# Proceedings of the 2<sup>nd</sup> International Workshop on Fluorosis Prevention and Defluoridation of Water



Nazreth, Ethiopia November 19-25, 1997

Edited by: Eli Dahi & Joan Maj Nielsen

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#### Preface

In October 18-22, 1995, **The First International Workshop on Fluorosis Prevention and Defluoridation of Water** was held in Ngurdoto, The Arusha Region, Tanzania. The workshop was sponsored mainly by **The Danish International Development Agency DANIDA**, under the **Enhancement of Research Capacity programme, Enreca**. Thirty participants took part in the workshop, where 28 papers were presented and discussed. Eight countries, out of which 5 developing countries, were presented in the workshop.

During the first workshop it was decided to approach **The International Society for Fluoride Research, ISFR**, for collaboration on publication of the proceedings and on setting up an International Organising Committee for future arrangements of the Workshop. The proposals were kindly accepted by ISFR.

As a result of this collaboration, this **Second International Workshop on Fluorosis and Defluoridation of Water** was organised in November 19-25 in Nazreth, Ethiopia.

On behalf of the International Organising Committee I would like to address thanks to the **Local Organising Committee** for identification of excellent workshop environment and for assistance to participants on visa formalities. Furthermore thanks to **Danida-Enreca** to **Norad** and to **UNDP**, for sponsoring respectively 15, 9 and 3 workshop participants. It is only due to these contributions that the workshop could be arranged successfully in a fluoride affected region in a developing country and bringing together professionals and field workers from other fluoride affected developing countries.

It is intended that other workshops will follow this workshop, seeking for more sustainable technologies and appropriate solutions to the fluoride problems. Thus the **Third International Workshop on Fluorosis and Defluoridation of Water** is envisaged to take place in Thailand, year 2000.

Eli Dahi Chairman of the International Committee.

## CONTENTS

# Session I: Epidemiology

Dental fluorosis in subjects exposed to fluoride containing drinking water at different
A Bårdsen and K Bjorvatn
Fluoride and fluorosis: The status on research in South Africa <i>A J Louw and U M E Chickte</i>
Dental fluorosis in relation to altitude and fluoride in drinking water in Uganda <i>M C Rwenyonyi, K Bjorvatn, J M Birkeland &amp; O Haugejorden</i> 23-29
Dental fluorosis with special reference to incisors and molars of Tanzanian school children
A K Awadia, K Bjorvatn, J M Birkeland & O Haugejorden
Prevalence of skeletal fluorosis among retiring employees of Wonji Shoa Sugar Factory
G Shifera, Z Melaku, G Aseffa and R Tekle-Haimano
Prevalence of dental fluorosis in the Wonji Shoa Sugar Estate W Fantaye, G Shifera, and R Tekle-Haimanot, Ethiopia
Prevalence of low back pain at an agro-industrial community in the Rift Valley Z Mekalu, R Tekle-Haimanot and G Shifera

# Session II: Occurrence and Methodology

Fluoride contamination and mineralogical composition of East African Magad	i
(Trona)	
J M Nielsen and E Dahi 50-5	6
A simple method for determination of the total fluoride intake <i>G Karthikeyan, Anitha Pius and B V Appa Rao</i>	2
Analytical problems in assessment of fluoride in food <i>M K Malde, K Bjorvatn and K Julshamn</i>	6
Assessment of the fluoride content of relevant weaning food items in Western Ugand <i>M Wandera, M K Malde and K Bjorvatn</i>	a '0

Fluoride and silicon content in drinking water	
S Bapurao	

# Session III: Defluoridation; Laboratory Experiences

Defluoridation properties of activated alumina G Karthikeyan, B V Apparao and S Meenakshi
Kinetics of sorption fluoride on calcined magnesite in batch J J Singano, D A Mashauri, F W Mtalo and E Dahi
Bone charring by calcination <i>W Puangpinyo and N Osiriphan</i>
Effect of calcium addition on the defluoridation capacity of bone char <i>P Jacobsen and E Dah</i>
Defluoridation of drinking water by use of clay/soil K Bjorvatn, A Bårdsen and R Tekle-Haimano
Significance of elevation on the fluoride binding capacity of Ethiopian soils A Kvalheim, K Bjorvatn, A Bårdsen, R Tekle-Haimanot
Household purification of fluoride contaminated Magadi (Trona) J M Nielsen and E Dahi110-117
Economical technology of fluoride removal using fishbone charcoal columns at domestic level D S Bhargava 118-122
Biodefluoridation of fluoride containing water by a fungal biosorbent <i>N Lakshamiah, P K Paranjape and P M Mohan</i> 123-126

## Session IV: Defluoridation; Field Experiences

technology

C Venkobachar, L Iyengar and A K Mudga ......138-145

Low cost domestic defluoridation	
J P Padmasir	. 146-150
Charcoal packed furnace for tow-tech charring of bone <i>P lacobsen and F Dabi</i>	151-155
Done alon board busket deflueridator in Tenzonia bouesholde	. 101 100
<i>P Jacobsen and E Dahi</i>	. 156-159
A review of the defluoridation program of drinking water supplies of an Estate	Ethiopian
G Shifera, and R Tekle-Haimanot	. 160-167

# Session V: Discussion Papers

-175
ject,
-181
sha,
-190
-197
j



# Session I Epidemiology

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# DENTAL FLUOROSIS IN SUBJECTS EXPOSED TO FLUORIDE CONTAINING DRINKING WATER AT DIFFERENT AGE

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#### Bergen, Norway

SUMMARY: The present study was planned in order to gain more information on the agerelated susceptibility to dental fluorosis. Fourty children, who had been life-long consumers of water from high-fluoride wells in the county of Hordaland, Western Norway, were identified, together with a similar number of their older siblings, who had been exposed to high-fluoride water at ages varying from 6 months to 6 years. After informed consent had been given, each child and one of the parents were interviewed. Then the children were examined for dental fluorosis. Scorings were given according to the TF-index. Reliability tests were performed. The fluoride content of the relevant drinking water ranged from 0.5 to 7.2 mg/L. Of the 80 children examined, 66 had the upper central incisors erupted. Of these 66, 22, or one third, were without any sign of dental fluorosis. The rest had TF-scores in the range of 1-5. The children were divided into groups, according to the age at which they were exposed to high-fluoride water. As compared to the whole group, the odds ratio for a child to have a maxillary incisor with fluorosis were 7.63 (95% CI = 2.28, 25.58) when the exposure to high-fluoride water took place during the first year of life. These findings indicate that the first year of life is the most important period for the development of dental fluorosis in the maxillary central incisors. An inverse relationship seems to exist between the severity of dental fluorosis and the age at which exposure took place.

Key words: Dental fluorosis; Exposure; Fluoride; Window of maximum sensitivity

#### INTRODUCTION

The fact that excess fluoride intake causes dental fluorosis in developing teeth is well documented.<sup>1-3</sup> However, studies concerning the induction of dental fluorosis are relatively few, and findings are inconsistent. It has been indicated that on a worldwide basis the prevalence of dental fluorosis is increasing, even in low-fluoride areas. To provide better guidelines for the use of fluoride in caries prevention, and also to avoid negative focusing on fluoride, it is of importance to know the biological aspects of dental fluorosis, and the process leading to its induction.

Most major water-works in Norway rely on low-fluoride surface water reservoirs.<sup>4,5</sup> The possibility of artificial water fluoridation was discussed in the 1960'ies, but due to political reasons fluoridation of drinking water has never been implemented in Norway.<sup>6-8</sup> In stead, realising the cariostatic properties of fluoride,<sup>9-11</sup> national caries prevention programs have focused on the use of alternative fluoride sources such as fluoride containing toothpaste, fluoride tablets, fluoride mouthrinses and professionally applied topical fluoride agents.

Fluoride toothpaste is presently used by 92 % of Norwegian children in the age of 7-15 years, and by 88 % of the total population.<sup>12</sup> The use of fluoride tablets has been recommended for all children from the age of 6 months.<sup>13</sup> Until recently (1996) fluoride tablets have been distributed in child-health care clinics as well as by the school dental service, on request - free of charge. Fluoride tablets and fluoride rinses have been used by 32 % of children in the age of 7-15 years, and by 9 % of the total

population.<sup>12</sup> Sixty percent of the Norwegian school children (7 to 15 years) have been exposed to local professional fluoride application on a yearly basis.<sup>14</sup>

The use of and choices in fluoride supplements in Norway has increased; so has the consumption of groundwater.<sup>15</sup> Recent investigations have shown that the Norwegian hard-rock groundwater may contain high concentrations of fluoride.<sup>16-18</sup> As fluoride analysis is not automatically included in the testing of new wells, young consumers of the ground waters may, unexpectedly, develop dental fluorosis.

The present study was planned in order to gain more information on the age-related susceptibility to dental fluorosis.

#### **MATERIAL & METHODS**

Based on information obtained in previous studies,<sup>18-19</sup> 40 children who had been lifelong consumers of water from high-fluoride (> 0.5 mg F/L) groundwater wells in the county of Hordaland, Western Norway, were identified, together with a similar number of their older siblings, who had been exposed to high-fluoride water at ages varying from 6 months to 6 years.

The fluoride level of the wells was determined, based on the analysis of at least two water samples within a year, and none of the measurements showing a fluoride value < 0.5 mg/L.

Informed consent to partake was given by the participants' parents, or by the participants them selves if 18 years of age, before clinical examination.

**Clinical Examination.** Clinical examinations were conducted at the county dental health service clinics, while the participant was seated in a dental unit under standard clinical illumination. Teeth were cleaned with pumice and rotating rubber cups, rinsed with water and dried (air-jet) for approximately 30 seconds, before the examination. Each participant was examined for dental fluorosis, and scorings were recorded according to the modified TF-index.<sup>3,20</sup> All fully erupted teeth were given a score. Dental fluorosis was assessed by one examiner (AB) without previous knowledge of the children's fluoride exposure or time of residency in the area. Clinical photographs were taken of the anterior teeth in all the participants, using a Dental Eye camera (Yashica) and Kodak colour slides (EPN 200).

Reliability tests for the scoring of dental fluorosis were carried out, and showed excellent agreement (Kappa = 0.85, p < 0.01).<sup>21</sup> No statistically significant systematic error was found using Student's T-test for paired observations (p = 0.49).

**Questionnaire.** Before clinical examination, the participants and their parent(s) were interviewed. For participants without any parent present, the questionnaire was sent home to be completed by the parents. Following information was requested: area of residence and length of residency as well as type of water supply. Other questions were related to start and duration of the use of fluoride supplements, commencement and use of fluoride toothpaste and oral hygiene. Information on the frequency of intake of various beverages; history of breast- and baby bottle feeding as well as previous and present diet was also obtained. Information concerning traumatic injuries to the tooth/teeth or their predecessors was taken from the participant's dental record. Some of the obtained information is shown in Table 1, given as divided (mostly dichotomised) variables.

**Data Analysis.** The data were coded, computerised and analysed using the Statistical Program for Social Science (SPSS-PC Inc.).<sup>22</sup> Descriptive analyses were performed and frequency distributions compared using chi-square tests (significance level set at 5%). The Pearson correlation coefficient was used to look for direct relation between severity of dental fluorosis on the maxillary permanent central incisors and the age (in months) when exposed to high-fluoride water, as well as between TF-score on the same tooth-group and fluoride content in the high-fluoride water.

The variables (dependent and independent) used in this study were divided - most of them dichotomised - in order to facilitate interpretation of the results (Table 1). Multiple logistic regression analysis including independent variables having a statistically significant bivariate effect, was carried out to assess the effect of each variable after adjusting for the effect of all others in the model. The 95% confidence interval of the odds ratio (OR) was calculated using Wolf's equation.<sup>23</sup>

TABLE 1. Frequency distribut	tion on the divided variables.				
Variable		n*	%		
Sex	Female	30	46		
	Male	36	54		
Use of breast milk substitutes	No	22	33		
Use of breast mink substitutes	Yes	41	62		
F-tablet intake	Less than once a day	25	38		
	Once a day or more	41	62		
Use of F-toothpaste	Once a day or less	19	29		
	More than once a day	47	71		
Age at start using F-tablets	One year or younger	30	46		
	Older than one year	20	30		
Age at start using $\mathbf{E}$ toothpaste	One year or younger	33	50		
Age at start using 1-toothpaste	Older than one year	33	50		
Amount of fluoride used on the	Up to 1/3 of the length of the head of	2	3		
toothbrush	the toothbrush	2	5		
	More than 1/3 of the length of the	64	07		
	head of the toothbrush	04	91		
Age when introduced to high	0 - 12 month	33	50		
fluoride groundwater	13 - 24 month	11	17		
nuonde groundwater	25 - 36 month	8	12		
	37 - 84 month	14	21		
*Number of subjects answering the relevant question (range: 50-66)					

#### RESULTS

**Participation.** A total of 30 families were involved in the study, each with 2 to 4 children. Of the selected group of 80 individuals, 66 had their upper central incisors present without any filling, orthodontic brackets or any history of traumatic injuries to the tooth/teeth or the predecessors (83%). The age of participants ranged from 8 to 18 years. No significant difference in gender distribution were observed (Table 1).

**Fluoride Exposure.** The mean fluoride content of the drinking waters was 2.2 mg/L, and the fluoride content of the relevant drinking water sources ranged from 0.5 to 7.2 mg/L ( $25^{th}$  percentile; 0.7 mg F/L, and  $75^{th}$  percentile 3.3 mg F/L).

**Dental Fluorosis.** In the present study, 44 out of 66 participants (67 %) had TF-scores in the range of 1 to 5 on the maxillary permanent central incisors. The distribution of TF-scores according to age when exposed to high-fluoride groundwater is shown in Table 2. Some of the children who had been exposed to high-fluoride water only after 1 year of age had milder forms of dental fluorosis (TF-score 1 and 2), while eight out of nine children who had TF-scores 3-5 had been exposed to high-fluoride water before the age of 9 months. The exception was an individual who had been exposed to high-fluoride water only at the age of 14 months.

A negative correlation was found between the severity of dental fluorosis on the maxillary permanent central incisors and the age - in months - when the individuals first were exposed to high-fluoride water (r = -0.54, p < 0.01). No statistically significant correlation was found between the TF-score on the maxillary permanent central incisors and the fluoride content of the drinking water (r = 0.09, p = 0.48).

Three of the independent variable from Table 1 had a significant bivariate effect on the prevalence of dental fluorosis: (i) age when exposed to high-fluoride water, (ii) intake of fluoride tablets and (iii) frequency of the use of fluoridated toothpaste.

	TF-score							
Age							То	tal
when exposed to high-fluoride water	0	1	2	3	4	5	1 -5	%
0 – 12 month	3	14	8	4	1	3	30	91
13 - 24 month	4	6	-	1	-	-	7	64
25 - 36 month	2	5	1	-	-	-	6	75
37 - 84 month	13	1	-	-	-	-	1	7

**TABLE 1**. TF-score on maxillary permanent central incisors according to age when introduced to high-fluoride groundwater.

**TABLE 2.** Regression coefficient and odds ratios for participants (n = 66) with the dependent variable: Dental fluorosis or not on the maxillary permanent central incisors (coded; 0 = TF-score 0, 1 = TF-score  $\ge 1$ ).

1	,	1			
Independent variables		Regression	Odds ratio	95 % confidence	
		coefficient	Tatio	inter var	
	0 - 12 month vs. the average of all groups	2.03	7.63	2.28 - 25.58	
Age when introduced	13 - 24 month vs. the				
to high fluoride	average of all groups	0.44	1.56	0.43 - 5.63	
groundwater	25 - 36 month vs. the	0.88	2.40	0.40 11.00	
	average of all groups	0.88	2.40	0.48 - 11.88	
F-tablet intake	≥ once a day	1.01	0.15	0.02 1.25	
	vs. < once a day	-1.91	0.15	0.02 - 1.55	
	> once a day	0.42	0.66	0.11 2.07	
Use of r-toothpaste	vs. ≤ once a day	-0.42	0.00	0.11 - 3.97	

The final multiple logistic regression model for fluorosis on the maxillary permanent central incisors is shown in Table 3. In this analysis with TF-score as the dependent

variable (0 = TF-score 0, 1 = TF-score  $\geq$  1) and the above three variables as the independent variables, only one variable had a statistically significant risk of giving dental fluorosis. Participants which were exposed to high-fluoride water within the first 12 months of life showed an odds ratio of 7.63 (95% CI = 2.28 - 25.58) for dental fluorosis ( $\geq$  TF-score 1) on the maxillary permanent central incisors as compared to the average risk for the whole group.

#### DISCUSSION

Several reviews have been written regarding the mechanisms by which fluoride can affect mineralising tissues.<sup>24-26</sup> The authors seem to agree that the severity of enamel fluorosis depends on the amount of fluoride ingested, but the duration of exposure and critical stage of amelogenesis is not established.

Even before fluoride was found to be the causative element for dental fluorosis, estimates for the period of greatest susceptibility of enamel damage to the maxillary central incisors were implicitly established by McKay<sup>27, 28</sup> who noted that the risk of mottling enamel to maxillary central incisors ceased after the age of five years. At the same time, Ainsworth<sup>29</sup> carried out a study in Essex, England, and concluded that upper central incisors were not susceptible to mottling after the age of three years. Larsen *et al*<sup>30</sup> support Ainsworth's findings, and find the dental enamel to be most susceptible to fluoride in late secretory- and early maturation phase. Ishi and Suckling<sup>31</sup> and Evans and Stam<sup>32</sup> found the development of dental fluorosis to be an enamel maturation phenomena. Then Holm and Andersen<sup>33</sup> and Ismail and Messer<sup>34</sup> found the enamel secretion phase to be of most importance for the development of dental fluorosis. DenBesten and Thariani<sup>35</sup> concluded that it is likely that enamel fluorosis can result from a number of mechanisms; animal and human studies indicate that the transition/early maturation stage is particularly susceptible to fluoride. But exposure to high levels of fluoride during the secretory stage may also increase the risk of fluorosis by increasing the fluoride concentrations locally during enamel development.

#### Conclusion

These findings indicate that the first year of life is the most important period for the development of dental fluorosis in the maxillary permanent central incisors. An inverse relationship seems to exist between the severity of dental fluorosis and the age at which exposure took place.

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SUMMARY: The public health aspects of fluoride and the issues of fluoridation and defluoridation of drinking water in South Africa have been recorded, debated and discussed since 1935. The levels of fluoride in the different areas in South Africa were recorded, e.g. 0.3 - 35 mg/L, as compared to observations of dental caries in these areas. The prevalence of dental caries was at least twice as high amongst children in low fluoride areas compared with children in areas with fluoride levels between 1.1 and 1.8 mg/L. Also the presence of fluorosis amongst children in high fluoride areas was reported and the areas of endemic fluorosis were identified. Fluorosis prevalence was reported to be up to 100 % in certain areas. Severe fluorosis was found to be a predisposing cause of dental caries. The prevalence varies according to the fluoride content of the drinking water. A surprisingly high fluorosis prevalence 33.5 % has been reported in association with relatively low F concentrations (0.54 mg/L and even 0.4 mg/L). The relationship between fluorosis prevalence and fluoride content of the drinking water was positive but the spatial variation in the prevalence was not fully explained by the variation in the fluoride content of the drinking water. Current studies with the objective of arriving at a proposal in terms of what should be regarded as an optimal level of fluoride indicate that the optimal fluoride level in terms of caries prevention and fluorosis for the prevailing conditions is lower than generally accepted. In order to minimise dental fluorosis, a level of 0.54 mg/L appears acceptable. In the event of concentration fluctuations the content could be controlled so as never to rise above 0.7 mg/L.

Key words: Optimum Fluoride Concentration; Fluorosis; Dental Caries; South Africa.

#### INTRODUCTION

The issues of fluoridation, fluorides and fluorosis in South Africa have been recorded, debated and discussed since 1935. These have raised emotions and invectives from antagonists and protagonists. During the past decades the general public has become familiar with fluorides because of the continuing debate, mainly on the issue of water fluoridation to prevent dental caries. The concentration of fluoride in the drinking water is therefore considered of substantial significance from a public health point of view. While at low concentrations it is beneficial in preventing dental caries, higher concentrations of fluoride in the drinking water are detrimental to health.<sup>1</sup> The effects of excess fluoride (F) in the drinking water are easily observed on the teeth and can be seen radiographically on skeletal and soft tissue. If drinking water containing more than 1 mg F/L is consumed during the period of permanent teeth calcification, it can lead to dental fluorosis. Severity of dental fluorosis is related to the fluoride concentration in the water and varies from mottling of enamel to gross calcification defects which weaken enamel and can eventually lead to loss of teeth.<sup>2</sup>

Other effects from high intake of fluoride are: 1) bone changes in the form of exostosis when water containing 8-20 mg F/L is consumed over a long period of time; 2) crippling fluorosis in the form of painful skeletal rigidity and deformities when water containing more than 20 mg F/L is consumed over a period of 20 or more years.<sup>3</sup> This report will present an overview of research status of fluoride and fluorosis in South Africa.

#### **HISTORICAL PERSPECTIVE**

The research on water fluoridation, fluorides and fluorosis in South Africa can be categorised into three periods.<sup>4</sup> The first extends from 1935 to 1978. Maughan-Brown,<sup>5</sup> Staz<sup>6</sup> and Abrahams<sup>7</sup> reported the presence of fluorosis amongst children in high fluoride areas. The most comprehensive work was undertaken by Ockerse<sup>8-10</sup> and Ockerse and Meyer<sup>11</sup> delineating the areas of endemic fluorosis, the levels of fluoride in the different areas in South Africa and the observations of dental caries in these areas. Steyn and Reinach<sup>12</sup> and Steyn<sup>13,14</sup> devoted most of their efforts to the toxic effects of fluoride on human beings. It was Ockerse<sup>8-10</sup> however, who contributed to the body of knowledge on the relationship of endemic fluorosis (mottled enamel) and high levels of naturally occurring fluoride in the drinking water in many parts of Southern Africa. Ockerse<sup>8</sup> further demonstrated that schoolchildren residing in areas with varying levels of fluoride in the drinking water showed different levels of dental caries. Ockerse<sup>15</sup> reported that the prevalence of dental caries was at least twice as high amongst children in low fluoride areas compared with children in areas with fluoride levels between 1.1 to 1.8 mg/L.

Abrahams<sup>7</sup> confirmed much of Ockerse's findings in his studies in the North Western Cape. The work of Ockerse and Staz had prompted the Council for Scientific and Industrial Research (CSIR) to consider an investigation into the desirability of systemic water fluoridation.<sup>16</sup> Staz<sup>17</sup> reported that the CSIR: "records its approval of the suggestions to add fluorides to community water supplies as a positive preventive health measure to reduce the ravages of dental caries". In 1966 the report by the Commission of Inquiry into the Fluoridation of Water<sup>18</sup> recommended that: a) steps should be taken to encourage, advise and assist local authorities to fluoridate the water supplies of their communities as soon as possible and b) local fluoridation schemes should aim to achieve an optimal concentration of fluoride in the drinking water for the prevalent climatic conditions according to the criteria laid down. After publication of the report no action was taken by the government. During the 1970's a number of studies further explored the relationship between fluoride areas in South Africa.<sup>19-22</sup>

The second period extends from 1978 to 1989 during which time Taljaard<sup>23</sup> reported on the views of the profession and the Department of Health's future policy, which triggered public debate from those who opposed the implementation of water fluoridation. This public debate prompted the Secretary of Health to organise a National Symposium on Water fluoridation in 1979 in Pretoria.<sup>24</sup> The symposium ended inconclusively with no clear mandate to the government to implement water fluoridation. This was a watershed period for the pro-fluoridation lobby in South Africa. However, the resilience of the dental profession encouraged considerable further research into the levels of fluoride in drinking water in South Africa.<sup>25-28</sup> Simultaneously, the Medical Research Council and other organisations supported further research on fluorides in South Africa.<sup>25,27-31</sup> During this period some studies further explored the issues of fluorosis as a sequel to the ingestion of fluoride through drinking water sources.<sup>32-34</sup>

The third period stretches from 1990 to date. Following political changes the issue of water fluoridation, with its accompanying questions on fluorosis and fluoride in general was taken up once more. In 1991 the Medical Research Council organised a symposium on water fluoridation at which meeting a working group was formed to promote water fluoridation.<sup>35</sup> The Several political organisations<sup>36</sup> proposed the implementation of water fluoridation as a primary health care measure. In 1995, the Oral Health Committee

appointed by the Ministry of Health<sup>37</sup> recommended that the government implement water fluoridation as part of the Reconstruction and Development Plan.<sup>38</sup> A Sub-Committee on Water fluoridation was subsequently set up to oversee the implementation of water fluoridation in South Africa.

#### SOME PROMINENT RESEARCH FINDINGS

Table 1 reflects a summary of prominent research findings.

In the Northern Cape province Ockerse<sup>8</sup> found a much higher prevalence of dental caries in Upington compared to Kenhardt and Pofadder areas. Fluorosis prevalence also varied according to the fluoride content of the drinking water. He concluded that with fluorosis prevalence a 100 % in certain areas in and around Kenhardt and Pofadder, fluorosis is so severe that it is a predisposing cause of dental caries.

In the Pilanesberg area (North West Province) Ockerse and Meyer<sup>11</sup> found fluoride in the drinking water to vary between 0.33-35 mg/L. They also found climatic condition influences on the fluoride levels i.e. lower after heavy rain. This could also explain why at higher levels of fluoride 10.45 mg/L only moderate and not as expected severe fluorosis was observed (lower fluoride levels during early calcification).

TABLE 1. Prominent Research Findings						
References	Age	Country/ Region	Fluoride (mg/L)	Index	Fluoros. (%)	Moderate Severe,%
Ockerse & Meyer <sup>11</sup>	6-15	Pilanesberg	0.33-35	Dean	49	57
	6-17	Upington	0.38	Dean	16	3
Ockerse <sup>8</sup>	6-16	Kenhardt	6.8	Dean	100	70
	6-16	Pofadder	2.5 (av)	Dean	94	53
Bischoff <i>et al</i> <sup>19</sup>	14-23	Saulspoort	0.4-6	Dean	83	60
Van d Marrya at al <sup>20</sup>		Saulspoort (H)	0.4-6	Dean	83	60
v all u. Mel we el ul		Mabeskraal (L)	0.02-0.2	Dean	11.4	22
Retief <i>et al</i> <sup>22</sup>	14-17	Kenhardt	3.2	Dean	94	58
		Northwest		Dean		20
Zietsman <sup>31</sup>	5-20	Province	0.5 - 1.6	and	53	
		(5 villages)		TF		
Lewis <i>et al</i> <sup>32</sup>	6 18	KwaNdebele (H)	8 - 9	Dean	88	54
Lewis & Chikte <sup>33</sup>	0-18	KwaNdebele (L)	0.6 - 1.6		90	3

The higher than expected prevalence of fluorosis at Saulspoort<sup>19</sup> compared to findings in Austria was ascribed to the effect that higher mean maximum daily temperatures have on water consumption and subsequent incidence of fluorosis. Females were found to have less fluorosis than males. This was due to physical factors where females tried to remove the unsightly stains

TABLE 2. Pe	rceptions of	parents
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Questions	Response	Sub	Optimal	Supra
		(%)	(%)	(%)
Aware stains	Yes	69	62	92
Other	No	31	38	8
Of concern	Yes	54	54	78
	No	13	9	14
Aware stains	Yes	38	45	83
Own child(ren)	No	62	55	17
Of concern	Yes	33	39	75
	No	5	6	10

through manual abrasion with sand or ash and not due to internal genetic differences.

Van der Merwe  $et al^{20}$ also observed higher fluorosis а prevalence (11.4%) than generally would be expected at Mabeskraal a low F area (0.02-0.2 mg F/L). Two possible explanations were given: a) poor diet which was found to be a doubtful explanation and b) a possibility that the mottling observed was not dental fluorosis. The study also confirmed a higher caries level at the low fluoride area

TABLE 3. P children.	erceptions	of 12	- 15 yea	ars old
Question	Response	Sub	Optimal	Supra
		(%)	(%)	(%)
Aware stains	Yes	44	61	84
Other	No	56	39	16
of Concern	Yes	31	46	58
	No	14	15	26
Aware stains	Yes	15	45	69
Own hild(ren)	No	85	55	31
Of Concern	Yes	10	34	53
	No	5	10	16

of Malbeskraal compared to Saulspoort the high fluoride area.

These findings were also confirmed by Retief *et al*<sup>21</sup> in Kenhardt. They observed no differences of fluoride levels between the two maxillary incisors, nor did they find any differences for gender. This study in Kenhardt also indicated that in a high fluoride area such as this the severity of the fluorosis may be a predisposing factor to caries development. There was also a tendency for caries to decrease with increased enamel fluoride concentration in pupils with no or mild fluorosis.

Looking at spatial variation of fluorosis and fluoride content of water in an endemic area (5 villages in the North West Province) Zietsman<sup>31</sup> found a surprisingly high fluorosis prevalence 33.5 % in association with absolutely and relatively low F concentrations (0.54 mg/L and even 0.4 mg/L). The relationship between fluorosis prevalence and fluoride content of the drinking water was positive but the spatial variation in the prevalence was not fully explained by the variation in the fluoride content of the drinking water. It would seem as though a high prevalence was caused by the absence of water with low F content (<0.4 mg/L), rather than being either the presence of some sources with very high F content or a high mean F content.

Lewis *et al*<sup>32</sup> found similar prevalences of fluorosis in two areas with significantly different levels of fluoride in the drinking water, but significant differences existed in severity.

#### **CURRENT STUDIES**

Wide spread studies are currently being conducted among various communities in the Northern Cape Province with varying levels of fluoride in the drinking water with the

objective of arriving at a proposal in terms of what should be regarded as an optimal level of fluoride.

One of these studies also looked at the perceptions of the people in these communities regarding the appearance of fluoride exposed individuals, as well as the relationship of nutritional status and fluorosis.

Table 2 reflects some of the findings in terms of perceptions. Among the parents, the awareness of staining on the teeth of other individuals as well as on the teeth of their own children is highest in the supra-optimal communities. The concern parents had about the stains was also highest in the high fluoride areas. The same tendency was observed

among 12-15 years old children (Table 3). The caries experience (DMFT) was lower in the 15-year-olds from the optimal F areas but showed an increase for both 12- and 15-

year-olds from the optimal to the supraoptimal area. The 12-year-olds from the sub-optimal and optimal areas are very similar (Table 4). This may already be an indication that cariostatic benefit from the sub-optimal fluoride levels mav be comparable to the generally accepted optimal levels of around 1 mg/L. It may also be an indication that the optimal fluoride level in terms of caries prevention and fluorosis for the prevailing conditions is lower than generally accepted. This thinking is in agreement with Du Plessis<sup>34,39</sup> stating that a level of 0.54 mg/L appears acceptable and in the event of fluctuations if the fluoride content could be controlled so as never to rise above 0.7 mg/L, dental fluorosis should not be a problem.

**TABLE 4.** DMFT by fluoride level and age.

ugo.					
Sub-optimal					
Age	n	D	М	F	DMFT
6	38	0.10	0	0	0.10
12	120	0.46	0.13	0.09	0.68
15	73	1.81	0.34	0.01	2.16
Optimal	1				
Age	n	D	М	F	DMFT
6	27	0.07	0	0	0.07
12	80	0.50	0.23	0	0.73
15	75	1.29	0.21	0.07	1.57
Supra-Optimal					
Age	n	D	М	F	DMFT
6	47	0.11	0	0	0.11
12	130	1.12	0.32	0.03	1.47
15	104	2.64	0.59	0.12	3.35

#### DEFLUORIDATION

A draft report dated 29 May 1997 on the 'Feasibility of Fluoridation of the Water Supplies of Medium and Small Towns in South Africa' was compiled for 'The National Fluoridation Committee' of the Department of Health, Republic of South Africa by Dr S O'Hickey. Although the main objective of this report was to make recommendations on the feasibility of water fluoridation it also stated that dental fluorosis is a public health problem in some areas where there is an excess of fluoride, above the optimal range in the drinking water. Defluoridation in such circumstances is strongly recommended. It is also stated that defluoridation technology/equipment can be fitted to a single tap or home. This is of particular importance for small towns and especially small rural communities.

Defluoridated water is available in very few areas and the use of such water by the local people has been remarkably low and no reasons for this phenomenon seem to be forthcoming. The author of the above mentioned report on tasting the defluoridated water found it to have a slight but definitely unpleasant metallic taste which could be caused by prolonged storage in defluoridaters due to lack of use. This is clearly something which will need to be taken into account when defluoridation is perused on a more widespread scale. In terms of the South African Water Quality Guidelines<sup>40</sup> it is recommended that the concentration of fluoride in potable water should never exceed 4 mg/L due to the likelihood of skeletal fluorosis with crippling as well as tooth loss. By implication this is understood to presumably mean that in the absence of an alternative water source potable water exceeding 4 mg/L should be defluoridated. However, taking into account the fluorosis findings from local studies<sup>8,19,21</sup> provisional findings of studies currently in progress, as well as empirical observations the authors of this paper are of

the opinion that defluoridation should be considered at a lower level of 3 mg/L. The main reason for this opinion is due to the severity of fluorosis that is prevalent at this level which is not only an aesthetic problem, but may also be a predisposing factor to caries.<sup>8,21</sup> If the maximum allowable limit for fluoride in potable water of 1.5 mg/L as recommended by the South African Bureau of Standards<sup>41</sup> and Committee for Scientific Industrial Research (CSIR)<sup>42</sup> is adhered to, than the cut-off levels for considering defluoridation should be even lower. This is also in agreement with the guidelines of the World Health Organisation (WHO).<sup>43</sup>

The work done thus far on defluoridation in South Africa,<sup>44</sup> albeit very scanty favours two processes: (a) activated alumina adsorption methods and (b) reverse osmosis (membrane separation method). The activated alumina appears most attractive because alumina is somewhat specific for fluoride and has a relatively high fluoride exchange capacity. Regeneration, which can be performed with caustic soda, is fairly straightforward and the process is reliable, safe and relatively simple to use. Cases have also been demonstrated where fluorides in the concentration range of approximately 4-20 mg/L in borehole waters could be reduced to potable standards with activated alumina treatment. Reverse osmosis has also been demonstrated to reduce fluoride from approximately 12 mg/L to potable standards.

#### CONCLUSION

Fluorosis definitely appears to be a public health problem in certain areas in South Africa which require the attention of authorities at the various levels of government. This attention will need to consider defluoridation of water especially in areas where alternative water sources are not available. By implication this calls for defluoridation demonstration programmes establishing the efficacy, effectiveness and feasibility of defluoridation.

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## DENTAL FLUOROSIS IN RELATION TO ALTITUDE AND FLUORIDE IN DRINKING WATER IN UGANDA

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SUMMARY: The prevalence and severity of dental fluorosis among 10-14 year old children (n = 294) in four rural areas of Western Uganda were recorded using the TF index. The children, who were permanent residents in the areas, were exposed to drinking water containing either 0.5 or 2.5 mgF/L and were living at altitudes of 900 m and 2200 m (0.5 mgF/L) or 1750 m and 2800 m (2.5 mgF/L). In the 0.5 mgF/L areas, 25 % of the children (n = 81) living at 900 m had dental fluorosis as compared to 45% of the children (n = 82) at 2200 m above sea level ( $\chi^2$  =7.48). In the 2.5 mg F/L areas, 67% of the children (n = 67) living at 1750 m had dental fluorosis as compared to 84% of the children (n = 64) at 2800 m above sea level ( $\chi^2$ =5.25). Regression analyses showed that the fluoride intake from the drinking water explained most of the severity of dental fluorosis. Altitude had a significant (P < 0.05) effect, but explained only 2-3% of the variance in dental fluorosis.

Key words: Dental fluorosis; Prevalence; Altitude; Magadi.

#### INTRODUCTION

The association between dental fluorosis and the fluoride content of the drinking was established already 60 years ago and is still valid.<sup>1</sup> Thus there is no doubt that the endemic dental fluorosis found in the African Rift Valley primarily is caused by the high fluoride concentration in the drinking water.<sup>2-5</sup> However, several studies have come up with results that cannot be explained solely on the basis of the fluoride content of the ingested water.<sup>6-13</sup> Besides the fluoride in the drinking water, factors such as temperature,<sup>14-17</sup> altitude of residence,<sup>9,18</sup> diet,<sup>9,13,19</sup> nutritional status,<sup>20,21</sup> and dentifrice<sup>22-28</sup> have been reported to influence dental fluorosis.

Most of the reports on dental fluorosis in the Rift Valley area are from Kenya, Tanzania and Ethiopia. Only one study on the prevalence and severity of dental fluorosis in Uganda seems to have been carried out,<sup>2</sup> and great variation in the prevalence of dental fluorosis was found, even in areas with the same fluoride concentration in drinking water. That study was performed in 4 districts. Variations in ethnic and social background, diet, and altitude may have contributed to the heterogeneous findings.

Due to these heterogeneous results the aim of this paper is to report the prevalence and severity of dental fluorosis among Ugandan children, who are relying on either high- or low-fluoride levels in the drinking water, and who are living at different altitudes.

#### MATERIALS AND METHODS

The survey was conducted in Western Uganda, which geologically is a part of the great African Rift Valley. Two village schools were selected in the Kisoro district; Mutolere (1,750 m above sea level) and Kabindi (2,800 m), and two villages in the Kasese district; Mpondwe (900 m) and Kyabayenze (2,200 m). Mutolere and Kabindi

had piped water from the same source. Also Mpondwe and Kyabayenze shared a common water supply. The fluoride contents of the waters were unknown at the time of examination in 1996. The piped water systems had been operating since 1962 (Kisoro) and from 1982 (Kasese).

A total of 294 schoolchildren (Mutolere = 67, Kabindi = 64, Mpondwe = 81 and Kyabayenze = 82) were randomly selected for the examination (Figure 1). Children included in the study had to fulfill the following criteria: aged 10-14 years, born and raised in the village, absence from the village for not more than 1 month a year during the first 6 years of life and use of drinking water from the same source during the first 6 years of life.

Permission to carry out the study was obtained from the relevant health and school authorities, and informed consent was given by the parents of the children. Clinical examination was done by the principal investigator (MCR) under field conditions following the criteria specified by WHO.<sup>29</sup> By the clinical examination, the child was sitting on an ordinary chair outside the school building with indirect natural daylight as the source of illumination. Cotton rolls were used to wipe the teeth dry and control saliva. Assessment of dental fluorosis was done according to the Thylstrup and Fejerskov index (TFI) modified in 1988.<sup>30</sup>

After the clinical examination, the child and his/her mother were interviewed according to a structured questionnaire in order to get information on residence, dietary habits during the early childhood, oral hygiene practices. Based on the amount of liquid, cups per day, consumed and the fluoride level in the drinking water, the fluoride exposure during early childhood was estimated.

Samples of drinking water and  $magadi^{I}$  were collected and analyzed for fluoride. Chisquare ( $\chi^2$ ) statistics were used to assess significant differences between groups, and multiple regression analyses to determine factors related to dental fluorosis. The significant level was set at 5%.

#### RESULTS

The fluoride concentration of the drinking water in Kisoro (Mutolere and Kabindi) was 2.5 mg F/L, and in Kasese (Mpondwe and Kyabayenze) 0.5 mg F/L, cf. Figure 1. The fluoride concentration in the magadi samples ranged from 2.8 to 17.1 (mean 8.5) mg F/kg, cf. Table 1. The distribution of children according to sex showed no significant difference in each of the respective areas (Figure 1). There was no significant difference ( $\chi^2 = 3.0$ , df = 1)

<b>TABLE 1</b> Fluoride concentration inmagadi from Uganda.				
	mg F⁻/kg	g Magadi		
Sample	Sample	Average		
1	2.8			
2	5.5	8.5		
3	17.1			

between males and females in the prevalence of dental fluorosis.

In areas with 0.5 mg F/L in the drinking water, 25 % of the children at 900 m had dental fluorosis as compared to 45 % at 2,200 m above sea level ( $\chi^2 = 7.48$ , df = 1). In areas with 2.5 mg F/L in the drinking water, 67 % of the children had dental fluorosis

<sup>&</sup>lt;sup>1</sup> The Ugandan Magadi (Kutwe salt) is a Trona collected from shores of Lake Kutwe. The salt is very popular used in the local households as tenderizer and taste improver in food.







**FIGURE 2**. Prevalence of dental fluorosis (permanent teeth) according to altitude and fluoride in the drinking water.

at 1,750 m as compared to 84% of the children at 2,800 m above sea level ( $\chi^2 = 5.25$ , df = 1), cf. Figure 2.

The increasing prevalence by altitude and fluoride in water was also reflected in increased severity of dental fluorosis with altitude and fluoride concentration in the drinking water, cf. Figure 3.

The proportion of children with a daily water intake of  $\ge 1.75$  liters was higher ( $\chi^2 = 8.4$ , df = 1) in Kisoro as compared to Kasese, cf. Figure 4. Regression analyses showed that the fluoride exposure from the drinking water explained 68 % of the dental fluorosis in the Kisoro and 31 % in Kasese. Altitude only explained 2 % and 3 % in the respective districts. However, the effect of altitude was significant (P < 0.05,





Table 2). Magadi had no significant effect on the prevalence of dental fluorosis ( $\chi^2 = 2.7$ , df = 1, Table 3). According to the mothers, the average consumption of magadi was 2 g twice a week.

#### DISCUSSION

The prevalence and severity of dental fluorosis showed a direct relationship with the concentration of fluoride in the drinking waters and with altitude, cf. Figure 2. The differences in the fluoride exposure explained more of the variations in the prevalence of dental fluorosis in the 2.5 mg F/L area than in the low fluoride area. Children in the high fluoride area consumed more liquid than in low fluoride area. However, the high fluoride level in the water rather than the difference in water intake may account for the greater explanatory power of the fluoride exposure in the high fluoride area.

<b>TABLE 2.</b> Variables related to the prevalence of dental fluorosis according to district.				
District	Variable	Regression	Prediction	P-value
		Coefficient	(%)	
Kisoro	<sup>1</sup> F exp. from water	0.83	67.7	0.001
2.5 mg F/L	Altitude	0.13	1.5	0.01
Kasese	<sup>1</sup> F exp. from water	0.55	30.6	0.001
0.5 mg F/L	Altitude	0.19	3.3	0.005
<sup>1</sup> The fluoride exposure (mg) from water is the product of volume of water (I) taken per (a 3)				

years old) child per day and the concentration of F (mg/L) in drinking water.

**TABLE 3.** Use of Magadi according to children with and without dental fluorosis.

Use of magadi	Children with fluorosis	Children without fluorosis
Yes	130	124
No	26	14
$(\gamma^2 = 2.7, df = 1)$		

When simple analyses were only carried out, the prevalence of dental fluorosis was significantly related to altitude. Regression analyses, however, showed only a limited, but significant effect of altitude. This finding strengthened results from Tanzania. They claim that the effect of altitude, first reported from Kenya,<sup>18</sup> could rather be explained by the use of magadi in food preparation.<sup>9</sup> In our study about 2.0 g magadi is consumed per child twice a week, according to the mothers' estimates. Provided, a mean concentration of 8.5 mg F/kg magadi, compared to 36 - 6800 mg F/kg in Tanzania, a child will only consume about 0.03 mg F/week from the magadi. This is about 1/10 of the amount ingested daily from the drinking water in a 0.5 mg F/L area and thus the magadi probably contributes little to the severity of dental fluorosis in our material. The prevalence of dental fluorosis in our study was relatively low as compared to results from other African studies with similar range of fluoride concentration in the drinking water.<sup>2,6,9,11,12,31</sup> The different prevalences may partly be explained on the basis of factors such as temperature, altitude, differences in life styles and dietary habits such as the use of magadi. Moreover, other possible explanations are the criteria used for recording fluorosis, conditions under which the examinations were done inter-examiner variations, and analyses of the data.

