo district, Kenya.

Available at: <http://epubl.luth.se/1402-1617/2005/198/LTU-EX-05198-SE.pdf>

This is a Master of Science thesis carried out in Nakuru and Baringo Districts in the Rift Valley Province in Kenya. It gives information on fluoride concentration testing and GIS mapping, as well as concentrations of fluoride in ground waters in this region of Kenya.

PART 2: FLUORIDE HEALTH IMPACTS

CATEGORY 1 – PUBLISHED PAPERS

Community Perception on Fluoride and Related Health Problems in a Fluorotic Area in Ethiopia

Reference: Melaku Z. et al. (2000). Community Perception on Fluoride and Related Health Problems in a Fluorotic Area in Ethiopia. 3rd International Workshop on Fluorosis Prevention and Defluoridation of Water, Session 1 Epidemiology: 10.

Available at: www.de-fluoride.net/3rdproceedings/14-21.pdf

This paper is a summary of a survey conducted to describe the knowledge, attitude and perception regarding fluoride and related health problems of the community of Wonji-Shoa in Ethiopia. Six focus groups of 8 to 10 participants were identified to represent the various segments of the population in the area. Then a series of six focus groups discussions were carried out in a community setting led by the investigators.

The results showed that the health consequences of consuming untreated water are fairly understood but there is a knowledge gap and misperceptions concerning fluoride and its health consequences. The study recommends providing health education to the community affected by fluoride with emphasis to the women from the lower socioeconomic segment.

Effect of Tamarind Ingestion on Fluoride Excretion in Humans

Reference: Khandare, A. L., Rao, G. S. and N. Lakshmaiah (2002). Effect of Tamarind Ingestion on fluoride excretion in humans. European Journal of Clinical Nutrition 56, No. 1: 82.

Available at: www.nature.com/ejcn/journal/v56/n1/full/1601287a.html

The objective of this study is to evaluate the effect of tamarind (*Tamarindus indicus*) ingestion on excretion of fluoride in school children. It is a randomized diet-control study. 18 healthy boys completed the study and each of them consumed 10g tamarind daily with lunch for 18 days. The nutrient composition of the daily diet was constant throughout the experimental period. It was found that “tamarind intake led to significant increase ($P < 0.001$) in the excretion of fluoride in 24h urine (4.8 ± 0.22 mg/day) as compared to excretion on control diet (3.5 ± 0.22 mg/day). However, excretion of magnesium and zinc decreased significantly”. “Excretion of calcium and phosphorous were not significantly different while creatinine excretion decreased with tamarind intake”. The study concludes that tamarind intake is likely to help in delaying progression of fluorosis by enhancing urinary excretion of fluoride.

Fluoride in Drinking-Water

Reference: Fawell, J., Bailey, K., Chilton, J., Dahi, E., Fewtrell, L. and Y. Magara (2006). Fluoride in Drinking-Water: Chapter 3 Human Health Effects and Chapter 4 Guidelines and Standards. IWA Publishing, on behalf of the World Health Organization.

Available at:

www.who.int/water_sanitation_health/publications/fluoride_drinking_water/en/index.html

The third and fourth chapters of this document discuss the effects of fluoride on human health and of fluoride guidelines and standards in drinking water. Key information includes the following:

- Adverse effects of fluoride intake range from mild dental fluorosis to crippling skeletal fluorosis
- 75-90% of ingested fluoride is absorbed and 99% of the body burden of fluoride is retained in calcium rich areas such as bone and teeth
- Dental fluorosis:
 - The margin between the beneficial effects of fluoride and the occurrence of dental fluorosis is small and public health programmes seek to retain a suitable balance between the two (IPCS, 2002)
 - Dental fluorosis increases as fluoride concentration increases
 - Dental fluorosis ranges in appearance from scarcely discernible to a marked staining or pitting of the teeth in severe forms. It is caused by an elevated fluoride level in, or adjacent to, the developing enamel (Whitford, 1997). Dental fluorosis cannot develop in adults; dental fluorosis in an adult is a result of high exposure during childhood.
- Skeletal fluorosis:
 - Associated with the consumption of drinking water with high concentrations of fluoride and of other sources of fluoride (e.g. food, air, dental products)
 - Crippling fluorosis is associated with higher levels of fluoride, and can result from osteosclerosis, ligamentous and tendinous calcification and extreme bone deformity
- According to different studies, exposure of mothers to fluoride in drinking water and adverse pregnancy outcome has no increased risk of either spontaneous abortion or congenital malformations

Guidelines for Drinking-Water Quality

Reference: World Health Organization (2006). Chapter 12 Chemical Fact Sheets, Guidelines for Drinking-Water Quality.

Available at: www.who.int/water_sanitation_health/dwq/gdwq3rev/en/index.html

- The chemicals of greatest health concern in some natural waters are usually excess natural fluoride, nitrate/nitrite and arsenic.
- There is no difference in absorption between natural or added fluoride in drinking water.
- Absorbed fluoride is rapidly distributed through the body, where it is incorporated into teeth and bones, with virtually no storage in soft tissues. Fluoride in teeth and bone can be mobilized after external exposure has ceased or been reduced. Fluoride is excreted via urine, faeces and sweat.