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## DENTAL FLUOROSIS WITH SPECIAL REFERENCE TO INCISORS AND MOLARS OF TANZANIAN SCHOOL CHILDREN

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SUMMARY: The prevalence and severity of dental fluorosis are investigated in (maxillary) central incisors and first molars in 192 children (6-16 years of age) born and raised in two neighbouring municipal communities, Arusha and Moshi. The fluoride content of the community water supply in Moshi was 0.4 mg F/L as compared to 3.8 mg F/L in Arusha. Randomly selected children from 8 different schools were examined according to field conditions. Fluorosis was found to be endemic in both communities: 91 % of the children in Arusha had a Thylstrup/Fejerskov index of  $\geq$  1, as compared to 65 % in Moshi. In Arusha 90% of the upper central incisors and 68 % of the upper first molars had fluorosis scores  $\geq$  1. Similarly, in Moshi, fluorosis prevalence was 57 % and 51 % in, respectively, central incisors and first molars. Also, the severity of fluorosis was higher in Arusha than in Moshi: 70 % of the Incisors in Arusha had a TF score  $\geq$  4 as compared to 27 % in Moshi. Similar scores for first molars were 32 % and 19 %. Our findings underscore the importance of the fluoride content of drinking water in the development of dental fluorosis. However, the present study does not support the commonly held opinion that dental fluorosis is more prevalent in the six-year molars than in the central incisors<sup>3</sup> in fluorosis endemic areas.

Key words: Dental fluorosis; Upper central incisors; Upper first molars; Tanzania.

#### INTRODUCTION

A positive relationship is normally found between the fluoride content of drinking water and the prevalence and severity of dental fluorosis. However, wide individual variations in severity as well as distribution of dental fluorosis may be seen within the same area.<sup>1,2</sup> No tooth is immune to dental fluorosis, but all teeth are not equally affected. According to Fejerskov et al.<sup>3</sup>, teeth that develop early in life, such as incisors and permanent first molars are the least affected.<sup>3,4</sup> One would expect that the two groups of teeth, mineralising at more or less the same time, would be equally affected by the intake of high-fluoride water. However, even among this group of early mineralising teeth, the relative severity of fluorosis may differ: according to a study from Uganda, the maxillary incisors had the highest scores,<sup>2</sup> whereas a similar study in Kenya reported the first molars to be most affected.<sup>5</sup> Various explanations have been put forward to explain the observed variability; such as individual patterns of mineralisation, varying thickness of the enamel in the affected teeth etc.<sup>3,4</sup>

The present study is part of a greater project to assess oral health, especially dental fluorosis, in selected areas in the East African Rift Valley and to elucidate the importance of nutritional and other relevant factors. We here report on our findings regarding dental fluorosis in permanent central incisors and molars in school children in two Tanzanian communities with, respectively, high and low fluoride content in the drinking water.

#### MATERIALS AND METHODS

Two neighbouring municipalities in the Rift Valley area of Northern Tanzania, Arusha and Moshi, were chosen for the study. Arusha is located at approximately 1500 m above sea level, whereas Moshi is at slightly lower elevation. Ethnicity and standard of living were similar, and both

TABLE '	1.	Distrik	oution	of	subjects
according	to	age,	area	and	fluoride
concentrat	ion	in the	drinkir	ig wa	ter.
A	<b>A</b>	le -	М	1-1	

Age	Arusha	Moshi	Total
years	3.8 mg/ L	0.4 mg/ L	Total
6-9	46	9	55
10-16	74	63	137
Total	120	72	192

communities had public water works serving the whole population.

Approximately 30 children, 6-16 years of age, were randomly selected from each of 8 urban schools (5 in Arusha and 3 in Moshi), cf. Table 1. The selection was based on the schools' attendance register. Only children born and raised in the respective areas were included in the study. Using a structured questionnaire, subjects and accompanying parents were interviewed on diet, dietary habits and living conditions. Special emphasis was put on questions regarding the intake of fluoride containing food items such as **tea** and **magadi**.

All oral examinations were carried out by the principal investigator (AKA) under field conditions<sup>6</sup> in the shade outside the schools, while the children were seated on a school chair. The teeth were cleaned and dried using gauze and cotton rolls. The buccal surfaces of the permanent teeth were inspected for dental fluorosis, scorings were recorded using the Thylstrup-Fejerskov index (TF-index).<sup>7</sup> In order to make comparison with earlier studies, scorings were also recorded according to Dean's Index.<sup>8</sup> In case of doubt, lowest score was given. Teeth with less than 50% of the buccal surface visible, were excluded.

Drinking water samples were collected, and analysed for fluoride by the use of the ion selective electrodes, according to standard procedures (ORION 9600 BN). The SPSS program was used for data entry and statistical analyses.

#### RESULTS

The fluoride content of the relevant water sources was 3.8 mg/L in Arusha and 0.4 mg/L in Moshi.

The prevalence of dental fluorosis in the two cities was 91% and 65% in Arusha and Moshi, respectively.

As demonstrated in Figure 1, the prevalence of dental fluorosis - in both communities - was higher in maxillary central incisors than in first molars. The difference was statistically significant in Arusha (paired t-test t = 5.17, df = 119, p = 0.001), but not in Moshi (t = 1.58, df = 71, p = 0.118 NS).





Also, as shown in Figure 2, the **severity** of fluorosis was more pronounced in incisors than in molars. A significant difference was, again, found in Arusha (paired t-test, t = 8.35, df = 100, p = 0.001), whereas, in Moshi the difference between the prevalence of severely fluorosed incisors and 1st molars (TF=  $\geq$ 4) was insignificant, according to the t-test (t = 1.42, df =71, p = 0.159 NS).

As compared to individuals who started to drink tea during the first year of life as well as to those who started to take tea only during the third year of life, significantly higher prevalence of dental fluorosis was recorded in individuals who started to drink tea while they were between 1 and 2 years of age (cf. Figure 3). (Arusha,  $\chi^2 = 37.8$ , df=2, p=0.0001; Moshi,  $\chi^2 = 15.3$ , df=2, p=0.0005).

Magadi was found to be a common food additive in both Moshi and Arusha. In Moshi, 70 % of those who had dental fluorosis consumed magadi while only 40 % did in Arusha (cf. Figure 4). The data, however, included no information on the time of introduction to the diet of the child.

#### DISCUSSION

Considering the low fluoride content of the drinking water, the prevalence of fluorosis might seem unexpectedly high in Moshi. This may partly be explained by the frequent use of magadi; the local brand of which has a high content of



fluoride.<sup>9</sup> Our findings are in agreement with what has previously been reported from the same area,<sup>10</sup> and the prevalence might even be considered low in comparison to what has been reported from two low-fluoride areas in Kenya<sup>11</sup> and in the Sudan.<sup>12</sup>

The difference in prevalence and severity of fluorosis between maxillary central incisors and first molars was found to be significant only in high-fluoride Arusha. A similar but weaker trend was seen in low-fluoride Moshi. This pattern of dental fluorosis is different from what has been presented e.g. by Fejerskov *et al*,<sup>3</sup> Manji *et al*,<sup>5,11</sup> van Palestein *et al*,<sup>4</sup> but is in agreement with Møller *et al*.<sup>2</sup>

No convincing explanation has been offered for the conflicting findings. It should be noted, however, that, according to Watson and Lowrey,<sup>13</sup> the mineralization of the first molars will start at birth, while the incisors may start three to four months later. Depending upon the weaning time and the introduction to high-fluoride water, tea and possibly magadi, this small difference in time, may play a decisive role in development of dental fluorosis. Further studies of the early feeding habits in Arusha and Moshi should be conducted.

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# SKELETAL FLUOROSIS AMONG RETIRING EMPLOYEES OF WONJI SHOA SUGAR FACTORY

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SUMMARY: A retrospective study was done involving 263 retiring employees of Wonji Shoa Sugar Factory, an estate located in the Ethiopian Rift Valley and which is known for the high fluoride levels in its domestic water supplies. The radiological prevalence of skeletal fluorosis in the study sample was found to be 70.3 %. Skeletal fluorosis was associated with male gender (p<0.05) and also the prevalence was higher among factory and agricultural workers than among administrative workers (p<0.05). The association between male gender and fluorosis persisted within strata of occupation, indicating that the association was not mediated by occupation. The prevalence of clinical signs suggestive of skeletal fluorosis was markedly lower than the radiological prevalence and raged between 20.6 % and 40.2 % for the whole sample, indicating that many cases were asymptomatic. Further clinical epidemiological studies are suggested and strengthening of the existing defluoridation program is recommended.

Key words: Skeletal fluorosis; Occupational factors; Gender factors; Rift Valley; Ethiopia.

#### INTRODUCTION

Individuals depending on water supplies with fluoride levels greater than 3-6 mg/L or ingesting more than 10-20 mg fluoride daily are likely to develop skeletal fluorosis after 10-20 years of exposure.<sup>1,2</sup> Early skeletal involvement by fluorosis is not clinically obvious even though radiological changes are discernible in the skeleton at early stages.<sup>3</sup> In later stages skeletal fluorosis is manifest with restriction of movement of the spine and of the joints of the limbs and with neurological complications. Radiologically, skeletal fluorosis is characterised by increased radiodensity, by osteophyte formation, and by calcification of ligaments, tendons and interosseous membranes. The most obvious changes are seen in the spine and pelvis.<sup>4,5</sup> Although the entire spine may be affected, pronounced changes occur in the lumbar spine.<sup>5</sup>

Endemic skeletal fluorosis is widespread in the East African Rift Valley due to high levels of fluoride in the water sources.<sup>6</sup> Wonji Shoa Sugar Factory (WSSF) is an agroindustrial estate of 5000 hectares in central Ethiopia located in the Rift Valley. It was established in 1954. The estate presently has about 8000 employees working in three sectors-factory, agriculture, and administrative office. Most of the factory and administrations workers live in the two factory villages and most of the agricultural workers reside in the 14 plantation villages. The community of the estate has depended on well water, which has been known to have high fluoride levels. It has a dual water supply of raw water for washing purposes and defluoridated water for drinking and cooking purposes. Defluoridated water was made available to the factory villages since 1962 and to all plantation villages since 1976.

In 1972, the existence of spinal fluorosis among the workers of the estate was discovered. Between 1976 and 1984, 244 workers retired before reaching the official

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age of retirement, i.e. 55 years, due to disable spinal fluorosis.<sup>6</sup> At the time of retirement the employees of WSSF are required to undergo radiological examinations of the lumbar spine, and estimation of fluorosis-related disability is made by an independent medical board based on clinical and radiological findings. The purpose of the present survey is to estimate the present prevalence of skeletal fluorosis among the adult population of the estate by retrospectively examining the data of employees who had retired over a two year period.

#### MATERIALS AND METHODS

The survey was conducted in October 1997 and involved all employees of WSSF who retired between January 1, 1995 and December 31, 1996. Demographic and clinical data were obtained from the report forms of the examining medical board. The X-rays of the lumbar spine (PA and lateral view) were blindly examined by a radiologist (one of the investigators) and graded according to Roholm's classification which identifies three stages of skeletal fluorosis; Stage I, II, and III.<sup>7</sup> A total of 308 workers retired during the two year period. Completed medical board report forms and X-rays of the lumbar spine were available for 263 (85.4%) of the retirees and all of these were included in the survey.

#### RESULTS

**Demographic data.** Only one person (0.4 % of the sample) was aged 50 years, all the rest being 55 years old at the time of retirement. Of the sample, only 13 subjects (4.9 %) were females. The duration service in the estate was above 20 years for 261 retirees (99.2 % of the total) and was 16-20 years for the remaining 2 retirees (0.8 %). Among the study subjects, 106 individuals (40.3 %) worked in the factory sector, 125 individuals (47.5 %) worked in the agricultural sector and 32 individuals (12.2 %) worked in the administration sector. The last one included the workers who worked in personnel, finance, material supply, medical, etc. departments.

**Clinical data.** According to the fluorosis-related disability estimation made by the examining medical board, 146 individuals (55.5 % of the sample) were found to have no disability. The remaining 117 individuals (44.5 % of the sample) were assessed as having some degree of disability. The prevalence of five clinical signs that may be related to skeletal fluorosis and which were looked for by the medical board is presented in Table 1. The prevalence of those clinical signs in the whole sample ranged between 20.6 % and 40.2 %.

Factory who retired during 1995 and 1996.								
Clinical Signs	Present			Absent	Te	Total		
Chinical Signs	No	%	No	%	No	%		
Kyphosis	54	20.6	208	79.4	262	100.0		
Impaired walking	62	23.9	197	76.1	259	100.0		
Impaired squatting	101	39.1	157	60.9	258	100.0		
Impaired neck mobility	77	30.0	180	70.0	257	100.0		
Impaired lumbar mobility	105	40.2	156	59.8	261	100.0		

**TABLE 1.** Prevalence of five clinical signs among employees of Wonji Shoa Sugar Factory who retired during 1995 and 1996.

**Radiological findings.** Of the 263 subjects studied, 185 individuals (70.3 % of the total) had radiological signs of spinal fluorosis. Among those affected, 113 individuals (61.1 %) had stage I fluorosis, 42 individuals (22.7 %) had stage II fluorosis and 30 individuals (16.2 %) had stage III fluorosis. The only individuals who retired at the stage of less than 55 years had stage II fluorosis. Of the two individuals who had served in the estate for less than 20 years, one was radiologically free of skeletal fluorosis but the other one had stage III fluorosis.

The distribution of the three stages of skeletal fluorosis among the study subjects by sex is shown in Table 2. The radiological prevalence of all the stages of skeletal fluorosis was 72.0 % (180 individuals out of 250) among the men. The radiological prevalence of all stages of skeletal fluorosis was 38.5 % (5 individuals out of 13) among the women. The difference in the prevalence of skeletal fluorosis between the two sexes was statistically significant (p=0.02, Fisher's exact test). Furthermore, all women who were found to have fluorosis had stage I fluorosis.

**TABLE 2.** Distribution by sex of the three stages of skeletal fluorosis among employees of Wonji Shoa Sugar Factory who retired during 1995 and 1996. (figures in parentheses represent percentages of the row totals).

C	Radiolog	ically fluor	otic		Radiologically	$T_{abal}(0)$
Sex	Stage I	Stage II	Stage III	All stages	non-fluorotic	1 otal (%)
Male	108	42	30	180 (72.0)	70 (28.0)	250 (100)
Female	5	0	0	5 (38.5)	8 (61.5)	13 (100)
Total	113	42	30	185 (70.3)	78 (29.7)	263 (100)

The distribution of the three stages of skeletal fluorosis among the study subjects by work is shown in Table 3. It can be seen that the prevalence of all stages of skeletal fluorosis was 77.4 % among the factory workers, 72.0 % among agricultural workers and 40.6 % among administration workers. The difference was statistically significant (Chi-square test; p=0.0003). Among the 13 administration workers who were found to have skeletal fluorosis, 12 had stage I fluorosis while only one had stage II fluorosis, none having stage III fluorosis.

**TABLE 3.** Distribution by work place of the three stages of skeletal fluorosis among employees of Wonji Shoa Sugar Factory who retired during 1995 and 1996 (figures in parentheses represent percentages of row totals).

					·	
Work Dlago		Radiolog	ically Fluor	Radiologically	Total (7)	
work race	Stage I	Stage II	Stage III	All stages	non-fluorotic	10tal (70)
Factory	37	25	20	82 (77.4)	24 (22.6)	106 (100)
Agriculture	64	16	10	90 (72.0)	35 (28.0)	125 (100)
Administration	12	1	0	13 (40.6)	19 (59.4)	32 (100)
Total	113	42	30	185 (70.3)	78 (29.7)	263 (100)

The occurrence of four clinical signs and of fluorosis-related disability in accordance with the existence of fluorosis among the study subjects is shown in Table 4. Among the fluorotic individuals, the prevalence of four clinical signs that could be related to skeletal fluorosis (kyphosis, impaired neck mobility, impaired squatting and impaired lumbar mobility), ranged between 23.4 % and 47.3 %.

1996.						
Clinical sign		Radiologicall	y fluorotic	Radiologically non-fluorotic		
Chincal sign		No	%	No	%	
	Present	43	23.4	11	14.1	
Kyphosis	Absent	141	76.6	67	85.9	
	Total	184	100.0	78	100.0	
Impaired	Present	82	45.3	19	24.7	
Squatting	Absent	99	54.7	58	75.6	
	Total	181	100.0	77	100.0	
Impaired neck	Present	67	36.8	10	13.3	
mobility	Absent	115	63.2	65	86.7	
moonity	Total	182	100.0	75	100.0	
Impaired	Present	87	47.3	18	23.4	
lumbar mobility	Absent	97	52.7	59	76.6	
	Total	184	100.0	77	100.0	
Disability	Present	87	47.0	59	75.6	
	Absent	98	53.0	19	24.4	
	Total	185	100.0	78	100.0	

**TABLE 4.** Occurrence of four clinical signs and of disability among fluorotic and nonfluorotic individuals who retired from Wonji Shoa Sugar Factory during 1995 and 1996.

#### DISCUSSION

The high radiological prevalence of skeletal fluorosis in the sample (70.3 %) suggests that the condition is prevalent among the adults residents of the estate. About three-fifth of those affected had stage I fluorosis, the rest having the more severe stages of fluorosis. A survey conducted in 1979-80 and involving 530 workers aged 45-55 years found radiological evident skeletal fluorosis in 46 % of them (personal communication, Wonji Hospital). The lower prevalence in that survey could be due to differing criteria of radiological classification and to difference in duration of residence in the estate of the study subjects.

In the present study skeletal fluorosis was associated with male gender. This association persisted within strata occupation (administrative and non-administrative), indicating that the association between male gender and fluorosis was not mediated by occupation. The number of women in the survey was very small to make sound conclusions but the lower prevalence of fluorosis among the women studied was not because of engagement in administrative work. Lower prevalence of skeletal fluorosis in females than in males was previously reported by a study in India.<sup>3</sup>

The higher prevalence of skeletal fluorosis among factory and agricultural workers than among administrative workers could be due to agronomic factors which predispose workers to varying amounts of water ingestion. This might also explain the lower prevalence of the more severe stages of fluorosis among administrative workers.

The prevalence of certain clinical signs suggestive to skeletal fluorosis was low in the present study as compared to the radiological prevalence, and ranged between 20.6 % and 40.2 % for the whole sample, and between 23.4 % and 47.3 % among the fluorotic subjects. This suggests that many individuals had asymptomatic skeletal fluorosis. A survey conducted in 1990 and involving 328 adults had found physical

impairments indicative of skeletal fluorosis in 63 % of those who resided in the area for greater than 20 years.<sup>8</sup>

It is suggested that elaborate clinical and epidemiological surveys be carried out to better understand the exact magnitude and nature if the problem and potential risk factors that may predispose to the development of skeletal fluorosis. It is recommended that revival and strengthening of the existing defluoridation program of drinking water supplies of the estate be undertaken urgently.

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## PREVALENCE OF DENTAL FLUOROSIS IN THE WONJI SHOA SUGAR ESTATE

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ABSTRACT: Wonji Shoa Sugar Estate is a large compound in central Ethiopia known for the high fluoride levels in its drinking water supplies. A defluoridation programme of drinking water in a parallel supply has been going on for the last 35 years. Three samples with a total of 234 children aged 8-12 years from four villages of the estate were examined for dental fluorosis. Mouth prevalence rates of 95.7 %, 92.4 % and 71.8 % were found respectively for Wonji village, Shoa village and villages M and O for all grades of dental fluorosis. The variations in the occurrence and severity of dental fluorosis between the four villages are discussed and the impact of the defluoridation programme is appraised. The poor impact of the defluoridation programme is attributed to low reliability of the defluoridation and inconsequent consumption of defluoridated water.

Key words: Dental Fluorosis; Defluoridation programme; Rift Valley; Ethiopia.

#### INTRODUCTION

Fluoride is an essential trace mineral that is present in trace amounts in every human tissue but becomes concentrated in bones and teeth.<sup>1,2</sup> Fluoride in foods and water is easily observed by way of the portal system. From the amount of ingested, about half is retained in bones and teeth and the rest is excreted in urine.<sup>1,3,4</sup>

Ingestion of excessive fluoride during the period of tooth development causes dental fluorosis. The pathological changes of dental fluorosis involve hypoplasia and/or hypocalcification of the enamel.<sup>1,5</sup> Hypoplasia which is due to poor formation of the organic matrix of the enamel or missing chunks. Hypocalcification results in lack of luster and is manifest clinically as white, yellow or brown lines or areas (opacites).

Dental fluorosis can occur both in deciduous teeth and in the permanent teeth, the latter being more affected.<sup>6</sup> The extent of fluoride uptake in different parts of the dentation and skeleton depends upon the amount ingested and absorbed by the organism, the duration of fluoride exposure and the type, the region and the metabolic activity of the tissue concerned. Dental fluorosis is a good indicator of exposure to excessive amounts of fluoride. It can be easily studied by examining the permanent incisors. The critical age of calcification of the enamel of the permanent incisors is 3-4 years. The permanent incisors erupt during age of 7-9 years.<sup>7</sup>

Endemic fluorosis is widely distributed through out the world. Its occurrence has been reported in the USA, China, England, India, South Africa, New Zealand, Sweden, Morocco, and Cuba.<sup>8</sup> It is wide-spread in the East African Rift Valley, including the Ethiopian Rift Valley due to the high levels of fluoride in water sources.<sup>9</sup> Wonji Shoa Sugar Estate (WSSE), established in 1954 and located in central Ethiopia within the Rift Valley, has been known to have high levels of fluoride in its water supplies. The community residing in the two factory villages (Wonji and Shoa) and 14 plantation

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villages scattered throughout the estate has depended on well water having high fluoride levels for domestic purposes.

In the two factory villages of WSSE, a defluoridation programme of drinking and cooking water supplies has been going on since 1962. In the plantation villages, a defluoridation programme was launched in 1976 but has been discontinued for the last two years. Raw water has been supplied separately to all factory and plantation villages right from the beginning. So there has been a dual supply of treated and raw water. In some plantation villages, when well water is unavailable from taps, the residents consume water from the nearby river, Awash River.

After the defluoridation programme was started, a survey of dental fluorosis was carried out in 1985 in four villages of the estate. The purpose was to examine the impact of the defluoridation activities on the prevalence of dental fluorosis.<sup>5</sup> The purpose of the present survey is to estimate the present prevalence of dental fluorosis in the same four villages and make an assessment of the outcome of the defluoridation programme in the light of the earlier studies.

## MATERIALS AND METHODS

The target population was all children aged 8-12 years, residing in one of the four villages Wonji, Shoa, Village M or Village O, and having 6 or more permanent anterior teeth erupted. All eligible children learning in Grades 1-3 of the two of the three elementary schools in Wonji villages and one of the two elementary schools in Shoa villages were registered, producing a sample of 70 children for Wonji villages and a sample of 79 children for Shoa village, respectively. For villages M and O all eligible children were registered and a random sample of 85 children was selected.

Each child's guardian was interviewed and every child had examination of the anterior teeth under natural light by one of the investigators. Dental fluorosis for each examined tooth was graded according to the classification criteria of Dean and modified by Møller.<sup>6,7,10,11,12</sup>

Normal: The enamel is translucent, smooth and presents a glossy appearance.

- Very mild: Small paper white areas on the tooth involving less than 25 % of the surface.
- Mild: Opaque areas involving up to 50 % of the surface.
- Moderate: The whole of the enamel surface may be affected with chalky white areas or yellowish or brown staining; surface subject to attrition become worn.
- Severe: The enamel is grossly defective, opaque, pitted, stained brown and brittle, and may have a corroded appearance.

Data were collected in July 1997. Data were processed using a computer. Prevalence rates of dental fluorosis were calculated in two ways:

- Mouth prevalence rate as the percentage of children having at least one tooth affected.
- Tooth prevalence rate as the percentage of affected teeth.

## RESULTS

In total 234 children were included in the survey. Baseline characteristics of the respondents are given in Table 1 and 2.

Ethopia 1997.							
Consumption	None		So	ome times	Frequently		
Consumption	No	%	No	%	No	%	
Sweets	15	6.4	215	91.9	4	1.7	
Sugar cane	7	3.0	97	41.5	130	55.6	

**TABLE 1.** Consumption of sweets and sugar cane among study subjects, Wonji, Ethiopia 1997.

The male and female sex ratio level 1:1.2. Most of the respondents (88.5 %) were born in the villages of the state and 94.0 % had lived in the estate during the early childhood (i.e. up to age of 4 years). The average monthly family income was 101-600 Birr for 74.4 % of the children. Most of the children (93.6 %) have been consuming sweets sometimes or frequently and 97.0 % have been consuming sugar cane sometimes or frequently.

The results of the examination of the teeth are summarised in Table 3. The tooth prevalence rates of dental fluorosis for each of the three samples are presented in Table 4. The mouth prevalence rates of dental fluorosis for each sample are given in Table 5. The table shows the mouth prevalence rates of all grades of dental fluorosis (very mild to severe) and also for all grades of dental fluorosis excluding the very mild cases.

For the sample from Wonji villages, only 3 children (4.3 %) has all anterior teeth free form dental fluorosis and all of these children were born outside the estate. From the sample of Shoa village, only 6 children (7.6 %) had all anterior teeth free from dental fluorosis. Among these 6 children, four were born outside the estate. In the sample from village M and O, 24 children (28.25) had all their anterior teeth unaffected with fluorosis. Among these, 22 were born and grown up in the estate.

TABLE 2. Baselir	ne characte	eristic of			
respondents					
n=234	No	%			
Sex					
Male	106	45.3			
Female	128	54.7			
Place of Birth					
Estate	207	88.5			
Other	27	11.5			
<b>Residence</b> (childhood) <sup>*</sup>					
Estate	220	94.0			
Other	14	6.0			
Present Residence					
Shoa	79	33.8			
Wonji	70	29-9			
Camp 11	43	18.4			
Camp 12	42	17.9			
Average monthly income					
$\leq 100$	16	6.8			
101-300	93	39.7			
301-600	81	34.6			
> 600	44	18.8			
* Up to age 4					

#### DISCUSSION

The results of this survey showed high prevalence rates of dental fluorosis in the four villages of WSSE studied particularly in Wonji and Shoa villages. The prevalence rates are comparable to those found in previous studies. One study in 1977 showed and 87% mouth prevalence of dental fluorosis among children 5-10 years old.<sup>12</sup> Another study involving 1456 individuals in 14 communities of central part of the Ethiopian Rift Valley found prevalence rates of dental fluorosis ranging between 69% and 98%, the average rate being 84%.<sup>13</sup>

A survey conducted in 1985 in WSSE, and involving the examination of the permanent incisors in 8 years old children found mouth prevalence rates of 64 % for Wonji village, 75 % for Shoa village and 34 % for village M and O.<sup>5</sup> The corresponding tooth prevalence rates were 49 %, 56 % and 18 % respectively. Among adults 20-25 years old a mouth prevalence rate of 77 % and a tooth prevalence rate of 69 % were found. That survey used a grading of dental fluorosis different from the one that was used in this survey but it can be seen that still there are high rates of dental fluorosis in the estate.

IADLL	<b>LE 5.</b> Results of dental examination among respondents, wonji, Ethopia, 1997.											
Туре	Degree of tooth damage (problem), n=234											
of	Nor	rmal	Very	y mild	N	Iild	Mo	derate	Se	vere	D	MF
Tooth	No	%	No	%	No	%	No	%	No	%	No	%
11	37	15.8	48	20.5	27	11.5	82	35.0	40	17.1	-	-
12	57	24.4	38	16.2	29	12.4	78	33.3	11	4.7	21	9.0
21	41	17.5	47	20.1	28	12.0	78	33.3	39	16.7	1	0.4
22	55	23.5	35	15.0	32	13.7	77	32.9	10	4.3	25	10.7
31	86	36.8	52	22.2	48	20.5	42	17.1	5	2.1	1	0.4
32	86	36.8	51	21.8	49	20.9	40	17.1	4	1.7	4	1.7
41	86	36.8	53	22.6	48	20.5	42	17.9	5	2.1	-	-
42	85	36.3	53	22.6	47	20.1	40	17.1	4	1.7	5	2.1

TABLE 3.	Results of dental examination among respondents, Wonji, Ethiopia, 1997.
Туре	Degree of tooth damage (problem), n=234

Variabla		Pla	ce of residence	
variable	Wonji	Shoa	Camp 11	Camp 12
Tooth examined	560	632	344	336
Tooth with fluorosis	482	512	130	161
Tooth prevalence (%)	86.1	81.0	37.8	47.9

TABLE 5. Mouth prevalence rates of dental fluorosis in three samples from Wonji Shoa Sugar Estate.

Village	Sample size	All grades	All grades excluding very mild cases
Wonji	70	95.7 %	84.3 %
Shoa	79	92.4 %	86.2 %
M & O	85	71.8 %	27.1 %

The fluoride levels in the various water supplies of the estate during the one year period preceding this survey (July 1996 to June 1997), as documented in the record book of the laboratory in Wonji Hospital (the hospital of WSSE) are present in Table 6. In Wonii and

Shoa villages where the defluoridation plants were operational, it can be seen that the levels of fluoride in treated water were at times above the safe level. In villages M and O, which are supplied by water from a common well the defluoridation plants were not functioning and the levels of fluoride in the raw water were similar to those in treated water from Wonji and Shoa villages.

The prevalence rates of dental fluorosis in Wonji and Shoa villages were found to be high despite the availability of treated water. This appears largely to be due to the dual nature of the water supply, with the supply of raw water with high fluoride levels being readily accessible. It is very likely that children in these villages consumed significant quantities of raw water. Moreover, the level of fluoride in treated water was at times high in villages M and O the prevalence rate of dental fluorosis is lower

and most cases of dental fluorosis are very mild. The reason for this is partly because of the low level of fluoride in the raw water supply of the villages and partly because the residents in these two villages too often consumed water from Awash River when tap water from the well was available, as observed during the conduct of this survey.

It can be concluded that the impact of the defluoridation programme on the occurrence of dental fluorosis in the estate is negligible at present. The situation was found to be much better in the areas were the raw water had low fluoride levels and where the residents were additionally consuming river water.

<b>TABLE 6.</b> Fluoride levels in the water supplies
of the four villages of WSSF during the period
July 1996 to June 1997.

Village	mg F/L	mg F/L
	treated water	raw water
Wonji	< 0.10 - 1.8	6.0 - 12.5
Shoa	0.24 - 4.0	0.37 - 6.0
M and O	-	0.80 - 2.2

#### ACKNOWLEDGEMENTS

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## PREVALENCE OF LOW BACK PAIN AT AN AGRO-INDUSTRIAL COMMUNITY IN THE RIFT VALLEY

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ABSTRACT: A descriptive cross-sectional study is carried out at the Wonji Shoa Sugar Estate in the Rift Valley region of Ethiopia. A total of 1939 adults were surveyed with respect to Low Back Pain, LBP. The overall prevalence of LBP is 21.7 %. The prevalence is only slightly higher in males than in females. The highest frequency of low back pain (28.7 %) was found in the age group 50 years and above. The frequency of LBP is associated with duration of residence in the fluorotic Estate. 25.4 % of the Estate population seem to be completely ignorant the fluoride problems and the on going defluoridation program.

Keywords: Low back pain; Fluoride; Defluoridation program; Rift Valley; Ethiopia

#### INTRODUCTION

Endemic fluorosis is widespread in the East African Rift system including the Ethiopian Rift Valley, where it is associated with high fluoride content in drinking water in areas of acidic volcanic rocks. Fluoride concentration between 1 and 20 mg/L have been reported from areas of endemic dental and skeletal fluorosis in the Ethiopian Rift Valley.<sup>1</sup>

Prolonged and excessive fluoride ingestion after ten to twenty years results in the development of skeletal fluorosis.<sup>2</sup> Early skeletal involvement by fluorosis is not clinically obvious even though radiographical changes are discernible in the skeleton at early stages. In the later stages. Skeletal fluorosis manifests with restriction of movement of the spine and of the joints of the extremities and with neurological complications as a results of compression of the spinal cord and the spinal nerves.

The spine being one of the areas which is frequently affected by skeletal fluorosis, we have carried out this study to determine the prevalence of low back pain in an area in the Ethiopian Rift Valley with a high fluoride content and compared it with a similar study carried out in an area outside the Rift Valley. Additionally, the knowledge of the community about fluoride and its consequences has been evaluated.

#### MATERIALS AND METHODS

This is a descriptive cross-sectional study, which was carried out at the Wonji and Shoa villages of the Wonji Shoa Sugar Estate (WSSE). A major agro-industrial community situated in the Rift Valley region of Ethiopia, between July 1997 and October 1997.

**Target Population:** All adults aged 18 or above and residing in the Wonji and Shoa villages of the WSSE at the time of the survey.

**Sampling:** Four villages, amongst nine were selected by simple random sampling using a lottery method.

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**Method of data collection:** Data was collected, using a pre-tested Questionnaire by trained interviewers on house-to-house basis under the supervision of one of the investigators.

A total of 1939 adults were included in the survey. Baseline characteristics of the respondents are given in Table 1. The overall response rate is 92.3 %.

<b>TABLE 1.</b> Characteristics of study population and frequency of Low Back Pain.								
Stud	ly Population		With 1	LBP	Withou	Without LBP		
Characteristics	No.	%	No.	%	No.	%		
Age (years)								
< 20	266	13.7	42	15.6	240	84.4		
20-29	465	24.0	103	22.1	362	77.9		
30-39	617	31.8	162	26.3	455	73.7		
40-49	368	19.0	50	13.6	318	86.4		
> 50	223	11.5	64	28.7	159	71.3		
Sex								
Male	1105	56.6	213	19.3	892	80.7		
Female	842	43.3	138	16.4	704	83.6		
Duration of reside	nce (years)							
< 10	369	19.0	42	11.3	327	88.7		
11-19	644	33.3	96	14.9	549	85.1		
> 20	926	47.7	283	30.6	643	69.4		
Total	1939	100.0	421	21.7	1519	78.3		

#### RESULTS

The frequency of low back pain is given in Table 1. The overall prevalence of low back pain in the study population is 21.7 %. The prevalence of low back pain is slightly higher in males than in females (19.3 % vs. 16.4 %) but this was not found statistically significant. The highest frequency of low back pain (28.7 %) was found in the age group 50 years and above followed by those between 30 and 39 years (26.3). With regard to duration of residence the highest prevalence was noted in those who have been residing in the area for more than 20 years (30.6 %), and the lowest in those who have been residing in the area for less than 10 years (11.3 %).

Table 2 summarises the low back pain frequency in the different villages. The difference in the prevalence of low back pain amongst the different villages included in the study was not found statistically significant.

The overall awareness of the community about the presence of excess fluoride in drinking water in the areas and its consequences was examined. 74.6 % were aware, 25.4 % were not.

Low back pain is common in the general population resulting suffering, in appreciable disability, and social costs in developed countries,<sup>3</sup> whereas very little is known about the epidemiology of low back pain in the developing The prevalence of countries.

TABLE 2.	Frequency	of pa	tients	with	low	back
pain by villa	ige.					

p	•••		
Village	Population	%	with
		LBP	
Wonji F	610	24	.0
Wonji G	547	19	0.3
Shoa 1	358	23	5.4
Shoa 4	424	20	0.0
Total	1939	21	.7

low back pain reported from developed countries is varied from depending on the type of population studied and the methodology. There are no studies from other developing countries for comparison, but studies from USA indicate that the prevalence in whites is one and half times greater than the prevalence among African-Americans.

In this survey the overall prevalence rate of low back pain was 21.7 %, which is much higher than the results of a similar community. In a study carried out at Maskan and Mareko district, in an area which is situated outside of the Rift Valley region where the prevalence rate of low back pain was found to be 5.7 % (unpublished data).

Since there is no difference in terms of other variables such as sex, history of past trauma and type of bed used in these two surveys, the most probable explanation for the higher prevalence of low back pain in this survey is the high fluoride content of drinking water in the area resulting in deposition of fluoride in the lumbar spine and the surrounding ligaments. This is supported by the finding of a higher prevalence (30.6 %) in those residing in the area for more than 20 years compared to (11.3 %) those living in the area for less than 10 years. These findings are similar to the results of a study carried out in Wonji Sugar Estate in 1990. In which amongst 328 adults examined, physical impairment indicative of skeletal fluorosis was found in 6.7 % of those who resided in the area for less than 10 years and in 63 % of those resided in the area for less than 20 years.

Similar to other studies on LBP, in this study women and men are affected with low back pain with approximately equal frequency. Our finding of a decline in frequency of low back pain amongst middle age is consistent with the findings of previous studies.<sup>5</sup>

The awareness to the fluoride problem, where 25.4 % of the estate population seems to be completely ignorant, warrants strengthening the existing health education system at the Sugar Estate to increase the awareness in the public about this important health problem. Probably this will have a great impact on the proper utilization and success of existing and future defluoridation programs and the mitigation of low back pain problems.

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Session II

**Occurrence & Methodology** 

## FLUORIDE CONTAMINATION AND MINERALOGICAL COMPOSITION OF EAST AFRICAN MAGADI (TRONA)

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SUMMARY: Magadi from Lake Magadi, Kenya, Lake Natron, Tanzania, Lake Katwe, Uganda and lake El Atrun, Sudan, are analysed along with efflorescent crust magadi from the surface soil from Northern Tanzania. The fluoride and carbonates concentrations are measured chemically and the mineralogical composition is determined through X-ray diffraction analysis. Magadi from Lake Natron and Lake Magadi are found to be very similar consisting mainly of trona  $(CO_3^2 + HCO_3 > 10.4 \text{ meq/g magadi})$ , mixed with halite and either kogarkoite or villiaumite respectively. This is resulting in fluoride concentrations up to 8.7 mg F/g magadi. The scooped magadi is less pure with respect to trona, but its fluoride content is of the same order of magnitude (0.23-5.1 mg F/g magadi). The scooped magadi consists of trona  $(CO_3^2 + HCO_3 = 3.5 - 9.5 \text{ meq/g magadi})$  with different mixtures of halite, quartz, villiaumite, kogarkoite, and thermonatrite. No fluoride containing minerals are identified in magadi from Uganda and Sudan, probably due to the very low fluoride concentrations, = 0.02 and < 0.24 mg/g magadi respectively. The Sudanese magadi consists of different mixtures of trona, halite, and quartz resulting in a variation its alkaline strength  $(CO_3^2 + HCO_3^2 = 4.6-11.9 \text{ meq/g magadi})$ . The magadi from Lake Katwe consists of trona  $(CO_3^2 + HCO_3 = 7.0 \text{ meq/g magadi})$  mixed with burkeite and halite.

**Key words:** Trona; Magadi; Soda; African lakes; Fluoride contamination; Villiaumite; Kogarkoite; Alkaline lakes.

#### INTRODUCTION

Trona, Na<sub>2</sub>CO<sub>3</sub>·NaHCO<sub>3</sub>·2H<sub>2</sub>O, is a commonly used salt in several countries in East, West, and Central Africa and it is the second most commonly used salt in Nigerian homes.<sup>1</sup> In East Africa trona is locally called *magadi* and the name probably originated from the Masai word magad meaning bitter. The main use of magadi is cooking tough food materials such as beans and maize utilising its ability to fasten the softening and the digestive property of the food during cooking. In addition magadi is used as a prophylactic agent and a feed supplement to cattle and goats. Furthermore, in some places, it is ground with tobacco in the preparation of snuff.<sup>2-13</sup>

In the Eastern Rift Valley magadi is formed, as the so-called crystalline magadi, in the alkaline lakes due to chemical weathering of rock minerals and high evaporation of the lake waters. Magadi is also formed, as the so-called scooped magadi, on the surface soil formed due to capillary evaporation of soil water.

Analyses of both crystalline and scooped magadi from Nigeria and Ghana have shown that crystalline magadi consists essentially of trona mixed with minor contents of halite (NaCl). The scooped magadi is rich in trona but it also contains admixtures of quartz, clays, chlorides, and sulphates.<sup>1,2,12</sup>

Investigations have shown that magadi from the alkaline lakes and from the surface soil in the Eastern Rift Valley is often heavily contaminated with fluoride.<sup>3,6-9,14,15</sup> The high fluoride concentration in the Eastern Rift Valley is strongly related to weathering of the volcanic rocks rich with fluoride and alkalis as found in the Rift

Valley areas.<sup>14,16,17</sup> The ingestion of fluoride through magadi may in certain cases be significant in comparison to the total intake of fluoride<sup>7</sup>, having in mind that the acceptable maximum daily fluoride intake given by WHO is 4.0 mg F/person/day.

It is the aim of this study to elucidate the differences in fluoride concentration in magadi from the Eastern and the Western Rift and the deposition of different fluoride containing minerals from the alkaline lake brines. The differences in the mineralogical composition caused by the differences in the composition of the lake brines will also be discussed.

#### **Materials & Methods**

Magadi samples from Lake Natron, Tanzania (n=9), Lake Magadi, Kenya (n=15), Lake Katwe, Uganda (n=1), El Atrun, Sudan (n=5) and efflorescent crust from Northern Tanzania (n=9) have been collected at the lakes or bought at market places in Magadi town, Kenya, in Khartoum, Sudan and in cities in Uganda and Tanzania. The origin of the magadi bought at the markets was determined according to the information given by the dealers (for location of the alkaline lakes see Figure 1). The magadi samples were crushed and homogenised in a mortar. An amount of 2.00 g magadi powder was dissolved in 100.0 ml distilled water. Hereafter the fluoride and carbonate concentrations were measured in the solutions. Fluoride concentrations were measured in samples from Uganda and Sudan. Fluoride concentrations of Tanzanian and Kenyan magadi are taken from Nielsen and Dahi.<sup>15</sup>

**Fluoride measurements:** The fluoride concentrations were measured using a Radiometer F1052 fluoride electrode and a Metrohm Ag/AgCl reference electrode with a sleeve type diaphragm connected to a Metrohm potentiometer (692 pH/Ion Meter). An aliquot of 10.0 ml of the sample solutions was mixed with 10.0 ml CDTA-tisab and the fluoride concentration was measured using the calibration method as described in the Standard Methods.<sup>18</sup>

**Carbonate measurements:** The carbonate  $(CO_3^{2^-}+HCO_3^-)$  concentrations were measured using the end point titration method according to the Standard Methods<sup>18</sup>, where the end point pH = 4.5 was used. An aliquot of 5.00 ml of the magadi solutions (2.00 g/100.0 ml) was diluted to 50.0 ml and titrated automatically with 0.1 N H<sub>2</sub>SO<sub>4</sub> using a Metrohm pH-electrode connected to a Metrohm 719S Titrino. The values of pH and the added amount of acid were recorded at 5 seconds intervals on a PC. The calibration of the pH-electrode was done using Metrohm buffer solutions pH = 4.0 and pH = 9.0.

**X-ray diffraction:** Selected magadi samples were crushed to powder and analysed by X-ray diffraction using a Philips PW1050 automated diffractometer equipped with a graphite monochromator, an automatic divergence slit, and a 0.10° receiving slit. The radiation was CuK $\alpha$  and the XRD spectra were recorded as step scans in the interval 5°<20<65° using a step size of 0.10° and a count time of 2.0 sec/step. The reflections with I>3 $\sigma$ (I) were recorded as observed.





lakes see Figure 1).		
Origin of Sample	n	Minerals detected
Lake Magadi	3	Trona, halite, villiaumite
Lake Natron	5	Trona, halite, kogarkoite, thermonatrite
Lake Katwe	1	Trona, halite, burkeite
Sudan	4	Trona, halite, quartz
Northern Tanzania, scooped	6	Trona, halite, quartz, villiaumite, kogarkoite,
		thermonatrite

**TABLE 1.** Minerals identified by XRD in selected magadi from Africa (for location of lakes see Figure 1).

#### RESULTS

The results of the measurements of the fluoride and the carbonate concentrations in magadi can be seen in Figure 2, where the carbonate concentration is plotted versus the logarithmic fluoride concentration. The results from the XRD analyses can be found in Table 1, where the identified minerals in selected magadi samples are stated.

#### DISCUSSION

It is seen from the results in Figure 2 that there is a wide variation in the carbonate concentration of the magadi. The carbonate concentration in the magadi is mainly due to the content of trona but also due to the contents of thermonatrite,  $Na_2CO_3 \cdot H_2O$ , and burkeite, Na<sub>2</sub>CO<sub>3</sub>·2Na<sub>2</sub>SO<sub>4</sub>, cf. Table 1. The highest carbonate concentrations, 10.4-13.1 meq/g magadi are found in magadi from Lake Magadi and Lake Natron. These magadi samples are almost equal to pure trona indicated by the dashed line in Figure 2. The scooped magadi is less pure with respect to trona as the crystalline magadi from Lake Magadi and Lake Natron. The carbonate concentration in the scooped magadi is also subject to considerable variation, 3.5-9.5 meq/g magadi. The scooped magadi is mainly contaminated with halite (NaCl) and quartz (SiO<sub>2</sub>). Makanjuola and Beetlestone<sup>1</sup> found out that scooped magadi from Nigeria consists of trona mixed with sand and clay. The magadi from Lake Katwe was bought at the local market and it is probably from the trona harvest. The magadi is not pure trona, the carbonate concentration being as low as 7.0 meq/g magadi. This is caused by contamination of halite and burkeite cf. Table 1. The carbonate concentration of the Sudanese magadi, 4.6-12.0 meq/g magadi, is subject to considerable variation like scooped magadi. The Sudanese magadi also contains impurities such as halite and quartz. However, magadi from Sudan is very different from the other magadi samples, it is not an evaporite from an existing lake or the surface soil but a salt deposit found in the mountains.

The fluoride concentration in the magadi is subject to considerable variation ranging from 0.02 to 8.7 mg F/g magadi, cf. Figure 2. The concentration of Tanzanian and Kenyan magadi is comparable to what has been reported by other researchers who have analysed magadi from Kenya and Tanzania.<sup>3,7,9,14</sup> The highest fluoride concentrations are found in magadi from Lake Natron and Lake Magadi, up to 8.1 and 8.7 mg F/g magadi, respectively. The high fluoride concentration in the Kenyan and Tanzanian magadi is caused by the high fluoride content of the volcanic rocks found in the Eastern Rift Valley.<sup>16</sup> The fluoride containing mineral, villiaumite (NaF), has been identified by X-ray diffraction in one of the magadi samples from Lake Magadi,

cf. Table 1. Observations of villiaumite have also been reported by Baker.<sup>3</sup> The double salt, kogarkoite (Na<sub>2</sub>SO<sub>4</sub>·NaF) is identified in three of the magadi samples from Lake Natron. Darragi<sup>19</sup> also identified kogarkoite in the salt crust of Lake Natron by X-ray diffraction and scanning electron microscope observations. The presence of this rare mineral is not known in evaporites of the alkaline lakes in the African Rift Valley, but it has been observed in thermal deposits (84°C) at Mt. Princeton Hot Springs, Colorado<sup>20</sup> and in the Lovozero Massif, Kola Peninsula in nepheline syenite pegmatite.<sup>21</sup> The deposition of kogarkoite in Lake Natron, in contrary to villiaumite in Lake Magadi, is caused by a relatively higher sulphate concentration in the lake brine, of Lake Natron compared to Lake Magadi.<sup>22</sup> The fluoride concentration of the scooped magadi is in the same order of magnitude, 0.2 -5.1 mg F/g magadi as the crystalline magadi from Lake Magadi and Lake Natron even though the carbonate concentration is significantly lower. Fluoride containing minerals have been identified both as villiaumite and kogarkoite in the scooped magadi. The lowest fluoride concentrations are found in the Sudanese rock samples, 0.03-0.24 mg F/g magadi and in the evaporite from Lake Katwe, 0.02 mg F/g magadi. The fluoride concentration of the magadi from Lake Katwe is very low compared to the concentration of magadi from Tanzania and Kenya. This is caused by the differences in fluoride concentration of the lake brines. Deelstra<sup>23</sup> found out that the saline lake waters of the Western Rift contain much smaller amounts of fluoride than the lake waters of the Eastern Rift. No reflections from fluoride containing minerals have been observed on the XRD data for the Sudanese and Ugandan magadi. This is probably due to the very low fluoride concentrations. Fluoride containing minerals have not been identified in all the magadi samples from Lake Natron and Lake Magadi but it is assumed that the measured fluoride is present as villiaumite or kogarkoite and not as fluorite  $(CaF_2)$  in these calcium poor environments.

Halite (NaCl) is identified in all the magadi samples. The evaporite from Lake Katwe contains much more halite than magadi from Lake Natron and Lake Magadi. This is related to a higher chloride concentration of the brines. Brines from Lake Katwe have an equivalent Cl/TAL ratio of  $2-3^{24}$  whereas the equivalent Cl/TAL ratio of Lake Magadi and Lake Natron brines is  $0.23-1.0.^{3,25,26}$ 

In the extensive phase relations reported by Teeple<sup>27</sup> burkeite is found to be a phase in the system Na-Cl-SO<sub>4</sub>-CO<sub>3</sub>-H<sub>2</sub>O but burkeite is only identified in the magadi from Lake Katwe not in magadi from Lake Magadi or Lake Natron.

The quartz  $(SiO_2)$  identified in the scooped magadi is probably due to mixing with soil when collecting the efflorescent crust, and the quartz in the Sudanese magadi is assumed to origin from rock minerals.

Thermonatrite is observed in magadi from Lake Natron and in the scooped magadi. Eugster<sup>28</sup> has observed thermonatrite in shallow surface pools during the late stages of evaporation of Lake Magadi brines. Thermonatrite is normally deposited after the precipitation of trona if the brine keeps on being under-saturated with respect to atmospheric  $CO_2$ .<sup>29</sup>

The analyses of magadi from Tanzania and Kenya show that these samples contain high amounts of fluoride. Thus, the use of magadi heavily contaminated with fluoride as a tenderiser may in certain cases result in very high fluoride intake, even higher

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# A SIMPLE METHOD FOR DETERMINATION OF TOTAL FLUORIDE INTAKE

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SUMMARY: Determination of fluoride intake by human subjects must include the fluoride contribution through water and food. Fluoride intake through water means fluoride through drinking water and also through water used for cooking food. Thus fluoride intake through food must exclude the fluoride through water used for cooking, as the amount of water used for cooking depends on the diet pattern of the people. On this basis, the total fluoride intake through water is determined in 3 fluorotic and 2 non-fluorotic areas. The fluoride intake through water is found less than that through food in the non-fluorotic areas and even in one low fluorotic area. People of highly endemic fluorotic areas however, are exposed to greater fluoride intake through water than through food.

KEY WORDS: Fluoride intake; Water fluoride; Food fluoride; Fluorosis; Endemic areas.

## INTRODUCTION

The prevalence of fluorosis in an endemic area is normally interpreted on the basis of the percentage of fluorosis and the fluoride levels of the drinking water sources of the region. The fluoride concentration in the water samples of a region has necessarily to be reliable in order to establish the degree of fluorosis. The contribution of fluoride through food, water used for drinking and cooking and also through beverages, if any, constitute the total fluoride intake by the subjects.

While explaining the dietary intake of fluoride, either food alone or food and beverages at meal time or food, beverages and water at and between meals are to be considered as sources of fluoride. An easy and reliable method is employed successfully to determine the contribution of water and food to the total fluoride intake by the subjects of selected areas and the details of this method are reported here.

There have been several reports<sup>1-3</sup> in the literature about the role of fluoride through food and water and also the total fluoride intake by human subjects in explaining the prevalence of fluorosis in fluorotic areas. Evidence for quantitative attempts to develop a methodology to evaluate the contribution of water and food to the total daily intake of fluoride is lacking. The fluoride present in water used for the preparation of food and beverages should not be ignored eventhough the concentration of water is fairly low. In the absence of such precautions the results obtained may attribute to incorrect conclusions about the actual values of fluoride through food and water. Therefore the total fluoride intake must include the contribution of fluoride from food, water and also from other food drinks if any, the subjects consume during a prescribed period of study. A new workable methodology was developed to determine the contribution of fluoride through water and food in order to find out the total fluoride intake and the details are reported in the following pages.

## MATERIALS AND METHODS

**Sampling:** 80 human subjects were selected from five areas. Two areas were identified as control areas, C-I & CII with no fluorosis of any kind. The three others were identified as fluorotic areas of this investigation, F-I, F-II & F-III. The criteria adopted for the selection of areas were the same as reported earlier by the authors.<sup>4</sup> Selection of families and subjects was done using purposive sampling technique<sup>5</sup> from the five mentioned areas. Collection of samples of food and water consumed by the subjects was done for four alternate days. The exact quantity of diet and volume of drinking water consumed by each subject in a day were obtained separately in high-density polyethylene containers. The solid part and liquid part of the diet were analysed to determine the fluoride concentrations. The liquid diet was filtered and whatever solids remained in it was added to the solid diet. The solid diet was weighed and ground to a homogeneous mixture.

**Analysis**. The liquid diet and water were measured separately. 50 ml of the liquid diet sample was taken in a distillation flask. Sufficient AnalaR silver sulphate was added to precipitate the chlorides present. 50 ml of AnalaR 60 % perchloric acid was added and the distillate collected between 135 - 139 °C was neutralised using 0.2 N NaOH and analysed for fluoride.

Formula	Constituents	Defin	itions	Unit
$I_{\rm T} = I_{\rm DW} + I_{\rm D}$	$I_{DW} = C_W \cdot V_W$	$I_{T}$ $I_{D}$ $I_{DW}$ $C_{W}$ $V_{W}$	Total daily intake per person, Intake through diet, Intake through drinking water, Concentration of F in water, Volume of consumed water	$\begin{array}{c} mg \cdot c^{-1} \cdot d^{-1} \\ mg \cdot c^{-1} \cdot d^{-1} \\ mg \cdot c^{-1} \cdot d^{-1} \\ mg / L \\ L \cdot c^{-1} \cdot d^{-1} \end{array}$
$I_{\rm D} = I_{\rm LD} + I_{\rm SD}$	$\begin{split} M_D &\approx V_{LD} + M_{SD} \\ I_{LD} &= C_{LD} \cdot V_{LD} \\ \end{split} \\ I_{SD} &= f_{SD} \cdot M_{SD} \end{split}$	$\begin{array}{c} M_{\rm D} \\ I_{\rm LD} \\ C_{\rm LD} \\ V_{\rm LD} \\ I_{\rm SD} \\ f_{\rm SD} \\ M_{\rm SD} \end{array}$	Total amount of diet, * Intake through liquid diet, Conc. of F in liquid diet, Volume of liquid part of diet, Intake through solid diet, Fraction of F in solid part of diet, Mass of solid part of diet,	$\frac{kg \cdot c^{-1} \cdot d^{-1}}{mg \cdot c^{-1} \cdot d^{-1}}$ mg/L L · c <sup>-1</sup> · d <sup>-1</sup> mg · c <sup>-1</sup> · d <sup>-1</sup> mg/kg kg · c^{-1} · d^{-1}
$I_{W} = I_{DW} + I_{CW}$		$\begin{array}{l} I_W\\ I_{DW}\\ I_{CW} \end{array}$	Total daily intake through water, Intake through drinking water, Intake through cooking water,	$mg \cdot c^{-1} \cdot d^{-1}$ $mg \cdot c^{-1} \cdot d^{-1}$ $mg \cdot c^{-1} \cdot d^{-1}$
$\begin{split} I_{\mathrm{T}} &= I_{\mathrm{F}} + I_{\mathrm{W}} \\ I_{\mathrm{F}} &= I_{\mathrm{T}} - I_{\mathrm{W}} \end{split}$		$\mathbf{I}_{\mathrm{F}}$	Intake through food alone,	$mg \cdot c^{-1} \cdot d^{-1}$

TABLE 1. Utilised relationships and definitions.

<sup>\*</sup> The water used for cooking was found to constitute 80 % of the total diet or for all practical purposes 1 litre of water per person per day.

**TABLE 2**. Fluoride intake through water and food/day by the selected human subjects in the control areas. Figures in columns 2- 6 are in mg F per capita per day.  $M \sim Male$ . F  $\sim$  Female. C  $\sim$  Child.

C.	hight	Intake th	rough					
Su	bject	Drink.	Cooking	Water	Food	Total	Water	Food
INC	).	Water	Water	vv ater	roou		%	%
Co	ntrol A	vrea I (C-I No	n-fluorotic)					
1	(M)	0.84	0.71	1 55	3 73	5 28	29.4	70.6
2.	$(\mathbf{F})$	0.70	0.54	1.24	3.06	4.30	28.8	71.2
3	$(\mathbf{C})$	0.41	0.35	0.77	1 91	2.68	28.7	71.3
4.	$(\mathbf{C})$	0.37	0.25	0.62	1.44	2.06	30.1	69.9
5.	$(\mathbf{M})$	0.89	0.70	1.59	3.90	5.49	29.0	71.0
6.	(F)	0.68	0.51	1.19	3.06	4.25	28.0	72.0
7.	$(\mathbf{C})$	0.50	0.43	0.93	2.27	3.20	29.1	70.9
8.	$(\mathbf{C})$	0.44	0.38	0.82	1.63	2.45	33.5	66.5
9.	(M)	0.92	0.91	1.83	3.85	5.68	32.2	67.8
10.	(F)	0.80	0.72	1.52	3.09	4.61	33.0	67.0
11.	$(\mathbf{C})$	0.63	0.54	1.17	2.28	3.45	33.9	66.1
12.	$(\mathbf{C})$	0.59	0.48	1.07	1.84	2.91	36.8	63.2
13.	(M)	1.00	0.89	1.89	3.97	5.86	32.3	67.7
14.	(F)	0.83	0.71	1.54	3.05	4.59	33.6	66.4
15.	$(\mathbf{C})$	0.65	0.57	1.22	2.53	3.75	32.5	67.5
16.	$(\mathbf{C})$	0.67	0.62	1.29	2.66	3.95	32.7	67.3
	(-)							
	Ave	0.68	0.58	1.27	2.77	4.03	31.5	68.5
	Contr	ol Area II (C-	II Non-fluor	otic)				
17.	(M)	1.20	0.91	2.11	3.63	5.74	36.8	63.2
18.	(F)	1.00	0.70	1.70	2.84	4.54	37.4	62.6
19.	$(\mathbf{C})$	0.72	0.67	1.39	2.30	3.69	37.7	62.3
20.	$(\mathbf{C})$	0.67	0.58	1.25	1.98	3.23	38.7	61.3
$\frac{-0.}{21.}$	$(\mathbf{M})$	1.26	1.14	2.40	3.64	6.04	39.7	60.3
22.	(F)	1.09	0.89	1.93	3.04	4.97	38.8	61.2
23.	$(\mathbf{C})$	0.82	0.71	1.53	2.48	4.01	38.2	61.8
24.	$(\mathbf{C})$	0.72	0.61	1.32	2.27	3.59	36.8	63.2
25.	$(\mathbf{M})$	1.21	0.89	2.10	3.87	5.97	35.2	64.8
26.	(F)	0.99	0.74	1.73	3.01	4.74	36.5	63.5
27.	(C)	0.75	0.64	1.39	2.68	4.07	34.2	65.8
28.	(C)	0.58	0.47	1.05	1.98	3.03	34.7	65.3
29.	$(\mathbf{M})$	1.22	1.14	2.30	3.81	6.17	38.2	61.8
30.	(F)	0.98	0.89	1.87	3.21	5.08	36.8	63.2
31.	(C)	0.67	0.59	1.20	1.85	3.11	40.5	59.5
32.	(C)	0.62	0.40	1.08	1.58	2.60	40.6	59.4
Av	ve	0.91	0.75	1.65	2.76	4.41	37.6	62.5

IVI & IVIAIC. I	Temale. C	on onitio.	T	ntake throu	ugh:		
Subject	Drink	Cooking	1		Total	Water	Food
No.	Water	Water	Water	Food	Total	%	%
Fluorotic Area	I (F-I)						
33. (M)	1.47	1.26	2.73	8.35	11.08	24.6	75.4
34. (F)	1.18	0.97	2.15	6.95	9.10	23.6	76.4
35. (C)	1.00	0.81	1.81	5.52	7.33	21.7	75.3
36. (C)	0.83	0.70	1.53	4.42	5.95	25.7	74.3
37. (M)	2.15	1.61	3.76	7.31	11.07	34.0	66.0
38. (F)	1.77	1.21	2.98	5.36	8.34	35.7	64.3
39. (C)	1.03	0.92	1.95	3.55	5.50	35.5	64.5
40. (C)	0.98	0.64	1.62	2.80	4.42	36.7	63.3
41. (M)	2.06	1.66	3.72	8.71	12.43	29.9	70.1
42. (F)	1.69	1.30	2.99	6.76	9.75	30.7	69.3
43. (C)	1.29	1.03	2.32	5.77	8.09	28.7	71.3
44. (C)	1.14	0.94	2.08	5.06	7.14	29.1	70.9
45. (M)	1.48	1.29	2.77	8.54	11.31	24.5	75.5
46. (F)	1.15	0.94	2.01	7.14	9.23	22.6	77.4
47. (C)	0.88	0.71	1.59	4.90	6.49	24.5	75.5
48. (C)	0.74	0.57	1.31	3.64	4.95	26.5	73.5
Ave.	1.30	1.04	2.33	5.92	8.26	28.4	71.4
Eluorotic Area	II (F-II)						
49 (M)	5 20	2.96	916	8 61	17 77	51.5	48 5
50 (F)	4 28	3.08	7 36	6.21	13 57	54.2	45.8
50. $(\Gamma)$	3.18	2.66	5.84	5 76	11.60	50.3	49.7
51. (C)	2 94	2.00	5 27	4.82	10.09	52.2	47.8
53 (M)	4 56	4 07	8.63	6.28	14 91	57.9	42.1
54 (F)	3 59	3.18	677	5.68	12.45	54.4	45.6
55 (C)	2.29	1 76	4.05	3 10	7 15	56.6	43.4
56. (C)	2.49	1.99	4.48	3.63	8.11	55.2	44.8
57 (M)	4 56	3 90	8 46	8.72	17.18	49.2	50.8
58. (F)	3.51	3.04	6.55	7.93	14.48	45.2	54.8
59. (C)	3.12	2.72	5.84	5.48	11.32	51.6	48.4
60. (C)	2.85	2.27	5.12	5.18	10.30	49.7	50.3
61. (M)	5.26	3.89	9.15	7.09	16.24	56.3	43.7
62. (F)	4.27	3.10	7.37	5.62	12.99	56.7	43.3
63. (C)	3.01	2.14	5.15	4.71	9.86	52.2	47.8
64. (C)	3.19	2.71	5.90	4.78	10.68	55.2	44.8
Ave.	3.64	2.86	6.57	5.85	12.42	53.0	47.0

**TABLE 3.** Fluoride intake through water and food/day by the selected human subjects in the fluorotic areas. Figures in columns 2- 6 are in mg F per capita per day.  $M \sim Male, F \sim Female, C \sim Child$ .

TABLE 3 (continued)								
Fluorotic Area III (F-III)								
65. (M)	15.68	13.61	29.29	3.14	32.43	90.3	9.7	
66. (F)	12.36	10.14	22.50	3.66	26.16	86.0	14.0	
67. (F)	14.49	9.07	23.56	3.27	26.83	87.8	12.2	
68. (M)	11.55	11.26	22.81	2.42	25.23	90.4	9.6	
69. (F)	11.17	9.04	20.21	3.68	23.83	84.6	15.4	
70. (M)	16.17	13.33	29.50	3.35	32.85	89.8	10.2	
71. (F)	13.27	10.42	23.69	3.20	26.89	88.1	11.9	
72. (F)	11.45	8.74	20.19	3.22	23.41	86.2	13.8	
73. (C)	8.37	6.61	14.98	3.33	18.31	81.8	18.2	
74. (C)	7.11	5.85	12.96	2.49	15.45	83.9	16.1	
75. (C)	10.33	9.30	19.63	3.68	23.31	84.2	15.8	
76. (C)	9.87	9.04	18.91	3.72	22.63	83.6	16.4	
77. (C)	8.65	6.80	15.45	3.28	18.73	82.5	17.5	
78. (C)	9.66	8.01	17.67	3.44	21.11	83.7	16.3	
79. (C)	8.86	7.39	16.25	3.51	19.76	82.2	17.8	
80. (C)	10.47	8.62	19.09	4.02	23.11	82.6	17.4	
Ave.	11.22	9.20	20.42	3.34	23.75	85.5	14.5	

100 g of solid diet sample was taken for analysis. The procedure for analysis was based on the classical Willard-Winter distillation method<sup>6</sup> and the distillate was analysed for fluoride.

The fluoride contents of drinking water and food parts were determined using fluoride ion selective electrode and an expandable ion analyser EA 920 Orion USA<sup>7</sup>.

**Calculation of total intake per day:** The total intake of fluoride in mg per person per day,  $I_T$ , is the sum of the intakes from drinking water,  $I_{DW}$ , and from the diet,  $I_D$ . The total intake through water,  $I_W$ , is however the sum of intakes from drinking water,  $I_{DW}$ , and from cooking water  $I_{CW}$ . The intake through food,  $I_F$ , alone may be calculated as the difference between the total intake,  $I_T$ , and the intake through cooking and drinking water,  $I_W$ . The intake through diet,  $I_D$ , is the sum of intakes through liquid part of diet,  $I_{LD}$ , and solid part,  $I_{SD}$ . These relationships are indicated in table 1.

#### **RESULTS AND DISCUSSION**

The proposed methodology was utilised by the investigators to determine the fluoride intake through water and food per day by the selected human subjects of the five areas. The results obtained along with the percentage intake of fluoride through water and food separately are presented in tables 2 and 3.

From table 2 &3 it is seen that the contribution of fluoride intake through water is on an average much less in control and low fluoride areas C-I, C-II & F-I, compared to the fluorotic areas, respectively 35 and 57 % of the total intake. Accordingly the food contribution is found to be 65 and 43 % of the total intake in the control areas and in the fluorotic areas respectively. Thus, on average basis, food fluoride contributed more towards total fluoride intake in the non-fluorotic and low fluoride areas, while water fluoride contributed more in the high fluoride areas F-II & F-III, Figure 1. This means that the contribution of fluoride through water is, in some fluorotic areas. less than that through food. On the other hand fluorotic areas II and III are highly endemic as reported earlier by the authors<sup>4</sup>. The fluoride level of drinking water in area III varies between 4 to 9 mg/L. Our results show that the fluoride contribution through water varies from 82 to 90 percent while the same through food is only between 10 and 18 percent.

Thus our results confirm

that people of highly endemic fluorotic areas are exposed to greater fluoride intake through water and this is responsible for the increased prevalence of fluorosis. This simple method is employed successfully to identify the dominant factor(s) causing fluoride toxicity of the areas.

100

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# **FIGURE 1.** Average contributions of fluoride intakes though water and food in the studied fluorotic and control areas.

## ANALYTICAL PROBLEMS IN ASSESSMENT OF FLUORIDE IN FOOD

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SUMMARY: Different conventional digestion methods for the analysis of fluoride in organic material have given ambiguous results. Better, standardised procedures are needed. The present paper discusses some of the fallacies in the analysis of fluoride in solid food, and points out methods to overcome the problems.

Key words: Fluoride analysis; Food; Standard reference material.

#### INTRODUCTION

Recent concern for the total ingestion of fluoride has brought new emphasis on solid food as a possibly significant fluoride source. Various multidisciplinary researches have provided a vast information on the topic. However, the information is often ambiguous. Our pilot studies on oyster tissue, the only standardised and certified reference material that at the time was available for fluoride analysis, may exemplify the ambiguity. The oyster tissue was dissolved by the use of nitric acid and perchloric acid and the solvent was neutralised with sodium hydroxide and trisodium citrate.<sup>1</sup> According to this method, the fluoride concentration in the oyster tissue is of 24 mg/kg (Table 1). The fluoride concentration given by the producer, National Institute of Standards and Technology (NIST), was 240 mg F/kg. In order to have our findings controlled, oyster samples were sent over to a reference laboratory, which in the first testing, using acid digestion, came up with a fluoride concentration of 2.3 mg/kg. In a second testing, using the ashing method, the fluoride concentration was measured to be 250 mg/kg. Thus, in three analyses we obtained results that differed two orders in magnitude; the ashing method being closest to the value given by NIST.

**TABLE 1.** Fluoride concentration in oyster tissue (1566a) in mg/kg, as informed and determined by National Institute of Standards and Technology (NIST), using three different methods for digestion of the tissue material. In all cases the fluoride ion-selective electrode was used to determine fluoride in the final solution.

Informative value	Acid digestion method	Acid digestion method	Ashing method
NIST	University of Bergen	Reference laboratory	Reference laboratory
240	24	2.3	250

#### MATERIALS AND METHODS

As the ashing method seamed to give the most reliable results when analysing oyster tissue, we decided to use this method for further studies. In order to evaluate the accuracy of the method, i. e. the agreement between the sample's true content of fluoride and the result of the analysis, we wanted to verify the method through testing of a certified reference material. A reference material or an in-house control material (material prepared in the laboratory) should have a fluoride concentration similar to the concentration expected in the authentic sample. At present, very few reference

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materials certified for fluoride are available. We managed to find four products in addition to the oyster tissue (1566a, from National Institute of Standards and Technology): prawns (GBW 08572), fly ash (GBW 0807) and two brands of maize (GBW 08506, National Research Centre for Certified Reference Materials, China<sup>2</sup>). Unfortunately, both oyster tissue and maize are presently commercially unavailable. We have, consequently, chosen to test two in-house control materials; tea (African Pride, Tanzania Tea Blender LTD., Dar es Salaam, Tanzania) and cod (0271 cod powder, Rieber & Søn A/S, Bergen, Norway), in addition to the prawns.

Dried samples of 0.2 to 2.0 g, depending on the expected fluoride content, were weighed and put into 70 ml nickel crucibles. The samples were covered with 6 ml 8 M sodium hydroxide solution. After the slurries had been evaporated to dryness on a hot plate, the crucibles were covered and combusted in an oven at 525°C. After cooling, distilled water was added and the crucibles were once more put on a hot plate in order to better dissolute the fusion cake. The sample solution was neutralised by nitric acid, TISAB was and the fluoride concentration was measured using an ion selective electrode (Orion 9609) connected to potentiometer (Orion, model 920A).

Linearity of the standard response curve, the limit of determination, the recovery and repeatability were recorded.

## **RESULTS AND DISCUSSION**

**Linearity of the standard curve.** In Table 2 the r-values from linear regression analysis are given. Satisfactory results were observed with concentrations as low as 0.01 mg F/L and the best results were obtained when using the standards based on blank samples. The r-values describe the linear relationship between mV-readings and fluoride concentration (log F) in the standard solutions. Measuring response was determined using six fluoride concentrations, 0.0001, 0.001, 0.01, 0.1, 1.0 and 10.0 mg F/L covering both sides of the active working range. The standard solutions were prepared by adding sodium fluoride standard (100 mg/L) to blank samples (series 1 and 2 in table 2) or distilled water (series 3 and 4 in Table 2). The reason for using a blank sample to prepare the standards was to obtain the same matrix in the standard solutions as in the samples.

**Limit of determination.** We tested two different fluoride ion-selective electrodes (A and B) using the same blank solutions. For electrode A, the limit of determination was 0.02 mg F/L, while for electrode B the limit was 0.1 mg F/L. The limit of determination is the lowest amount of fluoride in a sample that can be quantitatively determined with a certain confidence. The limit is calculated as 10 times the standard deviation of the average of the blank samples (N>20). The results illustrate that there can be great differences between electrodes. Electrodes should, therefore, be carefully tested before being used.

**Repeatability.** In Table 3 the repeatability of three different samples is shown. The relative standard deviation was 8, 13 and 20 % for the oyster tissue, prawns and cod, respectively. This is not satisfactory for the two lowest concentrations. The prawns were certified to 5.31 mg F/kg resulting in a relative error of 9 %. An estimate of the repeatability of a method is obtained when identical test portions are analysed in the same laboratory, using the same equipment and within a short period of time. We

made samples from the same batch, and we used materials ranging over different concentration levels since the precision depends very much on the concentration of the analyte. It should however be noted that the N for oyster tissue and prawns are low and that the analysis of the cod tissue was not obtained in a single day.

**TABLE 2.** Linear regression analysis of mV-readings vs. log F of 3, 4, 5 or 6 measuring points of the standard curve using fluoride ion-selective electrode. Measuring point 1, 2, 3, 4, 5 and 6 are concentrations of 10, 1.0, 0.1, 0.01, 0.001 and 0.0001 mg F/L, respectively. Series 1 and 2 are made from 100 mg/L NaF-solution diluted in blank samples (NaOH, HCO<sub>3</sub> and distilled water) while series 3 and 4 are diluted to correct concentration with distilled water.

Standards	Series 1	Series 2	Series 3	Series 4
	r-values	r-values	r-values	r-values
1, 2, 3, 4, 5 and 6	- 0.9037	- 0.8987	- 0.9112	- 0.9258
1, 2, 3, 4 and 5	- 0.9443	- 0.9420	- 0.9598	- 0.9637
1, 2, 3 and 4	- 0.9812	- 0.9810	- 0.9927	- 0.9907
1, 2 and 3	- 0.9990	- 0.9987	- 0.9999	- 0.9999

<b>TABLE 3.</b> Repeatability of different samples.						
Sample type	Ν	Concentration mg/kg SD		RSD %	Relative error %	
Oyster tissue	5	250	20	8	-	
Prawns	4	4.8	0.6	12	9	
Cod	26	41	8	20	-	

**Recovery.** In Table 4 the results from the recovery test are shown. We found a recovery of 105 and 95 % in the tea and cod samples, respectively. The recovery is thus within acceptable limits. A known amount of fluoride was added and analysed in order to check the possible interference with the analyte from any compounds in the sample. The amount added should correspond to the level normally present in the sample material and the recovery should be in the range 80-110 %.<sup>3</sup>

<b>TABLE 4.</b> Recovery of fluoride (%) in tea and cod samples.						
Tissue	Ν	g	Added F, μl (100 mg/L)	F in sample mg/kg	Added F mg/kg	Recovery %
Tea	5	0.2000	280	124	$238 \pm 7$	105±7
Cod	5	0.5000	300	37.2	$67.6\pm2.6$	95±8

There obviously is need for documentation of the reliability of analytical procedures for the determination of fluoride in foods. To be able to compare results from different studies, analyses that give the total fluoride concentration should be preferred. This is because different acids and different mixtures of acids may dissolve the fluoride in the organic matter to various degrees. The absorption and bioavailability of fluoride from different foods are influenced by several factors e.g. the mineral content of the diet<sup>4,5</sup> and pH of the gastric juices.<sup>6</sup> Therefore it is not possible to extrapolate results obtained by either acid digestion or methods giving the

total fluoride concentration to available fluoride in human beings. Analytical determination of fluoride should preferably be followed up by bioavailability experiments.

There is a great need for a standardised reference material with both low-fluoride and high-fluoride content based on cereals or vegetables. This to have some sort of external test of the methods used. The recovery is often given as a proof for the reliability of a method. A satisfactory recovery tells us that there are no interfering agents and that the fluoride added has been detected. It is important to remember that the recovery tells nothing about how much of the total fluoride in a sample that has been analysed.

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## ASSESSMENT OF THE FLUORIDE CONTENT OF WEANING FOOD ITEMS IN WESTERN UGANDA.

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SUMMARY: The fluoride intake through weaning food was assessed with reference to the Kasese district in Western Uganda. Thirty one mothers with children less than five years of age participated in the study and provided food samples. The fluoride content of the food items on dry basis weigh basis varied from 1.8 mg F/kg to 24 mg F/kg with a median of 2.7 mg F/kg. The highest concentrations were found in millet and sorghum. As these cereals are part of the standard diet, it is discussed that staple weaning food may contribute considerably to the total fluoride load in toddlers in the Kasese district. The findings are in agreement with the fact that fluorosis may be prevalent in areas of low fluoride content in the water.

Key words: Fluoride; Weaning; Food, Fluorosis, Western Uganda, Millet, Sorghum.

#### INTRODUCTION

An excessive intake of fluoride during early childhood may cause damage to the formation and calcification of teeth; resulting in dental fluorosis. Dental fluorosis is endemic in the African Rift Valley, including certain areas of Uganda.<sup>1</sup> In areas with high fluoride content in the drinking water, water is considered to be the main source of ingested fluoride.<sup>2</sup> Other ingredients of the diet may, however, contribute to the total daily fluoride intake, and need to be investigated.<sup>3</sup> The present study was made to assess the fluoride content of staple weaning foods in the Kasese district of Western Uganda, an area known for high prevalence of dental fluorosis in areas of both low (0.6 mg/L) and high (2-3 mg/L) water fluoride levels.<sup>1</sup> The weaning food is defined as that food consumed by children during the period the child is introduced to foods other than breast milk.<sup>4</sup>

#### MATERIALS AND METHODS

In collaboration with local community health workers, 31 homes were randomly selected for the investigation. The basis for inclusion was the presence of at least one child below the age of five. The mothers (age 18 to 42), who were interviewed following a standard questionnaire, provided samples of the weaning food used in the household, as well as samples of drinking water.

The mothers also provided the investigators with an estimate of the average amount of food a child ate or drank at a meal. The estimates were measured using a top balance scale for solids and a measuring cylinder for liquids.

The food items were sun-dried and brought to University of Bergen, Norway for analysis. Fluoride analyses were performed by use of a combined fluoride electrode (Orion 96 09 00) connected to an Orion 920A ISE Meter. An ashing method described by Malde *et al*<sup>5</sup> was used to assay the fluoride in the food items.

#### RESULTS

Water. The fluoride concentration of 13 samples ranged from 0.2 mg/L to 0.7 mg/L. The mean value was calculated as 0.4 mg/L (s.d = 0.17).

**Weaning period.** Weaning of Kasese children started on an average at 4 months of age (range 1-6) and lasted until the children were 22 months (range 8 - 40).

Weaning food. The food samples obtained from the mothers consisted of cereals, legumes, bananas, roots

the principal for	ood to which a child			
was weaned.				
Food	Number of children <sup>*</sup>			
Millet	23			
Banana	15			
Roots (tubers)	7			
Others <sup>**</sup>	12			
<sup>*</sup> A child can be weaned to more than one				
food type.				
**				

TABLE 1. Food type mentioned as

\*\* Some food types were mentioned only once, this is a total of all those mentioned less than 5 times.

(tubers), and leafy vegetables. The food items that were more commonly mentioned in the interviews are given in Table 1.

The fluoride content of the food ranged from 1.8 mg/kg to 24 mg/kg dry weight (d.w.) cf. Table 2. The median concentration was 2.7 mg/kg. Lowest value of 1.8 mg/kg was found in legumes (beans and groundnuts) and highest of 24 mg/kg was found in the cereal millet. Based on the estimates given by the mothers, the fluoride content of the average child's daily food intake was calculated for the various age groups- as shown in Table 3.

<b>TABLE 2.</b> Mean fluoride concentrations from food groups.					
Food Group	Number of Semples	Mean F conc	Standard		
roou oroup	Number of Samples	mg/kg (d.w.)	deviation		
Legumes	11	1.8	0.4		
Banana	4	2.1	0.5		
Leafy vegetables	5	2.7	0.6		
Roots/Tubers:	6	1.9	0.4		
	2	2.0	0.5		
	2	2.3	0.3		
Cereal <sup>*</sup> : Sorghum	1	5.6			
Millet	1	24.0			
* Samples were measured in duplicate to obtain the result with standard deviations of: millet					
= 1.5, Sorghum $= 6.1$ .					

**TABLE 3.** Theoretical children intakes of fluoride though porridge based on Sorghum or millet containing respectively 5.6 and 24 mg F<sup>-</sup>/kg. It is assumed that the porridge is prepared by using of 250 g dry weight /L.

Age (months)	Ν	Average consumption (mL/day) <sup>1</sup>	Sorghum <sup>2</sup> 5.6 mg F⁻/kg	Millet <sup>2</sup> 24 mg F <sup>-</sup> /kg	Upper <sup>3</sup> limit fluoride intake (mg F <sup>-</sup> /day)
<6	13	600	0.84	3.6	0.33
7-12	17	900	1.26	5.4	0.40
13 - 36	48	900	1.26	5.4	0.65
<sup>1</sup> Estimates made by mothers					
<sup>2</sup> 250 g of flour are used to prepare 1 L of porridge					
<sup>3</sup> Upper limit based on Calorie needs and recommended intake of 0.05 mg per 100 kcal. <sup>6</sup>					

#### DISCUSSION

The average fluoride concentration of the water samples from the 31 households was 0.4 mg/L (range 0.2 - 0.7). According to accepted standards an upper level of 0.7 mg/L is recommended for tropical countries.<sup>7</sup> We should, consequently, expect to see little or no dental fluorosis in this population. However, a recent study by Rwenyonyi *et al*<sup>8</sup> found a dental fluorosis prevalence of 25 % in low-water-fluoride areas (0.5 mg F/L) in the Kasese district. A previous study by Møller *et al*<sup>1</sup> reported even higher prevalence of dental fluorosis (72.7 %) at a water fluoride level of 0.6 mg/L in the same area. The high prevalence of dental fluorosis in these low fluoride areas seems to indicate the presence of alternative fluoride sources perhaps present in the diet.

The weaning food in rural Africa comprises of locally available products. Thus, it is the traditional staple food that is made into weaning food.<sup>9</sup> Analyses have shown that *commercially available* weaning foods may substantially add to the fluoride load of infants and toddlers.<sup>10,11</sup> There is, however, very little work done previously focusing on unprocessed foods (such as these used in Kasese) as to their possible role as a source of fluoride in the diet of infants and toddlers.

The median fluoride concentration found in Kasese, 2.7 mg F/kg), is well above what is considered *low* (0.1-2.5 mg F/kg) and above what is reportedly the level found in unprocessed foods.<sup>6</sup> The Kasese weaning foods may, consequently, contribute considerably to the total fluoride intake in children, especially where sorghum and millet are used. The influence of nutritional fluoride may start early, as the mean age for weaning of children of the Kasese district is 4 months.

The bio-availibility and amount of daily intake of fluoride from the relevant food items needs to be further examined. Further studies should also be done to asses the possible contribution of tea and trona in areas with endemic dental fluorosis.

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# FLUORIDE AND SILICON CONTENT IN DRINKING WATER

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SUMMARY: Though the fluoride content of drinking water has been implicated as the main cause of fluorosis, the severity and incidence of the disease do not always correlate with the fluoride content of drinking water. It has been suggested that other components in the water and/or in the diet may enhance or protect from fluoride toxicity. Silicon has been suggested to enhance the fluoride toxicity. In this study 227 water samples from fluorotic and non-fluorotic areas in India are analysed for silicon and fluoride contents. Water samples containing less than 2 mg F/L show significant positive correlation between the fluoride and the silicon content. Water samples containing more than 2 mg F/L sow a significant negative correlation between the fluoride was observed in four none fluorotic areas, while there was no correlation in samples from fluorotic areas. It is discussed that the silicon content of water from bore well and water from open wells was not found statistically significant.

Key words: Fluoride; Silicon; Drinking water; Fluorosis; Fluorotic areas, India.

#### INTRODUCTION

Endemic fluorosis has been a major public health problem in India as well as in many other countries. Although the fluoride content of drinking water is considered to be the most important factor responsible for endemic fluorosis, results of many studies have shown that the incidence and severity of the disease does not always run parallel with the levels of fluoride in drinking water.<sup>1,2,3</sup> Therefore, it was suggested that some other factors present in water and/or diets would enhance the fluoride toxicity.

Reddy and Srikantia<sup>4</sup> observed that high protein, adequate vitamin C and calcium could prevent the appearance of symptoms of fluorosis in experimental animals. Lakshmaiah and Srikantia<sup>5</sup> have observed that the fluoride retention was significantly higher on sorghum based diet than those based on rice. Even general under-nutrition has been shown to enhance the deleterious effects of fluoride ingestion in cattle.<sup>6</sup>

Recently Kaminisky and co-workers<sup>7</sup> observed that radiographically detectable steosclerosis due to chronic exposure to high fluoride (8 mg/L) in drinking water, was not associated with clinical symptoms. This has further strengthened the idea that there are other factors in water and/or diet that would enhance or decrease fluoride toxicity. Studies carried out at the National Institute of Nutrition, Hyderabad, have shown that the silicon content of the diet is one such factor.<sup>8</sup> In those studies, it was observed that there was a significant increase in fluoride content of femur of animals fed high silicate diet. Silicon has been shown to be an essential element for chicks and rats<sup>9,10</sup> and it is also known to be essential for bone mineralization.

Since it is a common observation that elements supplied through drinking water are absorbed to a large extent, we were interested to know the silicon content of drinking water samples from fluorotic areas. To the author's knowledge, there is only one Russian report<sup>11</sup> linking silicon content of drinking water samples to hardness of

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water. Keeping in view of these studies, a detailed investigation is planned with the following objectives:

- To find out whether there exists any relationship between fluoride and silicon content of drinking water samples collected from different areas.
- Whether the fluoride content of deep bore wells is different from the surface water or water from dug wells.
- Whether silicon content of drinking water follows any particular trend with varying fluoride content in fluorotic and non-fluorotic areas.

<b>TABLE 1.</b> Number of water samples and their locations.					
Sample Lo	cation	No. of	Samples from		
		samples	bore wells		
Kakatiya N	lagar	11	7		
Prashant N	agar	34	14		
Seethaphal	mandi	41	15		
Nacharam		29	10		
North East	ern parts of India	38	0		
Samples from villages in Nalgonda (fluorotic area):					
I.	Samastha Narayanpuram	13	12		
II.	Shivannagudem, Ananthampet	36	36		
III.	Yeragandlapalli	12	12		
IV.	Indurthi	13	13		

## MATERIALS AND METHODS

Two hundred and twenty seven drinking water samples were collected. Of these 74 samples were from endemic fluorotic villages in Nalgonda district of Andhra Pradesh State, 38 from North-eastern part of India and the remaining 115 samples were from surrounding areas of Hyderabad city, table 1.

In all water samples, the fluoride content was measured using Orion fluoride ion specific electrode and silicon by a plasma atomic emission spectrometer with 251.611 nm emission line.<sup>12</sup> The recoveries of added elements, fluoride and silicon, were nearly 100 % and replicates agreed very well. Analysis of standard reference materials showed a good agreement with the reported values.

As there were distinctly two patterns in the distribution of fluoride values in relation to silicon values, the water samples were divided into 2 groups. The first group of 146 water samples with less than 2 mg F/L and the second group of 81 water samples with 2.0 mg F/L or more. By regression equations it was evaluated whether the silicon content could be expressed as a function of the fluoride content in these 2 groups was evaluated. The statistical fitness was evaluated using the student's 't' test and the correlation coefficients were calculated between fluoride and silicon contents.<sup>14</sup>

## RESULTS

**Bore well vs. surface water.** The distribution of the drinking water samples is given in table 1. About 52 % of the samples were from bore wells. There was no statistically significant difference in the mean fluoride content of water samples from bore well and open wells.

Silicon and fluoride contents of water F < 2 mg/L. The relationship between the contents of silicon and fluoride for the first group of samples is given in Figure 1. There is a significant positive correlation between the two parameters (P<0.001) (r=+0.7297). Since 1 mg/L fluoride content is suggested as upper limit for drinking water in tropical countries the corresponding silicon content appears to be 28 mg/L. According to the observed trend every increase in 0.5 mg F/L, the increase in silicon content is about 10 mg/L.







Silicon and fluoride contents in water with  $F \ge 2$  mg/L. Figure 2 shows the obtained relationship of silicon to fluoride content in water samples with more than or equal to 2 mg F/L. There is a significant negative correlation (r = -0.2894, p<0.01)

between fluoride and silicon content. The decline in silicon content in relation to fluoride content appeared to be 1.2:1.

**Region-wise analysis.** The results were also analysed from another angle. The water samples were from 6 distinctly different areas. The relationship between fluoride and silicon content is calculated for all 6 locations. It was observed that wherever the fluoride content was high, i.e. fluorotic area, there was no relationship between fluoride and silicon content. Further, wherever fluoride values were low, i.e. non-fluorotic area, there was a significant positive correlation.