- Low concentrations provide protection against dental caries, both in children and in adults.
 - The fluoride guideline value is set at 1.5 mg/L. Epidemiological evidence that concentrations above this value carry an increasing risk of dental fluorosis, and progressively higher concentrations lead to increasing risks of skeletal fluorosis. The value is higher than that recommended for artificial fluoridation of water supplies, which is usually 0.5–1.0 mg/litre.
 - In setting national standards for fluoride or in evaluating the possible health consequences of exposure to fluoride, it is essential to consider the intake of water by the population of interest and the intake of fluoride from other sources (e.g., from food, air and dental preparations). Where the intakes from other sources are likely to approach, or be greater than, 6 mg/day, it would be appropriate to consider setting standards at a lower concentration than the guideline value.
 - In areas with high natural fluoride levels in drinking-water, the guideline value may be difficult to achieve, in some circumstances, with the treatment technology available.
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Integrated Fluorosis Mitigation

Reference: Sukumar, D. (2007). Integrated Fluorosis Mitigation: Guidance Manual. National Environmental Engineering Research Institute (NEERI), UNICEF, Regional Medical Research Centre for Tribals, Public Health Engineering Department, India. Available at: <http://neeri.res.in/pdf/integrated.pdf>

This manual presents a brief review of fluoride pollution and existing fluorosis mitigation approaches and a comprehensive approach on Integrated Fluorosis Mitigation (IFM). The highlights include:

- Nutritional aspect:
 - Chinoy et al. (2000) indicated that protein supplementation has a beneficial effect on reducing the fluoride induced liver toxicity and is necessary for recovery from fluoride toxicity
 - Community based studies strongly suggest that calcium status modifies the type of bone changes seen in fluorosis (Krishnamachari et al., 1986 and Chakma et al., 2000)
 - Clinical trials carried out with therapeutic supplementation of micronutrients in fluorosis affected children of Rajasthan also showed beneficial effect on skeletal deformities (Gupta et al., 1994).
 - Reversal of fluoride induced cell injury through nutritional supplementation of calcium, vitamin C and antioxidants have been reported from Delhi
 - Over 90% of the persons affected with severe skeletal fluorosis, bone disease and deformities belong to the low socio-economic group, where calcium intake status is low (Teotia et al., 1984)
 - A study in China shows 43.8% prevalence of skeletal fluorosis with normal nutrition while 69.2% in those with malnutrition, clearly underlining the role of nutritional aspects (Liang et al., 1997)

- Concept of Integrated Fluorosis Mitigation (IFM):
 - Needs multidimensional approach starting with proper identification of the problem and its causes
 - Scientific tools such as Quantitative Chemical Risk Assessment (QCRA) are used for assessment of health impacts due to excessive fluoride intake in relation with the nutritional aspects
 - Water quality monitoring and surveys along with reported dental/skeletal fluorosis symptoms lead to the identification of fluoride contamination. This should be followed by an integrated fluorosis mitigation strategy involving water management solutions, improved water defluoridation techniques and nutritional supplementation (example of Chakoda Bhaji (*Cassia tora*), a vegetable commonly found in rural area, very high in calcium content, reported that its consumption has resulted in reversal of fluorosis (Chakma et al., 2000)).
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PART 3: FLUORIDE REMOVAL TECHNOLOGIES

CATEGORY 1 – PUBLISHED PAPERS

Appropriate Defluoridation Technology for Use in Fluorotic Areas in Tanzania

Reference: Mjengera, H., and G. Mkongo (2003). Appropriate Defluoridation Technology for Use in Fluorotic Areas in Tanzania. *Physics and Chemistry of the Earth* 28, No. 20-27: 1097–1104.

Available at: www.sciencedirect.com

In this study done in Tanzania, the bone-char, alum/lime, and the combination of the two methods have been tested at both household and institutional levels. Different sizes of the kiln were fabricated and tested with the bone char methods, and crushing and sieving devices were developed.

Results show that small particles sizes are more efficient in term of fluoride removal, but the use of very fine particles sizes of bone char is not recommended since it clogs easily. Also, filter columns packed with heat-activated bones were found to be more effective than fill and draw bucket type. The water produced with the bone char media had a residual fluoride concentration of less than 0.1 mg/litre from an initial fluoride concentration of 12.0 mg/litre. Regarding alum and lime for fluoride removal, a plant with an automatic chemical dosing and mixing system for use at institutional levels was developed and tested. The water produced had a fluoride concentration of 2-3 mg/litre from an initial concentration of 12 mg/litre.

This document also gives a detailed explanation of how to prepare the bone char media.

Defluoridation of Water at High pH with Use of Brushite, Calcium Hydroxide, and Bone Char

Reference: Larsen, M. J., Pearce, E. I. F. and S. J. Jensen (1993) Defluoridation of Water at High pH with Use of Brushite, Calcium Hydroxide, and Bone Char. *Journal of Dental Research* 72, No. 11 (1993): 1519–1525

Available at: <http://jdr.sagepub.com/cgi/content/abstract/72/11/1519>

This is a study of the bone-char method efficiency improvement in fluoride removal by pre-treating the water with brushite and calcium hydroxide. Different amounts of brushite, calcium hydroxide, and bone char were suspended in batches (100 mL) of distilled water containing 0.53 mmol/litre fluoride for 24 hours. The pH, concentrations of fluoride, calcium, phosphate and degrees of saturation with respect to brushite, hydroxyapatite, and fluorapatite were determined.