#### DISCUSSION

The results presented in this study are different from those of Teotia and Teotia<sup>13</sup> who suggested that the fluoride content of deep bore wells was lower than those from the dug wells. Recently Kaminisky and co-workers,<sup>7</sup> have reported that there were no clinical symptoms of fluorosis in population groups who consumed for several years drinking water with 8 mg F/L. It is reported that the fluoride content of femur of animals fed high silicate diet was significantly high.<sup>8</sup> It is a common observation that in India the symptoms of fluorosis are seen in areas where the fluoride content of drinking water is 3 to 4 mg/L. It is possible that even the low fluoride values may become toxic when their silicon content is high. However, in situations where the fluoride content is high, e, g. 6 to 8 mg/L, the low silicon content may act as protective factor and prevent the manifestation of the disease. Reddy and Srikantia<sup>4</sup> observed that high Ca, vitamin C and protein protected the experimental animals from fluorosis.

There appears to be some limit for the silicon solubility in water samples (maximum about 50 mg/L). At higher concentration silicon compounds polymerise and become insoluble.

There are no reports in India on the silicon content of water samples. In one report from Russia the silicon content was about 25 mg/L with hardness less than 2.5 meq/L.<sup>11</sup> Since it is suggested that high silicon content may enhance the fluoride toxicity, it appears that 28 mg Si/L in waters containing 1 mg F/L is optimal. The silicon content of water and food samples might play an important role in fluorosis in some areas in India where the fluoride content of drinking water is moderately high.

The region-wise analysis of the data showed that silicon and fluoride content of water samples from north eastern parts of India are very low.

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# Session III

# Defluoridation; Laboratory Experiences

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# DEFLUORIDATION PROPERTIES OF ACTIVATED ALUMINA

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SUMMARY: Laboratory studies were conducted to assess the chemical behaviour of activated alumina as a defluoridating material by the batch equilibration method. Defluoridation is found to require a minimum of 20 minutes contact time. The capacity is found to be between 3 mg/g in alkaline water and 20 mgF/g in acidic water. At pH 7, the defluoridation capacity was found to be 5.6 mg/g. The capacity decreases with increased bicarbonate concentration, but is found to be independent of the presence of other anions like chloride and sulphate. The saturated medium could be regenerated by 2 % hydrochloric acid, 2 % sodium hydroxide or 1 % sulphuric acid. The fluoride removal obeyed Langmuir's adsorption isotherm indicating that the forces of adsorption are governed by chemi-sorption.

**Key words:** Activated alumina; Alumina; Fluoride; Defluoridation capacity; Batch equilibration; Regeneration; Sorption mechanism.

#### INTRODUCTION

Defluoridation of water is one of the alternatives already adopted for provision of safe drinking water in some fluorotic areas. A large number of materials have been identified as potential defluoridating materials, functioning through ion exchange, adsorption and chemical processes. Each method has its limitations as well as its merits.

Activated alumina is one of the most popular and cost effective materials used for defluoridation of water.<sup>1-6</sup> However very little information is available about the chemical behaviour of activated alumina in the defluoridation process. This study is carried out to establish the dependence of various factors governing the defluoridation property of activated alumina. The effect of different parameters on the defluoridation capacity has been experimentally verified. Based on the results a plausible mechanism of the fluoride removal by activated alumina is proposed.

## MATERIALS AND METHODS

Defluoridation experiments using activated alumina LR grade supplied by s.d. Fine Chemicals Pvt. Ltd., Boisar, India were carried out by batch equilibration method. Besides the determination of defluoridation capacity of activated alumina, the effect of the variables such as contact time, particle size and other parameters like pH, concentration of fluoride ion, temperature and the presence of chloride, sulphate and bicarbonate ions were experimentally verified.

The concentration of fluoride was measured using expandable ion analyser EA 920, the fluoride ion sensitive electrode 9409 and the reference electrode (all Orion USA make).<sup>7</sup> The pH measurements were done using an analog pH meter 324 of Systronics India make. The concentrations of chloride and bicarbonate ions were determined by the standard titrimetric method.<sup>8</sup> The concentration of sulphate ion was determined by turbidimetric method using Spectronic 20 spectrophotometer (Bausch and Lomb).<sup>8</sup>

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Regeneration experiments were carried out using regenerants like hydrochloric acid, sulphuric acid and sodium hydroxide.

Defluoridation capacity of activated alumina was calculated in mg F per g by dividing the amount of removed fluoride by the added amount of the medium.

The effect of contact period on the efficiency of activated alumina was determined by keeping the following parameters constant:

- The particle size
- pH at 7.0
- The fluoride ion concentration of raw water sample at 10 mg/L
- The concentration of chloride, sulphate, and bicarbonate ions as nil.

Activated alumina of different particle sizes were prepared using scientific test sieves of ELITE scientific instruments Co., Bombay. The experiments were conducted by keeping the concentration of fluoride at 10 mg/L, in the absence of chloride, sulphate, and bicarbonate ions. Temperature was retained at 30°C and the pH was maintained at 7. The effect of pH on the defluoridation capacity was determined at the following five pH levels viz., 3, 5, 7, 9, and 12. All weighings were carried out using a Mettler AE 240 electronic balance having an accuracy of 0.02 mg.

#### **RESULTS AND DISCUSSION**

**Contact time, particle size and temperature.** The defluoridation capacity of activated alumina reached saturation after a period of 20 minutes. That is, the minimum period of contact required for maximum defluoridation is 20 minutes. There is an increase in the defluoridation capacity with a decrease in the particle size as expected, since the process is governed by adsorption. The defluoridation capacity of activated alumina is independent of temperature.

**Effect of fluoride and other ions.** The adsorption of fluoride on activated alumina obeyed Langmuir's adsorption isotherm indicating that the forces of adsorption are governed by chemi-sorption.

Chloride and sulphate ions have very little effect on the fluoride removal capacity of activated alumina, whereas bicarbonate ions behave differently. Figure 1 shows the influence of bicarbonate ions. Activated alumina adsorbs bicarbonate ions also in addition to fluoride. The observed dependence of bicarbonate ions on the defluoridation capacity of the material is due to the specificity factor, which distinguishes between fluoride and other ions such as chloride and sulphate besides bicarbonate. The adsorption of anions by activated alumina takes place in the following order:

 $F^{-} > OH^{-} >> HCO_{3}^{-} >>> Cl^{-} = SO_{4}^{2-}$ 

**Effect of pH.** Figure 2 shows the marked dependence of pH on the defluoridation efficiency of the adsorbent. At pH 7, it has the defluoridation capacity of 5.6 mg F/g activated alumina which increased nearly four times to a value of 20.4 mg F/g at pH 3. It decreases to 3.0 mg F/g activated alumina at higher alkaline range. It is worth noticing the extremely high defluoridation capacity at pH 3, a property, which could make activated alumina a very promising material. The reasons for this marked influence of pH are discussed in detail in the later part of this paper.

**Regeneration.** Results of elution capacity using sulphuric acid, hydrochloric acid and sodium hydroxide indicate that 2 % hydrochloric acid, 2 % sodium hydroxide and 1 % sulphuric acid are good regenerants.

**Defluoridation** mechanism. Defluoridation by activated alumina is explained in terms of adsorption of fluoride ions at the activated alumina/solution interface. Stern model of the alumina/solution interface is shown in Figure 3 and the mechanism of adsorption by activated alumina is explained on the basis of the double layer theory.

The zero charge potential for activated alumina is at pH 9 and the surface charge of activated alumina may be controlled by the potential determining ion. The potential determining ion in this case is the hydrogen ion.<sup>9,10</sup> This means that when the concentration of hydrogen ion is greater, i.e. if the pH is less than 9, the alumina surface acquires a positive charge and this surface adsorbs negatively charged fluoride ions from the solution. Therefore at low pH range the positive charge on the surface of activated alumina increases and adsorption of fluoride becomes more pronounced.

Thus one type of the force responsible for adsorption of fluoride on activated







alumina is certainly the columbic forces between the positively charged aluminium oxide and negatively charged fluoride ions. Nevertheless, the charge alone does not determine the adsorption process in the double layer. Another factor responsible is the specificity. This 'specificity' factor distinguishes between the fluoride and two ions, chloride and sulphate. The charged aluminium oxide favours fluoride ion and hence there is no adsorption of chloride and sulphate ions. However, activated alumina also adsorbs bicarbonate ions to some extent though the order is  $F^- > HCO_3^-$ . This is probably the reason for the decrease in the defluoridation capacity of activated alumina at increased concentrations of bicarbonate ions.

At pH greater than 9, when the activated alumina acquires a negative charge, there is still sufficient defluoridation capacity which cannot be explained in terms of forces of chemisorption. Fluoride removal by activated alumina may be purely governed by physisorption at higher pH ranges. In fact adsorption through van der Waals type of forces may be taking place to some extent throughout the pH This argument range. is supported by the fact that the defluoridation capacity is reached only after a



minimum of contact time of 20 minutes. Chemi-sorption also starts taking place with decreasing pH and the extent of chemi-sorption increases at a much higher rate. This explains the very steep increase of defluoridation capacity of activated alumina from pH 5 to pH 3. When fluoride is eluted by 2 % sodium hydroxide solution the activated alumina acquires a very high negative charge and the adsorbed fluoride ions are repelled from the surface and are diffused into sodium hydroxide solution.

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# KINETICS OF SORPTION OF FLUORIDE ON CALCINED MAGNESITE IN BATCH

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SUMMARY: A series of sorption of fluoride on calcined magnesite are obtained from thermostatic pH-chemostat and jar test experiments. The fluoride removal is observed not to start instantly, as normal, but after a lag time of up to ½ an hour of contact time. A model for sorption kinetics is developed, based on first-order reaction with respect to the concentration of fluoride. The rate constant is directly proportional to the dosage. The model takes into accounts the lag time observed. The kinetical model can be described for any given dosage and initial fluoride concentration in the water. The reaction rate parameter, K, varies however slightly for different initial concentrations of fluoride in the water and different dosage of calcined magnesia. These relationships are described separately by two linear equations. It is discussed that the observed lag time is due to the fact that magnesia cannot remove fluoride without hydrolysis, and it is probably this hydrolysis which constraint the start of the removal.

**Key Words:** Fluoride Removal; Defluoridation; Drinking Water Treatment; Sorption Kinetics; Magnesite; Calcination; Modelling.

#### INTRODUCTION

The use of drinking water containing fluoride at high levels, which affect the public health, is a serious problem for communities in fluorotic areas in many developing countries. A number of defluoridation methods have been developed and tested.<sup>1</sup> These include the bone char, the contact precipitation, the Nalgonda technique, the activated alumina and the clay sorption techniques, just to mention the prominent ones. This paper presents the kinetical aspects of defluoridation by means of calcined magnesite.

Already in the 1930's the magnesium oxide, i.e. calcined magnesia, has been proposed for the removal of fluoride from drinking water.<sup>12</sup> Though it has not been successfully implemented so far, the calcined magnesia is one of the most promising defluoridating agents, especially in countries like Tanzania, where magnesite can be harvested from locally available mines.<sup>2-6</sup>

In spite of reported high fluoride removal capacity,<sup>6</sup> the basic batch kinetics of the fluoride uptake by magnesia has not been established yet. A literature survey has revealed that only a few batch experiments using jar test apparatus have been reported.<sup>3,5,7</sup> In most cases the results show the fluoride concentration versus time. Apparently there is no correlation between the surface area of the calcined magnesite and the fluoride removal capacity.<sup>3,7</sup> There seems to be a correlation between the dosage of calcined magnesite and the fluoride removed from the water.<sup>3</sup> Some results show that the fluoride uptake, for a dosage of 2 g/L, initial concentration of 20 mg/L and after a contact time of 12 hours, is almost negligible for smaller particle media, 0.063 mm, meanwhile there is a substantial fluoride uptake by bigger grain particle media, even up to 24 hours.<sup>3</sup> One of the developed models predicts that fluoride uptake is a slow and a continuous process which is taking place even after a contact time of 52 hours for dosage of 7.0 g/L, initial fluoride concentration of 20 mg/L and grain size of 2.0-4.0 mm.<sup>5</sup> On the contrary, another model predicts that the equilibrium concentration in the aqueous phase would be achieved after a contact

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time of 15 hours with varying sorption constant K for dosage of 8.0 g/L, initial fluoride concentration of 20.0 mg/L and grain size of 0.25-0.5 mm.<sup>3</sup> The magnesia used in these studies, i.e. Nielsen<sup>3</sup> and Jackson and co-workers<sup>5</sup> was calcined at 700°C for a duration of 4.5 hours.

An advanced mathematical model was developed by Bregnhøj and Dahi<sup>9,10</sup> to describe the uptake kinetics on bone char media. The model was tested for validity on calcined magnesite, but it was concluded that it does not fit well.<sup>8</sup> The reaction rate parameter, K, varied between 0.00076 and 0.0029 L/(mg.h<sup>0.5</sup>) for a capacity,  $f^*$  as high as 56 mg/g.<sup>8</sup>

The objective of this paper is to present a new series of sorption data and to propose a more-simple model for kinetics of fluoride uptake by calcined magnesite in agreement with the obtained experimental data.

# MATERIALS AND METHODS

Two types of experiments are carried out; one using jar test equipment and another using a chemostat regulated at constant temperature and pH.

Preparation of Magnesia. Magnesite was collected 230 km east of Arusha town at Chambogo mines in Same district, the Kilimanjaro region in northern part of Tanzania. The material was crushed to small grain sizes of 0.063 - 2 mm on an electrical crushing machine and then calcined in a furnace. The temperature was raised at a rate of 4°C per minute from room temperature to 700°C, and then kept constant for 4.5 hours. The oven was then allowed to cool to room temperature before opening. The electrical furnace used was of type Scandia micro programmable TC - 15/45 and of type Nabertherm, model L9/R. After calcination the magnesia was sorted out in a test sieve shaker type Endecotts LTD EFL2 mk3 and type Vibro (VM1) to different sizes fractions. The grain size 0.063 - 0.125 mm diameter was used. The specific surface area was tested according to BET method.

Jar test experiments. The jar test experiments were carried out by setting six 1 litre beakers each containing equal quantity of raw water under a stirring paddle. The jar test apparatus was of type Phipps and Birds Stirrer 7790-402. Two different series of experiments were made. The first set of experiments was made using a constant initial fluoride concentration of 20 mg/L for different dosage of 0.5 - 4 g/L. The second set was made at constant dosage of 0.5 g media per litre for different initial fluoride concentrations of 5 - 31 mg/L. Stirring was carried out at 25 RPM. Five mL samples were taken at the different contact times, up to 6 hours. The samples were filtered through 0.45  $\mu$ m membrane filters from Schleider and Schuel, before measurement of the fluoride concentrations.

pH-chemostat. The chemostat experiments were carried out using a Dosimat 715, 614 Impulsomat, Water-bath themostated at 25°C type Julabo U3, two 691 pH Meters Metrohm type, burettes, multiple stirrer, 1 litre beaker, pH and fluoride electrodes. The pH was kept constant at 10.5 at different given initial fluoride concentrations. The solutions were kept stirred using a paddle rotated at about 100 RPM to keep the magnesia particles evenly suspended in the solution. Before starting each experiment, the thermostat, pH and fluoride electrodes were submerged in the fluoride solution in the beaker. Readings of pH and mV were recorded. The electrodes used were Radiometer F1052 fluoride electrode and Metrohm Ag/AgCl reference electrode with sleeve type diaphragm connected to a Metrohm 691 pH-meter. pH was monitored using a Metrohm 691 pH meter and combined pH electrode. The Impulsomat was set correctly before adding magnesia to the fluoride solution in the beakers. The dosimat, which was connected to the impulsomat, was feeding a 0.25 M nitric acid solution to the beaker, automatically as pH tends to raise above 10.5. Controlling pH below 10.5 was made manually by adding 0.25 M potassium hydroxide using a glass burette, supplied with a peristaltic pump. The experiment was carried out for a duration of 4 hours. Samples were taken before starting, after 15 minutes and at the end of each experiment. The samples were filtered using a SM 16510/11 polycarbonate filter holder through 0.22  $\mu$ m membrane filter paper from Schleider and Schuel. The samples were tested for fluoride, total alkalinity and hardness. The results of pH, mV readings and net acid added were recorded at different intervals. Each experiment was carried out in 2 replicates.

Fluoride and pH-measurements. The fluoride concentrations were measured directly using Radiometer F1052 fluoride electrode and Metrohm Ag/AgCl reference electrode with sleeve type diaphragm connected to a Metrohm 691 pH-meter. pH was monitored using a Metrohm 691 pH meter and combined pH electrode. Both pH and fluoride concentration were measured according to Standard Methods.<sup>11</sup>

#### RESULTS

The results from the jar tests series of experiments are presented in Figure 1 A & B. The rate of fluoride removal is plotted versus contact time in order to illustrate the observed lag time before the uptake of fluoride starts taking place.

# MODEL DEVELOPMENT

**Order of reaction.** As the rate of removal is not following a continuously decreasing pattern, the data obtained would not be following a simple kinetic of a well defined order. Thus plotting the data as  $\ln(F_{R,t}/F_{R,0})$ , as  $1/F_{R,t}$  and as  $F_{R,t}^{1-n}$  have confirmed that the order of the reaction, if any, is not of  $1^{st}$  or  $2^{nd}$  order. It varies between 1.2 - 1.8, and it does not take into account the lag time. It was concluded that the reaction pattern is complex and the reaction order changes within the same run. However, for engineering practise, since the rate of reaction is high within the first 2 hours and it fits the first order pattern within this time, it was found appropriate to adopt a simple  $1^{st}$  order kinetics, but where the model takes into account the lag time as observed.

In an advanced description of the sorption kinetics, the rate of the reaction is dependent on the residual fluoride concentration in the water as well as the simultaneous residual of defluoridation capacity of the sorbing medium. A literature study has revealed that such derivations would always result in complex expressions containing multiple variables and constants, cf. e.g. Nielsen 1993.<sup>3</sup> In this study, therefore, an attempt is made to develop a simple model, which describes the change of fluoride concentration in water as a function of time. The model should however is pragmatic and compromises about the precision of fitting the pattern in order to limit the expression using as few variables and rate parameters as possible.

Model description. The model adopted is as below:

 $F_t = F_0 \exp[-K \cdot X_T \cdot (t - t_{lag})]$ 



dosage of magnesia (A; initial conc. 20 mg/L) and initial fluoride concentrations (B; dosage is 0.5 g/L). The media are calcined at 700°C for 4.5 hours. The grain size is 0.063 - 0.125 mm.

Thus the model takes into account:

- The rate of removal follows a simple 1<sup>st</sup> order kinetics with respect to the residual fluoride concentration, F<sub>T</sub>.
- The rate constant being k is not a constant, but is dependent on the dosage, X<sub>T</sub>.
- k is approximately directly proportional to the dosage  $X_T$ , thus  $k = K \cdot X_T$ .
- The reaction does not start at t = 0, but at  $t = t_{lag}$ .

# DISCUSSION

**Derived parameters.** Figure 2 A shows that addition of 0.5 g/L to water containing 20 mgF/L resulted in a concentration of less than 5 after a contact time of 6 hours.

This means a removal capacity of about 30 mg/g, which is extremely high compared to other mineral media and even to bone char.<sup>1</sup>

The experimental and estimated model results are presented graphically in Figure 2. It shows that the model reproduces the trend of the experimental data very well for both at high as well as low dosage.



**FIGURE 2** Comparison of experimental data and the developed model results for jar test experiments. Initial fluoride concentration in A is 20 mg/L. Dosage of magnesia in B was 0.5 g/L. The media is calcined at 700°C for duration of 4.5 hours, grain size 0.063 - 0.125 mm.

The estimated values of K are presented in table 1, including the results observed from different dosage, different initial fluoride concentrations and the chemostat series where pH was controlled at 10.5.

From the deviation of the estimated K, respectively 8.4, 14.1 and 33.9 % of the average value, it may be concluded that the model is most sensitive towards variations in the initial fluoride concentration, especially when pH is controlled where estimation of an average K becomes of no use.

This is in agreement with the above-mentioned fact that the process is not a simple 1<sup>st</sup> order reaction. Figure 3 illustrates the trends of K-values when estimated for different dosage and initial fluoride concentration.

TABLE 1. Estimated reaction values, IV, of the proposed model.								
Batches of different dosage			Batches of different initial F			Chemostat $pH = 10.5$		
F <sub>0</sub>	XT	К	F <sub>0</sub>	X <sub>T</sub>	K	F <sub>0</sub>	XT	K
mg F/L	g/L	L·g <sup>-1</sup> ·h <sup>-1</sup>	mg F/L	g/L	L·g <sup>-1</sup> ·h <sup>-1</sup>	mg F/L	( <b>g</b> )	L·g <sup>-1</sup> ·h <sup>-1</sup>
20.1	0.5	0.696	5.04	0.5	0.693	10.6	0.5	0.36
20.1	1.0	0.658	10.1	0.5	0.652	11.3	0.5	0.32
20.1	1.5	0.715	15.9	0.5	0.731	11.7	0.5	0.33
20.1	2.0	0.632	21.8	0.5	0.599	20.0	0.5	0.41
20.1	3.0	0.561	25.7	0.5	0.572	21.7	0.5	0.66
20.1	4.0	0.633	31.4	0.5	0.489			
SUM $(F-F_{est})^2 = 41.5$		SUM $(F-F_{est})^2 = 30.1$		SUM $(S-S_{est})^2 = 14.5$				
Average K 0.649					0.623			0.416

TABLE 1. Estimated reaction values, K, of the proposed model.



**FIGURE 3.** Estimated trends of K-value dependency on the dosage, left, and the initial fluoride concentration, right.

Also the  $t_{lag}$  value is found to be dependent on the dosage as well as the alkalinity of the water. The lower the dosage and the higher the alkalinity, the higher is the value of lag time. For dosage of 0.5 g/L to water of no alkalinity, of the  $t_{lag}$  was found to be 15 minutes. The same dosage to natural water with initial alkalinity the lagging was about 30 minutes.

The developed model fits to both jar and pH-chemostat experiments though the two types of experiments represent different approaches. The pH-chemostat experiments represent a controlled experiments in the laboratory, while jar test experiments represent the experiments which can be done practically in the field.

The observed lag time has not been reported in connection with other defluoridation media. On the contrary the defluoridation process is known to start instantly at maximum rate.<sup>10</sup> The delay in case of magnesia, is believed to be due to hydrolysis processes which apparently have to take place, prior to defluoridation. This may indicate that magnesia by it self can not remove fluoride. Its hydrolysis product on the other hand is highly efficient in taking up the fluoride from the water.

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## LIST OF SYMBOLS

- t time, (h).
- $t_{lag}$  lag time before the reaction starts taking place, (h).
- $X_T$  concentration of sorption media or the dosage, (g·L<sup>-1</sup>)
- K reaction rate parameter,  $(L \cdot g^{-1} \cdot h^{-1})$
- k reaction rate constant or specific rate,  $(h^{-1})$  where n is order of reaction.

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# PREPARATION OF BONE CHAR BY CALCINATION

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SUMMARY: Preparation of bone char in ceramic kilns as used in the northern part of Thailand is normally a calcination rather than a charring process. This laboratory study shows that firing the cleaned bones up to 500°C and then gradually opening the kiln gate and letting it cool down results in bone char of a good quality and a with maximum fluoride removal efficiency of 91.8 %. At higher temperature of calcination, the percentage was less, decreasing with increasing temperature. At 800°C the percentage of fluoride removed was less than 50. For comparison, the efficiency of normal ICOH bone char, which is heated in an electric furnace at 600°C for 20 minutes, was only 69.4 %. Thus calcination in ceramic kiln may be a promising process for use by local communities.

Key words: Bone charring; calcination, local preparation, Thailand.

# **INTRODUCTION**

The ICOH Defluoridator, which has been available for defluoridation in Northern Thailand for several years, is based on bone char burnt at 600°C for 20 minutes in an electric furnace. Its defluoridation properties are attributed to adsorption and ion exchange of the water fluoride. Its efficiency depends on the initial fluoride concentration, the amount of bone char available and amount of water treated and the rate of water flow. The bone media of the ICOH defluoridators have to be changed every 1 - 3 months. Nowadays, ICOH is no longer able to supply ready-made filters to communities and communities cannot produce such a bone char themselves because of the high cost of electric furnaces as well as pollution to the environment of the bone charring process.

In 1995, three high-school children living in high fluoride areas in Lamphun Province tried to solve this problem in their community by charring cattle bones on a stove by the calcination process, but with uncontrolled temperature. They used the black coloured bone char as a filter medium in their defluoridators and measured the residual fluoride in the water after filtration. Their experiences demonstrated good efficiency in fluoride removal.<sup>1</sup> However one disadvantage of this bone char was that the water, after filtration, had a bad smell and was yellow in colour, i.e. unfit for drinking. To treat the bad odour and yellow water, 1 litre of activated carbon was used to make it clear and odourless. This would increase the cost of filtration and necessitate cleaning the activated carbon every 3 - 4 weeks and completely changing it every 3 months. However, the community can carry out this method of bone char production, if the process could be carried out at optimal temperature, providing high removal capacity of the medium and good water quality for drinking. Thus the calcination of bones is an interesting process, which may be the process of choice for communities in their local preparation of defluoridation media.

From many scientific studies reviewed, the two charring techniques are described:

- 1. **Calcination** is known as a process of high temperature heating in the presence of atmospheric oxygen. The end product being pure bone mineral, a compound related to hydroxyapatite. All organic material are combusted to  $CO_2$ .
- 2. **Pyrolysis** is a charring process with no or very limited access to oxygen. In the organic phase the bone is converted into inorganic carbon and graphite, which makes this bone char black whatever charring temperature is used.

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However, the fluoride uptake is mostly ascribed to the bone mineral phase (apatite).<sup>2,3</sup>

Recently, a study presented by Dahi and Bregnhøj<sup>3</sup> showed that when bone has free access to oxygen in the atmosphere (calcination), the defluoridation capacity is drastically reduced when heated at temperatures higher than 550°C. In the same range pH and alkalinity of defluoridated water increases. They found that pyrolysis provides the best bone char defluoridation capacity demonstrated by a linear correlation between surface area and defluoridation capacity.<sup>3</sup> According to Mjengera et al.<sup>4</sup> bone charred at 600°C for a duration of 20 minutes has an optimal fluoride removal capacity.

## MATERIALS AND METHODS

**Calcination.** Twenty kilograms of cattle bone were cleaned, all meat remnants, lipids and tendons being removed, and then dried in the sun for 2-3 days. Batches of 3 kg at a time were heated on the plate of a cross draft kiln with a thermocouple. Atmospheric air was flushed into the kiln chamber from room temperature for approximately 1 hour. Then the firing was stopped and the kiln-gate opened gradually to cool down. The process was performed at 300, 400, 500, 600, 700, 800°C for 1.5 - 2 hours. This yielded 6 batches of bone char weighing 1 kg each. The bone char was ground in a mortar and sieved to obtain 3 - 5 mm grain size.

**Conventional calcination.** For comparison purposes bone char was also prepared as normally used in the ICOH defluoridator, i. e. calcined up to 500°C, then firing stopped.



**Bone char properties.** The colours of the bone char batches were recorded. The defluoridation capability of the bone char batches was tested by placing 80 g in 3 plastic columns imitating defluoridators. The plastic column set-up is shown in Figure 1. The columns were loaded at the rates of 1 or 4 L/h using water containing respectively 2.56 and 2 mg fluoride/L. Treated water samples were collected every 5 respectively 10 minutes of filtration. The fluoride concentrations were measured using Ion Selective Electrode Orion 720A and Orion 720.

**Defluoridation capability.** The mean residual fluoride for each kind of bone char tested in the columns was calculated. The mean percentage of fluoride removed was used as an expression of defluoridation capability.

#### RESULTS

**Colour of bone char.** The colour of bone calcined at the different from low to high temperatures was different, changing respectively from black to brown, grey and white. For each batch the fraction of the different colours was determined, Table 1.

TABLE 1. Physical property of bone calcined at different temperatures						
Temp.	Popo colour	Property of treated water				
°C	Bone colour	Colour	Smell	Taste		
300	black : brown (4:1)	Dark yellow	Bad odour	Undrinkable		
400	black : brown (4:1)	Dark yellow	Bad odour	Undrinkable		
500	black : brown (3:1)	Light yellow to clear	None	Normal		
600	black : grey (4:1)	Clear	None	Normal		
700	black : grey (3:1)	Clear	None	Normal		
800	Black : grey : white (3:2:1)	Clear	None	Normal		

**TABLE 1.** Physical property of bone calcined at different temperatures

**TABLE 1.** Fluoride removal (%) in case of filtration through columns of bone char calcined up to different temperatures.

Bone char 80 g, flow rate 1 L/h,  $S_0=2.56$  mg F/L

Temp (°C)	Fluoride removal %
500	91.8
600	75.0
700	62.5
800	45.3

Organoleptic Property of treated water. The appearance as well as the smell and taste of the treated water was evaluated as shown in Table 1.

Defluoridation of water. Bone calcined up to 500°C then stopping firing, had the highest mean percentage of fluoride removed (91.8 %). At temperature higher of calcination, the percentage was less decreasing with temperature. increasing At 800°C the percentage of fluoride removed was less than 50 %, Table 2, Figure 2.



**FIGURE 2.** Average residual fluoride concentration after filtration by calcined bone char at different temperatures (bone char 80 g, flow rate 1 L/h,  $S_0$ =2.56 mg F/L)



The bone char normally used in the ICOH defluoridators, i.e. heated at 600°C in electric furnace for 20 minutes, was found to remove 69.4 % of the water fluoride.

Thus compared with the "normal" bone char, the bone calcined as described above at 500°C, with 88.9 % removal, had 28 % higher defluoridation capacity.

#### DISCUSSION

Because the burning bone was also fired by its organic components, it continued burning for a while after the burner had been extinguished in the kiln. During that time the calcination temperature would increase if no heat was released. Hence, to stop the increase of calcination temperature, the kiln gate was immediately and gradually opened after firing stopped.

The results of this study seems to correspond to that of Dahi and Bregnhøj<sup>3</sup> who concluded that the defluoridation capacity is drastically reduced between 550°C and 600°C, if we assume the fluoride removal efficiency shown in this study. However, the optimal calcination temperature presented (500°C) may be different from their experiment, according to different procedures. According to this study however, maximum removal capacity was obtained by firing the bones from room temperature up to 500°C in 1.5 - 2 hours then stopping firing and gradually opening the kiln gate to let the calcined bone char cool down. Moreover, the treated water by calcined bone char at optimal temperature had good properties of the treated water.

Further studies should be carried out to confirm the efficiency and clarify other parameters in defluoridation by calcined bone char, before bringing it to communities as fluoride filter for drinking water. However the results obtained so far seem to show that the calcination procedure followed may be appropriate for production of bone char by local communities

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# EFFECT OF CALCIUM ADDITION ON THE DEFLUORIDATION CAPACITY OF BONE CHAR

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SUMMARY: Dosage of small amounts of calcium chloride to fluoride water prior to contact with bone char which has already been saturated with fluoride is shown to provide an additional fluoride removal capacity. The additionally obtained removal capacity increases with slower filtration velocities and increasing calcium dosage. A filtration velocity of 0.07 m/h, corresponding to a contact time of about 2 hours, and a dosage of 100 mgCa/L, are shown to provide an additional removal capacity of about 3 mgF/g, i.e. almost a doubling of the genuine removal capacity of the bone char. The process is shown to be capable of reducing the fluoride concentration form 10 to about 0.5 mgF/L. The additionally saturated column is shown to be regenerated by simple adjustment of the pH of the water to 11 and allowing to flow for a few bed volumes. The useful regeneration capacity, where the fluoride concentration is reduced from 10 to less than 1.5 mg/L is determined to be about 1.0 mg/g.

**Key words:** Bone char; Fluoride; Calcium; Defluoridation capacity; Regeneration; Contact precipitation.

#### INTRODUCTION

Several methods have been studied for defluoridation of drinking water. Few of them have been tested and utilized in practice. One of the most studied and practiced method is the use of bone char in sorption filters. Bone char is an excellent selective sorbing agent for fluoride. Its capacity to absorb the fluoride is relatively high, 2-6 mg/g.<sup>1</sup> Furthermore, if bone char is prepared properly it would, apart from removing the fluoride, improve the raw water quality in general.<sup>2</sup>

However, the use of bone char in a simple sorption process has a couple of serious limitations. Firstly the change of the medium upon saturation is a cumbersome job. Secondly the good quality bone char may be unaffordable to most local peoples in fluorotic areas. Thirdly, the availability of good quality bone char is limited, if not non-existing, in most of these areas. Many efforts have therefore been brought about in order to develop methods by which the usability of the bone char could be improved. Three of these methods may be distinguished: The regeneration, the surface coating and the contact precipitation.

The *regeneration* method has been used at a large scale in Briton, USA, where a plant was operated for many years.<sup>3</sup> Thus it is proven that bone char can be saturated and regenerated for several hundreds cycles. The regeneration is carried out by the use of a 1 % alkaline solution, where the process is assumed to be an ion exchange between the fluoride ions and the hydroxyl ions:

 $BC-F^- + OH^- \Rightarrow BC-OH^- + F^-$ 

The *surface coating* has been studied in the laboratory by Christoffersen et al..<sup>4</sup> According to this method, fluoride saturated bone char, if immersed in an acidic solution of calcium and phosphate or of bone char powder, would take up a fresh layer of hydroxyapatite on its surface. The so surface coated bone char will behave as fresh bone char and be able to absorb a new amount of fluoride. The saturation and

surface coating processes has been demonstrated to function for several cycles, of declining capacities though. The bone char powder is normally produced as a waste material in the preparation of grained bone char.

The *contact precipitation* method has been reported recently. It has been tested for defluoridation of drinking water in a primary school in Tanzania for a couple of years.<sup>5</sup> According to this method, appropriate amounts of calcium and phosphate compounds are added to the raw water prior to its flow through the fluoride saturated bone char filter. The bone char, being rich with fluorapatite, would catalyze the precipitation of fluoride mainly in agreement with the following equation:

$$10 \text{ Ca}^{++} + 6 \text{ PO}_4^{---} + 2 \text{ F}^- \Rightarrow \text{Ca}_{10}(\text{PO}_4)_6\text{F}_2$$

It has been proved that raw water containing approximately 10 mg/L fluoride could be treated on long term basis at an efficiency of 95 %, i.e. residual concentration of about 0.5 mg/L, by addition of 120 mg/L calcium and 150 mg PO<sub>4</sub>/L phosphate. The main mechanism behind the contact precipitation is attributed to the production of fluorapatite. However, if all the removed fluoride was precipitated as pure fluorapatite, higher dosage of phosphate would have been needed. It has therefore been concluded that the process, at least at the employed relatively high initial fluoride concentration, may involve some precipitation of calcium fluoride.<sup>5</sup>

The purpose of this study is to investigate the processes taking place when calcium alone is added to the fluoride water prior to flow through the fluoride saturated bone char column.

#### METHODS

**Fluoride water.** Tap water from the water supply at Ngurdoto, Tanzania, was used through out the experiments. The fluoride concentration, which was between 9.5 and 11.5 mg/L of drinking water, was standardized to 10 mg F/L of raw water for treatment. This was done either by addition of sodium fluoride in stock solution or by dilution with rainwater.

**Saturated bone char.** Cattle bones collected at village level were charred in a locally developed kiln packed with charcoal. The charring took about 15 hours, during which the access of atmospheric oxygen was restricted and the temperature controlled at between 400 and 500°C. The charred bones were crushed and sieved into grain size 0.5 -1.5 mm. Three kg of bone char medium were added to 9 liters of a solution containing 1gram fluoride per one liter of drinking water. The mix was stirred regularly for a period of 3 weeks. At the end the water had a fluoride content of 21 mg/L i.e. the bone char had absorbed 3 mg F/g bone char. This saturated bone char was drained, dried and then used through out this study.

**Batch experiments.** Batch experiments were carried out in plastic bottles containing one liter of fluoride water. In order to account for blind effects the experiment was carried out in triple, one where calcium but not saturated bone char was added, one where saturated bone char alone was added and one where both calcium and saturated bone char were added. The bottles were shaken by turning upside down 10 times and left idle for 8 hour. This was repeated 6 times in order to provide a total contact time of 48 hours, before the residual fluoride concentrations were measured.

**Column experiments.** Glass columns of 30 mm inner diameter and 300 mm height were packed with 106 mL or 80 g saturated bone char, the height of the bed being 150

mm. The height of the supernatant water was 50 mm. The columns were loaded with the fluoride tap water at rates of 50, 200 and 800 mL/hr, corresponding respectively to 2.1 h, 32 min. and 8 min. hydraulic retention times or to 1.13, 0.28 and 0.07 m/h flow velocities. The flow was controlled using an inlet of silicon-tubing and a regulating clamp in combination with a dripping device. The calcium chloride and or sodium hydroxide were added by, respectively during water treatment and regeneration of media. The outlet water was collected in batches and the average fluoride concentration was measured batch wise.

**Fluoride and pH measurements.** The fluoride concentrations were measured using a fluoride selective electrode (Metrohm 6.0502.150) and an Ag/AgCl reference electrode (Metrohm 6.0726.100) connected to a Metrohm 704 pH-meter. Five mL of samples was mixed with 5 mL of TISAB and compared with standard solutions of 1 to 10 mg F/L. pH was measured using a Metrohm electrode 6.0220.100.

# RESULTS

Table 1 shows the effect of addition of calcium to fluoride water containing saturated bone char. It is seen that the dosage of calcium results in additional uptake of fluoride, but only in the presence of saturated bone char.

The column experiments of

different flow rates of fluoride Figure 1. Flow rates of 50, 200 and 800 mL/h are utilized. These correspond to hydraulic retention times of 2.1 hours, 32 minutes and 8 minutes or to flow-velocities of 0.07, 0.28 and 1.13 m/h respectively.

In the third campaign of experiments the same columns and water are used. The flow rate was fixed to 200 mL/h, but the dosage of calcium was different, respectively 0, 25 and 50 mg/L. The results are shown in figure 2.

The fourth set of experiment aimed at cycling between treat-ment of the fluoride water and regenerating the **TABLE 1.** Batch experiment where calcium is added to water containing10 mg/L fluoride with and without the presence of saturated bone char. Contact time is 48 hours.

Added meterials	Flask	Flask	Flask
Auteu mater lais	Ι	II	III
CaCl <sub>2</sub> in mgCa/L :	360	0	360
Saturated bone char in g/L:	0	2	2
Fluoride residual (mg/L)	10	8.2	4.5
Add. F- removal cap. mg/g	0	0.9	2.8

different flow rates of fluoride water added 100 mg/L calcium are shown in



**FIGURE 1.** Column experiments where 10 mg/L fluoride water is added 100 mg/L calcium and the mix is passing the saturated bone char columns at three different rates of flow. Hydraulic retention time is indicated.

columns by use of sodium hydroxide. The results are presented in Figure 3.

#### DISCUSSION

It is well known that calcium can precipitate fluoride as calcium fluoride,  $CaF_2$ , the solubility constant Ks being 1.92 10<sup>-11</sup>. The results of the batch experiments shown in table 1 illustrate that calcium and fluoride may remain dissolved in the water, after a contact time of 48 hour, even if the solubility product is exceeded by not less than 130 times. The results are in agreement with the fact that the fluoride can not be removed instantly from the water by simple addition of large quantities of calcium compounds.

The results of the batch experiment illustrate furthermore the difficulty in defining the saturation point of bone char. Though the bone char was



different amounts of calcium are added to 10 mg/L fluoride water. The columns containing saturated bone char are loaded with the water at a fixed flow velocity of 0.28 m/h corresponding to retention time of 32 minutes.

saturated after contact time of three weeks, ending in a steady residual fluoride concentration of 21 mg/L, it was able, at a later stage, to remove fluoride from water containing 10 mg/L. Though this additional removal capacity is limited, 0.9 mg/g, it illustrates the complexity of the bone char saturation processes. The experiments demonstrate however, that addition of calcium increases this additional removal capacity significantly. The additional removal capacity gained by addition of calcium is measured to be 2.8 mg/g, i. e. about doubling the genuine removal capacity of the bone char.

Fluoride is also removed from the water, as the fluoride water, after addition of 100 mgCa/L, passes through a saturated bone char column. Figure 1 shows that the bone char removal capacity increases with increasing hydraulic retention time, i.e. slower filtration velocities. If the breakpoint is defined to 1.5 mgF/L in the treated water, the additional removal capacities obtained in the columns are 0.8, 1.6 and 3 mg/g respectively for the flow velocities of 1.15, 0.28 and 0.07 m/h. The slowest flow utilized, which is giving the highest augmentation in the calcium-bone char removal capacity, corresponds to contact time of approximately one hour. Faster flow probably results in higher escape of the added calcium. As compared to the batch experiments, this may indicate that the observed removal process is not a simple filtration of already precipitated calcium fluoride, but that the precipitation, whether calcium fluoride or not, is facilitated by the saturated bone char in the column. A contact time of at least 1 hour, i.e. a hydraulic retention time of at least 2 hours, is required for the process to take place.

The results shown in Figure 2 confirm that the saturated bone char by it self is able to absorb more fluoride, but at very low removal efficiencies, i.e. 50 % or less. Again if an effluent concentration of 1.5 mgF/L is defined as acceptable water quality criteria,



the number of treated bed volumes were 0, 40 and 120, for dosage of respectively 0, 25 and 50 mgCa/L. These represent additional efficient removal capacities of 0, 0.5 and 1.5 mg/g. It is therefore concluded that the amount of added calcium is crucial for the efficiency of the removal. The data are not conclusive about the optimum dosage. At flow of 200 mL/h, i.e. hydraulic retention time of 32 minutes, similar capacities, 1.5 and 1.6 mg/g, are obtained at dosage of respectively 50 and 100 mgCa/L.

The above mentioned results show clearly that the fluoride saturated bone char, if added calcium is able to absorb more fluoride, but only to become saturated again at higher capacity level. The results of the regeneration experiment, Figure 3, show that interruption of the treatment and adjusting pH to 11 elutes the additionally absorbed fluoride and renews the capability of the column to treat more water. The treatment capacity of the column is shown to be about 320, 150, 140 and 160 bed volumes. Counting on average effluent fluoride concentration of approximately 3 mg/l the removal capacity of the column in its four operation periods is respectively 3.0, 1.4, 1.3 and 1.4 mg/g. This indicates that the regeneration may be operated for equidistant cycles, i.e. at technical use, but at an efficient capacity of less than 1.4 mg/g. If the above mentioned criteria of 1.5 mg/L is defined as acceptable, then regenerative fluoride removal capacity gained by addition of calcium, i.e. 2.8 mg/g, and the genuine capacity of the bone char, i.e. 2-4 mg/g.

The results presented in this study are in agreement with the previous studies of Christoffersen et al. 1991,<sup>4</sup> Larsen and Pearce 1992,<sup>6</sup> Bregnhøj 1995<sup>1</sup> and Dahi 1996,<sup>5</sup>

all showing that calcium compounds are able to increase the fluoride removal capacity of the bone char. Yet the results are not conclusive about the mechanisms of the removal observed. It is known that bone char is not a perfect compound of hydroxy- and fluorapatite. The bone structures, especially after processing, storage or drying may be in deficit of calcium. If this calcium deficiency is covered through addition of e.g. calcium chloride, the bone char will be able to remove more fluoride from the water. The additional removal observed in this study could be attributed to coverage of this calcium deficit through contact precipitation of calcium fluoride and/or fluorapatite. It is unlikely that calcium fluoride is the only precipitate produced, as the residual fluoride concentration is much below the equilibrium concentration of saturation. It can however not be excluded that a simple ion exchange is playing a major role in the observed regeneration of the saturated bone char columns.

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# DEFLUORIDATION OF DRINKING WATER BY THE USE OF CLAY/SOIL

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SUMMARY: The present is part of a greater project assessing the fluoride-binding effect of soils from different parts of the world. In this part the clay/soil samples from Ethiopia are studied with respect to capacity and the effect of heating and stirring. The F-binding capacity of an «optimal» soil sample, laterite from Northern Cameroon is assessed by the use of sequential filtration. Five soil samples, collected from the high-land areas around Addis Ababa, reduced the fluoride content of the water from about 15 to 1 mg/L, at dosage levels of about 100 g/L. Pre-drying at 50 °C resulted in similar efficiency as preheating at 250°C. Soil samples from high-fluoride areas of the Ethiopian Rift Valley showed much lower and even negative removal efficiencies. Stirring speeded up the process, but it did not influence the finally obtained removal capacity. A gradual decline in F-removal efficiency was seen in the laterite used for sequential filtration. The average removal efficiency under the selected sequential filtration conditions was about 58 %, ranging from 90 % during the first sequences down to practically nil in the 30<sup>th</sup> runs. With an initial fluoride concentration of 15 mg/L, the total fluoride binding capacity of laterite from Cameroon was found to be approximately 0.5 mg/g. It is concluded that the high-land soil, due to easy availability and low cost, may be useful for removal of excessive fluoride from drinking water at household and village levels.

**Key words:** Fluoride removal; Lateritic soils; Clay firing; Stirring; Fluoride binding capacity; Ethiopian soil.

## INTRODUCTION

Harmful effects of fluoride intake are primarily, but not exclusively, linked to high levels of fluoride in the drinking water. As high-fluoride groundwater is often the only available and reliable source of water in low-precipitation fluorotic areas such as the East African Rift Valley, simple methods for low-cost defluoridation of drinking water are urgently needed.<sup>1</sup>

A series of methods have been suggested for the defluoridation of drinking water, but the *ideal system* may not yet have been found. Any health measure should, in order to be implemented, be effective, inexpensive and simple, and also *socially and ethically acceptable*. Thus, well-known, locally available defluoridation media such as clay and soil are of much interest in the high-fluoride areas of East Africa.

Fluoride is a natural constituent of the biosphere, and, consequently, of clay and soil. The inherent fluoride content, however, varies greatly from one area to another. Geology as well as the local topography<sup>1</sup> and depth of the soil layer<sup>2</sup> are factors to be considered in the quest for soils suitable for defluoridation purposes.

Clay-pots have, especially in the past, been widely used for the transport and storage of drinking water. Inexpensive, locally made ceramics may improve the quality of the water: apart from keeping water relatively cool, the clay pots may reduce the fluoride content of the water.<sup>3</sup> The fluoride binding effect of clay-ware is influenced by factors such as physico-chemical composition of the clay and the firing temperature in the kiln.<sup>4</sup> The aim of the present study was:

- to assess the fluoride binding effect of various Ethiopian soils, sampled, respectively, in mountainous and (relatively) low-land areas of the Rift Valley
- to test the possibility of using low-fired soils or soils with no previous firing for defluoridation of drinking water
- to study the effect of *stirring*, and, finally
- to assess the fluoride binding capacity in an «optimal» soil, laterite from Northern Cameroon, West Africa.

Findings related to the relative elevation of soil samples are more specifically reported elsewhere.<sup>5</sup> Here are presented results concerning preheating of the soil, stirring/no stirring of the water/soil systems, and also findings regarding the fluoride binding capacity of lateritic soil samples from Cameroon.

## MATERIALS AND METHODS

**Soil samples.** Ten topsoil samples (mostly red, lateritic clay) were collected in Ethiopia. Four of them, i.e. no 1 - 4, were from the mountainous areas around Addis Ababa. The six others, i.e. no 5- 10, were from the high-fluoride areas of the Rift Valley. In addition, samples of lateritic soil were collected from the high-land savannah of Adamoua, Northern Cameroon.

**Pre-treatment.** The Ethiopian samples were heated for about 3 hours at 250°C in an electrical furnace. Alternatively the samples were dried at 50°C for a similar period of time. Aliquots of 30-45 g were prepared from each of the soil samples. The Cameroon samples were left unheated.

**Fluoride water.** Fluoride solutions were prepared by dissolving 100 mg NaF in 3.0 L distilled, de-ionised water, giving a fluoride concentration of approximately 15 mg/L. Alternatively groundwater containing 7.4 mg F/L and NaF solutions containing 8.7 mg F/L were used.

**Batch kinetic experiments.** Soil samples were added to plastic containers and mixed with fluoride solutions in a weight/volume ratio of 1:10. The containers were closed by tight-fitting lids and placed on an electrical shaker (50 motions/min). At given different contact times, 5 mL samples were taken and analysed for the residual fluoride concentration in the water.

**Capacity measurement.** 100 g of untreated laterite was placed in a funnel - on filter paper circles - and used for sequential filtration of 4.8 L of the 15 mg F/L NaF solution described above. The filtration was carried out sequentially, where 200 mL aliquots were filtered at a time in total 24 times. The fluoride concentration was measured in each of the 24 filtrates. Three parallel test series were conducted.

**Fluoride measurements.** The fluoride concentrations were measured by the use of an Orion combination F electrode (9600900) connected to an Orion Research microprocessor ionalyzer/900.

#### RESULTS

**Various soils.** A sharp decline in the fluoride concentration was seen in all NaF solutions exposed to soils from the Ethiopian highlands. During the first hour roughly 50 % of the fluoride ions were removed from the water samples (Figures 1 and 2). In 24 hours the fluoride concentration was reduced to about 1 mg/L in all the four samples.

Only minor differences were observed in the fluoride binding patterns of soils fired at 250°C as compared the similar patterns of soils heated to 50°C only (Figure 1 versus Figure 2).

The fluoride removal effect of the soils from the Rift Valley was very limited; the average fluoride concentration in solutions after 24 hours of exposure 10.5 mg/L, was individual samples varying from 4.85 to 15.9 mg F/L; the last soil actually causing sample an increase in the fluoride content of the NaF solution (Figure 3).

Effect of stirring. The effect of stirring on fluoride removal in natural and artificially made high-fluoride waters was tested in medium efficient Ethiopian soil samples. As demonstrated by Table 1, stirring speeded up the initial fluoride binding process, but did not alter the efficiency on long-time. The fluoride removing effect, given in percentage of original fluoride concentration, was similar in artificial and naturally occurring high-fluoride waters.

Fluoride binding capacity. A in fluoride sharp reduction concentration was observed in passing through water the column made up of laterite from Cameroon. During the first few runs. the reduction was approximately 90 %. A gradual decline in the effect of filtration took place, but even after the 18<sup>th</sup> filtration, i.e.3600-mL of water containing 14.7 mg F/L, more than 50 % of the fluoride was removed by the laterite, cf. Figure 4.











**TABLE 1.** Defluoridation by the use of laterite, in artificially made high-fluoride, respectively naturally occurring waters, with or without stirring. Results given in percentage of original fluoride

Type of water	Stirring -	Fluoride removal in % after exposure time					
Type of water	Suring -	2 h	4 h	8 h	24 h	7 days	
NaF solution	Yes	48.7	64.4	77.6	87.2	94.0	
NaF solution	No	19.2	25.4	40.7	65.2	95.0	
Groundwater	Yes	50.3	66.8	76.4	84.2	93.0	
Groundwater	No	25.9	32.7	44.1	50.0	93.0	

concentrations 8.7 mg F/L, respectively 7.4 mg F/L

The experiment was terminated after 24 runs. By extrapolation one should expect the laterite filter (100 g) to be saturated by fluoride after approximately 6 more runs, i.e. after the filtration of 6000 L of fluoride solution containing 14.7 mg/L. The average removal of fluoride in the water would then be indicated in the graph as the adsorption of the  $15^{th}$  aliquot, i.e. 8.5 mg/L. Thus the trend shows that 100 g laterite may take up not less than 50 mg of fluoride, at an average removal efficiency of 58 %. The binding capacity, under such conditions would be slightly more than 0.5 mg/g.





# DISCUSSION

The fluoride which enters the hydrological system may, to a large extent, be traced back to volcanic activities associated with rift formation and chemical weathering of volcanic rocks.<sup>6</sup> The African Rift System is dominated by alkaline base-rocks, richer in Na and F than analogous rocks in other regions of the world<sup>7</sup>, and high-fluoride groundwater is a rule, rather than an exception in the Rift Valley. Dental and skeletal fluorosis, consequently, are endemic in the region and there is an acute need for simple and inexpensive methods for defluoridation of water.<sup>8</sup>

Clay pots, made from the local clay, have been tested for fluoride removal e.g. by Ndegwa.<sup>3</sup> Most writers have concluded that the method is too slow and that clay-ware has too little fluoride removing capacity.<sup>9</sup> As shown in our experiments, however, the capacity and the speed depend on the selection of clay/soil, and on the practical procedures involved in the fluoride removing process.

Recently, Zewge and Moges<sup>10</sup> investigated brick and pot chips as defluoridating media, and reported fluoride binding capacities up to 0.56 mg/g, which is similar to what we found in our column studies. This is still a *low* capacity, as compared to *ando soil*, a relatively young, black soil prevalent e.g. in Kenya<sup>11</sup> or *contact precipitation with sodium dihydrogen phosphate and calcium chloride*, as described by Dahi.<sup>12</sup> However, lateritic clay in most places is available at practically no cost, and by the use of simple batch-type methods drinking water with an acceptable fluoride concentration may be provided in less than 24 hours. By the use of stirring, the fluoride binding process may be speeded up. Likewise, a columnar type of soil/clay filter may produce low-fluoride water rather quickly.

Firing sterilises the clay, and reduces the turbidity induced by the defluoridation process. From an environmental point of view, however, firing should be avoided, as it requires energy and may lead to further deforestation and ecological strain in vulnerable areas such as the East African Rift Valley. Our finding that no firing of the soil is needed for defluoridation purposes would seem to make the proposed method even simpler. However, the possibility of micro-biological contamination of the soil, and, consequently, of the drinking water, should not be overlooked. Further studies are needed to assess this aspect.

Most of our experiments have been conducted using artificially made NaF solutions. According to the results presented in Table 1, defluoridation of, respectively, NaF solutions and chemically more complicated high-fluoride groundwater, seems to follow the same pattern.

Lateritic soils may not be «best» solution to the treatment of high-fluoride waters. Soil is, however, easily available in the problem-areas, the method is socially and environmentally acceptable, and implementation requires a minimum of equipment and technical skill. Even with its relatively low fluoride binding capacity, 1 kg of laterite should - according to our findings - be able to reduce the fluoride content of 100 L water from, e.g. 6 mg/L to less than 1 mg/L. Hence, the use of local clay or soil could possibly represent a simple and inexpensive way of improving the quality of drinking water in high-fluoride areas; at household level and at village level.

## ACKNOWLEDGEMENT

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# SIGNIFICANCE OF ELEVATION ON FLUORIDE BINDING CAPACITY OF ETHIOPIAN SOILS

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SUMMARY: The present study tests the hypothesis that elevation might be a factor to be considered in the search for the soil best fit for fluoride removal in high-F waters. Fifteen soil samples (mostly reddish ferruginous laterite) were collected from surface soil in the Ethiopian Rift Valley area above Addis Ababa, at elevations between 2000 and 3,000 m. The samples were heated at 500°C for three hours, whereupon the fluoride binding capacity was tested by immersing aliquots of 10 g fired soil in 100 ml NaF solution. The residual F-concentrations were checked versus time. Soils from the lower areas of the Rift Valley were fluoriderich, and tended to release fluoride, even to 40 mg F/L solutions. At initial concentration of 5 mg/L, addition of 100 mg soil to 1 litre of the water could almost double the initial fluoride concentration, i.e. "removal" efficiency of -78 %. On the other hand, soil samples from elevated areas could remove fluoride from the aqueous NaF solutions. On an average, the removal efficiencies were of the same magnitude, 67 and 68 % from waters containing respectively 5 and 40 mg/L. This corresponds to removal capacities of respectively 0.03 and 0.3 mg/g. It is discussed that soils from the Ethiopian Highland may be used for inexpensive moderate removal of excessive fluoride from the drinking water.

Key words: Fluoride; Defluoridation; Lateritic soils; Elevation; Ethiopia; Rift Valley.

## INTRODUCTION

The volcanic base-rock in the African Rift system is predominantly alkaline, and rich in e.g. sodium and fluoride. The soil produced by the weathering of these rocks is similarly rich in fluoride. After precipitation, however, rainwater leaches fluoride from soils as well as from crystalline rocks.<sup>1</sup> The surface waters of East Africa, therefore, usually have high fluoride concentrations.<sup>2</sup> Furthermore, since the hydrochemistry of aquifers is strongly influenced by the surrounding lithology, the fluoride content of ground water of Rift Valley is high, frequently to the extent that the waters are rendered unfit for human consumption.<sup>3,4</sup>

In discussing the occurrence of fluoride in the environment, a WHO expert committee (1994) concluded that «Waters with high fluoride content are usually found at the foot of high mountains» and, conversely, «In high mountains, the fluoride content of the soil is usually low».<sup>5</sup> The experts offered no explanation for these facts. However, over the millennia, fluoride may have been washed out of the topsoil in high places. Rivers and creeks have transported the element to the low lands, where it, due to evaporation, has enriched soil and local groundwaters. This might particularly be the case in the African Rift Valley, where low precipitation and high temperature often drain the mountain-rivers before they reach the ocean.

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High fluoride drinking water is a serious health problem in the Rift Valley as well as in other arid and semi-arid areas in Africa and elsewhere. Simple, low-cost defluoridation methods are urgently needed. Consequently, we decided to test the following hypotheses:

Fluoride-depleted topsoil may be able to reabsorb  $F^{-}$ , and, thereby, remove excessive fluoride from high-fluoride water.

The fluoride binding capacity of the soil may depend on e.g. the relative elevation of the sites from where the soils were sampled.

### MATERIAL AND METHODS

**Soil samples.** A series of 15 soil samples were collected for the study. The sampling sites were

chosen based on their relative elevation: Starting at Lake Shala, at the "bottom" of the Rift Valley, 2,000 m above seal level, soil samples were collected from sites of increasing elevation, up to an altitude of approximately 3,000 m, in the mountains around Addis Ababa. As exact data for the altitude of some of the sites are not known, elevation was indicated according to an increasing, relative scale, 1-10 (Table 1). The distance between the lowest and the highest point was around 200 km.

The samples were taken from the surface layers of the soil. Most samples were reddish ferruginous laterite; i.e. soils formed in tropical regions by decomposition of under-laying rocks. In some sites the laterite was covered by a layer of black, humus-like earth. The soil samples were collected in plastic bags, labelled, and brought to Bergen, Norway, for further processing. The soils were heated at 500°C for three hours, and aliquots of 10 g soil were prepared from each sample.

**Fluoride binding capacity.** The fluoride binding capacity of the various soils was assessed by immersing the 10 g soil samples into aliquots of 100 ml NaF, which had been prepared to contain 5, 10, 20 and 40 mg F/L.

The test samples (100 ml NaF solutions with 10 g soil) were kept unstirred at room temperature in lidded plastic vials. After 1, 2, 6, 12 and 24 hours, 5 ml liquid was taken from each of the (sixty) vials, and analyses were made by the use of a fluoride selective electrode (ORION comb. fluoride electrode 960900), according to standard methods.<sup>6</sup>

The results were entered in an SPSS-statistical programme.

#### RESULTS

Figure 1 and 2 demonstrate the results of fluoride analyses over a 24 h period in solutions of, respectively, 5 and 40 mg F/L, after the immersion of soil from the three lowest, respectively, highest sites. The immersion of soil in all cases caused changes in fluoride concentration of the NaF solutions. In most cases the fluoride concentration was decreasing.

<b>TABLE 1.</b> Location of sampled
soils, according to elevation.

Relative	A #00
elevation	Area
1.	Lake Shala
2.	Lake Langano
3.	Lake Ziway
4.	Wonji
5.	Мојо
6.	Debre Zeyt
6.	Akaki
7.	Borago River
8.	Borago Mountain (1)
8.	Borago Mountain (2)
9.	Kotebe Kara
10	Lambaret

However, soil samples from the lake areas, i.e. the lowest parts of the Rift Valley, tended to release fluoride into the solution, thereby increasing the concentration of fluoride. This was particularly notable in the solutions with lower concentrations of fluoride, i.e. 5-10 mg F/L, Figure 1.

Based on the results at 24 h, the reduction of fluoride, expressed in mg/L, was plotted against the relative elevation of the soil sites. Similar patterns were found in all the tested solutions: Soil from higher altitudes absorbed more fluoride than did soils from the areas at a lower elevation. There seemed to be no systematic difference in fluoride absorbing capacity between black and red soils.

#### DISCUSSION

The fact that clay and clay-ware as well as various soils may be used for defluoridation of water is well known.<sup>7-9</sup> In most cases, however, investigators have concluded that the F binding capacity of these materials is - for practical purposes - too low.<sup>10</sup> The reason for the negative conclusion may be that the defluoridation agents are made from high-fluoride clay/ pottery. The defluoridating agent may, consequently, have started out more or less saturated with fluoride. The average fluoride binding capacities observed in this study are shown in table 1.



**FIGURE 1.** Fluoride concentration in water vs. time, after exposure to soils collected at mountains areas and low parts of the Ethiopian Rift Valley, respectively. (Mean of three samples and standard deviation)



**FIGURE 2.** Fluoride concentration in water vs. time, after exposure to soils collected at mountain areas and low parts of the Ethiopian Rift Valley, respectively. (Mean of three samples and standard deviation)

TABI	E 1. Aver	age o	data	of fluor	ide remo	val as ca	lculated f	or three	replica	a sa	mple	эs
from	Ethiopian	soil	at di	ifferent	altitude.	Negative	removal	means	that t	he	soil	is
leach	ing fluoride	e to th	e fluc	oride wa	ater. Dosa	age is 100	g soil/L.					

Relative height in m	200	)0	30	00
Initial F-conc., mg/L	40	5	40	5
F-conc. after 24 contact time	41.9	9.4	13.0	1.6
F-removal efficiency in %	-5	-87	68	67
Soil removal capacity	-0.02	-0.04	0.27	0.03

It should be emphasised that there is a wide variation in the fluoride binding capacity also of high-altitude soils. In fact, the altitude per se is probably irrelevant. The crucial point might seem to be whether precipitation and surface water have depleted or enriched the soil as to fluoride.

The *best* defluoridating agent may not yet have been found through this study, but, according to the present findings, black and red soils from the Ethiopian Highland may be used for inexpensive moderate removal of excessive fluoride from the drinking water.

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## HOUSEHOLD PURIFICATION OF FLUORIDE CONTAMINATED MAGADI (TRONA)

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SUMMARY: Purification of fluoride contaminated magadi is studied using bone char sorption and calcium precipitation. The bone char treatment is found to be workable both in columns and in batches where the magadi is dissolved in water prior to treatment. The concentrations in the solutions were 89 g magadi/L and 95 and 400 mg F/L respectively in natural and synthetic solutions. The fluoride removal capacities observed were 4.6 mg F/g bone char for the column system and 2.7 mg F/g bone char for the batch system in case of synthetic magadi solution. It is however concluded that the batch system is the best treatment method. A procedure for purification of fluoride contaminated magadi at household level is described.

Key words: Trona; Magadi; Fluoride; Fluoride removal; Magadi purification; Household treatment.

#### INTRODUCTION

The World Health Organisation's guideline for the maximum fluoride concentration in drinking water is 1.5 mg/L. However, WHO stresses that the climatic conditions and daily water intake should be taken into consideration when adopting fluoride standards for countries. Especially in fluorosis afflicted areas in East Africa drinking water is not the only source of fluoride intake. The naturally occurring mineral trona  $(Na_2CO_3\cdot NaHCO_3\cdot 2H_2O)$ , locally called magadi, is known to contain relatively high amounts of fluoride.<sup>1,2</sup> Magadi is used in the household mainly as a tenderiser to shorten the cooking time of legumes and vegetables. The frequency of magadi use for food preparation in households in Arusha Region, Northern Tanzania ranges from daily use to a couple of times per week.<sup>3-6</sup> The fluoride content of magadi is subject to considerable variation. In Tanzanian magadi, concentrations from 0.1 mg F/g magadi to 18 mg/g have been reported.<sup>4,6-8</sup> The fluoride concentration in magadi originating from Lake Magadi and harvested by the Magadi Soda Company also varies.<sup>8</sup> However, a concentration of 4.0 mg F/g magadi has been reported to be typical.<sup>9</sup>

Since drinking water in most cases is the major contributor to the total fluoride intake efforts have been put in development of methods for defluoridation of drinking water. Promising methods which are now in use in Northern Tanzania are the contact precipitation technique and the bone char filter technique.<sup>10</sup> The human fluoride intake through magadi may under certain circumstances be significant, even compared to the intake through water and other sources.<sup>3,4</sup> The aim of this investigation is to develop a method for purification of fluoride contaminated magadi. For this purpose two processes are tested: A bone char sorption process in batch as well as in column and a precipitation process based on addition of calcium chloride.

#### MATERIALS AND METHODS

**Column Experiments.** Synthetic magadi solution and natural magadi solution were filtered through two glass column filters in series, the first containing bone char, the second containing activated carbon. Gravel was placed at the inlet and the outlet of both columns. The filters were connected with a plastic tube and closed with stoppers of rubber. Distilled water was filtered through the columns to clean the filter materials. Hereafter the filter columns were filled with magadi solution and left for 24 hours before

the filtration was started. The flow through the filters was kept constant during the whole experiment. Samples from the outlet of both the bone char filter and the activated carbon filter were taken at regular intervals of 6 to 8 hours. The column design parameters are listed in Table 1 and the design is illustrated in Figure 1.

Jar test experiments using bone char. Experiments were carried out in one litre plastic beakers containing 500 ml synthetic magadi solution. Amounts of 50 and 100 g bone char were added, respectively. The solutions were stirred in the Jar test apparatus (Phipps & Birds Stirrer 7790-402) at a speed of 50 and 100 rpm, respectively. After 24 hours of stirring samples were taken, filtered through a 0.45  $\mu$ m Minisart GF filter and fluoride concentration and alkalinity were tested.

Demonsterre	T	Synthe sol	tic magadi lution	Natural magadi solution	
rarameters	Unit	Bone Char	Activated Carbon	Bone Char	Activated Carbon
Cross sectional area, A	cm <sup>2</sup>	7.07	7.07	7.07	7.07
Length of filter bed, L	m	0.80	0.80	0.40	0.40
Surface loading, Darcy velocity, $v_s$	m/hour	0.10	0.10	0.06	0.06
Porosity of filter bed, $\varepsilon$	-	0.55	0.62	0.55	0.62
Contact time, T <sub>C</sub>	hours	4.3	4.9	3.9	4.4
Grain size of filter bed, GS	mm	2.0-2.8	1.0-3.0	2.0-2.8	1.0-3.0
Mass of filter bed, M	g	335	150	170	75

**TABLE 1.** Design parameters for filtration of synthetic and natural magadi solutions through bone char and activated carbon filters

**Precipitation experiments.** Experiments were carried out in one litre plastic beakers containing 500 ml synthetic magadi solution. Aliquots of 1.00, 2.00, 3.00, and 4.00 ml of calcium chloride solution were added, respectively. The solutions were stirred in the Jar test apparatus (Phipps & Birds Stirrer 7790-402) at a speed of 50 rpm for 5 minutes. After 1 hour of settling the solutions were decanted. Samples were taken, filtered through a 0.45  $\mu$ m Minisart GF filter and fluoride concentration and alkalinity were tested.

**Bone char preparation.** The bone material was delivered from the Danish bone meal factory DAKA, where the bone material has been boiled, washed, dried and crushed into small particles  $d_{BC}$ <4.0mm. The bone material was pyrolysed in a programmable ceramic oven (Scandia Oven, Type SK 355) in a closed steel container, where the access of atmospheric oxygen was restricted. The temperature was raised at a rate of 4°C/minute from room temperature to 600°C and then kept constant for 10 hours. The charred bone material was allowed to cool down to room temperature before the oven was opened. The bone char was divided into fractions  $d_{BC}$ <0.50 mm, 0.50-1.0 mm, 1.0-1.4 mm, 1.4-2.0 mm, 2.0-2.8 mm, and  $d_{BC}$  >2.8 mm using a test sieve shaker (Endecotts EFL2 mk3). The grain size 2.0-2.8 mm fraction was selected and used as bone char in all experiments. The specific surface area of the bone char was measured according to the BET method and found to be 116 m<sup>2</sup>/g.

**Calcium chloride solution.** The calcium chloride solution was prepared by addition of analytical  $CaCl_2$  to distilled water with a dosage of 2.5 mol/L.

**Synthetic magadi solution.** The synthetic magadi solution was prepared by addition of analytical chemicals to distilled water as follows: 0.393 mol/L Na<sub>2</sub>CO<sub>3</sub>, 0.393 mol/L NaHCO<sub>3</sub>, 21.1 mmol/L NaF, and 28.6 mmol/L NaCl. The dosage of sodium carbonate and sodium bicarbonate equals a concentration of 89 g/L pure trona.

**Natural magadi solution.** Magadi collected at Lake Natron, Tanzania was dissolved in distilled water (89 g/L) and the concentration of carbonates and fluoride equals a concentration of 1100 meq/L and 5.0 mmol/L, respectively.

**Fluoride and alkalinity measurements.** The fluoride concentrations were measured using a Metrohm fluoride ion selective electrode and a Metrohm Ag/AgCl reference electrode with a sleeve type diaphragm connected to a Metrohm potentiometer (692 pH/Ion Meter). Aliquots of 5.0 mL of the sample were mixed with 5.0 mL distilled water and 10.0 mL CDTA-tisab and the fluoride concentrations were measured using the standard addition procedure, according to Standard Methods.<sup>11</sup>

The alkalinity was measured using the end point titration procedure according to Standard Methods.<sup>11</sup> The end points were pH=8.3 and pH=4.5. Samples of 1.00 mL were diluted to



for column design experimental set-up for column design experiments in case of synthetic and natural magadi solutions.

50.0 mL and titrated automatically with 0.1 N  $H_2SO_4$  at a maximum titration rate of 1.00 mL/min. pH values were monitored continuously (5 seconds interval) using a Metrohm pH-electrode connected to a Metrohm 719S Titrino (potentiometer/dosimat). The values of pH and the amount of acid added were recorded on a PC. The calibration of the pH-electrode was carried out using Metrohm buffer solutions having pH=4.0 and pH=9.0.

#### RESULTS

The results of the measurements of the total alkalinity and the fluoride concentration in the effluent solution from the bone char columns and from the activated carbon columns are shown in Figure 2. The upper figure concerns the treatment of the natural magadi solution while the lower one concerns the synthetic solution. The results are plotted, for both the fluoride and the total alkalinity, as the ratio between the concentrations of the outlets and the concentration of the inlet to the bone char column. The abscissa is drawn as the fluoride loading on the bone char column. For the activated carbon column, the initial pore water is however neglected, as it has not been treated in the bone char column.

During the filtration the removed fluoride is taken up by the bone char. In Figure 3 the estimated fluoride concentration in the bone char,  $f_t$  is plotted versus the fluoride loading on the column. The fluoride concentration in the bone char is calculated as the sum of the

differences between the inlet and the effluent concentrations,  $S_{i}$ - $S_{e,t}$ , in the different samples multiplied by the differential part volumes of treated solutions,  $\Delta V_k$ , divided by the mass of bone char,  $M_{BC}$  in the column:

$$f_{t} = \frac{1}{M_{BC}} \sum_{k=0}^{V_{t}} (S_{i} - S_{e,t}) \cdot \Delta V_{k}$$

Table 2 shows the experimental data and the results from the jar test experiments.  $X_{BC}$  is the amount of bone char added to 1 litre of synthetic magadi solution and the  $\Delta TAL$  is the change in alkalinity between the treated and the untreated magadi solution. Table 3 shows the experimental data and the results from the bone char column experiment where synthetic magadi solution is treated. The dosage of bone char,  $X_{BC}$ , is calculated as the amount of bone char in the column divided by the amount of treated water at a selected effluent fluoride concentration corresponding to different filtration times. The average effluent fluoride concentration, Se.average, is calculated as the sum of the effluent concentrations, S<sub>e,t</sub>, in the different samples multiplied by the differential part volumes of treated solutions,  $\Delta V_k$ , divided



by the cumulative volume of treated solution, V<sub>t</sub>, at the sampling time:

$$S_{e,average} = \overline{S_e} = \frac{1}{V_t} \sum_{k=0}^{V_t} S_{e,t} \cdot \Delta V_k$$

It was investigated to what extent the addition of  $CaCl_2$  to magadi solution would reduce the fluoride contamination. The results of the measurements of fluoride and carbonates concentrations can be seen in Figure 4. The reductions in carbonate and bicarbonate alkalinity and fluoride concentration are plotted versus the amount of calcium added. It is concluded that the idea of using  $CaCl_2$  for removal of fluoride from highly alkaline solutions is not feasible. Even though the solubility product for fluorite (CaF<sub>2</sub>) is

exceeded,  $K_{\text{fluorite}}=10^{-10.57}$ at 25°C, the reduction in the concentration fluoride is minor compared to the reduction carbonate in aragonite alkalinity, i.e.  $(CaCO_3)$  is more likely to precipitate than fluorite.

#### Discussion

In Figure 2 it is observed that bone char is able to fluoride remove from alkaline highly waters (magadi solutions) without any major changes in the total alkalinity of the treated solution. However, focusing the initial few on bedvolumes of treated solution there seems to be a minor but significant removal of alkalinity in the bone char column. The alkalinity removed in case of natural and synthetic magadi solutions are equal to 308 meq and 125 meq, respectively. This removal is probably caused bv precipitation of CaCO<sub>3</sub>, as the bone char is known to contain varying amounts of free calcium. The same phenomenon is observed in the jar test experiments. The total alkalinity of the treated solution is lower than the initial total alkalinity. It is



versus the fluoride loading for column experiments in case of natural and synthetic magadi solutions containing 89 g magadi/L.



**FIGURE 4.** The reduction in carbonate and bicarbonate alkalinity and fluoride concentration versus the addition of calcium in precipitation experiments in case of synthetic magadi solution containing 89 g magadi/L.

seen from Table 2 that the higher the dosage of bone char, the higher is the reduction of the total alkalinity.

From Figure 3 it can be seen that the concentration of fluoride in the bone char increases linearly until the effluent fluoride concentration starts to increase, i.e. the break point. This takes place in the bone char columns after loading of 300 and 1200 mg F<sup>-</sup> from the natural and synthetic magadi solutions, respectively. Figure 3 shows that achieved bone char capacities in case of natural and synthetic magadi solutions are 2.8 and 4.6 mg F<sup>-</sup>/g BC as the inlet fluoride concentrations are 95 and 400 mg F/L, respectively.

Batch	X <sub>BC</sub>	Stirring	Se	f	faverage	ΔTAL
test no.	g/L	rpm	mg/L	mg/g	mg/g	meq/L
1	50	50	234	3.24	3.20	39.0
2	50	100	238	3.16		32.0
3	100	50	126	2.74	2.69	104
4	100	100	137	2.63		65.0

TABLE 3. Experimental data and results from jar test experiments in case of synthetic magadi solution containing 89 g magadi/L and 400 mg F/L.

**TABLE 4.** Experimental data and results from bone char column experiment in case of synthetic magadi solution containing 89 g magadi/L and 400 mg F/L. The figures in bold,  $X_{BC}$  should be compared with the same figures in table 2.

V	X <sub>BC</sub>	Se	f <sub>t</sub>	S <sub>e,average</sub>
1	g/L	mg/L	mg/g	mg/L
3.35	100	177	3.55	45.0
6.72	50	367	4.46	177

The results are in agreement with the sorption isotherms theory, where the maximum sorption capacity is higher, the higher the inlet concentration.

When using the bone char as a fluoride removal media in the jar test experiments, it has been necessary to add more than 100 g/L bone char to the synthetic magadi solution to obtain an acceptable residual fluoride concentration of less than 1 mg F/g magadi. An addition of 100 g BC per litre synthetic magadi solution resulted in a residual fluoride concentration of 1.4 mg/g magadi.

Tests were conducted to elucidate if the mixing of bone char and magadi solution is of any significance. Twenty five grams of bone char were added flasks containing 250 ml synthetic magadi solution and shaken for 0, 5, 10, 15, and 30 minutes, respectively. After 24 hours of contact time the residual fluoride concentrations were measured to be within the same range, 151-168 mg F/L. From these results it is concluded that when sufficient contact time is provided, the mixing is of minor importance.

When comparing the column and the batch test's possibilities for being utilised at household level in developing countries the column method has 2 main drawbacks. The operation of the columns is quite troublesome, as the flow must be controlled at a very low rate. Furthermore, the breakpoint has to be checked which is an almost impossible task at household level. On the same line the batch treatment results in approximately 20 % lower sorption capacity and hence a higher consumption of bone char, cf. Table 2 and 3. However, this drawback seems to be minor compared to the major advantage of simple operation, where no breakpoint needs to be checked. Compared with the column method the batch method is much more preferable for use in developing countries, especially at household level.

In defluoridation of drinking water small grain sizes have shown higher efficiency compared to larger grain sizes.<sup>12</sup> However, in this study not less than 30 % higher defluoridation efficiency is achieved in batch tests when using large grain sizes, the initial concentration of fluoride being 400 mg/L. The batch experiments showed that the fluoride removal is about 70 % when using bone char grains of 2.0-2.8 mm compared to

60 % for grains of 0.5-1.0 mm. Thus, it is important to avoid crushing the bone char into small grains when dealing with solutions containing high fluoride concentrations. This is quite opposite the recommendation in normal water defluoridation.

This study shows that defluoridation plants based on bone char for treatment of magadi solutions may be efficient both as batch systems and as column systems. When taking the drawbacks of the systems into consideration it is concluded that the batch system is superior to the column system at household level. The set-up of the batch system may be recommended as follows:

- About 100 mL of clean drinking water is transferred into a 200 mL container.
- A small amount of magadi, ca. 10 g, is added and the solution is stirred until complete dissolution apart from possible dirt.
- About 1.5 times as much of bone char (ca. 15 g) is added.
- The mix is stirred gently a few times and left overnight.
- The next day the purified magadi solution is decanted and then ready for use.

As the solution is durable larger batches can be prepared for use over a week or even a month.

Raw natural magadi, especially scooped magadi, may contain insoluble fractions: clay particles, organic matter and alike. The utilised settling over night was observed to result in a good separation of this dirt.

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## ECONOMICAL TECHNOLOGY OF FLUORIDE REMOVAL USING FISHBONE CHARCOAL COLUMNS AT DOMESTIC LEVEL

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SUMMARY: Columns containing 45 g fishbone charcoal were tested in laboratory column bed with a continuous feed of water containing 2.5-20 mg fluoride /L. The useful capacity of the medium was defined as the volume of treated water or the amount of fluoride removed before the breakthrough at concentrations of 1 mg/L. It was observed that influent water containing 5 mgF/L resulted in useful volumes of 9.5 to 3 L, depending on the loading utilised (about 0.3 - 1.7 mL/min/g). For a loading of 0.3 mL/min/g between 3 and 19 L of water could be treated to concentrations less than 1.0 mg/L, depending on the initial fluoride concentration utilised (2.5 - 20 mg/L). Useful removal capacities between 0.3 and 1.4 mg/g were obtained, the highest capacity being observed for lowest flow rate and highest initial fluoride concentration. Total prising of the process including purchase of fishbone, transportation, charring etc. indicates that the method may be workable and economical at individual domestic levels in seashore areas where fishbone is largely available.

**Key words:** Fluoride removal; Fishbone charcoal; Defluoridation columns; Domestic level; Economical defluoridation technology.

#### INTRODUCTION

Fluoride in drinking water can cause beneficial or detrimental effects depending on its concentration and the total amount ingested. The beneficial effect of fluoride in small quantities (up to an amount of 1.0 mg/L) lies in its capability to increase tooth resistance against dental caries, which is thus prevented. Fluoride acts by improving the acid resistance of the enamel reacting with it to form fluorapatite, which is less soluble than hydroxyapatite. Higher concentration of fluoride can cause dental or skeletal fluorosis.<sup>1</sup> Fluorosis (a disease, often escaping detection) tends to discolour teeth (feeling of having cavities all around, so need filling) and the affected teeth can turn brittle and break. The disease can attack neck, spine, pelvic and shoulder joints and small joints of hands and feet. In some parts of the world water with fluoride content less than 20 to 30 mg/L is not available. The Indian drinking water supply standards recommend an acceptable fluoride concentration of 1.0 mg/L. A concentration of 1.5 mg/L is allowable for potable waters.<sup>2</sup> There is thus, a dire need to develop some economical technology applicable at domestic levels to reduce the fluoride concentration from very high concentrations to acceptable concentrations in drinking water. The various defluoridating materials mainly used include animal bone and bone charcoal, synthetic tricalcium phosphate, activated carbon, activated alumina, synthetic ion exchangers, alum, lime, etc. A comprehensive review of the defluoridation methods as studied by various researchers<sup>1,3,4</sup> has been presented elsewhere. Bone charcoal prepared from animal bones has been successfully used in many full-scale installations for defluoridation of drinking water in Southern California.<sup>5</sup>

It will be shown later in the `Materials and Methods' section that fishbone can be a cheap material in fish cultivation areas. Relatively little research has been done on the subject of fluoride removal capacity of fishbone charcoal through column studies. The design of fluoride removal processes is, in many cases, largely empirical and based on past experience.<sup>6</sup>

In a continuous flow, fixed-bed column operation, the efficiency and system cost depend on the removal capacity of the media, i.e., the amount of solute adsorbed per gram of the

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adsorbent. This capacity is a function of several factors such as flow rate, column bed depth, initial solute concentration, pH, temperature, and desired quality of the treated water. Effects of these parameters have been studied and presented elsewhere.<sup>7</sup> In this paper, an attempt has been made to evolve a fixed bed column of fishbone charcoal usable at domestic levels to economically treat waters containing very high fluoride concentrations.

#### MATERIALS AND METHODS

Fishbone charcoal was prepared by carbonising the cleaned and pulverised fishbone in an electric furnace in a closed retort at 1000°C for 2.0 hours. The cooled material was then sieved to get the required size having geometrical mean diameter of 0.549 mm (0.355 – 0.850 mm size range). This size is close to the one used in field application in Southern California.<sup>5</sup> The material was thoroughly washed with distilled water, oven dried at 103°C, desiccated, and stored in air-tight containers. The specific surface area and density of the material were determined to be 85 m<sup>2</sup>/g (as per ISS<sup>8</sup>) and 1.8 g/cm<sup>3</sup> respectively. These properties, however, depend on the kind of bones and the scheme of charcoal preparation.

The fluoride test solutions of different initial fluoride concentrations ( $C_0$ ) were prepared by adding appropriate amounts of anhydrous sodium fluoride to tap water. The tap water used to identify field conditions had a pH of 7.9-8.1; total dissolved solids of 170 mg/L; total hardness (as CaCO<sub>3</sub>) of 112 mg/L; calcium (as Ca<sup>++</sup>) of 44 mg/L; chlorides (as CI<sup>-</sup>) of 10 mg/L; sulphate (as SO<sub>4</sub><sup>--</sup>) of 1.5 mg/L; alkalinity (in CaCO<sub>3</sub>) of 75 mg/L; and fluoride (as F<sup>-</sup>) of less than 0.10 mg/L. A possible interfering effects of competing ions including calcium (Ca<sup>++</sup>) present in the stated tap water, on fluoride adsorption was investigated through batch adsorption tests from trial jar tests, and the interference was found to be negligible and insignificant.<sup>7</sup>

Fixed bed column studies were conducted with 2.5 cm diameter glass column filled with fishbone charcoal. Four sets of column studies were performed. In each set, for a given initial fluoride concentration ( $C_0 = 2.5, 5.0, 10.0, \text{ or } 20.0 \text{ mg/L}$ ) the downward flow rates were varied and maintained at 15 mL/min (3.06 mL/min/cm<sup>2</sup>), 30 mL/min (6.12 mL/min/cm<sup>2</sup>), 50 mL/min (10.2 mL/min/cm<sup>2</sup>), and 75 mL/min (15.3 mL/min/cm<sup>2</sup>) corresponding to each of the various C values. Additional runs were also conducted with fishbone charcoal filled to a column bed depth (D) of 15 cm with (i) the initial fluoride concentration of the test solution, kept constant at 5 mg/L and the flow rates maintained at either 15 mL/min, 25 mL/min, 50 mL/min or 75 mL/min, and (ii) a constant flow of 15 mL/min maintained and the initial fluoride concentration of the test solution kept at 2.5 mg/L, 6.0 mg/L, 10.0 mg/L, or 20 mg/L.

The fishbone charcoal was filled in glass columns to the required depths in such a way that the bulk density of the filled adsorbent material was 0.61 g/cm<sup>2</sup>. For example, a 15 cm column bed depth of fishbone charcoal weighed 45 g such that  $(45/(\pi/4 \cdot 2.5^2 \cdot 15) = 0.61 \text{ mg/cm}^2)$ . The top of the column was connected to a constant head-maintaining tank, which was charged by an overhead tank containing the feed solution.

In each of these tests, the effluent concentration was monitored at different times and samples were analysed for fluoride concentration using the specific ion electrode (Orion Ion Analyser, Model 901).

• 15

▲ 25

△ 50

75

40

Flow

mL/min

30

Volume of treated solution, L

5.316x + 10.82

The various runs were terminated when the effluent fluoride concentration at the bottom of the column beds exceeded 1 mg/L (the permissible concentration, designated as the break-through concentration). The volume of the effluent treated prior to the breakconcentration, through was designated as the `useful (or effective) treated effluent volume.

## RESULTS

The experimental observations are plotted in Figures 1 to 4.

The approximate cost of 167 kg of fishbone from Goa (India) seashore (including collection, cleaning. transport. manual pulverisation, etc.) was Rs. 167.00. The approximate cost of carbonisation is an electric furnace (36 kW for 2 hours, i.e., 72 units) including overheads and labour costs, was Rs. 216.00. About 40% material weight was lost during carbonisation. Thus, about 100 kg (167x0.6) of the fishbone charcoal cost Rs. 383.00 (167 216), i.e., approximately Rs. 4.00 (US \$ 0.20) per kg cost of the fishbone charcoal.

## DISCUSSION

**Breakthrough vs. flow rate.** The breakthrough curves for the initial fluoride concentration ( $C_0$ ) of 5.00 mg/L corresponding to the various influent flow rates (Q = 15, 25, 50 and 75 mL/min) are depicted in

cm, absorbent weight W=45 g, initial influent fluoride concentration  $C_0=5$  mg/L,and pH= 8.0 (tap water base).

10

20

FIGURE 1. Break through curves corresponding

to different flows with column bed depth D=15

Treated water volume, L

Effluent fluoride conc., mg/L

6

5

4

3

2

1

0

10.0

8.0

6.0

0



Figure 1. The retention time or hydraulic residence time corresponding to the flow rates (Q) of 15, 25, 50 and 75 mL/min are 4.91, 2.95, 1.47 and 0.98 min, respectively.

The volume of treated effluent (V) at the different flow rates at the chosen effluent fluoride concentration at breakpoint,  $C_e$  of 1.0 mg/L is read as 9.45, 8.25, 3.27 and 2.87 L corresponding to the flow rates of 15, 25, 50 and 75 mL/min, respectively.

The variation of treated effluent volume (V) with respect to the flow rate per unit weight of adsorbent (Q<sub>w</sub>) is shown in Figure 2. The volume of treated effluent decreases with an increase in flow rate per gram of the adsorbent. This is because, at higher flow rates, the decreased available retention time (or hydraulic residence time) of solution in the column may be inadequate. The optimum time of retention required for completing the sorption reaction between the solute and the media, which is likely to result in longer operation period until the breakthrough at  $C_e = 1.0 \text{ mg/L}$  takes place, may require lower flow rates.

Mass balance calculations were done to determine the amount of fluoride removed (F) at different flow rates (Figure 1) till the breakthrough concentration point  $(C_e = 1.0 \text{ mg/L})$ . The effluent concentration the up to breakthrough point varied from 0 to 1.0 mg/L as seen in Figure 1. During the breakthrough time, an average breakthrough effluent fluoride concentration (Ce-av) of 0.5 mg/L was assumed for the time interval in which C varied from 0 to 1.0 mg/L instead of using the procedure of integration breakthrough of curve or considering small time intervals. The amount of fluoride removed



**FIGURE 3.** Breakthrough curves (C vs L) corresponding to different initial fluoride concentrations ( $C_0$ ) with column bed depth (D) of 15 cm, absorbent weight (W) of 45 g, flow (Q) of 15 mL/min, and pH of 8.0 (tap water base).



for flow rates of 15, 25, 50, and 75 mL/min is estimated to be 42.5 mg, 37.1 mg, 14.7 mg, and 12.8 mg, respectively. These correspond to removal capacities of approximately 1, 0.8, 0.4 and 0.3 mg/g respectively. The higher the flow rate the lesser the fluoride removal capacity, due to shorter available contact time (or hydraulic residence time).