Bone char used alone took up fluoride slowly and inefficiently. The addition of brushite and calcium hydroxide resulted in high concentrations of calcium and phosphate, making the solutions highly supersaturated with respect to fluorapatite, and led to a 20-fold increase in fluoride removal from the water. The combined use of all three salts left low concentrations of phosphate in solution and optimized the fluoride uptake capacity. Repeated use of the same bone char for 18 consecutive runs demonstrated that uptake of fluoride by the bone char was improved by repeated use, provided that brushite and

calcium hydroxide were added. Therefore, addition of the two salts to the water may prolong the life of the bone char indefinitely, ensure the removal of fluoride, and thus avoid the problem of determining when the bone char is exhausted.

In conclusion, the bone-char defluoridation technique can be improved by addition of brushite and calcium hydroxide to the water.

Fluoride in Drinking-Water

Reference: Fawell, J., Bailey, K., Chilton, J., Dahi, E., Fewtrell, L. and Y. Magara (2006). Fluoride in Drinking-Water: Chapter 5 Removal of Excessive Fluoride. IWA Publishing, on behalf of the World Health Organization.

Available at:

www.who.int/water_sanitation_health/publications/fluoride_drinking_water/en/index.html

Chapter 5 provides a comprehensive description of existing household water treatment technologies for fluoride removal. It shows a decision tree for appropriate action in relation to elevated concentrations of fluoride in water sources and explains that “the provision of safe, low fluoride water from alternative sources should be investigated as the first option”. However, the five main household treatment technologies for fluoride removal are described as follows:

- **Bone Charcoal:** Blackish, porous, granular media. Its major components are calcium phosphate (57-80%), calcium carbonate (6-10%) and activated carbon (7-10%). Three most common types of domestic bone charcoal filters are: drum, double bucket, and column. Regeneration of the medium can be done by allowing equilibrium with 1% solution of sodium hydroxide and washing or neutralizing the surplus caustic soda (AWWA, 1971). Use of bone charcoal can be difficult to accept with some local customs and beliefs.
- **Contact Precipitation:** Fluoride is removed by adding calcium and phosphate compounds and bringing the water in contact with an already saturated bone charcoal medium.
- **Nalgonda:** Aluminium sulfate based coagulation-flocculation sedimentation. Aluminium sulfate is dissolved and added to the water with sufficient stirring to ensure initial complete mixing. Decanting can be done and filtration is needed to ensure none of the toxic sludge particles escape with the treated water. Sludge requires proper disposal.
- **Activated Alumina:** Aluminium oxide (Al_2O_3) grains are prepared to have an absorptive surface. The water passes through a packed column of activated alumina and pollutants and other components are absorbed onto the surface of the grains. It is recommended to avoid total saturation of the medium. Regeneration can be done by exposing the medium to 4% caustic soda (NaOH) in batch or by flow through column. The process produces caustic wastewater contaminated with fluoride that requires proper disposal.
- **Clay:** is an earthy sedimentary material composed mainly of fine particles of hydrous aluminium silicates and other minerals and impurities. Both clay powder and fired clay are capable of absorbing fluoride as well as other pollutants from water.

Further details are provided on the technical configuration, design criteria and sludge disposal (when applicable).

The last part of this chapter is a detailed explanation of the evaluation and selection of different treatment technologies.

Fluoride Removal Studies in Water Using Natural Materials: Technical Note

Reference: Chidambaram S., Ramanathan, A. L. and S. Vasudevan (2004). Fluoride Removal Studies in Water Using Natural Materials: Technical Note. Water SA 29, no. 3: 339.

Available at: www.wrc.org.za/archives/watersa%20archive/2003/july/14.pdf

The study tested the efficiency of natural materials for fluoride removal. Tested materials were: red soil, charcoal, brick, fly-ash and serpentine. Each material was set up in a column and the defluoridation capacities of these materials were studied with respect to time. Ten mg/litre of fluoride was passed through the columns and the variation of fluoride removal for a known rate of flow was studied. The removal rate of fluoride was tested using 25 grams of the material collected in different flasks. All the materials were not pretreated and the size fractions taken were of 2.5 ϕ mesh fraction. The flow rate of 1.5 mL/minute was found to be satisfactory during preliminary trials. The results are as follows:

- Untreated charcoal seems to have no effect on the concentrations of fluoride with time and it adsorbs 0.5 mg/litre of fluoride
 - The brick and fly-ash typically show a sudden removal of fluoride from 10 mg/litre to 5 mg/litre; 50% reduction of concentration takes place within 30 minutes
 - Red soil adsorbs 9 mg/litre of fluoride immediately after 15 min. After 30 min the fluoride decreases from 1.0 to 0.1 mg/litre and keeps on reducing with time up to 0.02 mg/litre in 90 to 120 minutes.
 - In serpentine, the fluoride removal is maximal in 0 to 15 minutes, i.e. 3.7 mg/litre.
 - Red soil has good removal capacity because it contains both the oxides of iron and aluminium which are good fluoride removers, but it needs regeneration after 15 minutes
-

Groundwater Fluoride Levels in Villages of Southern Malawi and Removal Studies Using Bauxite

Reference: Sajidu, S. M. I., Masamba, W. R. L., Thole, B. and J.F. Mwatseteza, J. F. (2008). Groundwater Fluoride Levels in Villages of Southern Malawi and Removal Studies Using Bauxite. International Journal of Physical Sciences Vol. 3 (1), pp. 001-011.

Available at: www.academicjournals.org/IJPS/PDF/Pdf2008/Jan/Sajidu%20et%20al.pdf

This paper presents findings about fluoride levels of some sources in selected areas of Southern Malawi and the potential of locally available bauxite in water defluoridation at laboratory scale. Powder X-ray diffraction characterization of the raw bauxite showed gibbsite and kaolinite as the major components. The bauxite was ground into powder form and was calcined in air using a muffle furnace at temperatures of 200, 300, 400, and 500°C to acquire different phases of bauxite for experimentation so as to determine the phase with the highest defluoridation capacity. The samples were held at each reaction temperature for 2 hours and quench-cooled to room temperature. Defluoridation capacity determinations were carried out by mixing 200 mL of 8 mg/litre fluoride solution

with 2.5 g of defluoridating material (bauxite or a calcined phase of bauxite) and shaking for 30 minutes. Fluoride concentration in the solution was monitored hourly for 12 hours.

Results show that calcinations are not necessary and that the raw bauxite has a capacity of 93.8% at a dose of 2.5 g/200mL of 8mg/L F⁻ solution. However, the bauxite coloured the water to earth brown making the water unaesthetic for drinking. The colour could easily be clarified by using activated carbons or *Moringa oleifera* seed powder.

Low-Cost Defluoridation of Water Using Broken Bricks

Reference: Wijesundara T. (2004). Low-Cost Defluoridation of Water Using Broken Bricks. 30th WEDC International Conference.

Available at:

The National Water Supply & Drainage Board of Sri Lanka developed a low cost household technology to remove excess fluoride in drinking water to avoid endemic fluorosis. Broken pieces of freshly burnt bricks were used as filter media in these units. A kinetic model for fluoride uptake in the filter with broken bricks as defluoridating agent was considered and the model parameters were estimated using experimental data. Data concerning the uptake of fluoride on broken brick pieces were obtained from water in batch in the defluoridator. The study concludes that broken bricks could be used as filter media for concentration of fluoride in raw water around 2 mg/litre.

Nomograph for Defluoridation of Water in Batch Using Fish Bone Char

Reference: Bhargava D.S. (2000). Nomograph for Defluoridation of Water in Batch Using Fish Bone Char. 3rd International Workshop on Fluorosis Prevention and Defluoridation of Water, Session 1 Epidemiology: 69.

Available at: www.de-fluoride.net/3rdproceedings/73-79.pdf

A nomograph is established based on literature data on laboratory studies on batch defluoridation of water using fishbone charcoal. It concludes removal capacities between 0.05 and 0.65 mg/g. The nomograph can be used to estimate the dose of fish bone char and stirring time required to defluoridate water containing up to 30 mg/litre down to acceptable level of 1.5mg/litre.

Sorption Study for Defluoridation by Bone Char

Reference: Watanesk S. and R. Watanesk (2000). Sorption Study for Defluoridation by Bone Char. 3rd International Workshop on Fluorosis Prevention and Defluoridation of Water, Session 1 Epidemiology: 80.

Available at: www.de-fluoride.net/3rdproceedings/84-88.pdf

This paper is a study on sorption of fluoride by bone char for defluoridation purpose. It was done in laboratory batch experiments. Results include the following:

- The sorption isotherm at a low concentration range of fluoride tended to be a momentarily sorption type
- The sorption of fluoride was found not to increase significantly with the increase in temperature

- The fluoride level could be reduced by single batch equilibration treatment with the efficiency of about 65 % after equilibrating with 0.100 g of bone char at pH 7 for 9 hours (from waters samples containing 10-13mg/litre of fluoride)
-

Technologies for Fluoride Removal

Reference: Lyengar L. (2002). Technologies for Fluoride Removal. Small Community Water Supplies: Technology, People and Partnership, TP 40, Chapter 22.
Available at: www.irc.nl/page/1917

This document presents different technologies for fluoride removal. It categorizes defluoridation methods according to the main removal mechanisms:

- Chemical additive methods: Nalgonda technique
 - Contact precipitation methods: saturated bone char
 - Adsorption/ion exchange methods: bone char, activated alumina, calcined clay
-

The Development of Bone Char-Based Filters for the Removal of Fluoride from Drinking Water

Reference: Korir H., Mueller, K., Korir, L., Kubai, J., Wanja, E., Wanjiku, N., Waweru, J., Mattle, M.J., Osterwatder, L. and C.A. Johnson (2009). The Development of Bone Char-Based Filters for the Removal of Fluoride From Drinking Water. 34th WEDC International Conference, Addis Ababa, Ethiopia.

Available at: **Not available online yet (October 2009)**

This paper explains that the bone char filtration is highly efficient to remove fluoride from drinking water, but that its lifespan is limited. It presents laboratory results and field testing of a new fluoride removal technology, based on a combination of bone char and calcium-phosphate pellets to extend the lifespan of the bone char media. The pellets slowly release the required chemicals into the water for fluoride precipitation. They don't create additional maintenance requirements for the users.