**Breakthrough vs. initial Conc..** Breakthrough curves at the different initial fluoride concentration (C<sub>0</sub>) are shown in Figure 3. These curves are obtained corresponding to a constant flow rate of 15 mL/min. From Figure 3 the values of the useful volume (V) of the treated effluents corresponding to  $C_e = 1.00 \text{ mg/L}$  were estimated as 18.8, 8.1, 5.4

and 3.24 L corresponding to  $C_0$  values of 2.5, 6.0, 10.0 and 20.0 mg/L, respectively. The variation of volume of the treated effluent versus the initial fluoride concentration ( $C_0$ ) is shown in Figure 4. At the chosen breakpoint concentration of  $C_e = 1.0$  mg/L, the useful volume (V) of the treated effluent decreases with an increase in  $C_0$  values. This is due to the fact that for a given flow rate and quantity of the adsorbent, the adsorption or exchange sites of the adsorbent are exhausted earlier when a higher initial fluoride concentration influent is encountered. Therefore, the operation period until the breakthrough point is less. Mass balance calculations were carried out, as before, to determine the amount of fluoride removed at different initial fluoride concentrations (Figure 3). The amount of removed fluoride (F) at  $C_0$  of 2.5, 6.0, 10.0 and 20.0 mg/L was 28.2 mg, 44.6 mg, 51.30 mg, and 63.18 mg, respectively. These correspond to useful removal capacities of about 0.8, 1, 1.1 and 1.4 mg/g respectively. The breakthrough times are shorter for higher initial fluoride concentrations, but the amount of fluoride removal capacity of the medium, increases with  $C_0$ . This is probably due to higher concentration gradients at higher  $C_0$  values.

**Practical applications.** Fish bones can be considered to be a comparatively cheaper and effective material for the preparation of fishbone charcoal for defluoridation of drinking water as equally feasible for isolated communities of small sizes as well as at domestic level in the form of home filters.

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## BIODEFLUORIDATION OF FLUORIDE CONTAINING WATER BY A FUNGAL BIOSORBENT

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SUMMARY: Alkali-extracted mycelial biomass (biosorbent) from Aspergillus niger was effective in sequestering fluoride from fluoridecontaining waters. When kept in contact overnight, the biosorbent, used at a level of 3%, removed fluoride to an extent of 45-50% from water containing 5 mg F/L. The kinetics of fluoride removal exhibited a rapid phase of binding for a period of 1.0 hour and a slower phase of binding during the subsequent period. The extent of defluoridation was dependent on the initial pH of fluoride-containing water and decreased with increasing pH. The biosorbent could bind fluoride for three successive treatments tested. The potential use of this biosorbent for biodefluoridation is being explored.

Key words: Biodefluoridation; Fungal biosorbents; Aspergillus niger.

#### INTRODUCTION

Excessive fluoride in drinking water causes dental and skeletal fluorosis, which is encountered in endemic proportions in several parts of the world. Due to the unavailability of effective therapeutic measures, defluoridation of drinking water appears to be the best method for combating the disease. Several methods, using a variety of materials, have been suggested from time to time. These methods are basically of two types: those based on an exchange process or adsorption, and those based on addition of chemicals to the water being treated.<sup>1</sup> Most of these methods have one or more short-comings with regard to the defluoridation capacity, cost effectiveness, operation at community level and quality of treated water. In view of this, search for more suitable materials and methods are still in progress.

Attempts to harness microbial biomass for the purification of polluted water from industries appear promising.<sup>2-5</sup> Live and inactivated fungal biomass exhibits interesting metal binding properties due to the presence of functional groups like amino, amide, carboxyl, hydroxyl, sulfhydryl etc. in the cell walls. The biosorption involves direct exchange of toxic heavy metal ions with the resident Ca<sup>++</sup>/Mg<sup>++</sup> ions of the biosorbent.<sup>6</sup> Fluoride binding by the fungal biosorbents appears possible considering their Ca<sup>++</sup>/Mg<sup>++</sup> content and the known affinity of Ca<sup>++</sup> and Mg<sup>++</sup> for fluoride ions. The following experiments demonstrate the ability of fungal biosorbent to bind fluoride which we intend to term as "biodefluoridation".

#### MATERIALS AND METHODS

The Aspergillus niger strain used in these experiments is a laboratory isolate<sup>7</sup> which exhibits resistance to toxic levels of Cd<sup>++</sup> and accumulates Ca<sup>++</sup> and Mg<sup>++</sup>. The preparation of alkali extracted biomass of A.niger (biosorbent) was according to Akthar *et al.*<sup>7</sup> Finely powdered biosorbent was first washed with glass-distilled water and then

suspended in fluoride-containing waters. After centrifugation, the fluoride content of the supernatants was determined.

Fluoride was measured using fluoride ion specific electrode (Orion model 94-09 fluoride electrode). The total ionic strength adjustment buffer (TISAB-I) contained 58 g NaCl, 300 mg trisodium citrate and 58 ml gl.acetic acid in 1 litre adjusted to pH 5.0-5.5 with 5 N NaOH. For the experiments, the following methods were used:

**Fluoride binding by the biosorbent.** Fluoride containing glass-distilled water was used for IA-IVA while fluoridated tap water was used for IB-IVB. The Biosorbent (3 g) was suspended in 100 mL glass-distilled water or tap water and pH adjusted to 7.8, stirred and centrifuged. The supernatant was discarded and the process repeated two more times. The final pellet was suspended in 100 mL of fluoride containing water. After specified period of exposure, the suspension was centrifuged and the fluoride content of the supernatant determined. For each subsequent transfer, the pellet was suspended again in fluoride containing water and after specified periods of exposure, the suspension was centrifuged and the fluoride content of the supernatant determined.

**Time course of fluoride binding by the biosorbent.** The biosorbent was washed thrice with glass-distilled water or tap water as described in the previous experiments (Table 1) and suspended in 100 mL of fluoride containing glass-distilled water or tap water. Suitable aliquots were taken at different time intervals and the fluoride content determined. The starting fluoride content of fluoridated distilled water was 5.13 mg/L and that of fluoridated tap water was 5.32 mg/L.

**Effect of pH on fluoride binding by the biosorbent.** The biosorbent (3 g) was suspended in 100 mL of distilled water or tap water, the pH adjusted to the specified values and fluoride stock solution (1900 mg/L) added to get the final fluoride concentration. After two hours at room temperature, the suspensions were centrifuged and the fluoride content of the supernatants determined. IA to IIIA are with glass-distilled water and IB to IIIB are with tap water.

Effect of biosorbent concentration on fluoride binding. The indicated amounts of biosorbent were first suspended in 100 mL glass-distilled water, pH adjusted to 7.0, centrifuged and the supernatant discarded. The pellet was then suspended rapidly in 50 mL of fluoride solution (5.13 mg F/L in glass-distilled water), allowed to stay for 2 hours at room temperature, centrifuged and the fluoride content of the supernatant determined.

<b>TABLE 1.</b> Fluoride binding by the sorbent							
Subsequent	Duration of	Fluoride	Capacity	Binding			
No. of transfers	exposure	in water	(µg F /g	(%)			
	(hours)	(mg/L)	biosorbent)				
I A	18.0	4.94	82.1	50.0			
II A	18.0	5.13	75.8	44.4			
III A	18.0	5.13	67.8	39.8			
IV A	90.0	5.13	87.6	51.4			
I B	18.0	5.32	90.3	50.9			
II B	18.0	5.32	58.6	33.0			
III B	18.0	5.32	38.0	21.4			
IV B	90.0	5.32	103.0	58.0			

## RESULTS

**Fluoride binding by the biosorbent.** The fluoride binding ability of the biosorbent with successive transfers is shown in Table 1. Overnight exposure for a period of 18 hours was considered convenient for application at community level. With successive transfers the per cent binding diminished from 50 % to 40 % in glass-distilled water and 51 % to 21 % in tap water. The duration of exposure for the last transfer (IV A & IVB) was 90 hours which resulted in higher binding due to longer period of exposure. **Time course of fluoride binding by the biosorbent.** The kinetics of fluoride binding by the biosorbent from the medium exhibited a rapid phase in which 31 % binding was complete within one hour in glass-distilled water and 24 % in tap water. Subsequent binding was slow and much less as shown in Table 2.

Effect of pH on fluoride binding by biosorbent. The effect of initial pH on fluoride uptake by the fungal biosorbent is shown in Table 3. Over the pH range tested (pH 5.5 to pH 10.0) the extent of binding decreased with increase in the initial pH. The decrease in binding was from 52 % to 17 % in glass-distilled water and from 45 % to 26 % in tap water.

Effect of biosorbent concentration on fluoride binding. The extent of fluoride binding was linearly related to the amount of biosorbent over a range of 0.25 g to 4.00 g, cf. Table 4.

TABLE2.biosorbent	Time course	of fluorid	e binding	by the
Duration of	Capa	city	Bind	ing
Exposure	(µg F⁻/g bio	osorbent)	(%	)
(hours)	Distil.	Тар	Distil.	Tap
1.0	52.2	42.6	30.6	24.1
2.0	52.2	50.3	30.6	28.4
4.0	52.2	55.2	30.6	31.2
7.0	57.0	62.2	33.5	35.1
12.0	57.0	62.2	33.5	35.1
24.0	65.8	76.2	38.6	43.0

TABLE	3.	Effect	of	pН	on	fluoride	binding	by	the
biosorbe	nt								

No.	F <sup>-</sup> conc. (mg/L)	Adjusted initial pH	Binding (%)	Capacity (µg F <sup>-</sup> /g biosorbent)
IA	5.01	5.54	51.9	86.7
IIA	4.75	8.06	38.0	59.9
IIIA	5.0	10.00	17.0	28.4
IB	4.5	5.62	45.4	68.2
IIB	4.9	8.08	39.7	64.9
IIIB	5.2	10.10	26.3	45.5

## DISCUSSION

There has been considerable research activity in recent years on the use of biosorbents (bacterial, algal and fungal) for scavenging heavy metal cations from polluted waters. Reports regarding the use of biosorbents for removal of hazardous anions like fluorides, nitrates etc. are not available. The results reported here clearly demonstrate the ability of alkali-treated Aspergillus niger to bind fluoride (biodefluoridation). The biodefluoridation is rapid and diminishes with repeated cycles. Mass dependence and pH influence further substantiate the authenticity of the observed biodefluoridation by the biosorbent. Lower binding at higher pH could be due to competition between F and OH for fluoride binding sites.

The mechanism of fluoride binding by the biosorbent is not clear. The organic matrix of the biosorbent contains  $Ca^{++}$  and  $Mg^{++}$  ions. It is possible that the  $Ca^{++}$  and  $Mg^{++}$  ions are responsible for binding fluoride. The primary amino groups could also get protonated at

acidic pН and bind fluoride. Further work is needed on the mechanism of binding which would enable genetic and chemical manipulation of the fungal and other biosorbents for large scale defluoridation of water supplies.

binding.				
No.	Biosorbent (g/50 mL)	F <sup>-</sup> conc. (mg/L)	Removal	Binding
1	0.25	5.13	20.25	7.89
2	0.50	5.13	33.25	12.96
3	1.00	5.13	52.25	20.37
4	1.50	5.13	76.95	30.00
5	2.00	5.13	87.60	34.15
6	4 00	5 13	171.0	66.66

**TABLE 4.** Effect of biosorbent concentration on fluoride

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# Session IV

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## DEVELOPMENT OF THE CONTACT PRECIPITATION METHOD FOR APPROPRIATE DEFLUORIDATION OF WATER

#### E Dahi<sup>\*</sup>

SUMMARY: This paper describes the development of defluoridation of water by contact precipitation, where fluoride water is mixed with calcium and phosphate and brought in contact with bone char which is already saturated with fluoride. The process is studied in jar test, in manually stirred buckets and in continuously fed columns. Furthermore in a "fill, mix and filter" column both in laboratory and in a pilot plant for a period of two years. An appropriate setup is then designed and constructed using low-cost, corrosion-resistant and robust locally available materials. The removal efficiencies obtained in the batch systems were relatively low especially when manually stirred. The fill, mix and filter technique demonstrated surprisingly high removal efficiencies, 95-98 %, without any sign of break through or saturation, at dosage levels corresponding to calcium/phosphate/fluoride weigh ratio of 8.5/10.8/1. It is discussed that the main processes behind may be a crystal growth or a catalysed precipitation of fluorapatite and/or calcium fluoride, as the components are brought in close contact with fluoride saturated bone char.

**Key words:** Defluoridation; Drinking water; Contact precipitation; Bone char; Pilot plant; Tanzania; Removal mechanisms; Fill, mix and filter technique; Apatite; Crystal growth.

#### INTRODUCTION

Millions of people, mainly in developing countries, are known to suffer from fluorosis due to the high fluoride concentrations in their drinking water,<sup>1</sup> and numerous methods and media, including low cost ones, are known to be able to remove fluoride from water.<sup>2, 3</sup> Yet none of these methods has so far been reported to be carried out successfully as a routine in any developing country.<sup>4</sup> This oddity is probably attributed to the fact that the available methods suffer from one or more serious drawback(s), e.g. low removal efficiency, low removal capacity, complex preparation of medium, complex procedures of regeneration, the need of monitoring in order to avoid the break through and deteriorated treated water quality. It is worth mentioning that the most promising methods of today, e.g. the bone char sorption, the activated alumina ion exchange and the alum/lime co-precipitation, have all been well known already in the thirties.<sup>5, 6</sup>

This paper describes the scientific background and the development of a new method, ascribed as contact precipitation.

#### MATERIALS AND METHODS

**Fluoride Measurements.** The fluoride concentrations were measured using a Metrohm fluoride electrode and a Metrohm Ag/AgCl reference electrode with a sleeve type diaphragm connected to a Metrohm potentiometer (691 pH Meter). An aliquot of 10.0 mL of the sample solutions was mixed with 10.0 mL CDTA-tisab and the fluoride concentration was measured according to the Standard Methods.<sup>7</sup>

**Bone char preparation.** The bone material was delivered from the Danish bone meal factory DAKA, where the bone material has been boiled, washed, dried and crushed into small particles of grain size < 4.0 mm. The bone material was pyrolysed in a



programmable ceramic oven (Scandia Oven, Type SK 355) in a covered steel container, where the access of atmospheric oxygen was restricted. The temperature was raised at a rate of 4°C/minute from room temperature to 600°C and then kept constant for 4 hours. The charred bone material was allowed to cool down to room temperature before the oven was opened. Bone char of 2.0-2.8 mm grain size was sorted out using a test sieve shaker (Endecotts EFL2 mk3).

**Saturated bone char.** For the jar test, the bucket and the column experiments saturated bone char was prepared by packing the fresh bone char in a column of 50 cm height and circulating of 13 mg/L fluoride water. The sorption was continued until the effluent concentration was as high as the influent concentration, i. e. for a period of one week.

For the field testing large quantity of bone char was saturated by suspending the fresh bone char medium in a solution containing 1 g fluoride per litre of drinking water. The mix was stirred regularly for a period of 2 weeks. At the end the water had a fluoride content of about 18 mg/L. At this point of saturation the bone char had absorbed 2.5 mg F/g bone char. The bone char was transferred directly to the village school plant contact chamber and brought to equilibrium with the local drinking water containing 8.2 - 13 mg/L fluoride.

**Jar test setup.** One litre plastic beakers each containing 1 L Danish municipality water enriched with fluoride to contain 13 mg/L fluoride were stirred in the Jar test apparatus (Phipps & Birds Stirrer 7790-402) at a stirring frequency of 140 revolutions per minute. Different media were added at time nil with and without chemicals, i.e. calcium chloride and calcium monohydrogen phosphate. The level of dosage being 159 mg Ca/L and 226 mg PO<sub>4</sub>/L. Six batches were added respectively fluoride saturated bone char, marble, quarts sand, unsaturated (fresh) bone char and saturated bone char. All media were added at same volume dosage of 100 ml/L and of same grain size, 2.0-2.8 mm. In parallel one batch was left as a blank and stirred separately without any addition. After 2 hours of stirring samples were taken, filtered through a

 $0.45 \ \mu m$  Minisart GF filters before determination of the residual fluoride concentrations.

Bucket setup. Different volumes of natural piped water containing 15 mg F/L were poured into 20 L plastic buckets, as normally used in the households in the Arusha region of Tanzania. The buckets were supplied with taps at a level of 5 cm above the bottom in order to allow decanting of medium. for Α wooden device was used for mixing, as illustrated in Figure 1. Saturated bone char was added to the water, grain size 2.0 - 2.8 mm, 83 g/L. Then at time nil calcium chloride and calcium monohydrogen phosphate were added, the dosage level being 237 mg Ca/L and 337 mg PO<sub>4</sub>/L. The suspensions were stirred, about 100 RPM, manually for 3 minutes every



15 minutes. The residual fluoride concentrations were measured vs. time as mentioned above.

**Column setup.** Glass columns 1.9 cm in diameter and 50 cm in length were packed using 118 g of bone char, fluoride saturated as described above. The bone char had grain size 2.0-2.8 mm, porosity 0.65 and bulk density 0.83 g/mL. The columns were loaded with fluoride water containing 16 mgF/L at tow different flow rates, 0.19 and 0.095 L/h corresponding to flow velocities of 0.67 and 0.34 m/h and a hydraulic retention time of  $\frac{3}{4}$  and  $\frac{1}{2}$  hours respectively. One column was used as a reference without additions of chemicals in order to account for the residual sorption (or desorption) capacity of the saturated bone char. Two columns were loaded with the same water after addition of 237 mg Ca/L and 337 mg PO<sub>4</sub>/L. The flow was controlled by a peristaltic pump, which however was only run during working hours, 8 hours per day.

Fill and filter contact column. A combined raw water and filter bed column was set up for testing the significance of declining rate contact filtration in a simple "fill and contact filter" system, Figure 3. The column was packed with 20 cm gravel in the bottom, then with 3.28 kg of fluoride saturated bone char of grain size 1-2 mm. Eleven litres of water containing 20 mg/L fluoride were added each day to the compartment above the bone char, along with 470 mg/L anhydrous calcium chloride and 1100 mg/L calcium monohydrogen phosphate dihydrate, corresponding to dosage of 169 mg Ca/L and 415 g PO<sub>4</sub>/L. The water was allowed to flow slowly at declining rate until the raw water compartment of 11 L was emptied. In most cases the filtration time was between 10 and 20 minutes.



**School pilot plant.** A plant was sat up at the Ngurdoto Primary School, Arusha Region, Tanzania, to cover the need for drinking water of about 500 peoples during the day hours of the weekdays. The plant, Figure 4, consisted of one column 0.32 m in inner diameter and 1.4 m height. The column was packed with 20 L gravel, 0.25 m height and grain size 5-10 mm, and 18.7 kg of fluoride saturated bone char, 0.28 m in height and grain size 1-4 mm. The upper part of the column, 1.1 m in height, equal 100 L, was left as a raw water chamber in which the chemicals were added daily and mixed with the raw water. After mixing the water was allowed to flow at a declining slow rate through the saturated bone char compartment to a large clean water tank. This tank was supplied with a pipe and a tape at the schoolyard for convenient water intake by the peoples and the teachers.

One senior school pupil was employed as a caretaker and operated the plant as follows: The caretaker arranges for fetching the water in buckets every morning. Then starts closing the flow control valve completely. One and half litres of each of the two stock solutions containing Ca and  $PO_4$  are pumped to the raw water column after mixing with the raw water from the first two buckets. The remaining water is then



pumped to fill the raw water column, in total about seven buckets. As raw water is falling into the raw water column, the supernatant water is completely mixed. The flow control valve is then opened, but only to allow slow flow through the contact bed, the average filtration velocity not exceeding 1 m/h. A water sample is taken by the plant keeper for testing. This procedure is carried out once a day, apart from holidays. However, in between, the procedure is omitted or carried out twice in the same day in order to meet the demand for drinking water. The design and operation and maintenance procedures are described elsewhere in more details.<sup>8</sup>

**Stock Solutions.** The caretaker prepares the two stock solutions once every  $10^{\text{th}}$  operation day. Two special measuring cups are used for volumetric portioning of the chemicals, respectively 300 g calcium chloride (311 mL CC) and 150 g Sodiumdihydrogenphosphate (142 mL MSP). The chemicals used are both from the Swedish Kemira LTD. CC is fabricated as technical grade flakes containing 77-80 % calcium chloride MSP is fabricated as Bolifor Granular containing 24 % P and 20 % Na. The dosage used corresponds to 85 mg Ca/L and 110 mg PO<sub>4</sub>/L. The chemicals are dissolved separately, each in 15 L defluoridated water. The stock solutions containers along with the respective chemical bags and the measuring cups and cylinders are coloured respectively in red and green in order to minimise the risk of failure dosage.

**Appropriate design.** After successfully testing of the Ngurdoto pilot plant for a period of 2 years, a more appropriate design was developed and constructed in the Arusha region, using corrosion resistant, robust, low cost and locally available materials. The plant is used for demonstration purposes, Figure 5, the dimensions and dosage are given in Table 1.

5.								
Raw water chamber:			Contact bed:		Clean water reservoir:			
Width	cm	50	Diameter	cm 35	Width	cm	120	
Breadth	cm	80	Height	cm 42	Breadth	cm	80	
Height	cm	75	Volume	L 40	Height	cm	83	
Volume	L	300			Volume	L	800	
Bone Char			Calciumchloridede	hydrate	Sodiummonohydro	ogenphosj	ohate	
Weight	kg	34	Bulk density	kg/L 1.06	Bulk Density	kg/L	0.96	
Volume	L	40	Stock solution	g/L 20	Stock solution	g/L	10	
Grain size	mm	1-4	Dosage mgCa/	mgF 8	Dosage mg	Ca/mgF	10	
Porosity	-	0.65	mg chemical/L	15	mg che	emical/L	30	

**TABLE 1.** Design parameters of the contact precipitation process as shown in figure 5.

**TABLE 2**. Residual fluoride concentration in jar test, where water containing 13 mg F/L is added different media with and without dosage of calcium and phosphate. Dosage of medium 100 mL/L. Contact time is 2 hours, stirring 140 RPM.

Dooug	booldge of mediant free mere. Contact and is E hours, stiming 140 fit m.								
Batch	Medium	CaCl <sub>2</sub> ·2H <sub>2</sub> O <sup>*</sup>	CaHPO <sub>4</sub> ·2H <sub>2</sub> O	Residual F <sup>-</sup> , mg/L					
0	Not added	Not added	Not added	13.0					
1	Not added	159 mgCa/L	226 mgPO <sub>4</sub> /L	12.8					
2	Marble	159 mgCa/L	226 mgPO <sub>4</sub> /L	13.0					
3	Quarts sand	159 mgCa/L	226 mgPO <sub>4</sub> /L	12.2					
4	Fresh bone char	159 mgCa/L	226 mgPO <sub>4</sub> /L	0.3					
5	Saturated bone char	Not added	Not added	14.1					
6	Saturated bone char	159 mgCa/L	226 mgPO <sub>4</sub> /L	3.2					

\* The dosage of calcium is calculated as the sum of calcium originated from calcium chloride + the calcium originated from calcium monohydrogen phosphate.

#### RESULTS

Jar test experiments. The experimental results of the jar test are shown in Table 2. It is seen that fluoride is not removed if only saturated bone char is added to the water. It is also seen that the fluoride removal is calcium negligible in case and phosphate are added to fluoride water alone or along with marble or sand. Similarly other media like charcoal, bone ash were tested and found indifferent with respect to the fluoride removal. Only in case calcium and phosphate are added to fluoride water that contain saturated bone char the removal process seem to take place to a significant extent, 75 %. The removal is thus moderate compared to





defluoridation by means of fresh bone char, which at a dosage level of 83 g /L resulted in 98 % removal after 2 hours of contact time.

Bucket experiments. The significance of batch volume on the fluoride removal rate is shown in Figure 6. While the 15 and the 7.5 L batch experiments are carried out in the bucket setup, the 1 L batch experiment is carried out in the Jar test apparatus. The bucket technique resulted, after mixing time of 3 hours, 57 % removal, i.e. significant but far from sufficient. The removal was improved, to 80 and 90 %, by reducing the volume of water without any changes in the dosage or the amount of saturated bone char added.

Continuously fed column. The fluoride removal in the continuously fed saturated bone char columns is shown in figure 7. It is seen that what is supposed to be a saturated column may still be able to remove fluoride. The removal efficiency is 24 %. In the chemical added columns the removal efficiencies are much higher, i.e. 82 and 84 %. It has been demonstrated that the efficiency could be improved slightly by increasing the contact time in the column from 30 minutes to one hour by decreasing the flow rate.

Fill, mix and filter column. Figure 7 illustrates the fluoride removal obtained in the fill, mix and filter column. The fluoride removal is shown to be high, 83 % in the first filtration cycle, improving to 93.5 % in the second and to more than 97 % from the  $6^{th}$  cycle and hereafter. If the result from the first day is neglected, the removal, on an average, is 98 %. Furthermore, at the time where the experiment was brought to an end, after treatment of 60 bed volumes of water, there was no sign of breakthrough. Rather, the removal seems to improve as the filter gets mature. Due to chemical addition pH was increased from about 7 to about 8 in the raw water. However, pH was 7.29 in the treated water. Similarly, the water alkalinity was 5.3 me/L in the raw water chemical mix, but falling down to 3.3 me/L in the treated water. The calcium content was 96 mg/L on an average in the treated water, and that most of the added calcium is retained in the filter.

**Long term testing in pilot plant.** The plant operation was initiated on 6<sup>th</sup> November 1995 and terminated in October 1997. During this period one raw water column of water was treated every day, apart from holidays.



Thus the total effective operation period was 586 days. The raw water contained between 8.2 and 13 mg F/L, subject to seasonal fluctuation. The fluoride removal was as high as 92 % in the first filter operation day, improving to 94 % in the second day and then stabilising at about 95-98 %. At the time where the experiment was brought to an end, after treatment of 51.8 m<sup>3</sup> of water, there was no sign of breakthrough. Rather, the removal seemed to improve as the filter gets mature. Only a slightly increase in the filter resistance was observed. During long holidays the plant was standing idle, but this did not affect the organoleptic or microbial quality of the water.

#### DISCUSSION

The results obtained from the jar test experiments show clearly that the fresh bone char process may be of extremely high efficiency in batch systems, but this would at the expense the capacity. Table 2 shows that fresh bone char is able to remove 98 % of the initial fluoride concentration of 13 mg/L, but at a capacity level as low as 0.15 mg F/g. The fluoride removal in the contact precipitation process is 75 %, i.e. significant but insufficient.

The jar test results demonstrate that saturated bone char can not, by it self, remove fluoride from water. Furthermore, addition of calcium and phosphate to fluoride water, is not able to remove fluoride from the water unless the water and the chemical mix is brought in contact with fluoride saturated bone char. This catalysis effect of the saturated bone char seems to be unique compared to other media like sand, charcoal, marble or bone ash.

The bucket experiments demonstrate that the removal is not only dependent on the dosage of chemical and the amount of the saturated medium available, but also on the mixing that facilitate the contact between what is assumed to be reagent and the catalysis medium. Apparently this is another limitation of the contact precipitation in batch. Moderate intermittent mixing even for two hours is not sufficient to carry out the contact precipitation at optimum. Therefore it is concluded that the batch technique, especially if carried out in bucket, is not capable to facilitate efficient contact precipitation of the fluoride.

On the contrary to the batch system, the column system seems to facilitate the contact precipitation efficiently. The removal efficiencies obtained being on an average 82 % and 84 % in the continuously fed columns, 98 % in the fill, mix and filter lab column

and 95-98 % in the fill, mix and filter column of the school pilot plant. It is unclear whether the mix and flow patter can be held responsible for this difference in the results. It has to be mentioned that the phosphate compounds utilised, as stated under Materials and Methods, have been different.

Probably higher removal efficiency in the contact precipitation is obtained by using more soluble calcium and phosphate compounds. Sodium dihydrogen phosphate, or MSP, is preferred to be used at large scale due to high solubility, relative high phosphate content, relative neutrality and still reasonable price.

The mechanisms of the contact precipitation are yet not fully understood. One of the main questions, which was repeatedly discussed during the invention of the contact precipitation technique, is whether the observed removal reveals through a steady state condition, or through a non-steady state. In the first case the removal is likely to continue as long as the chemicals are added and the bottleneck in the process would probably be the decrease in filter permeability. The original permeability can then easily be re-established through rejuvenation, without the need to replace or to regenerate the filter medium. In second case the bone char would be moving towards a saturation of the bone char at a higher capacity level, and thus would need to be renewed or regenerated at final saturation. It is already well documented that bone char, which has been saturated with respect to a given concentration of fluoride in water, may prove to be able to uptake additional fluoride. Addition of calcium ions, drying, and resting, e.g. during night-time, are factors known to facilitate additional removal capacities.<sup>9, 10</sup>

The second question which had to be clarified was whether the observed removal is a kind of regeneration of the bone char as described in the literature,<sup>11, 12</sup> or it is a contact precipitation of the added chemical along with the fluoride from the water. According to Christoffersen et al.,<sup>12</sup> treatment of saturated bone char in a solution of calcium and phosphate regenerate the bone char by coating it with a fresh layer of hydroxyapatite.

The mechanism of regenerations through surface coating seems not to explain the contact precipitation satisfactorily. This is, among other things, because simultaneous addition of the chemicals is more efficient than pre-treatment of the bone char. It is clear that the fluoride/calcium mix is over-saturated with respect to calcium fluoride. However, the residual fluoride concentration in the treated water is brought down to a level where a simple precipitation of calcium fluoride can not explain the results.<sup>9</sup> It is also clear that the fluoride/calcium/phosphate mix is over-saturated with respect to fluorapatite. The results may thus be explained as a precipitation of fluorapatite, a process which is only taking place if catalysed in a saturated bone char bed, i.e. a kind of heterogeneous crystal growth phenomenon, where the adjacent water is rich with the precipitate components.

The calcium/phosphate/fluoride weigh ration is 10.5/15/1 in fluorapatite. But the added calcium/ added phosphate/removed fluoride ration has been 8.5/20.7/1 and 8.5/10.8/1 respectively in the lab and field long term testing, where the very high removal was obtained. Thus both calcium and (in the field testing) phosphate are added at lower levels than required for production of fluorapatite. If fluorapatite crystal growth is the mechanism behind contact precipitation it can not be the only one. Probably some of removal is taking place as a precipitation of calcium fluoride.

The contact precipitation method seems to be most promising, especially because it is environmentally safe, socially acceptable, economically affordable, highly efficient and reliable, without much need for skills and monitering.<sup>8</sup> However, the challenge in its implementation remains to be at least two sided: motivation of users and availability of the chemicals at user points.

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## HOUSEHOLD DEFLUORIDATION OF DRINKING WATER USING ACTIVATED ALUMINA

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SUMMARY: This paper deals with the development of a simple "Point of Use" domestic defluoridation unit (DDU) using indigenously manufactured activated alumina (AA) and its evaluation for adoption in rural areas in India. Different products of indigenously manufactured AAs were screened for fluoride uptake capacity. Using 3 kg of AA, around 500 L and 1500 L of safe water ( $F^{-} \leq 1.5 \text{ mg/L}$ ) could be produced when the raw water fluoride was 11 and 4 mg/L respectively (alkalinity 450 = mgCaCO<sub>3</sub>/L). Exhausted AA was regenerated with alkali and acid using the simple `Dip procedure'. After 30 cycles of regeneration, the decrease in capacity was only marginal. Effect of raw water constituents on fluoride uptake was studied in the laboratory using simulated as well as natural waters. Results indicated that there was a decrease in treated water volume as raw water alkalinity increased. Presence of 250 mg/L sulphate in raw water decreased the AA fluoride uptake capacity by 15 %. Nearly 400 DDUs have been distributed in tribal areas of Dungarpur district in Rajasthan, India for field evaluation. Reports received till now have been very encouraging.

**Key words:** Defluoridation; Activated Alumina; Alumina; Domestic Defluoridation Unit Regeneration.

#### INTRODUCTION

Fluorosis is a chronic menace affecting a large population world-wide. As per the last survey conducted by the Rajiv Gandhi National Drinking Water Mission (RGNDWM) in India, around 25 million people in 8100 villages using mainly ground water sources for culinary purposes, are exposed to health risks related to fluorosis. Taking the health effects into consideration, WHO has set the guidelines of 1.5 mg/L as the maximum permissible limit for fluoride in potable water.<sup>1</sup> Surface waters seldom contain fluoride beyond this level, whereas excess fluoride may be present in ground waters depending on the presence of fluoride rich minerals as well as hydrogeological conditions.

Defluoridation of drinking water has to be practiced, if ground water sources have fluoride levels beyond the recommended limit. Methods practiced for removal of excess fluoride can be divided broadly into two categories, namely precipitation and adsorption. Precipitation methods depend on the addition of chemicals to the raw water, which leads to the formation of fluoride precipitates or adsorption of fluoride onto the formed precipitate.<sup>2-6</sup> Lime and alum are used either individually or in combination. The Nalgonda Technique, as developed in India in 1975, involves the addition of alum and lime.<sup>5</sup> It has been used in domestic as well as at community levels in India.<sup>6</sup> Limitations of these methods are: the daily addition of chemicals; large volume of sludge production; and not effective with water sources having high total dissolved solids (TDS) and hardness.<sup>7</sup> In adsorption method, fluoride of raw water is retained on the adsorbent due to physical, chemical or ion exchange interactions. Although wide variety of adsorbents have been used for defluoridation,<sup>8-16</sup> activated alumina (AA) technology has been the method of choice in developed countries.<sup>12-13</sup> AA, Alcoa F-1 is used in most of these studies and many defluoridation

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plants based on this technology have been installed.<sup>17-18</sup> Laboratory studies on the fluoride uptake using indigenously manufactured AA, grade G-80 was reported by Sharma<sup>19</sup> and by Bulusu and Nawlakhe.<sup>20</sup>

The present study was undertaken to screen various grades of indigenously manufactured AA, development of a `point of use' defluoridation unit for domestic use and its evaluation for adoption in rural areas in India.

#### MATERIALS AND METHODS

Six grades of AA used for this study are as follows: G-87 and AD-101 supplied by Indian Petrochemicals Ltd (IPCL), Thane, India, pulverized and screened to an effective grain size of 0.3-0.85 mm. OA-25 (1) and OA-25 (2) supplied by M/S Oxide India Ltd., Durgapur, India. Particle size of the supplied material was 0.3-0.6 mm. AA-P (0.4-0.6 mm) supplied Pavan Industries. by Hyderabad, India. AA-B supplied by Indian Alumina Industry Hyderabad was spherical in shape with an average diameter of 1 mm. AA was washed with tap water to remove fine dust and then air dried before using for the defluoridation study.

**Domestic defluoridation unit (DDU).** The unit consisted of basically two chambers made of the material of choice such as stainless steel (SS), copolymer



plastic etc. Upper chamber was fitted with a microfilter as shown in Figure 1. This had an orifice at the bottom to give a flow rate of about 12 L/h. This chamber was charged with 3 kg of AA and the depth was 17 cm. Perforated plate made of SS was placed on the top of AA bed to facilitate uniform distribution of raw water containing fluoride onto the bed. Upper chamber was covered with the lid. Lower chamber was provided with a tap to withdraw the treated water. If desired, lower chamber can be replaced with an earthen pot in rural areas which not only lowers the initial cost but also keeps water cold in summer months.

Use of the filter is very simple. High fluoride bearing water is filled in the upper chamber. Water percolates through AA bed, where fluoride is adsorbed onto the adsorbent. Treated water, collected in the lower chamber, can be withdrawn as and when needed.

**Screening of AA products.** This study was conducted by taking 3 kg of different grades of AA in domestic defluoridation unit (DDU). The filter was operated similar

to its	expecte	ed use	in the
field.	Raw	water	was
filled	in	the	upper
chaml	ber i	intermi	ittently
(three	to four	times	a day)
and	the	f	luoride
conce	ntration	in trea	ated
water	was	perio	dically

TABLE 1. Tap water characteristics.							
Parameter	Unit	Value					
pH	-	7.8					
Alkalinity	mg CaCO <sub>3</sub> /L	400					
Total dissolved solids	mg/L	636					
Sulphate	mg/L	85					
Hardness	mg CaCO <sub>3</sub> /L	160					
Fluoride	mg/L	0.8					

determined. AA was taken as exhausted when the  $F^-$  concentration of the treated water exceeded 1.5 mg/L. Simulated raw water was prepared using IIT Kanpur tap water spiked with 10 mg/L fluoride. Tap water characteristics are presented in Table 1.

**Regeneration of AA.** The exhausted AA was regenerated using NaOH and  $H_2SO_4$  for successive reuse for defluoridation. AA was transferred into nylon mesh bag which was then dipped in 8 L of 0.25 N NaOH (10 g/L) for eight hours (preferably overnight). Occasional lifting and stirring facilitates better contact between the regenerant and AA. After draining NaOH from nylon bag, AA was washed with raw water and then transferred to a bucket containing 8 L of 0.4 N  $H_2SO_4$  for 4-6 hours. As in the case of alkali, lifting the bag and dipping again facilitates good contact of the regenerant with AA. Subsequent wash with raw water till the pH raises to 6-7 makes the AA ready for the next defluoridation cycle.

**Raw water characteristics vs. removal capacity.** This study was conducted in the laboratory using simulated as well as natural waters. Batch sorption tests were conducted for studying the effect of pH. Fluoride spiked distilled water (F = 20 mg/L) and pH adjusted to the desired value using either 0.1 N HCl or 0.1 N NaOH after adding 500 mg AA per 250 mL. The contents were agitated in a rotary shaker. After 4 hours of contact time, the sorbent was separated and fluoride concentration was estimated in the supernatant.

Effect of alkalinity, sulphate, hardness and initial fluoride concentration was evaluated using simulated as well as natural waters in DDU. Simulated water used was IIT Kanpur tap water spiked with 10 mg F<sup>-</sup> per litre and the desired quantity of other constituents. DDU was operated in similar way as described earlier. Volume of safe water produced was determined in each case.

**Field testing of the DDU.** Under a pilot scale fluorosis control project, in Dungarpur district in Rajasthan, 1945 users in 388 households in four villages are using Domestic Defluoridation Units (DDUs) since December, 1996. The distribution of DDUs was preceded by intensive awareness generation programme among community members about the ill health effects of high fluoride water and preventive measures, capacity building and baseline data collection including prevalence of dental and skeletal fluorosis and water quality of all drinking water sources. The project is being implemented by NGOs in close cooperation with district administration and active community participation.

The raw water characteristics of ground water in the four villages are as follows: Fluoride 2.5 -6.0 mg/L, Alkalinity 120 - 480 mg/L, TDS 390 - 1287 mg/L and pH 7.0-7.8. The performance of DDUs is being monitored on a monthly basis.

Activated Alumina. Fel! Bokmarket ar inte demnerat.											
Cuada	Grain size	Volu	Volume of water treated for different cycles, L								
Grade	( <b>mm</b> )	1	2	3	4	5	6	7	8	9	10
G-87 IPCl, Thane	0.30-0.85	500	475	550	475	450	490	460	400	400	525
AD-101 IPCL, Thane	0.30-0.85	850	725	700	640	460*	600	550	500	450	300
AA-B	1.0	500	330	350	475	410	450	450	400	290*	450
OA-25(1)	0.30-0.60	650	475	350	250	Operat	tion di	sconti	nued		
OA-25(2)	0.30-0.60	725	600	450	300	Operat	tion di	sconti	nued		
AA-P Operation	0.40-0.60	800	400	250*	600	450	350	350	300	300	250
*Operated on continuous flow mode using a pump.											

**TABLE 2.** Comparative performance of different indigenously manufactured Activated Alumina.Fel! Bokmärket är inte definierat.

**Fluoride Analysis.** Fluoride in water samples was estimated using SPANDS reagent<sup>21</sup> as well as orion portable ion meter and fluoride specific electrode following the procedure given in the manufacturer's manual. Alizarin visual method<sup>21</sup> was modified for its use in rural areas. Ten mL water sample was taken in a marked flat bottom 15 mL glass bottle. Ten to fifteen drops of commercially available fluoride reagent (supplied by Century PP Industries, Jodhpur, India) was added. Persistence of pink colour even after one hour, indicates that fluoride is within permissible levels. On the other hand, yellowish tinge indicates high fluoride. This is a qualitative test mainly aimed for use in rural areas.

# RESULTS

Screening AA products. Results of the screening study are presented in Exhausted Table 2. AA was regenerated and reused for ten defluoridation cycles to assess the regeneration and reuse efficiency which determines the field application potential of different grades of AA. Among the six grades used in the present study, OA - 25 (1) and OA-25 (2) exhibited a significant loss in treated water volume after each successive cycle. produced an average of G-87 450-500 L of safe water per cycle and exhibited minimum loss in fluoride uptake capacity. Although treated water volume was higher in



the first cycle with AD 101 as compared to G-87, there was a decrease during subsequent defluoridation cycles. Decrease of 15-20 % in treated water volume with AA-B as compared to G-87 can be attributed to its larger particle size of 1 mm. AA-P yielded 800 L of treated water in the first cycle, which was reduced to 400 L in second cycle itself. These results indicate, among the screened grades of indigenously available AA grades, G-87 and AD-101 both supplied by IPCL and AA-B supplied by AI, Hyderabad are better suited for field application. All further work was carried out with G-87.

**Water constituents vs. defluoridation capacity.** The fluoride uptake study in the pH range of 3-9 showed a decrease in the binding capacity of AA from pH 5 to 9 with no optimum pH, cf. Figure 2. When pH was raised from pH 5 to 8, fluoride uptake decreased by 12%.

Alkalinity increase from 400 to 600 and 800 mg/L decreased the treated water volume from 500 L to 430 L and 350 L respectively when simulated fluoride spiked water of 10 mg F/L was used as raw water. Results of sulphate and fluoride spiked simulated water showed that the presence of 250 mg/L sulphate in raw water reduces the fluoride uptake capacity by 15 %. Hardness up to 800 mg/L did not significantly affect the performance of DDU. Increase in fluoride concentration decreased the treated volume output, although, fluoride uptake capacity per kg AA increased (Table 3).

<b>TABLE 5.</b> Effect of Initial Fluoride Concentration on the Performance of DDU							
Initial Fluoride Concentration,	mg/L	3.8	6.8	10.8			
Vol. of Treated Water,	L	1550	870	500			
Fluoride Uptake Capacity	mg/g AA	1.55	1.74	1.83			

Natural waters having different concentrations of fluoride, alkalinity, sulphate and total dissolved solids were tested to determine the effect of these constituents on fluoride uptake capacity of AA. Characteristics of these waters and treated water volume are given in Table 4.

Results indicate, among characteristics of raw water considered in the present study, defluoridation cycle is significantly affected by the raw water alkalinity.