Experiments: PVC columns were filled with 260mL of filter material (220 g, 1-4 mm). The pellets (2-6.3 mm) were used for contact precipitation with a ratio of 3:1. Distilled water spiked with 6 mgF/litre was used as well as piped water from a Kenyan groundwater source containing 6.2 mgF/litre.

Results: Fluoride removal efficiency was very high for all columns in the beginning, 98%. As the filters become saturated the fluoride concentrations increase. The pellets can improve filter performance and lifespan of the filters is prolonged by a factor of 6 in the case of synthetic groundwater and a factor of 3 for the Kenyan groundwater.

Use of Activated Clay for Defluoridation of Water

Reference: Bjorvatn K. and A. Bardsen (1995). Use of Activated Clay for Defluoridation of Water. Ngurdoto, Tanzania.

Available at: www.de-fluoride.net/1stProceedTotPDF.pdf#page=51

The aim of this paper is to demonstrate defluoridation of water by the use of lateric red clay from Cameroon, West Africa. The study presents data obtained by the use of red soil (lateric clay), calcined at 570°C for three hours. Water containing fluoride at concentrations from 5.50-7.75 mg/litre to 12, 30 and 130 mg/litre (NaF⁻ solutions), were exposed to calcined laterite, w/v 1:10. Water samples were taken at 1, 2, 4, 12, 24, 48, 72 hours and after one week and analyzed. Rapid reduction of the fluoride concentration in the water was observed during the first few hours. A steady state was in most cases reached in 48 hours.

With one kind of clay, the fluoride concentration was reduced from 5.47 to 0.48 mg/litre in two hours, from 12.2 to 0.26 mg/litre in 12 hours and from 31.2 to 0.76 mg/litre in 12 hours. Water with 132 mg/litre had a fluoride concentration of 4.58 mg/litre after one week.

CATEGORY 2 – HIGH QUALITY UNPUBLISHED PAPERS**Adapting the Biosand Filter to Remove Fluoride: An Investigation into Alternative Filter Media**

Reference: Hillman, A. (2007). Adapting the Biosand Filter to Remove Fluoride: An Investigation into Alternative Filter Media. Masters of Science Thesis, Cranfield University, United Kingdom.

Available at:

https://dspace.lib.cranfield.ac.uk/bitstream/1826/2183/1/HILLMAN_WM_MSC_THESIS.pdf

The aim of this thesis was to investigate the possibility of modifying the biosand filter to enable it to remove fluoride as well as biological contamination. Bone charcoal, baked lateritic clay, and lateritic clay bricks were identified as promising materials for a fluoride adsorbing alternative to sand, and were examined in a U.K. laboratory to determine the fluoride removal capability under conditions required for biosand filtration and the ability to support the biolayer. The study also investigated the household production of bone charcoal and the preferences of a small community for either bone char filtration or the Nalgonda technique.

The fluoride filtration results showed that the alternative materials are rapidly exhausted and therefore, requiring replacement would entail frequent interruption to the biosand filter's filtration of biological contamination. Given the difficulty of locating and preparing the alternative media, the rapid exhaustion, and community preference for the Nalgonda technique, this study's main recommendation is that fluoride removal should be performed as a separate process to biosand filtration and preferably by the Nalgonda technique (for fluoride concentrations < 10 mg/litre).

Defluoridation of Water by *Moringa Oleifera* Seeds and Inorganic Salts

Reference: Goswami, Lakshyeswar (2004). Defluoridation of Water by *Moringa Oleifera* Seeds and Inorganic Salts. Indian Institute of Technology, Bombay.

Abstract available at:

<http://www.library.iitb.ac.in/~mnj/gsd/cgi-bin/library?e=d-00000-00---0MTECH--00-1--0-10-0---0---0prompt-10---4-----0-0l--11-en-50---20-about---00-3-1-00-0011-1-0utfZz-8-10&cl=CL2&d=HASH7e8b0a6dcbd2463d7a18b1&x=1>

The present study evaluates the effectiveness of the natural polyelectrolyte extracted from *Moringa oleifera* seeds by two different methods; MOE and MOE-sc. The highest removal efficiency achieved is 16.01% by MOE for a sample with initial fluoride concentration of 5.28mg/l. For MOE-sc the removal efficiency is 23.08% for a sample with initial fluoride concentration of 3.50 mg/litre. MOE as coagulant aid with other inorganic salts has shown hardly any improvement in defluoridation. Some inorganic salts have been included in the study. After adjustment of the initial pH, ferric chloride (FeCl₃) has shown a removal efficiency of 47.79% reducing the initial fluoride concentration from 2.15 to 1.12 mg/litre for a dose of 200mg/litre of FeCl₃.

CATEGORY 3 – INFORMAL REPORTS

CDN's Experiences in Producing Bone Char

Reference: Catholic Diocese of Nakuru, Water Quality and K. Müller (2007). CDN's Experiences in Producing Bone Char. Catholic Diocese of Nakuru, Water Quality (CDC WQ) and Swiss Federal Institute of Aquatic Science (EAWAG).

Available at:

www.eawag.ch/organisation/abteilungen/sandec/publikationen/publications_ws/downloads_ws/fluoride_charring.pdf

This document is a report from the Catholic Diocese of Nakuru, Kenya which gives detailed information on bone char specifications and manufacturing steps for use in a defluoridation filter. Bone char is an effective material for fluoride removal only if it is a high quality bone char. Unless carried out properly, it may result in a product of low defluoridation capacity and/or deteriorated water quality.

The color of the bone char has been proven to be a good indicator for its ability to remove fluoride:

- Grey-brownish bone char: highest fluoride removal
- Black bone char: bones still contain organic impurities, which cause odor and color
- White bone char: reduced fluoride removal capacity

The following manufacturing steps are also explained in the document:

- Charring
- Crushing sieving
- Washing
- Drying

The document also explains how to regenerate saturated bone char.

CDN's Defluoridation Experiences on a Community Scale

Reference: Catholic Diocese of Nakuru, Water Quality and K. Müller (2007). CDN's Defluoridation Experiences on a Community Scale. Catholic Diocese of Nakuru, Water Quality (CDC WQ) and Swiss Federal Institute of Aquatic Science (EAWAG).

Available at: www.wrqaawag.ch/publications/community

This document reports on the Catholic Diocese of Nakuru's field experience on community scale defluoridation. It presents three kinds of community filters (institutional filters, community filters, filters for water works) and gives details about their technical specifications and the filters design and operation. All filters are using bone char as a media for fluoride removal. They don't require daily maintenance but the bone char needs to be regenerated.

CDN's Defluoridation Experiences on a Household Scale

Reference: Catholic Diocese of Nakuru, Water Quality and K. Müller (2006). CDN's Defluoridation Experiences on a Household Scale. Catholic Diocese of Nakuru, Water Quality (CDC WQ) and Swiss Federal Institute of Aquatic Science (EAWAG).

Available at: www.wrq.eawag.ch/publications/household

This report describes two kinds of filters implemented in Kenya by the CDN for fluoride removal using bone char:

- **Defluoridation bucket:** A 20 litre bucket is filled with 12 litres of bone char (8.4 kg). A tap is fixed at the bottom of the bucket after adding a drainage pipe. A perforated concrete plate is placed on the surface of the bone char to prevent disturbance of the filter medium during addition of raw water. Estimated lifespan of the media can be made based on the fluoride concentration of the raw water, the water consumption, and the adsorption capacity of the bone char (2 mg F/g bone char according to Albertus et al (2000)). The first few batches of treated water need to be discarded due to elevated turbidity and color in the treated water, deriving from the bone char and the water level in the bucket should never drop below the bone char level. 20 minutes of contact time is necessary to guarantee efficient fluoride removal.
- **Combined filter:** Combination of two filters arising from the need to remove fluoride and microbiological contamination from water. The upper bucket for raw water has a ceramic candle containing silver nitrate and activated carbon. Then the water flows top-down through a PVC pipe filled with bone char then bottom-up through an inner bucket also containing bone char (6 litres). The contact time at the beginning of the filtration process is around 30 minutes (for raw water with 1 NTU).

In October 2006, 34 CDN's households' filters efficiencies were tested. The results are as follows:

- Average 90%, ranging from 32% to 99%
- 76% (19) of the household units had a removal efficiency >90%
- 32% (8) of the household units had a removal efficiency >99%

Unnamed Position Paper

Reference: Author Unknown.

Available at: www.tickertape.net/position.htm

The author spent a year at the M. S. Swaminathan Research Foundation (MSSRF) in India studying the problem of fluoride-affected drinking water and methods of natural product defluoridation. The author makes interesting comments about natural products for fluoride removal:

- No natural products tested provide a promising alternative to chemical defluoridation methods. Research yielded two intriguing candidates with significant defluoridation capacity, but neither seems well suited for village-scale or community-scale water defluoridation.
- The seeds of the drumstick tree (*Moringa oleifera*) were the first tested. The tree is common throughout India and Africa and grows well in arid conditions. Drumstick seeds act as a coagulant. They have long been a traditional method for purification

of turbid water in both India and Africa. A previous researcher at MSSRF had shown drumstick seeds to have remarkable defluoridation efficiency, higher than that of activated alumina. Unfortunately, these results proved irreproducible.

- The roots of vetiver grass (*Vetiveria zizanoides*) are another product that has traditionally been used for water purification. Water filtered through the roots is supposed to have a sweeter taste, probably due partly to a taste imparted by the roots and partly to a decreased salt content in the water. The roots were effective at defluoridation and could remove as much as 70% of the fluoride from a sample. The defluoridation efficiency was higher than activated alumina, and the price was comparable. Unfortunately, back-of-the-envelope calculation indicates that a family would need to raise acres of vetiver grass every year in order to provide enough material for defluoridation.

PART 4: SUMMARY OF PUBLICATIONS

Table 1: Fluoride, Occurrence and Chemical Specifications

Country	Author(s)	Organization(s)	Year	Category	Key Findings
-	Fawell J. Kirtley J. Bailey K.	World Health Organization	2006	Published Paper	<ul style="list-style-type: none"> • Fluorine is one of the most reactive of all chemical elements; • It has a strong tendency to acquire a negative charge and in solution and it forms F- ions (fluoride); • In groundwaters, low or high concentrations can occur depending on the nature of rocks and the occurrence of fluoride-bearing minerals; • The other sources of exposure to fluoride are: air, dental products, food and beverages other than water.
-	Qian S.J. et al.	UNICEF	1999	Published Paper	<ul style="list-style-type: none"> • In many countries, fluoride is added to the water supply and product like toothpaste. • Fluoride is also found in food and in the air, so the amount of fluoride people actually ingest may be higher than assumed. • In surface freshwater fluoride concentrations are usually low - 0.01 ppm to 0.3 ppm. • The fluoride concentrations in groundwater can range from well under 1 ppm to more than 35 ppm.
Kenya	Näslund J. Snell I.	World Health Organization	2005	Unpublished Paper	<ul style="list-style-type: none"> • Master of Science thesis carried out in Nakuru and Baringo Districts in the Rift Valley Province in Kenya. • It gives information on fluoride concentrations testing and GIS mapping as well as concentrations of fluoride in ground waters in this region of Kenya.