**TABLE 4.** Performance of DDU Treating Natural Waters from Rajasthan and Uttan Pradesh

			Vol.of						
Sit No	e Place of sample collection	F conc (mg/L)	alkalinity (mg/L CaCO <sub>3</sub> )	TDS (mg/L)	SO <sub>4</sub> <sup>2-</sup> (mg/L)	рН	Treated Water (L)		
1	Belsi, Unnao, UP	3.3	580	780	38	8.37	1300		
2	Nainpura, Jaipur, Rajasthan	3.7	1225	5880	1300	7.47	470		
3	Prahladpura, Jaipur, Rajasthan	4.4	1320	3080	168	7.17	540		
4	Padampura, Jaipur, Dungarpur	5.0	1100	1960	85	8.34	580		
5	Dungapura, Jaipur, Rajasthan	5.0	634	2100	-	7.50	590		
6	Padampura, Jaipur, Rajasthan	5.1	1100	2210	250	7.50	660		
7	Dungarpur, Rajasthan	6.0	350	1050	-	7.91	820		
8	Makkar, Unnao, UP	6.5	500	990	150	7.60	810		
9	Jaipur, Rajasthan	7.0	475	1200	24	8.09	460		
10	Maharajpura, Jaipur	8.3	775	3080	730	7.29	330		
11	Sivadaspura, Jaipur	9.7	1025	2310	16	7.36	300		
12	Sivadaspura, Jaipur	9.8	900	1610	37	8.46	310		
13	Padampura, Jaipur	17.0	990	1368	28	7.89	185		
- N	- Not Determined								

**Field Evaluation.** The key findings of the field performance monitoring of 388 DDUs till June 1997 are as follows:

- 1. Households are using DDU regularly and expressed satisfaction;
- 2. No major operational problems were reported excepting leakage from micro-filter joint due to improper tightening of wing nut;
- 4. It is feasible to carry out AA regeneration at the village level;
- 5. Households were using 25-30 litres of water per day. Many households complained of inadequate capacity (9 Litres) of upper and lower containers;
- 6. Out of 179 respondents in Rayana and Kankri villages, 178 reported increased appetite, 116 reported decrease in gas formation, 158 reported relief in joint pain, 14 reported decrease in thirst for water and 28 felt less tired. It is noteworthy that all households reported symptomatic relief; and
- 7. The per-capita cost is 4-6 U.S.\$ and cost per regeneration is 0.50 US\$ per DDU. The frequency of regeneration is once in 1.5 to 3 months depending on raw water characteristics.

#### DISCUSSION

Fluoride uptake capacity, acid resistively and attrition loss of AA are probably three important parameters which are to be considered for field application of this process. Fluoride uptake capacity after successive regeneration reflects the efficiency of regeneration as well as loss of efficiency due to acid treatment as it is one of the regenerants employed. Further, repeated reuse of AA without much decrease in fluoride uptake capacity also indicates that attrition loss is not significant.

Alcoa F-1 (Alcoa Company, USA) has been extensively used for defluoridation of drinking water in western countries.<sup>12,16,17</sup> Defluoridation plant at Barlett, Texas, USA, which used Alcoa F-1, was constructed in 1952 and continued to operate essentially with the original medium for 25 years.<sup>17</sup> Maier et al.<sup>12</sup> reported that the plant showed an exchange capacity of 1272 mg F/kg of AA, when the raw water contained 8 mg F/L. A high exchange capacity of 3680 mg/kg of AA was observed for the same material at defluoridation plant in Arizona, USA.<sup>18</sup> In this plant, raw water pH was decreased to 5.5 prior to defluoridation. Exhausted AA was regenerated by successive treatments of alkali and acid. Our results have shown that G-87 exhibits an average capacity of 1833 mg /kg of AA at raw water pH 8.2-8.4 when fluoride concentration was 10 mg/L. There was no loss up to 5 cycles whereas loss of 15 % was observed after 35 cycles (data not shown). This reflects the regeneration efficiency as well as minimum loss due to acid treatment and attrition after prolonged use. Bulusu and Nawlakhe<sup>22</sup> reported a decrease of 83 % in fluoride uptake capacity after 20 defluoridation cycles, which remained more or less constant even after 40 cycles. 0.1 N HCl was used as regenerant by these investigators. Clifford<sup>16</sup> has reported that essentially complete removal of fluoride from exhausted AA is possible by successive treatment of alkali and acid. Our results are similar to that of Clifford.<sup>16</sup> It is well documented in literature that pH, alkalinity and initial fluoride concentration of water significantly affect fluoride uptake capacity.<sup>13,23</sup> Figure 2 shows that G-87 does not exhibit discrete optimum pH. This is in contrast to the reported behaviour of Alcoa F-1,<sup>15</sup> which exhibits maximum uptake at pH 5.0. The decrease as pH is raised from 5 to 8 is around 12% with G-87, where as it is 40% with Alcoa F1. Application of defluoridation technology with Alcoa F1 generally involves prior acidification to pH 5 to achieve maximum fluoride uptake followed by the neutralization of treated water.<sup>18</sup> However, Hao and Huang<sup>15</sup> have shown that the soluble alumino fluoro complexes are formed at pH<6 resulting in the presence of aluminium ions in the treated water. Hence they suggest that it may be preferable to carry out defluoridation

at a pH>6. Barbier and Mazounie<sup>23</sup> reported that acidification to pH 5.0 is not economically feasible if the raw water alkalinity is high due to the resultant buffering characteristics. However, raw water pH could be decreased to 7 to minimise hydroxyl ion competition. Fluoride uptake capacity of 2.24 mg/g AA was observed by these investigators when the raw water fluoride concentration was 11.5 mg/L and alkalinity of 395 mg/L.

A tap attachable defluoridation home unit was developed by Svedberg<sup>24</sup> using commercially available AA which was pre-treated by the manufacturer for the specific application. An exchange capacity of 1000 mg F/L was observed for raw water with fluoride and alkalinity concentration ranging from 7-10 mg/L and 80-160 mg/L respectively at pH 8.2. Our results show that even at a pH 8.2-8.4 and an alkalinity of 400 mg/L fluoride exchange capacity of 1833 mg/kg AA was observed. Results with simulated as well as natural waters showed that increase in alkalinity decreases the fluoride uptake capacity of G-87. Similar behaviour was reported for other grades of AA. Bicarbonate ions like hydroxyl ions probably compete for fluoride binding sites of AA. However it should be mentioned that there was no significant decrease in the bicarbonate concentration after defluoridation.

Clifford<sup>16</sup> has reported that sulphate affected fluoride uptake capacity beyond 250 mg/L. Our results are also similar to this observation. Treated water volume was drastically reduced with natural water having high bicarbonate and sulphate levels.

From the results of this study it is clear that DDU is simple to use and its user acceptability is excellent. While it is too early to arrive at a conclusion on its long term sustainability in rural areas besieged with adverse indicators such as low income generation, illiteracy and a lack of awareness about the water quality and its impact on human health, the initial results are encouraging. The next twelve months will confirm if users contribute towards regeneration of AA and regeneration is carried out efficiently by a village entrepreneur with out external support. However, it is clear that awareness generation among users, local capacity building and involvement of NGOs and private sector in service delivery are essential components for ensuring sustainability of domestic defluoridation.

Furthermore it can be concluded that indigenously manufactured AA is well suited for fluoride removal from drinking water in domestic defluoridation units.

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# LOW COST DOMESTIC DEFLUORIDATION

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SUMMARY: A simple household defluoridator is designed as a column, provided with a funnel attached to a down pipe arrangement, in order to allow for upflow filtration of fluoride water through a medium of locally available brick pieces. The unit is made of PVC-pipes of 1 m length and 20 cm diameter. Alternatively of brick/cement concrete of larger diameter in order to obtain sufficient removal when treating water of relatively high fluoride content (1.4 - 8 mg/L). Removal efficiencies observed are between 85 at the start of the operation period and 25 % at the end of it. The operation periods are between 90 and 250 days, after which the medium is replaced. Awareness programmes were conducted for school children, health staff, pre-school teachers and government officers at grass root level. The beneficiaries themselves were trained to change the filter medium in time in order to get optimum operation and maintenance. Eight hundred defluoridators were distributed in stages after the awareness programmes and they are in operation in different villages. A survey showed that nearly 85 per cent of the defluoridators are in working order, indicating the sustainability of the developed technique.

Key words: Fluoride; Defluoridation; Fluorosis; Bricks; Clay; Upflow filtration; Sri Lanka.

#### INTRODUCTION

Dental and skeletal fluorosis is an endemic problem in certain parts of the world such as China,<sup>1</sup> India,<sup>2</sup> and the Rift Valley of Africa. In recent studies it was shown that more than forty per cent of the wells in the North Central Province of Sri Lanka has fluoride rich water. Fluoride content of more than 1 mg/L was considered as fluoride rich water. Dental fluorosis had been identified as an endemic problem in the dry zone areas of Sri Lanka. The unsightly brown discoloration of the teeth had led young children affected in villages to a severe psychological impact. In addition medical reports revealed that skeletal fluorosis patients have been identified especially in certain dry zone areas in Sri Lanka.

Presently several methods have been practised to defluoridate water but in developing countries, the application of these techniques has certain drawbacks at the time of implementation. In Sri Lanka several methods are available, using filter media such as Serpentine, activated Alumina, Alum, and Charred bone meal. In all these methods the main disadvantage is that the filter media used is not readily available for the affected community.

In contrast the filter medium used in this study is freshly burnt bricks, which is freely available in affected localities. In addition replacing the filter medium upon saturation in the proposed unit is a low cost easy operation, which can be carried out at domestic level with negligible maintenance cost and absence of negative impacts on the environment.



#### MATERIALS AND METHODS

**PVC-defluoridator.** The newly designed household defluoridator is made of a PVC pipe, 100 cm in height and 20 cm in diameter. The inner diameter is 2 cm. The filter has a circular perforated plate fixed at 5 cm from the bottom. The outlet is fixed 5 cm below the top of the filter. The filter unit is packed up to a height of 75 cm with broken pieces of freshly burnt bricks, grain size 8-16 mm. The filter medium used for the removal of fluoride is low temperature burnt clay pieces. The fluoride rich water is fed through a funnel like inlet pipe at the top (Figure 1).

Before the start of operation the filter unit is filled with fluoride rich water and kept for at least 12 hours. Thereafter, when fresh fluoride rich water is fed through the inlet pipe, an equal volume of defluoridated water comes out automatically through the outlet. The efficiency of the defluoridator was tested by analysing the fluoride contents in the inlet and outlet at various time intervals.<sup>3</sup>

**Brick/cement defluoridator.** In an alternative design, the defluoridator is made out of cement and bricks (Figure 2).

**Programme strategies.** The strategies worked out in the pilot defluoridation programme in Sri Lanka may be listed as follows:

Awareness programmes were carried out in Village Schools. The students were requested to bring water samples of their wells. Analysis of these samples helped in the identification of fluoride rich wells.

With the help of public health officers, families with children less than 5 years of age using high fluoride drinking water were selected.

Thereafter awareness programmes were carried out to the beneficiary families and the defluoridators were distributed with special emphasis made on the change of filter media at appropriate time intervals.

IABLE	IABLE I. Fluoride removal percentages of the defluoridators							
Unit No.	Period of the tested cycle	Cycle length, Days	Well * mg F <sup>-</sup> /L	Outlet ** mg F <sup>-</sup> /L	Fluoride removal, %			
06	94.09.08 - 95.05.15	250	1.74	0.83	65.5 - 21.7			
09	94.11.17 - 95.10.05	326	1.42	1.26	29.1 - 11.3			
11	95.03.20 - 95.08.30	160	1.86	1.23	76.2 - 35.3			
12	94.09.10 - 94.04.15	220	1.45	0.87	49.3 - 33.0			
13	94.11.17 - 95.03.20	124	1.60	0.77	82.7 - 40.3			
26	95.05.10 - 96.12.30	235	2.35	1.43	77.6 - 31.3			
41	95.01.02 - 95.03.28	87	3.68	2.30	76.7 - 29.2			
10	94.09.26 - 95.02.20	155	2.85	0.65	72.6 - 22.6			
14	95.05.21 - 96.02.07	270	2.43	1.47	? - 39.5			
29	95.04.21 - 95.08.25	120	2.30	1.40	53.9 - 37.2			
* Avera	* Average value. ** At the end of cycle							

TABLE 1. Fluoride removal percentages of the defluoridators

Several visits to these beneficiaries were done in the first year to guide them in operation and maintenance. During these visits water sampling and testing were done to evaluate the efficiency of the defluoridators.

A visit at least once in six months thereafter for a period of 5 years is planned to check the functioning of the defluoridators and to encourage the continuous use.

## RESULTS

The removal efficiency of the defluoridator is determined to be between about 85 % at the start of an operation period and down to 25 % at the end of it. In a survey, it was found that about 80 % of the defluoridators were in operation by the beneficiaries. Table 1 shows the percentage of fluoride removal in defluoridators run bv beneficiaries in one village. All the beneficiaries had changed their filter medium on time, showing that the beneficiaries could manage these defluoridators on their own. The life span of the brick medium depends upon the fluoride content of the well water. It also depends on the daily consumption pattern of the household and the size of the



brick pieces packed in the defluoridator. Column 4 in table 1 gives the fluoride

content of the defluoridated water at the time of changing the filter medium. It was observed

that the fluoride content of the defluoridator No. 41 was high in spite of changing the medium within 87 days. This is because of the relatively high content of fluoride (3.68 mg/L) in the well.

Figure 3 shows data of a defluoridator, unit 42, with a large diameter of 280 mm, thereby increasing the capacity of the defluoridator. This facilitates the treatment of water containing higher content of fluoride (5 mg/L). The beneficiary was provided with this unit and instructions were given on its usage. The consumption rate of this household was 8 litres of defluoridated water per day. Accordingly after every 70 to 100 days of operation the filter medium had to be changed to obtain the best efficiency as shown in Table 2 and Figure 3, graphs A,B,C, & D.

TABLE 2. Def	luoridator no. 42.				
Inlet fluoride	Period	Graphs	F content treated water (mg/L)		
(mg/L)		51 mp	Start	End	
5.0	7.8.94 – 15.11.94 (100 days)	А	1.00	2.00	
5.0	20.11.94 - 10.2.94 (70 days)	В	0.24	1.24	
5.0	11.2.95 – 20.4.95 (100 days)	С	0.50	1.85	
5.0	25.4.95 – 5.8.95 (70 days)	D	0.66	1.75	

# DISCUSSION

A larger defluoridator of capacity 50 x 50 x 100 cm<sup>3</sup> was built with cement mortar and bricks by a village mason in a household. The water samples were collected periodically. In this defluoridator high levels of fluoride can be defluoridated as indicated in Table 4. The average fluoride content of the water used in this defluoridator was 8.0 mg/L. The consumption rate of defluoridated water in this household was 15 litres per day. Table 4 gives the fluoride content of the unit.

This domestic defluoridator, Figure 1, was compared with the bone char method<sup>4</sup> practised in Sri Lanka.

The salient features of the two methods are given in Table 4.

The burnt clay brick have silicates, aluminates and hematites as main components. When brick chips are soaked in water for several hours, these oxides get converted to oxyhydroxides of iron, aluminium and silica. The Si-O Al-O bonds are much stronger than Fe-O bonds. The geochemistry of fluoride ion (ionic radius 0.136 nm) is similar to that of the hydroxyl ion (ionic radius 0.14 nm) and these can be easily exchanged between them. As it takes minimum of 4 hours for the ion-exchange to take place, it is advocated to draw the water in the morning and evening. The upward flow technique used, prevented the sand particles mixing with water.

<b>TABLE 3.</b> Performance of a							
brick/cement	brick/cement defluoridator.						
Volume=250	L F <sub>inlet</sub> = 8						
mg/L.							
Time, days	Effluent, mgF/L						
01	2.95						
07	1.30						
15	1.20						
30	1.06						
50	1.24						
60	1.46						
70	1.44						
80	1.50						
100	1 70						

methou.		
Description	Bone char method	Brick pieces method
Methodology	Adaptation form Thailand	Locally developed
Flow Direction	Downward flow	Upward flow
Filter medium	Medium has to be fabricated and supplied to the community	Broken brick is already available in the localities
Cost of replacement	US\$ 5.0	US\$ 1.0

**TABLE 4.** Comparison of the brick chips defluoridation with conventional bone char method.

The results show that the developed defluoridators do not remove fluoride from the completely. It is a known fact that certain amount of fluoride in the range 0.5 to 0.8 mg/L is required for human body.

The fluoride removal efficiency, as observed, varies in the range of 80 to 30 per cent. For an inlet concentration of 4 mg/L this corresponds to an average residual concentration of 2.2 mg/L. It is therefore inferred that even a high fluoride level of 8 mg/L could be brought down to less than 2 mg/L by increasing the unit diameter and thereby its efficiency and capacity (Table 3).

Thus the important feature of the developed filter unit is the low cost of replacement of filter medium and its availability in the localities. This probable explains why the survey carried out indicated that the adopted technique is acceptable and sustainable technique for use at domestic level. With the help of awareness programmes and in collaboration with health staff, the developed technique may alleviate fluorosis in Sri Lanka.

## ACKNOWLEDGEMENTS

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# CHARCOAL PACKED FURNACE FOR LOW-TECH CHARRING OF BONE

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SUMMARY: A low-tech furnace for charring of raw bone using char coal is developed and tested. The furnace consists of a standard oil drum, fitted with simple materials as available in every market in small towns in developing counties. 80 kg of raw bone and 6 kg of charcoal are used for production of 50 kg of charred whole bones. This yields about 30 kg of useful bone char grains. Special arrangement is developed in order to fire the produced evil-smelling gasses, without introducing oxic conditions in bone charring chamber. The furnace is scalable according to the capacity needed.

**Key words:** Charring of bone; Bone char preparation; Furnace; Low cost furnace; Low-tech furnace: Fluoride; Defluoridation; Developing countries.

#### INTRODUCTION

For decades, bone char has been manufactured in large industrial scale to be used mainly as a colour sorbing agent in preparation of suger.<sup>1</sup> Also for many years bone char has been known and used as a defluoridation agent in municipal water treatment.<sup>2</sup> Initially, it was considered to be less suitable for use by individual households,<sup>2</sup> but this point of view never became widely accepted. The bone charbased household defluoridator was proposed as early as 1961,<sup>3</sup> initially for use in New Zealand.<sup>4</sup> Later it was modified as the ICOH defluoridator for use in Thailand and advocated by WHO for use in households of developing countries.<sup>5</sup> Only recently it became known that the implementation of the ICOH defluoridator in Thailand is limited, both geographically and in numbers.<sup>6</sup> Attempts to implement the ICOH defluoridator in Tanzania have not been successful so far.<sup>7</sup>

All through its history,<sup>2-7</sup> the use of bone char in individual household has encountered three major mutually related problems:

- 1. The bone char is not comfortably available in markets for the potential users, especially in not in developing countries.<sup>4, 5</sup>
- 2. The bone char, especially when prepared locally, may impart the water quality by giving the water unpleasant taste and/or smell or colour. Alternatively the defluoridation capacity of the product may be imparted.<sup>4, 8</sup>
- 3. The local production of bone char may, it self, be repulsive due to emission of evil-smell, which at the final end may discourage people from local production of the medium and the use of the method.<sup>4, 6</sup>

In order to resolve these problems the Danida-Enreca assisted Defluoridation Technology Project in Ngurdoto, Arusha Region, Tanzania, adopted a programme for development of an appropriate furnace. The objective of this paper is to describe one of the developed approaches; a charcoal packed furnace for low-cost preparation of bone char.

#### THE FURNACE

**Charring process.** Charring of bone is normally a process of heating to high temperature under restricted access of atmospheric oxygen, i.e. pyrolysis. Under such a treatment the organic materials in the bone crack to low molecule volatile compounds which evaporate. The residual organic carbon mineralises to graphite.



The graphite remains in the porous apatite structure. Properly charred bone does not add colour, taste or smell to the water. In the contrary it is capable, apart from removing fluoride, to remove discoloration, and bad taste or smell from the water.<sup>8</sup> In the developed furnace the heat is generated in part through controlled combustion of packed charcoal, in part through controlled firing of the organic materials in the raw bone. Heated fresh air is admitted at the smoke outlet in order to fire the produced nasty gasses before passing out to the chimney.

**Construction.** The furnace is made of a standard oil drum, 68 cm in diameter, 86 cm in height. The drum is placed on a cement foundation surrounded by a cylinder made of 4 pieces of corrugated galvanised iron sheets, fixed by means of 4 loops of 3 mm galvanised wire. The space between the drum and the corrugated plate cylinder, 20

cm wide, is filled with porous gravel for insulation, Figure 1. The drum has two holes, centralised in the bottom and in the cover. The holes are 12 cm in diameter to allow respectively for air inlet and smoke outlet. A double cylinder chimney made of corrugated iron sheets is mounted around the drum cover hole. This arrangement makes is possible to insulate the top of the furnace by covering it with about 15 cm layer of porous rocky gravel.

**Furnace inside parts.** The bottom hole in the furnace is covered with a perforated upturned funnel-like device for even diffusion of the inlet air in the charcoal chamber. An upturned funnel-like grill is placed over the air diffusion device. The space between the perforated device and the grill is used as a char coal compartment. Two <sup>3</sup>/<sub>4</sub> inch galvanised iron pipes are caste in the cement foundation for fresh air inlet beneath the drum bottom hole. Another <sup>3</sup>/<sub>4</sub> pipe is bend up to the centre of the drum for firing the produced gasses before emission to the atmosphere. The three pipes can be plugged either totally or partly by placing smaller pipe pieces at their inlets in order to control the fresh air and the heated air inlets.

#### **METHODS**

**Furnace operation.** Five kg, about 17 L of charcoal are piled on the bottom of the furnace between the perforated device and the grill. The charcoal is ignited using a slop of kerosene. As soon as major part of the charcoal makes red-hot it is spread around in the bottom. Whole pieces of raw bones, about 80 kg, together with additional 1 kg of charcoal, are piled around the grill up to the top of the furnace. After 1-2 hours of firing, when the furnace starts smoking heavily, the furnace is covered, the chimney is mounted and the top gravel is placed. The reducing cylinders are put in the air inlet pipes to reduce the ventilation. Depending on the wind direction and strength eventually only one air inlet is sufficient. Hereafter, the furnace is left undisturbed. Emission of evil-smelling smoke is prevented or significantly reduces by removing the cylinder vent reducer for the central air injector. During overnight charring is brought to an end and the furnace cools from about 500 °C down to less than 100 °C. Only at this or later point the furnace is uncovered for removal of charred bone pieces. The cycle takes 24 hours or so.

**Temperature monitoring.** The temperature was monitored using probe electronic digital thermometers, TESTO 950, at three different points in the furnace:

- 1. The bottom edge, 2 cm inside the drum at a level of 18 cm from bottom.
- 2. The top edge; 2 cm inside the drum at a level of 68 cm from bottom.
- 3. The Top centre; beneath the chimney at a level of 68 cm from bottom.

**Bone char crashing.** The charred bones are sorted out. Brownish as well as white pieces are of no use. The black and blackish-grey pieces are crashed using a manually driven iron roller, 35 cm in diameter, 75 cm in width. Concrete cement is caste in the iron cylinder in order to obtain required weigh. The roller is driven on a special platform for collection of crashed bone char.

**Grain selection.** The crashed bone char is transferred to a triple tray sieve system. The trays are hanged for manual swinging. Too coarse and too fine grains are rejected. The bone char of 0.2 - 0.5 grain size is collected from the medium tray.

**Bone char quality.** The quality of the bone char was tested by measuring the residual fluoride concentration in a bottle test. 2 g of bone char were added to 600 mL of distilled water containing 20 mg/L fluoride. Each bottle is shacked by turning upside

down 10 times during 15 seconds, and then left undisturbed for 8 hours. This cycle is repeated 6 times. After a total contact time of 48 hours the residual fluoride concentration is measured, using a Metrohm electrode and TISAB addition. The supernatant water is furthermore used for organoleptic evaluation of platability and measurement of pH and the colour using a Hach portable kit DR/2000.



# RESULTS

**Consumption and yield**. The furnace consumed 6 kg of charcoal per batch and about  $80 \pm 20$  kg of raw bones. Experience has shown that yield of charred bone is about 60 %, out of which 2/3 can be sorted out as grained medium, i.e. about 30 kg bone char per batch.

**Temperature profiles**. Figure 2 illustrates temperature versus time in a selected charring cycle. It is seen that the temperature reaches 400 -500 and remains at this level for a period of about 9 hours. Figure 3 illustrates the repeatability of the heating procedure. The curve pattern from one batch to another is almost the same; 7 hours of temperature increase, 9 hours of stationary charring period at 400-500 °C and 9 hours of cooling down.

## DISCUSSION

The Defluoridation Technology Project has carried out several investigations on technologies, which may be used for preparation of bone char. There seems to be no

doubt that charcoal packed column is far the cheapest and most user-friendly technique that can serve rural communities, if bone char has to be prepared locally.

Another important advantage of the technique is its scalability. It has been demonstrated that the furnace can be made in smaller scale, suitable even for the individual households. Furthermore, the furnace could be built in much larger scale, for centralised production of bone char to be distributed on commercial basis to rural areas.

Finely it has to be mentioned that same technique, after some modification, is probably most useful for charring or calcination of other media, like f. ex. magnesia, activated alumina, clay and the like. The key parameters in such modification would be the charcoal/media ratio, the air vent and flow and the mode of furnace packing.

Both the construction and the operation and maintenance can be carried out by villagers. However, like in rural brick making in kilns, good workmanship and some experience are required in order to run the process with a minimum of smell problems and the maximum of yield of good quality bone char.

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# BONE CHAR BASED BUCKET DEFLUORIDATOR IN TANZANIAN HOUSEHOLDS

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SUMMARY: A household defluoridator, made of a 20 L plastic bucket and 10 kg of bone char, is tested and found efficient to remove fluoride at a capacity of 1.1 mg/g. On an average, the defluoridator reduced the original contents of 8.5 mgF/L to 0.37 mgF/L, i.e. 95.6 %, for a period of 2 months, where 32.5 L were treated every day. The defluoridator could be manufactured locally in Ngurdoto village, Arusha Region Tanzania for a price of about 10 US \$ per unit. The defluoridator is monitored as operated in 10 households. The defluoridator reduced the fluoride concentration from 10.5 mg/L to less than 1 mg/L for periods between 4 and 13 months. The users expressed their acceptability of the defluoridator and its performance.

**Key words:** Drinking water treatment; Fluoride; Defluoridation; Bone char; Field test, Household treatment.

#### INTRODUCTION

The use of bone char for defluoridation of drinking water at household level is known as a promising method for provision of safe water in fluorotic areas in developing countries.<sup>1</sup> It is also known that the non-availability and the non-acceptability of local production of bone char are main problems in implementation the process.<sup>2</sup>

Recently a charcoal packed kiln was developed at the Ngurdoto Defluoridation Research Station, Arusha Region, Tanzania, to produce bone char at low cost and with a minimum aesthetic problems.<sup>3</sup> As a follow up a simple bucket defluoridator, containing the locally produced bone char, was made available for sale in the Station. The objective of this paper is to describe



the experiences gained from monitoring 10 of the sold defluoridators as operated by the different households.

#### **METHODS**

**Bucket defluoridator.** The bucket defluoridator consists of a 20 litres cylinder plastic bucket as normally available in the market and used in households. A tap is placed 3-4 cm over the bottom. A perforated piece of PEL pipe is placed as a drain. Ten kg of bone char, grain size 1 to 4 mm is directly in the bucket. Hereafter raw water is added by the users until the bucket is full. Treated water is directly tapped for drinking and cooking in the household. The users are instructed to keep the bone char submerged, Figure 1.

**Lab testing.** The bucket defluoridator was tested at the Ngurdoto Defluoridation Research Station prior to its launching to households.

The unit was loaded with water containing 8.5 mg/L. The water was added in small portions, 3-5 L each time, about 8 times per day, i.e. about 32 L per day. The water collected in one day was mixed and tested for fluoride contents, pH, colour and taste and smell. The filter was in use for 2 months.

**Distribution to households.** The bucket defluoridator was normally prepared and sold by a trained villager for a price of 6000 Tz.Sh. (10 US\$), including the 10 kg of bone char. For the field testing however, 10 defluoridators were sold to different families in the Ngurdoto village at <sup>1</sup>/<sub>2</sub> price for, in return, to allow for monitoring its performance. Initially a meeting was held

- 1. How often do you use the filter?
  - □ Never
  - $\Box$  Some times
  - $\Box$  When it is not raining
  - □ Always
- 2. Do you have any problem with the filter?
  - □ Yes: Mention Which
  - □ No problems
- **3.** Do you have any problem with the taste of the water?
  - □ Yes
  - □ No

**TABLE 1.** The questionnaire used in the study.

with the families in order to explain the idea of field-testing.

**Field testing.** The selected households were visited 8 times during a period of 13 months, from September 1996 to October 1997. All visits were made without prior notice. During the visits the raw and treated water were checked for fluoride contents and the condition of the defluoridator was recorded.

Furthermore, the household was interviewed as given in Table 1.

**Fluoride measurement.** The fluoride concentration was measured using a fluoride selective electrode (Metrohm 6.0502.150) and an Ag/AgCl reference electrode (Metrohm 6.0726.100) connected to a Metrohm 704 pH-meter. 5 mL of sample were mixed with 5 mL of TISAB and compared with standards.



#### RESULTS

**Lab testing.** Figure 2 shows the results from the controlled monitoring of the prototype defluoridator. On an average the defluoridator could reduce the original content of 8.5 mgF/L in the raw water to 0.37 mgF/L in the treated water, i.e. 95.6 % r

the to household dehuondators.									
Unit	Users		Residual conc. in mgF/L at day no.:						
no.	no.	122	154	182	212	243	279	318	408
1	8	0.4	0.3	0.4	0.5	0.2	1.2	0.6	1.4
2	10	0.2	0.1	0.1	0.1	0.2	1.2	1.5	2.7
3	7	1.0	2.1	1.1	2.2	4.7	7.0	- <sup>d)</sup>	- <sup>d)</sup>
4	9	0.3	0.4	0.5	0.7	0.4	5.0	2.0	1.1
5	9	_ <sup>b)</sup>	0.1	0.2	0.5	0.2	1.8	0.9	0.7
6	10	0.2	0.2	0.1	0.1	- <sup>d)</sup>	0.6	0.8	$2.0^{\rm c}$
7	10	0.3	0.1	0.1	0.1	0.1	0.6	$0.3^{c}$	0.4 <sup>c)</sup>
8	4	0.7	1.0	2.1	1.5	1.7	2.2	$2.7^{\rm c}$	- <sup>c)</sup>
9	5	0.9	0.8	- <sup>d)</sup>	- <sup>d)</sup>	1.0	1.3	$1.1^{c)}$	$2.0^{\rm c}$
10	3	0.2	0.1	$0.1^{c)}$	$0.1^{c}$	0.8	- d	1.5	1.8
Averag	ge conc.:	0.47	0.52	0.52	0.64	0.93	2.32	1.27	1.51
Raw w	ater conc	.: 9.4	9.9	10.4	11.8	11.2	10.5	10.2	9.3
Aver.	Remov. %	<b>95.0</b>	94.7	95.0	94.6	91.7	77.9	87.5	83.8
a) Defluction was said to be in use always, exact from $b$ $c)$ and $d$									

TABLE 1. Monitoring of the fluoride concentration in the raw and outlet water from that 0 have abald defly aride

fluoridator was said to be in use always, apart from <sup>6</sup>, <sup>6</sup>

<sup>b)</sup> In use only some times. <sup>C)</sup> Using seasonal rain water.<sup>d)</sup> Defluoridator was not in use. removal efficiency. After 59 days of operation, where 32.5 L were treated every day, the residual concentration was still  $\leq 0.7$  mgF/L. The sorption capacity under these conditions was measured to be 1.1 mgF/g bone char. The treated water had pH between 9.0 and 8.3, on an average 8.56. During the 5 days of operation the treated water had a minor discoloration. Apart from the first operation day, the water was fully palatable.

TABLE 2. Answers to the question: How often do you use the household defluoridator?

Defluoridator in use ?	Not in use	Sometime	Not in rain season	Always	Total
No. Of visit	5	1	9	65	80
%	6	1	11	81	100

Efficiency in field. As the households were all feeding their defluoridators from the same piped water supply, the fluoride concentration in the raw water was the same in all defluoridators each monitering day. The concentration varied from one monitoring day to another as shown in Table 1. The table shows the efficiency of the removal during the monitoring period of 408 days. On an average the fluoride concentration was reduced from 10.3 to 1 mg/L, i.e. 90 %.

TABLE 3. Content of	of water in t	he defluorida	ator during i	nspection.	
Water content	Empty	1⁄4 full	½ full	<sup>3</sup> ⁄4 full	Full

Water content	Empty	<sup>1</sup> ⁄4 full	<sup>1</sup> / <sub>2</sub> full	3⁄4 full	Full	Total
No. Of inspections	2	8	20	26	16	80
% of inspections	2.5	10	25	32.5	20	100

Acceptability and use pattern. During the 408 days monitoring period, the households were inspected in total 80 times. Table 2 shows that the household answered that the units were not in use only 5 times. Table 3 shows how much water was found in the defluoridators at the time of inspection. The only complaints expressed was that the discharge from the tape was too slow.

#### DISCUSSION

In spite of the fact that the bone char was prepared locally, the quality of the treated water was in general good and acceptable to the users. The villagers had no difficulties in operating the filter and in keeping the filter bed submerged.

From the lab testing, where the loading was similar to that of the household, it was estimated that the removal capacity is expected to be high, resulting in less than 0.7 mg F/L, until a removal capacity of at least 1.1 mg/g is reached.

The household testing did not allow for precise calculation of the filter capacity. On an average the 10 units reduced raw water fluoride of 10.3 mg/L to less than 1.5 mg/L for period a period of 13 months. However there was a wide variation in their operation period, before saturation. If a treated water concentration of 1 mg/L is adopted as a point of saturation, the effective operation periods varied between 4 months and more than 13 months. The saturation point was reached after 4 months (1 unit), 5 months (1 unit), 8 months (1 unit), 9 months (1 unit), 11 months (2 units), 13 months (2 units). Probably this reflects the different loading and defluoridated water consumption patterns in the different household.

#### ACKNOWLEDGEMENT

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# A REVIEW OF THE DEFLUORIDATION PROGRAM OF DRINKING WATER SUPPLIES OF AN ETHIOPIAN ESTATE

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ABSTRACT: The Wonji Shoa Sugar Estate covers an area of 50 km<sup>2</sup> in central Ethiopia inhabited by 25,600 people. Dental and skeletal fluorosis were identified in the area in 1957 and 1972 respectively. Since then the Estate has been well known for its high fluoride levels in its water supplies. An evaluative study of the 35-year-old defluoridation program was conducted. Data were obtained from documentary sources and by making interviews, site visits and laboratory analyses. The community of the Estate has largely depended on a dual supply of well water - raw water and defluoridated water. The major problems noted were the lack of proper operation of the defluoridation plants, the lack of ready accessibility of the treated water supply and the lack of compliance by the community. The Estate's community is found to be quite aware of the fluorosis problems. Yet only 56.3 % of the community have access to defluoridated water and suggestions for improvement are made.

**Key words:** Ethiopia; Rift Valley; Fluorosis; Skeletal Fluorosis; Activated alumina; Defluoridation; Regeneration of media; Operation and maintenance.

#### INTRODUCTION

Ingestion of excessive amounts of fluoride has been known to be pathogenic to humans, particularly causing dental fluorosis in children and skeletal fluorosis in adults.<sup>1</sup> Communities depending on water supplies with fluoride levels in excess of 1.5 mg/L are at risk of fluorosis.<sup>2</sup> The safe level of fluoride in water supplies is lower in hot climates. In the East African Rift Valley there are high levels of fluoride both in ground water and in surface water due to the high fluoride content of the volcanic bedrock.<sup>3</sup>

**The Wonji Shoa Sugar Estate.** The Wonji Shoa Sugar Estate (WSSE) is a sugar producing estate (the official name of the company being Wonji Shoa Sugar Factory, WSSF) owned by the Ethiopian Government, and is located in central Ethiopia within the Rift Valley, 110 km south-east of Addis Ababa. At present the estate stretches over an area of 50 km<sup>2</sup>. The altitude is 1540 m above sea level. The annual rainfall rarely exceeds 800 mm. During 1996, the mean maximum daily temperature was 27.4°C and the mean minimum daily temperature was 16.4°C. The estate presently has about 8000 employees. Its total population according to a census conducted in 1995 was 24,206, the present estimated population being 25,581. The community lives in two relatively large factory villages, namely Wonji and Shoa, and in 14 plantation villages scattered throughout the estate. The estate cultivates its own sugar cane plantation is obtained from wells and is supplied through pipes. The provision of water supply to the residents of the estate is the responsibility of WSSF.

**Historical background.** The estate was established by a Dutch company and officially started work in 1954. The existence of "fluoride problem" in the estate was first recognised in 1957 when the children of the Dutch families had dental

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examination while holiday in Holland. In 1962, two defluoridation plants were installed in the two factory villages where the Dutch families lived. In 1972 the existence of skeletal fluorosis in the estate was discovered. Between 1974 and 1976, defluoridated water was made available to all villages of the estate. In 1975, the estate was visited by a Dutch dentist named Prof. Otto Backer-Dirks who had been involved in investigating the fluoride problem of the estate.

In 1976, the Ethiopian Government established a committee to examine the problem, but recommendations made by that committee were not implemented. Despite the launching of the defluoridation program it was noted the existence of fluorosis had continued. So, in 1984-1985 a feasibility study was conducted by an English company to provide low fluoride potable water to the estate's community.<sup>4</sup> The study recommended the utilisation of water from Awash River as a single water supply to the entire community for all domestic purposes after conventional water treatment plus defluoridation of the entire supply at a single new plant. However, that alternative was not implemented due to cost hindrances.

**Study objective.** The community of the estate has largely depended on well water for domestic needs. At times Awash water is also used for domestic consumption in some villages. The defluoridation program of WSSE is the only one in Ethiopia and has now been going on for 35 years. The purpose of this review was to make an evaluation of the performance and outcomes of this program.

## MATERIALS AND METHODS

Data for the review were collected during the period July to October 1997. Historical data were obtained from records and files of WSSF. Contemporary data were obtained by interviewing program staff and by making site visits. Present levels of fluoride in water sources were determined at the laboratory of the hospital of WSSF (Wonji Hospital).

## RESULTS

**The present water supply.** The domestic water supply involves a dual supply of raw well water for washing purposes and defluoridated well water for cooking and drinking purposes. At present there are 20 operational wells in the estate. The depth of the wells ranges from 12 m in village A (initially 50 m) to 64 m in village K (initially 110 m). Awash water is supplied to all neighbourhoods of both factory villages from standpipes for gardening purposes. The residents of the plantation villages have access to Awash water either directly from the river or from irrigation canals. In some of the plantation villages, Awash water is used for human consumption when tap water is not available. The level of fluoride in Awash water is 2-4 mg/L.<sup>4</sup>

**Factory villages.** There are 7 wells in Wonji village and 4 wells in Shoa village providing raw water through an interconnected piping system to each village. Here is a reservoir in each village. The supply is directly connected to certain houses and the factories, offices, etc. Other houses have one standpipe for 8-12 houses. The supply of raw water is almost always available in each village. There is one additional well in Wonji village solely used for the supply of defluoridated water. In Shoa village water to be defluoridated is obtained from one of the wells also supplying raw water to the

village. In each village the water is treated at a defluoridation plant, stored in a reservoir and supplied to the residents from standpipes, each standpipe being allocated to 50-60 houses. Some public buildings such as factories, offices, schools, hospital, bakery, recreational facilities, etc. have their own standpipes.

**Plantation villages.** There are 8 wells providing raw water to 13 plantation villages through separate piping systems. There are reservoirs and at each village raw water is supplied from standpipes and/or from a washing area. There is recurrent lack of raw water in the plantation villages due to interruption of electricity or to failure of pumps. During these periods the residents fetch water from nearby village or use Awash water. The 14<sup>th</sup> plantation village, village P, is supplied with raw water transported by a trailer from village E or Shoa.

Village Band E get the supply of defluoridated water from Wonji and Shoa defluoridation plants respectively. Each of these villages has a single standpipe for defluoridated water. There are ten defluoridation plants which were supposed to provide treated water to the remaining plantation villages. They get water from the wells also providing raw water to the villages. In each village there is a single standpipe for the supply of treated water (none in village P) connected directly to a defluoridation plant, without any reservoir involved.

<b>IABLE 1.</b> Levels of fluoride in raw water from various wells of the factory villages.					
Dete	Fluoride levels from wells (mg/L)				
Date	Wonji village	Shoa village			
1973	5.6 - 13.5	3.3 - 18.7			
Nov. 1976	2.9 - 19.2	3.4 - 13.0			
Oct. 1984	2.7 - 18.8	3.3 - 11.2			
Oct. 1997	$1.4 - 18.8^{*}$	2.2 - 15.6*			
	1 1 1 0 1 1007 1 1				

\*The individual levels of fluoride in Oct. 1997 were 1.4, 6.2, 14.0, 14.8, 15.6, 16.4 and 18.8 mg/L in Wonji and 2.2, 6.2, 10.0 and 15.6 mg/L in Shoa.

**Requirements of the defluoridation program.** The defluoridation of drinking water supplies has been necessary because the community of the estate has depended on well water, which has high levels of fluoride. At present all wells have fluoride levels above 0.8 mg/L which is the safe level for the estate taking into account the local climatic conditions (Table 1 and 2). So still the defluoridation program is requisite.

**Defluoridation plants.** There are 12 defluoridation plants in the estate. The two plants in the two factory villages were erected in 1962. Initially each plant had a single filtration tank but later a second tank was added. The media capacity of each tank is bout 370 kg. There is a reservoir for treated water at each plant. Both plants are more or less in good condition at present. The ten plants in the plantation villages were installed in 1976. At present all of them are neglected and are heavily affected by rust. The defluoridation technique employed is adsorption by activated alumina, which in turn is regenerated by flushing with caustic soda. In the past bone char was also used.

**Materials.** In the past there was problem of availability of the medium. At present there is no shortage of the supply of the activated alumina but sometimes poor quality water, containing coarse grains of alumina, is supplied. Also there is no shortage of caustic soda either. At times there is lack of necessary spare parts for the maintenance of the defluoridation plants.

. . . . .

**Personnel.** The formation of personnel consists of two operators and a driver. At present all positions are filled but the driver is not in the defluoridation team. The operators appear to be satisfactory skilled.

**Transportation.** There is one vehicle in the budget formation but at present the vehicle is assigned to other tasks and is not available for defluoridation activities. Similar shortage of transportation hindering regeneration and maintenance activities was present also in the past.

**Organisation.** In the past the water supply service of the estate was divided and was organised under three departments. But since 1976 the whole domestic water supply (both the supply of raw **TABLE 2.** Levels of fluoride in raw water from the various wells of the plantation villages.

Village	Dec. 1971	Oct.			
-		1997			
А	3.0	1.8			
В	8.0	3.6			
С	7.5	12.0			
D	4.0	5.0			
Е	7.5	12.0			
Κ	7.5	17.5			
L	7.5	$4.5^{*}$			
М	3.0	3.1			
* Average of measurements made					
during various occasions from					
January to October 1997.					

water and the supply of treated water) has been organised under one department (Shoa Factory Mechanical Workshop).

**Laboratory.** The laboratory of Wonji Hospital performs fluoride analysis of water samples and is responsible for quality control of the program. It uses OIRION Research Model 701 digital pH-meter purchased in 1974. The necessary materials are available at present but in the past there was a problem of the supply of electrodes.

#### Performance of activities.

**Media renewal.** The frequency of media renewal was to be based on the results of fluoride analyses of treated water samples. At the Shoa plant media renewal is done every 1½-3 months. At the Wonji plant this is done every 9-12 months. During the activated alumina renewal caking of the medium has been a major problem. At the plantation villages, media renewal was carried out at irregular intervals. For instance at one village the medium was renewed after 18 years. The renewal activities have been totally abandoned at the plantation villages for the last two years due to lack of transportation.

**Regeneration.** At each of the two present operational plants, regeneration is carried out for each of the two tanks after having served for 24 hours. The two tanks at each plant serve on a 24 hour shift. The steps of the regeneration procedure appear to be well known by the operators but there is no supervision. The operators have problems during regeneration because of not having proper protective devices. After introduction of caustic soda solution, flushing is done with raw hard water as a result of which calcium carbonate scales form on the activated alumina granules reducing the adsorption capacity. In the past carbon dioxide injection was used to reduce regeneration time. Regeneration activities were carried out at irregular intervals at the plantation villages in the past due to shortage of transportation and have been totally abandoned as of the end of 1995.

**Monitoring.** Weekly analyses of water samples from Wonji and Shoa defluoridation plants was started in 1969 and from the plantation villages in 1976. However, frequently water samples are not collected from several plantation villages due to lack of transport. Analytical results are delivered to the responsible department after a delay of 5-7 days. The results were to be used to evaluate the regeneration activities

and to decide whether or not to change the media. Regular monitoring of prevalence rates of fluorosis has not been done.