Table 2: Fluoride Health Impacts

Country	Author(s)	Organization(s)	Year	Category	Key Findings
Ethiopia	Melaku Z. et al.	3rd International Workshop on Fluorosis Prevention and Defluoridation of Water	2000	Published Paper	<ul style="list-style-type: none"> The health consequences of consuming untreated water are fairly understood but there is a knowledge gap and a wrong perception concerning fluoride and its health consequences The study recommends providing health education to the community affected by fluoride with emphasis to the women from the lower socioeconomic segment
-	Khandare A. L. Rao G.S. Lakshmaiah N.	European Journal of Clinical Nutrition	2002	Published Paper	<ul style="list-style-type: none"> The study concludes that tamarind intake is likely to help in delaying progression of fluorosis by enhancing urinary excretion of fluoride
-	Fawell J. Kirtley J. Bailey K.	World Health Organization	2006	Published Paper	<ul style="list-style-type: none"> The margin between the beneficial effects of fluoride and the occurrence of dental fluorosis is small and public health programmes seek to retain a suitable balance between the two (IPCS, 2002) Dental fluorosis ranges in appearance from scarcely discernible to a marked staining or pitting of the teeth in severe forms. It is caused by an elevated fluoride level in, or adjacent to, the developing enamel (Whitford, 1997). Skeletal fluorosis: Associated with the consumption of drinking water with high concentrations of fluoride and of other sources of fluoride (food, air, dental products) Crippling fluorosis: Associated with higher levels of fluoride, and can result from osteosclerosis, ligamentous and tendinous calcification and extreme bone deformity
-	Qian S.J. et al.	UNICEF	1999	Published Paper	<ul style="list-style-type: none"> Fluoride is an effective agent for preventing dental caries if taken in 'optimal' amounts. In warm climate, the optimal fluoride concentration in drinking water should remain below 1mg/L, while in cooler climates it could go up to 1.2mg/L. Fluorosis is endemic in at least 25 countries across the globe. It is estimated that 10 million of people are affected by fluorosis.
-	Sukumar D.	UNICEF	2007	Published Paper	<ul style="list-style-type: none"> Community based studies strongly suggest that calcium status modifies the type of bone changes seen in fluorosis (Krishnamachari et al., 1986 and Chakma et al., 2000). Over 90% of the persons affected with severe skeletal fluorosis, bone disease and deformities belong to the low socio-economic group, where calcium intake status is low (Teotia et al., 1984). Integrated fluorosis mitigation strategy

Table 3: Fluoride Removal Technologies

Country	Author(s)	Organization(s)	Year	Category	Key Findings
Tanzania	Mjengera, H. Mkongo G.	Physics and Chemistry of the Earth	2003	Published Paper	<ul style="list-style-type: none"> Two methods have been tested in both household and institutional levels. The water produced with the bone char media had a residual fluoride concentration of less than 0.1 mg/L from an initial fluoride concentration of 12.0 mg/L. Regarding alum and lime for fluoride removal from, a plant with an automatic chemical dosing and mixing system for use at institutional levels has been developed and tested. The water produced had a fluoride concentration of 2-3 mg/L from an initial concentration of 12 mg/L.
-	Larsen M.J. Pearce E.I.S. Jensen J.	Journal of Dental Research	1993	Published Paper	<ul style="list-style-type: none"> Different amounts of brushite, calcium hydroxide, and bone char were suspended in batches (100 mL) of distilled water containing 0.53 mmol/L fluoride for 24 hours. Bone char used alone took up fluoride slowly and inefficiently. The addition of brushite and calcium hydroxide resulted in high concentrations of calcium and phosphate, making the solutions highly supersaturated with respect to fluorapatite, and led to a 20-fold increase in fluoride removal from the water.
-	Fawell J. Kirtley J. Bailey K.	World Health Organization	2006	Published Paper	<ul style="list-style-type: none"> This document is a complete description of existing household water treatment for fluoride removal. It shows a decision tree for appropriate action in relation to elevated concentrations of fluoride in water sources and explains that "the provision of safe, low fluoride water from alternative sources should be investigated as the first option.
-	Chidambaram S. Ramanathan A.L. Vasudevan S.	-	2004	Published Paper	<ul style="list-style-type: none"> Untreated charcoal seems to have no effect on the concentrations of fluoride with time and it adsorbs 0.5 mg/L of fluoride. The brick and fly-ash show typically a sudden removal of fluoride from 10 mg/L to 5 mg/L. 50% reduction of concentration takes place within 30 minutes. Red soil adsorbs 9 mg/L of fluoride immediately after 15 min. After 30 minutes the fluoride decreases from 1.0 to 0.1 mg/L and keeps on reducing with time up to 0.02 mg/L in 90 to 120 minutes. In serpentine the fluoride removal is maximal in 0 to 15 minutes, i.e. 3.7mg/L; later the defluoridation capacity increases to 5.8 mg/L at 90 to 120 min gradually.
Malawi	Sajidu et al	International Journal of	2008	Published Paper	<ul style="list-style-type: none"> Results show that the raw bauxite has a capacity of 93.8 % at a dose of 2.5g/200 mL of 8 mg/L F⁻ solution.

Country	Author(s)	Organization(s)	Year	Category	Key Findings
		Physical Sciences			<ul style="list-style-type: none"> • However, the bauxite coloured the water to earth brown making the water unaesthetic for drinking. The colour could easily be clarified by using activated carbons or Moringa oleifera seed powder.
Sri Lanka	Wijesundara T.	30th WEDC International Conference	2004	Published Paper	<ul style="list-style-type: none"> • Broken pieces of freshly burnt bricks were used as filter media in these units. • The capacity parameter was estimated to 0.10 mg/g and the reaction rate parameter which is a function of initial concentration varied between 0.001 and 0.0005 L/ (mg.ho.5) for low and high initial concentration. • The study concludes that broken bricks could be used as filter media for concentration of fluoride in raw water around 2 mg/L.
-	Bhargava D.S.	3rd International Workshop on Fluorosis Prevention and Defluoridation of Water	2000	Published Paper	<ul style="list-style-type: none"> • The nomograph can be used for handy estimation of dose of fish bone char and stirring time required to defluoridate water containing up to 30 mg F/L down to acceptable level of 1.5 mg/L
-	Watanesk S. Watanesk R.	3rd International Workshop on Fluorosis Prevention and Defluoridation of Water	2000	Published Paper	<ul style="list-style-type: none"> • The sorption of fluoride by bone char was found not to increase significantly with the increase in temperature. • The fluoride level could be reduced by single batch equilibration treatment with the efficiency of about 65 % after equilibrating with 0.100 g of bone char at pH 7 for 9 hours (from waters samples containing 10-13 mg F/L).
-	Lyengar L.	-	2002	Published Paper	<ul style="list-style-type: none"> • Presentation of different existing technologies for fluoride removal. It categorizes defluoridation methods according to the main removal mechanisms
Ethiopia	Korir H. Mueller K. Korir L. Kubai J.W Wanja E. Wanjiku E. Waweru J. Mattle M.J. Osterwatder L. Johnson C.A.	34th WEDC International Conference, Addis Ababa, Ethiopia	2009	Published Paper	<ul style="list-style-type: none"> • Results: at the beginning fluoride removal efficiency was very high for all columns, 98%. As the filters become saturated the fluoride concentrations increase. The pellets can improve filter performance and lifespan of the filters is prolonged by a factor of 6 in the case of synthetic groundwater and a factor of 3 for the Kenyan groundwater.
Tanzania, Cameroon	Bjorvatn K. Bardsen A.	-	1995	Published Paper	<ul style="list-style-type: none"> • The aim of this paper is to demonstrate defluoridation of water by the use of lateric red clay from Cameroon, West Africa.
Ethiopia	Hillman A.	Cranfield University	2007	Unpublished Paper	<ul style="list-style-type: none"> • Fluoride removal should be performed as a separate process to Biosand filtration and preferably by the Nalgonda technique (for fluoride concentrations <10 mg/L).

Country	Author(s)	Organization(s)	Year	Category	Key Findings
India	Goswami Lakshyeswar	Indian Institute of Technology	2004	Unpublished Paper	<ul style="list-style-type: none"> The study evaluates the effectiveness of the natural polyelectrolyte extracted from <i>Moringa oleifera</i> seeds by two different methods. The highest removal efficiency achieved is 16.01%. After adjustment of the initial pH, ferric chloride (FeCl₃) has shown a removal efficiency of 47.79% reducing the initial fluoride concentration of 2.15 mg/L to 1.12 mg/L for a dose of 200 mg/L of FeCl₃.
Kenya	Müller K.	Catholic Diocese of Nakuru	2007	Informal Report	<ul style="list-style-type: none"> This document is a report from the Catholic Diocese of Nakuru (Kenya) which gives detailed information on bone char specifications and manufacturing steps for use in a defluoridation filter.
Kenya	Müller K.	Catholic Diocese of Nakuru	2007	Informal Report	<ul style="list-style-type: none"> CDN's field experience on community scale defluoridation. It presents three kinds of community filters (institutional filters, community filters, filters for water works) and gives details about their technical specifications and the filters design and operation.
Kenya	Müller K.	Catholic Diocese of Nakuru	2006	Informal Report	<ul style="list-style-type: none"> In October 2006, 34 CDN's households' filters efficiencies were tested: <ul style="list-style-type: none"> Results, removal efficiency: <ul style="list-style-type: none"> Average 90%, ranging from 32% to 99% 76% (19) of the household units had a removal efficiency >90% 32% (8) of the household units had a removal efficiency >99%
-	Author Unknown	-	-	Informal Report	<ul style="list-style-type: none"> No natural products tested provide a promising alternative to chemical defluoridation methods. Research yielded two intriguing candidates with significant defluoridation capacity, but neither seems well suited for village-scale or community-scale water defluoridation.