**Availability of treated water.** Before 1962 only raw water was available to the entire community of the estate. Between 1962 and 1974 defluoridated water was available to certain families in the two factory villages. In 1974 the supply of defluoridated water was extended to all families of the two factory villages and of plantation village B and E. In 1976, defluoridated water was made available to the remaining 12 plantation villages. This was going on until the end of 1995.

At present, defluoridated water is available only to the two factory villages and the two nearby plantation villages B and E. The resident of the remaining 12 plantation villages utilises raw well water (either from the standpipes of treated water assuming that it is defluoridated, or from the standpipes of raw water) and sometimes Awash water for domestic purposes - cooking, drinking and washing.

So, out of the estimated present population of the estate of 25581, only 14396 or 56.3 % has access to defluoridated water. There is some waiting at the standpipes as has also been the case in the past.<sup>4</sup> The waiting is greater at Shoa village because residents from the surrounding areas also share the service. At Wonji village at times there is total lack of treated water but this does not happen at Shoa.

### Degree of utilisation.

The community is well aware of the fluoride problem, both of dental fluorosis and of skeletal fluorosis. The standpipes supplying treated water are well identified but are not very readily accessible as each one is supposed to serve a large number of people.

It is a common observation that still children and also adults consume considerable amounts the more readily accessible raw water even in the villages which have the supply of treated water. At times, the limited supply of treated water is abused for washing purposes.

#### **Outcome evaluation.**

**Fluoride levels in drinking and cooking water.** Fluoride levels in water from the 12 defluoridation plants are given in Table 3. The levels of fluoride in treated water from the two presently operational defluoridation plants before and after regeneration are given in Table 4.

**Prevalence rates of fluorosis.** In 1975 mottling of the permanent teeth was found in 100% of 8 to 10 year-old-children examined in Wonji and Shoa villages (personal communication, Prof. O Backer-Dirks). A survey conducted in 1977 showed 87% mouth prevalence of dental fluorosis among children 5-10 years old and 31% prevalence among individuals 16-25 years old.<sup>5</sup>

Surveys conducted in 1985 gave the following results.<sup>4</sup> In one of the surveys involving examination of the permanent anterior teeth, mouth prevalence rates of dental fluorosis ranging between 34 % and 75 % were found among 8-yaer-old children residing in the various villages of the estate and a 77 % mouth prevalence of dental fluorosis was found among adults aged 20-25 years. In the other survey involving examination of the prevalence posterior teeth mouth prevalence rates of dental fluorosis of 39% and 54% were found among children aged 12-15 years in the two villages; a 74 % prevalence rate was found among adults 20-25 years old. A survey done in 1997 in children aged 8-12 years showed mouth prevalence rates of

dental fluorosis ranging between 71.4 % and 95.7% in the various villages; excluding the very mild cases the rates ranged from 27.1 % to 86.1 % (personal communication, Fantaye et al.).

In 1975 there were 60 bone fluorosis patients identified in the estate, most of them having only restriction of movement, but five had neurological complications. A survey conducted in 1979-1980 by Wonji Hospital involving 530 estate workers aged 45-55 years, found radiologically evident skeletal fluorosis in 46 % of them. In 1990, a survey showed that 63 % of the individuals who had resided in the estate for more than 20 years had physical impairments indicative of skeletal fluorosis.<sup>6</sup> A survey done in 1997 and involving the X-rays of 263 retiring employees of the estate showed radiographical signs of spinal fluorosis in 70.3 of the individuals (personal communication, Shifera et al.).

**TABLE 3.** Fluoride levels in treated water from the various defluoridation plants of Wonji Shoa Sugar Estate.

11011	Honji onou ougui Estato.								
Plant	76.11.27	79.10.04	88.04.21	88.08.12	89.02.02	89.10.09	94.09.29	95.10.06	97.10.31
Wonji	0.36	0.45	0.40	0.23	1.30	0.28	< 0.10	0.70	0.92
Shoa	0.21	0.45	0.32	0.82	1.30	0.66	0.45	1.50	0.90
А	-	0.20	0.34	0.25	0.50	0.62	0.15	0.95	*
С	0.40	-	2.80	6.01	4.00	7.7	2.90	4.20	*
D	0.11	0.18	1.61	6.01	2.80	2.5	0.75	1.80	*
F	0.20	0.18	1.00	6.01	-	-	0.45	1.50	*
Н	0.28	0.57	2.80	6.01	4.60	6.1	9.50	6.80	*
Κ	0.15	0.34	7.10	4.56	4.30	2.2	-	3.50	*
L	0.11	-	1.30	4.56	0.61	5.2	3.00	-	*
М	-	0.26	1.40	0.48	1.00	2.0	0.17	-	*
0	-	0.30	0.25	0.52	1.30	1.3	-	-	*
Y	-	-	3.34	2.40	6.10	-	-	7.00	*
* Wate	er samples	s not analy	sed becau	ise there w	as no trea	tment at t	he plant.		

**TABLE 4.** Fluoride levels from defluoridation plants of the two factory villages on Wonji Shoa Sugar Estate, October 1997.

Defluoridation		Fluoride levels (mg/L) in					
plant	Inlet	Outlet before regeneration	Outlet after regeneration				
Wonji	1.4	1.2	0.92				
Shoa	2.2	1.8	0.90				

#### DISCUSSION

The domestic water supply of WSSE involves a dual supply of tap water from wells - a readily accessible supply of raw water and a very less readily accessible supply of defluoridated water. In certain plantation villages there is, in addition, access to a third source of water - Awash River water. There is a wide variation in the level of fluoride among the various wells, ranging from 1.4 to 18.8 mg/L, all exceeding the estate's safe level of 0.8 mg/L. Nineteen of the twenty presently operational wells (95%) have fluoride levels greater than 1.5 mg/L and eleven wells (55% of the total) have fluoride levels greater or equal to 10 mg/L. Therefore the defluoridation program is still necessary if the estate's community is going to continue depending on well water.

It seems that the number of available treatment plants is sufficient to provide drinking and cooking water for the estate's community except in one of the factory villages. The main problem rather is the lack of transport, which has resulted in the total shut down of media renewal and regeneration activities at ten of the twelve (83.3 %) defluoridation plants for the last two years. Therefore it is necessary that a vehicle be assigned solely for water supply activities as it was in the past. The quality of the activated alumina supplied is sometimes poor and measures have to be taken by WSSF to ensure that good quality activated alumina is consistency supplied. It has to be emphasised that the defluoridation plants, which had demanded a lot of efforts, dialogue and investment to be installed, be given due maintenance and care.

It is necessary that the frequency of media renewal and generation be strictly geared to the levels of fluoride in treated water. For this purpose regular weekly and random samples of water have to be collected from all defluoridation plants. Laboratory analytical equipment has to be updated and results have to be reported overnight to the responsible department. Proper protective devices need to be provided to the operators and supervision of the regeneration activities also has to be exercised. Possible mechanisms to reduce calcium carbonate scale formation during regeneration have to be investigated. To increase the adsorption capacity and service period of the media acidification has to be practised.

At present only 4 of 16 villages of the estate or 56.3 % of the total population has access to defluoridated water. The residents in several plantation villages are using water with very high fluoride levels (> 10 mg/L in four villages) for cooking and drinking purposes. This situation has to be urgently changed and all villages have to have access to defluoridated water. On the other hand it has to be known that due to the better accessibility of raw water in relation to treated water, significant quantities of raw water appear to be consumed particularly by children in the villages where defluoridated water is also available. So measures have to be taken to educate parents and children to alter this behaviour or to make treated water more readily accessible. The water supply department has also to make sure that there is a continuous supply of tap water in all the villages so that residents will not be forced to consume less hygienic water from Awash River or from irrigation canals.

The defluoridation program was launched in the two factory villages 35 years ago and was extended to all villages of the estate 21 years ago. But still there are high levels of fluoride in the water available to a large part of the community and the prevalence rates of dental fluorosis and skeletal fluorosis are also high. The reason for the failure of the program does not seem to be the deficiency of the defluoridation technique employed. Rather the reasons for the failure of the program in producing the final desired outcome i.e. the prevention of the new occurrence of dental and skeletal fluorosis are:

- The lack of proper operation of the existing defluoridation plants.
- The absence of easy accessibility of the treated water supply.
- The lack of compliance as to water ingestion behaviour by the community.

It is suggested that the concerned act without delay to revive the defluoridation program using the same or other more convenient technique and to modify the behaviour of the community so that the disfiguring and the disabling problems of fluorosis are eliminated. Or WSSF has to be ready to re-examine the excellent feasibility study conducted earlier which had connected resorting from well water to river water.<sup>4</sup>

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Session V

# **Discussion Papers**

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# SOLVING THE FLUOROSIS PROBLEM IN A DEVELOPING COUNTRY

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SUMMARY: Although substantial professional inputs to solve the fluorosis problem in Thailand in the last decade have not yielded satisfactory results, the output, admittedly not in material forms, includes encouraging lessons that have an impact on our way of thinking in tackling the problem. The people's awareness which is the essential initial step, then their conceptual understanding of the situation, followed by their resourceful efforts, are the keys to steering on a successful course. This essay also addresses the roles of the professionals. It calls for a reversion from the liberal orthodoxy of 'people participation' in (non-people) development to 'professional participation' in people development. It is written with the hope that our experience can be food for thought for a new, if not novel, approach in solving problems in developing societies.

**Key words:** Fluorosis; Defluoridator; Appropriate technology; Developing country; Approaches to development; People's aspiration/action.

#### THE BACKGROUND

Chronic and endemic as the problem of fluorosis has been in the northern part of Thailand.<sup>1</sup> But only in the last decade has it been tackled, and real

success is not in sight. It is not the technical know-how<sup>2</sup> that is the main hurdle. On the contrary, the technology works well, but still the problem remains as can be seen from the outcome of a pilot study in two villages provided with ICOH defluoridators.<sup>3</sup> If the technology itself cannot bring about a viable solution, we need to go beyond it and it is to be argued that it is the people themselves who are decisive in successful development.

For the sake of argumentation the problem of fluorosis can be looked at from two angles: techno-medical and social. The toxicity of fluoride concentration is the former, the long duration of water consumption the latter. The approach to the former is to find ways and means to reduce the toxic content, whereas the latter is concerned with habitual use of water or way of life. The ways and means in question need to be technically efficient. But being technically sound is merely a part of the story. More importantly it is to be realized that no matter how sound the technological means are, they should be operated in the context of the people's way of life. And that is related to the whole social and cultural configuration.

# THE FOLK'S AND THE HEALTH PROFESSIONAL'S DEFINITION

In some high fluoride areas of Thailand 100 % of the people have dental fluorosis.<sup>4</sup> As they have been accustomed to dental fluorosis all their lives and over generations, it was not really regarded as a problem. Prior to the last two decades, when communities were still relatively isolated and the inhabitants not much exposed to outer communities, fluorosis was not seen as unnatural.<sup>5</sup> If people with white teeth

# Casca

So every bondman in his own hand bears the power to cancel his captivity.

Shakespeare, Julius Caesar

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happened to be around, the locals immediately noticed that they were not from their neighbourhoods.

Dental as well as skeletal fluorosis is, indisputably, considered as a problem from the medical standpoint. Ironically, however, as an endemic condition fluorosis is not emphasised in the dental and medical schools. The standard curriculum of most dental schools world-wide generally advocates the value of fluoride more than a precaution against it.<sup>6</sup>

As a matter of fact folk's awareness came not from medical authority but originated from social and cultural factors. The affected communities, which generally are in the rural areas, have increasingly come under the influence of urban values. The groups, whose awareness has been changed, are those with contact with others from outside. The majority of people, who are most concerned with the problem of fluorosis, are the new generations going to work or to school in towns. The visual mass media is also another source of people's awareness, since it gives "the standard of beauty". In the absence of other symptoms, it was originally taken to be only as a cosmetic problem, not a problem of health. As for skeletal fluorosis, due to its long-term consequences, it was not of immediate concern.

In the beginning people with fluorosis tried to help themselves in several ways: by the application of charcoal, the use of sandpaper and the employment of quack doctors. Sandpaper was used to erase the brown stain from tooth surfaces. A mobile "doctor" with certain elementary dental equipment went from one village to another to polish teeth for the villagers.<sup>5</sup> These methods undoubtedly could be very damaging, for they could cause sensitive teeth and lead to infection. A minority with economic seek professional dental means treatment,<sup>7</sup> e.g. tooth extraction, which is an unethical practice. or crown restoration which is costly. When the ICOH approached these communities

#### "The ICOH defluoridator"

through of has been а series modifications. Initially we started from the general idea of the bone char recipe. Many experiments were conducted to find the appropriate formula for bone char production, on one hand and the container thereof on the other. Eventually the whole design was carried out according to the principles of appropriate technology and efficiency to reduce the fluoride content from 5 to 0.5 mg/L, i.e. lower than stated in the WHO standard. After some modifications of materials and adjustments of its practicalities, the defluoridator was put into use in individual households.

and proposed a solution to fluorosis, they were extremely pleased, and eager to cooperate.<sup>2</sup>

#### THE ACTIVE/PASSIVE ROLES

The ICOH defluoridator, cf. textbox, was installed in 100 households in two villages in Chiang Mai province where the natural fluoride content in the water was between 1 to 6 mg/L. The ICOH provided the defluoridator free of charge. The project staff visited the villagers under study on a regular basis. Moreover the ICOH had trained village health volunteers to change the ready-to-use-filter bags. The bags were prepared by the ICOH and sold to the villagers at a subsidised price. In order to get villagers to use the defluoridator no special persuasion was necessary. The daily handling of the defluoridator and the periodic changing of the filter caused no problems for the villagers<sup>8</sup>. The twelve-month evaluation showed that more than 90 households still used the defluoridator.<sup>2</sup> It had become so popular that the people in the adjacent villages showed strong interest in obtaining the equipment or sharing the treated water.<sup>8</sup> After several years the ICOH, having difficulties in producing bone char, was no longer able to supply the-ready-to-use filters. But the people still continue to use the ICOH defluoridator without changing the filter. The project, useful insofar as the scientific experiment is concerned, was not so successful in terms of villagers' health. The fluorosis problem of the new generation remains unsolved.<sup>9</sup>

Nevertheless the whole project was not altogether in vain. Positive results have come from an unexpected quarter. As mentioned, certain villages were enthusiastic about the idea and the method. They asked for the installation of the defluoridator at their own cost. The villagers in this community employed it wisely. They stopped its usage after the supply of ready-to-use filter bags failed. The idea of making their own bone char was not congenial to their sense of propriety, hence not acceptable. Though by itself the ICOH equipment was not of sustainable value, the villagers had acquired conceptual understanding through its utilisation. Once the rationality of defluoridation was grasped, they turned to their own resources, intellectual, cultural as well as material. In the course of trial and error they arrived at a solution and built containers to collect rain water for consumption.

According to tradition the northerners are dependent not on rainwater, but on water from wells, for household consumption.<sup>5</sup> The idea of utilising rain water, though common among the people in the central plains and elsewhere, did not come right away as a natural solution to villagers inhabiting fluoride areas. Odd as it may seem, the peculiar barrier could not be attributed to the non-customary practice alone. More importantly it was due to lack of the understanding. When the insight is attained, the people themselves are better qualified to work out their own ways. However, even if a rain container is functional for the people of this village, it does not follow that it is to be applied or recommended to other villages without discretion, for it is rather expensive and it is unsuitable for traditional houses roofed with leaves. As far as this one village is concerned it is an alternative that they have adopted, and they have their own way of managing the budget. The level of community concern and folk's efforts is very clearly indicative of the folk as active actors. At present every household in this community has its own container. Dental fluorosis is no longer a problem of the new generation, all of whom now have normal teeth.<sup>10</sup>

Sociologically speaking a comparative view between the two experimental villages (Case A) and the other village that asked for the installation of the defluoridator at their own cost (Case B), is of considerable value. The approaches of the ICOH personnel were clearly different. In Case A, their approach was as implementers, while in Case B as catalysts and supporter. In the former case the villagers waited for the implementers to solve the problems for them; it was the ICOH s' job, not theirs. In the latter, the villagers were the initiators; whereas the ICOH was merely their consultant. In other words, though both of them shared the similarities of awareness and concern, the contrast lies in the roles of health personnel and of the folk. In Case A the ICOH personnel were active and the folk passive, whereas in Case B the reverse was the case. And the reversed roles of the actors determined the failure and the success in the different communities.

# THE CONCEPT OF APPROPRIATE TECHNOLOGY

The rationale of the design and of the implementation of the ICOH defluoridator is based on the principles of appropriate technology. But similar to other concepts, its meaning is subject to interpretation. Strictly or broadly interpreted, however, from experience drawn from this project, the concept of appropriate technology itself could be called in question. And it is perhaps now appropriate to go beyond it and move on to a new developmental approach.

As stated, the ICOH defluoridator, considered as a working apparatus, has a functional design. And as equipment, it could be made by the villagers themselves. The major parameter lies in the material of the filter. Neither health personnel nor villagers have come to terms with bone char. The production and the supply of bone char are, in fact, the essential considerations. The Provincial Health Office, which is in charge of people's health in the provinces, did not accept it as appropriate technology. They have found it too cumbersome to produce bone char. No one wishes to produce it due to its undesirable odour. The factories which produce bone char are always boycotted by their neighbours. The villagers themselves associate its production with the practice of funeral cremation. Scientifically and technically the bone char method is not only effective, but also appropriate and it is even economically conducive. But socially and culturally it is anathema. The ICOH defluoridator has proved to be "appropriate" only for research purposes.

#### "Appropriate"

means that besides being scientifically sound the technology is also acceptable to those who apply it and to those for whom it is used. This implies that technology should be in keeping with the local culture. It must be capable of being adapted and further developed if necessary. In addition, it should preferably be easily understood and applied by community health workers, and in some instances even by individuals in the community; although different forms of technology are appropriate at different stages of development, their simplicity is always desirable. The most productive approach for ensuring that appropriate technology is available is to start with the problem and then to seek, or if necessary develop, a technology which is relevant to local conditions and resources<sup>11</sup>."

Nonetheless one could argue that the concept of appropriate technology, applied in this case, is perhaps too strictly defined. For it takes only the technical aspects into account, whereas the broad definition, expounded by WHO, includes also the social and cultural dimensions. According to the WHO definition of appropriate technology:

These words are spoken to health personnel as the focal audience in authoritative voice with an advisory tone. It is unquestionably fine in the company of the development agencies with all their best intentions and experiences. But it implies that it is they who run the show, and the folk, the stakeholders themselves, are just the beneficiaries. Underlying the definition is the articulation of the personnel and implicitly the folk are in the back bench. As a matter of fact, it is unlikely that scientists will find what is appropriate for folk. "Culture", central in the definition, does not designate a definite meaning with consensus. On the contrary, it is an elusive term, and it involves with the whole way of life. We all consciously know, though occasionally are forgetful, that we do not have access to others' minds. Even anthropologists find it impossible to perceive "what the natives perceive".

Understanding the folk's outlook thus is a hermeneutic process.<sup>12</sup> How to make technology compatible with the local culture is not a matter of a get-it-right solution which is waiting to be discovered. It is a process in which all parties share at every step. That is to say, to tackle the problem from the scientific point of view is totally inadequate. The folk's point of view should constantly be regarded right from the outset. To put it in another way both the folk considerations in the field together with the scientific formulation should constitute a loyal partnership with the emphasis on the articulation of the folk. Paradoxically it may sound rather idealistic, but in point of facts it is pragmatic. The point is contrary to a rather common working method that the outcome from the laboratory blackboard or the meeting room is put to the test in the field. The introduction of bone char is an exemplary lesson of the implementation of scientific measures without a dialogic consultation in a democratic spirit between the technico-medical expertise and folk wisdom. Putting the above abstract definition against the concrete situation of our cases, one critical component absent from the meaning of appropriate technology is that it does not incorporate the stakeholder into every single phase of the decision-making process. The essence of the definition, put simply, privileges the personnel in their best endeavours to achieve the technology appropriate for the folk.

The facts and the points above perhaps could well illustrate that the WHO definition is insufficient. If the scientists tend to think for the villagers, their works will more likely be scientist-centric. The adoption of a technological design is to be openly debated and jointly determined by the villagers and the scientists side by side. Hence faith in technology as a means of curing problems needs to be unified with the consideration on villagers' value choice. Appropriate technology needs to be defined along the line of "folk-directed technology".

# WHICH WAY TO DEVELOPMENT?

We see our attempts to solve the fluorosis problem not as a specific isolated case but as a case in point of developmental thinking. The term "development" has become a stock in trade in various fields of knowledge. A number of its conceptual variations, throughout the latter half of this century, have appeared and disappeared from the academic market. They have been, for example, "integrated development", "community-based development", "people-centred development", " participatory development", "alternative development", "sustainable development", and many other minor versions. The trend, conceptualised very positively, is to increasingly incorporate people into the process. But the question remains not merely 'how' in the sense of working method, but more fundamentally it is about the world outlook to development which underpins the way of thinking.

Development, in the case of our solving fluorosis problems, requires the comprehensive concerted efforts of various contributors. They range from the international organisation such as WHO, the national health ministry, the state administrators under different authorities, health personnel, scientists and, last but not least, the folk. Each contributor has her/his respective function. The success story entails that 'the last in the list' must not, as commonly taken, be directed but they must direct their own course. This 'folk in command' outlook does not mean that we are to go to the extreme, namely by holding the people to be supreme and ultimate which is commonly advocated among some variations of populism. The roles of

professionals should be conducted in such ways as to be supportive of people's awareness and actions.

The liberal version of the orthodoxy of development has rendered one of its key concepts popular. It is the slogan of 'people participation'. Admirable as it may sound; however, with hindsight of our experience, our thinking points in another direction. That is, if the people's aspiration, the actual determinant in change, is to be articulated, probably a reversal in thinking is called for. The questions regarding the development for people and not for professionals then are: in place of 'people participation', should it not be 'professional participation'?. And should the technocrat/scientist not know her/his place so as to be not on top but on tap?

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# FLUOROSIS CONTROL IN THE RURAL DRINKING WATER SUPPLY AND SANITATION PROJECT, KARNATAKA, INDIA

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SUMMARY: The Danida assisted Rural Drinking Water Supply and Sanitation Project, Karnataka, India, faces a fluoride problem in some of its areas. 8-53 % of the water sources are estimated to have fluoride content between 1.5 and 3 mg/L. 2-5 % of the sources contain over 3 mg F/L. The project objectives fit well within the Government of India's overall aim to ensure a minimum of 40 litres per capita per day safe water to its rural population. The project classifies its areas in 4 categories: Non-problematic, Moderately fluoride affected, Fluoride affected and Severely fluoride affected. Different levels of interventions are adopted for each category.

Keywords: Fluorosis; India; Danida project; Defluoridation strategies.

#### INTRODUCTION

The Rural Drinking Water Supply and Sanitation (RDWSS) Project is a DANIDAassisted project aiming at improved and sustainable drinking water supply and sanitation in three districts of Karnataka India. The project has faced the fluoride problem, which occurs in some of its areas. This paper highlights some of the project's early findings and proposed problem solutions.

#### THE PROJECT

The RDWSSP is a DANIDA-assisted project aiming at improved and sustainable drinking water supply and sanitation in three districts of Karnataka, India. Among other things, the project envisages the promotion of a decentralised, demand-driven and participatory approach in project planning and implementation. This approach, though being largely focused on Village Councils (Gram Panchayats) as being the lowest, most appropriate level, will involve a wide range of both 'hard' and 'software' activities at other levels, i.e., village, district (taluk), and also state level. In addition to the state-level, departments and the respective district, taluk and gram-level Panchayat Raj Institutions (PRIs), the project organisation includes a Project Advisory Group (PAG) located in Bangalore, and one District Co-ordination Unit (DCU) in each the districts of Kolar, Chitradurga and Bijapur. These units, in addition to providing general advice and assistance in the project implementation, are considered to have an important responsibility in overall action research (R&D).

Based on the experiences of an earlier DANIDA-supported Integrated Rural Sanitation and Water Supply Project (1990-1996), and following the findings of preliminary 'quick' water quality surveys and studies in the project areas, the project partners identified the "development and testing of appropriate solutions to excessive fluoride contents in the groundwater" as one of the four critical, immediate objectives of the project.

## NATIONAL FLUOROSIS STRATEGY

During 1986, the Government of India (GOI) introduced the "Technology Mission on Safe Drinking Water" (changed to "Rajiv Gandhi National Drinking Water Mission" in the early 1990s) for providing 'potable' water to the people of rural India. As part

of this initiative, and to address the specific water quality problems in a focused manner, the Mission identified a number of sub-missions of which the Sub-mission on the "Control of Fluorosis" was one. The Sub-mission's activities started in 1987, with the aim to achieve the following, important objectives:

- to update and create awareness on the relation between fluoride and fluorosis;
- to facilitate/conduct health and water quality surveys in the affected areas; and
- to introduce ameliorative and preventive measures for prevention and control of fluorosis.

Ultimate goal of the activities has been; (i) to provide safe water (i.e., with fluoride levels not exceeding 1.5 mg/L); and (ii) to control and prevent fluorosis in endemic areas.

The above objectives/goals fit well within the GOI's overall aim to ensure adequate access for the rural population to a minimum supply of 40 liters per capita per day (lpcd) of safe water.

# PAST PROJECT EXPERIENCES

In the course of an earlier DANIDA-supported Integrated Rural Sanitation and Water Supply (IRS&WS) Project, which was implemented by the Government of Karnataka (GOK) between 1990 and 1996, considerable experience was gained with both the identification and addressing of fluoride problems in rural areas. In line with existing government guidelines the 'normal' approach in this earlier project was to establish new, if necessary distant, sources in those areas where no existing 'safe' sources could be identified (i.e., with F<1.5 mg /L).

Being faced with a situation in which even distant sources could often not be found, the project, towards the end of its implementation, embarked on a number of R&D activities which included several field-level orientation awareness camps. Moreover a detailed study related to the demand for, and the introduction and general utilization of, domestic defluoridation units based on the Nalgonda technique in rural households.<sup>2</sup> In addition, the project supported the installation of one community defluoridation plant, based on activated alumina filtration, in one of the habitations, while the local government sanctioned three similar plants in other villages.

A wide range of specific issues and problems related to local capacity and capabilities were encountered, e.g., to carry out water quality testing, to secure continued high quality manufacture, supply and distribution of spares, tools and chemicals, and to ensure proper, local-based management of the community plants. In addition an important finding was that the success of defluoridation and/or of any other fluoride/fluorosis control measure largely depends on the people's, often individual, knowledge, perceptions and beliefs with regard to fluorosis. Hence, one of the major conclusions was that increased efforts would be required to carry out large-scale awareness creation campaigns throughout the rural areas.

With regard to household and community defluoridation, the findings of the IRS&WS Project generally supported the prevailing view that:

<sup>&</sup>lt;sup>2</sup> A report on the Experiences with Household Defluoridation in Peddur and Kondamvarripalli Villages in Bagepalli Taluk, Kolar District, Carel P.M. de Groot and B.H. Vasudev

- household defluoridation may be considered an option for those households which occupants are, first of all, convinced that the practice will protect themselves and their families, and who, at the same time, can afford to buy, operate and maintain the (available) units; and
- community plants, or, for that matter any other O&M-intensive solution, should not be considered unless there adequate, reliable and sustainable arrangements are in place for the overall management of such systems.

# **FLUORIDE OCCURENCE**

Based on the findings of water quality studies carried out during the IRS&WS Project (1990-1996) as well as the current RDWSS Project (1996-1997), excessive fluoride levels (i.e., F>1.5 mg /L) in the groundwater is anticipated to prevail in several parts of the project areas, in particular in the two Districts of Kolar and Chitradurga. At the same time, it has been noted that the fluoride levels in these effected sources are moderately high, and do not generally exceed 3.0-3.5 mg /L.

**TABLE 1.** Percentage of water sources with different fluoride contents in the stated districts and percentage of villages with cases of fluorosis.

District	Taluk (samples)	% of Sources having conc. in mg/L			Villages with
		<1.5 mg /L	1.5-3.0 mg/L	>3.0 mg /L	Fluorosis cases
Kolar	Bangarpet (257)	85	15	0	15
	Gudibanda (58)	81	19	0	35
Chitradurga	Hiriyur (173)	45	53	2	28
	Hosadurga (209)	73	22	5	22
Bijapur	Bagalkot (100)	63	32	5	20
	Jamakhandi (73)	89	8	3	0

Table 1 presents the findings of 'quick' surveys in only six of the total 31 taluks in the project area. It can be seen that the total number of sources affected with fluoride ranges from about 11% (Jamakhandi) to 57% (Hiriyur). On the other hand, only 2-5% of the total number of sources have fluoride levels exceeding 3 mg/L.

## **CONTROL STRATEGY**

In order to allow the project to address the fluoride-related problems in a focused, cost-effective, appropriate and operational manner, it has been proposed to develop and introduce a strategy, based on the following, guiding principles<sup>3</sup>:

the project will carry out intensive and extensive fluorosis control programmes in Gram Panchayat where sources contain fluoride (>1.5 mg /L), and where it is not feasible to supply the area/habitations from nearby sources. This programme should include awareness campaigns, training and orientation of Gram Panchayat officials, and promotion of household defluoridation;

in villages/areas where the fluoride content exceeds 3.0 mg/L without feasible distant sources, the project will, in collaboration with the engineering sections and with possible assistance from other experts in the field, assess the solution(s) to be adopted on a case-by-case basis; and where water quality problems (fluoride, hardness,

<sup>&</sup>lt;sup>3</sup> this being a discussion paper, it is important to note here that both the 'guiding principles', as well as the ensuing strategy, are still under study, and that they are yet to be approved and endorsed by the project authorities in Karnataka.
salinity, nitrate, etc.) constitute a major impediment in ensuring a full (40-55 lpcd) supply of safe drinking water, including water for cooking, to the population, the project may - in close association with the Gram Panchayats and subject to extensive awareness creation programmes among the villagers - consider, plan and implement water supply works based on the principle of "dual supply", i.e., providing for at least 10 lpcd of safe drinking water, while allowing other (point) sources, not necessarily potable, to be used for other purposes such as washing, bathing, etc.

While recognising that adoption of the above principles would imply a compromise of the existing standards, as currently specified by the Government of India, it is assumed that they will enable the project to develop a strategy, as suggested in the sections below, which will be both implementable, as well as realistic in terms of funding requirements, technical feasibility, and sustainability. More specifically, it is assumed that the guiding principles will enable the project to:

- rank the problem areas in relation to (a) the extent/geographical distribution, and
   (b) the level/concentration, of the fluoride problem;
- prioritise its focus and activities on those areas with the most serious problems first (i.e., where fluoride exceeds 3.0 mg/L), while, on the other hand, leaving
- ample mandate and opportunity to carry out intensive fluorosis awareness campaigns and promotion of household defluoridation in the remaining fluoride-effected areas; and
- identify and promote appropriate, technically feasible, affordable, and sustainable solutions to ensure adequate and reliable water supply to the rural population (both for drinking as well as for other purposes), rather than being 'forced into' adopting technically sophisticated, high-cost, and O&M-intensive solutions which, under the prevailing rural conditions, tend to be non-sustainable any way.

### TABLE 2 Defined classification of the project areas.

## I Non-problematic

Yield and water quality data indicate that the project will be able to ensure a minimum of 40 lpcd of safe drinking water through rehabilitation and/or augmentation of the existing system/technology. Any other sources, though not necessarily potable, can be used for other purposes.

#### II Moderately fluoride effected

The data indicate that the project will be able to ensure 10 lpcd of safe drinking water through rehabilitation and/or augmentation of the existing system. Any other sources, though not necessarily potable, will make up for the balance of 30 lpcd, and can be used for other purposes.

### **III Fluoride affected**

The data indicate that fluoride levels in the area are generally in the range of 1-5- 3.0 mg/L, that it will not be possible to ensure a minimum of 10 lpcd of safe drinking water through the mere rehabilitation and/or augmentation of the existing system.

#### IV Severely fluoride effected

The data indicate that most of the sources in the area have fluoride levels exceeding 3.0 mg/L, and thus, that it will not be possible to ensure 10 lpcd of safe drinking water through the mere rehabilitation and/or augmentation of the existing system.

<b>Table 3</b> Area type and corresponding in	erventions
Area Classification Preferred Intervention/Solution	Alternative Intervention/Solution <sup>1)</sup>
I Non-problematic	
<ul> <li>Rehabilitation of existing supply;</li> <li>Information to users about which sources are fit for drinking and which , if any, are not is the only action required</li> </ul>	Alternative inventions not applicable; the preferred solution is assumed to be feasible.
II Medewatala flagaride officiated	
<ul> <li>Rehabilitation of existing supply is envisaged to ensure a minimum supply of 10 lpcd safe water:</li> </ul>	Alternative inventions not applicable; the preferred solution is assumed to be feasible.
<ul> <li>Physical identification of safe sources and those to be used for other purposes.</li> </ul>	
<ul> <li>Intensive and extensive campaigns at both habitation and Gram Panchayat level to create and sustain awareness about the water quality problems and to promote proper protection, use and maintenance of safe sources for drinking.</li> <li>III Fluoride affected</li> </ul>	
• Upgrading of the existing technology	In case the preferred option does not appear to be
• Identification and establishment of an	feasible, the following combination of actions
alternative, (more) distant source to	will be required:
ensure a minimum of 40 lpcd safe	Rehabilitate/augment the existing technology
drinking water.	utilising the "most potable" sources.
<ul> <li>Information to users about which sources are fit for drinking and which, if any, are not, is the only action required.</li> <li>IV Severely fluoride affected</li> </ul>	Identify and provide adequate protection measures for all safe sources (even though the combined capacity may be less than 10 lpcd) Intensive and extensive campaigns at both habitation and GP level to create and sustain awareness about the water quality problems and promote proper protection, use and maintenance of the (scarcely available) safe sources for drinking. Promote household defluoridation, a/o through the training/orientation of key motivators/staff in the GP and through the support of local manufacturing, supply, distribution, and maintenance systems.
<ul> <li>Ungrading of the existing technology</li> </ul>	In case the preferred option does not appear to be
<ul> <li>Identification and establishment of an alternative, (more) distant source to ensure a minimum of 40 lpcd safe drinking water.</li> <li>Information to users about which sources are fit for drinking and which . if any, are not is the only action</li> </ul>	feasible, other options will have to be considered on a <b>case-by-case basis</b> . Such intervention, which could include surface water intakes, regional schemes, community defluoridation, distribution and maintenance (i.e., active promotion) of Household Defluoridation, will have to be identified and
required.	implemented in close association with the relevant government departments and line agencies.
"to be considered only if/when the preferred	d option is not (or, does not turn out to be) feasible

# Table 3 Area type and corresponding interventions

## **AREA CLASSIFICATION**

Based on the project's past experiences, and taking into consideration both the existing GOI/RGNDWM guidelines, it is proposed to base the identification and classification of fluoride-effected areas on the following important parameters/factors:

- the quality of sources
- the (combined) capacity of the sources
- the distance of sources from the habitation
- the type of water supply scheme envisaged and
- the intended and/or potential use of the sources.

Bearing in mind the above parameters, and taking a habitation as the basis for the area assessment and classification, the categories are suggested as shown in Table 2.

## **PROPOSED INTERVENTIONS**

Based on the above classification, and taking into account the financial and organisational and institutional constraints faced in the field, both within the project organisation as well as among the implementing agencies, the project would subsequently be able to identify different sets of solutions/interventions for different problem areas. As further elaborated upon in Table 3, the interventions could be further sub-divided into the following two groups, i.e.:

- preferred solutions/interventions, which, in all cases, would imply the identification and establishment of alternative, if necessary distant, sources to ensure an adequate supply of 'safe' drinking; and
- alternative solutions for those case where no alternative 'safe' sources can be identified; depending on the extent and seriousness of the problem, such solutions could range from carrying out basic fluorosis awareness campaigns, to adopting technological solutions such as defluoridation, (semi-)regional schemes, surface water intakes with treatment, etc..

# INFORMATION ABOUT ORAL HEALTH AMONG WOMEN ATTENDING HEALTH CLINICS IN ARUSHA, TANZANIA.

# A N Åstrøm<sup>\*</sup>, A K Awadia<sup>\*</sup>, and O Chande<sup>\*\*</sup>

## Norway and Tanzania

Summary: 140 women from Arusha, of who 50 % were affected by dental fluorosis, were interviewed. Only 27.5 % had ever heard about fluoride and their knowledge of the links between exposure to fluoride and health injuries was found to be rather low. The participants reported to receive most information about tooth decay from the sources they trusted most, health personnel in terms of dental auxiliaries and Mother-Child Health aids. Perceived personal risk or perceived likelihood of having dental caries, loss of teeth, gum disease, children with pitted, coloured and fractured teeth or injuries to the bones were below but close to average. Furthermore, perceived personal risk of having server tooth decay was significantly influenced by the amount of information received from health personnel and by the degree of trust in information from these sources in multivariate analyses. This result underscores the important role of health workers as providers of oral health information.

Keywords: Fluoride; Fluorosis; Oral Health; Perceived risks; Health information; Arusha.

## INTRODUCTION

Due to high fluoride concentrations in drinking water, dental fluorosis is endemic in those parts of Northern Tanzania that belong to the East African Rift Valley.<sup>1</sup> Otherwise, oral diseases have traditionally not been among the most prominent health problems in Tanzania.<sup>2,3</sup> It is anticipated, however, that the caries prevalence will increase due to industrialization and economic progress with changed dietary habits in terms of increased sugar consumption and its refined by-products.<sup>4</sup> Apparently, there seems to be an urgent need for influencing dental care behaviors in the general population. Awareness and capability among the general population to take over a fair share of the responsibility for their own oral health will reduce the need for dental manpower and be in accordance with the self-reliance philosophy of this country.

Despite of considerable work done for oral health education in Tanzania, these activities have been evaluated to a lesser extent, both on the part of the promoters and the recipients <sup>5.</sup> Some studies performed soon after implementation of educational intervention have indicated poor oral health knowledge among primary school children.<sup>5,6</sup> Apparent gaps in oral health related knowledge and behaviour among children entering primary schools, indicate low parental contribution to their preschool children regarding oral health issues. This provides some information about the success of educational activities provided to mothers as a means of reaching their pre school children.<sup>7</sup>

Personal beliefs about susceptibility to harm and diseases are cited as factors in the failure to change behaviour by education.<sup>8,9</sup> Knowledge might not lead to a change in risky behaviour among those people who do not perceive their susceptibility of contracting disease as high.<sup>10</sup> Hence, for health education to persuade individuals to change behaviour, those who provide the information must understand how individuals perceive their own susceptibility. While it appears that knowledge alone does not predict behaviour, perceived personal risk might do so. Therefore it seems to be important to study how information and perceived personal risk are related to each other as well as to preventive behaviour.

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Previous investigations have suggested that people use direct experience to infer their perception of risk or susceptibility.<sup>11</sup> For instance, more risky behaviour in the past have usually resulted in higher levels of perceived risk, and ill people seem to feel more susceptible to health problems than to non-health problems.<sup>12,13</sup> Therefore, it was expected that behaviours leading to health and dental health problems and direct personal experience with health hazards result in higher risk appraisal.<sup>12</sup>

The present study set out to assess perceived personal risk/susceptibility with regard to a variety of oral health related problems in a sample of Tanzanian mothers differing in age, education and occupational status. The ways in which women's perceived personal susceptibility of having server tooth decay might be influenced by the amount of information received from various sources, trust in these informational sources and their level of oral health knowledge are also investigated. Finally, the women's personal experience with symptoms related to tooth decay and their frequency of sugar intake were examined as predictors of perceived personal risk of having server tooth decay. Such information might be valuable for oral health educators by providing a basis for the planning and improvement of further educational activities.

## MATERIALS AND METHODS

**Sample and survey instrument.** A convenient eligible sample 140 women attending the Mother and Child Health Clinic (MCH) at Mount Meru regional hospital in Arusha volunteered to participate in a structured interview performed by research staff. Most of the women were young, in fact 59.6 % was between 15 and 25 years old (range 15-40 years). A total of 81 % of the women investigated reported to be married. Furthermore, 65 % reported primary school, 25 % secondary school, and only 3 % university or college as their highest educational level. The most frequently reported type of employment was housewife, 61.9 %. Nevertheless, 40 % of the women reported to be engaged as farmers, teachers and clerks.

After one week, a total of twelve women were re-interviewed by the same researchers. Test - retest reliability scores were in the range Pearson's r=.80 to .90 concerning the central variables utilised in this study.

**Statistical analyses.** The Statistical Packages for Social Sciences (SPSS, version 7.5) were use for statistical analyses of the present data.

**Measures.** A comprehensive, structured questionnaire was included in this study and used as a controlled interview schedule. This research instrument, originally constructed in English was translated to Swahili. The research team, which comprised two experienced researchers, one of them recruited from the Arusha regional hospital, had a training session prior to the actual fieldwork.

In addition to assess information about demographic factors in terms of age, marital status, level of education, and current work, the respondents were assessed on the following measures:

Awareness of a substance called fluoride was assessed in terms of "Have you ever heard /learned about fluoride- a substance commonly found in drinking water, toothpaste and several food items?". Response categories were: 1= "yes", 2= "no".

Risk awareness of fluoride related injuries to health and oral health was measured by the following questions: "Have you ever heard that pitted, discoloured and fractured teeth may be caused by excessive exposure to fluoride?". Then "Have you ever heard that ingestion of high levels of fluoride from drinking water and food items might cause injuries in your bones?" Response categories for these two questions were 1 = "yes" and 2 = "no".

**Presence/absence of dental fluorosis.** A crude dental examination with regard to the presence/absence of dental fluorosis was performed in daylight by use of dental probe and mirror. This variable was coded 1= presence of dental fluorosis and 2= absence of dental fluorosis.

**Symptoms related to tooth decay** was based on a sum score derived from the following questions. "How often have you had toothache, bad breath, food impact and difficulties with chewing food items?" The response categories ranged from 1= "often" to 3= "seldom / never".

Amount of information from different sources was assessed by asking the women how much information about tooth decay, they had received from five different sources in terms of magazines/newspapers, dental auxiliaries, radio/TV and MCH aids, as well as how trusted were each source of information. The response categories ranged from 1= "very much" to 4= "non at all".

The amount of information about tooth decay (caries), received from MCH aids and dental auxiliaries and how trusted were these two sources of information were added into two sum scores yielding amount of information from **health workers** and trust in information from health workers. Furthermore, amount of information from radio/television and newspapers/magazines and trust in those sources were added into two sum scores yielding amount of information from **media** and trust in information from media.

**Level of oral health knowledge.** The participants also rated how well informed they were about tooth decay as compared to other mothers of their own age living nearby. Response categories ranged from 1= "better informed than other mothers living nearby" to 3 = "worse informed than other mothers living nearby"

**Risk behaviour related to tooth decay** was based on a sum score derived from three questions assessing the frequency of intake of soda, chocolate/sweets and cakes/biscuits. Response categories for each item ranged from: 1= "daily" to 3= "seldom never".

**Perceived personal risk** of having tooth decay, gum disease, children with pitted, coloured and fractured teeth, loss of teeth and injuries in bones was assessed by five separate questions. An example "How likely or unlikely do you think it is that you sometimes in your lifetime will acquire server tooth decay "Response categories on each question ranged from +2= "very likely" threw 0= "neither likely nor unlikely" to -2= "very unlikely".

### RESULTS

Awareness of a substance called fluoride. As shown in Table 1, only 31 % of the total sample had ever heard about fluoride and 62 % was not aware of the link between excessive ingestion of fluoride and oral health hazards. Finally, 95 % answered no when asked if they knew that ingestion of fluoride might cause injuries in the bones. Because most of the respondents had low awareness regarding fluoride and related oral health problems and the eligible sample was rather restricted, further

analyses of the predictors of women's perceived risk of having children with dental fluorosis and their perceived susceptibility of acquiring injuries in bones could not be performed.

<b>TADLE 1.</b> Frequency of distribution of response to questionnalite and testing	TABI	LE 1.	Frequency	of distribution	of response to	questionnaire	and testing
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	Yes		No		Total	
	Ν	%	Ν	%	Ν	%
Ever heard of fluoride	43	31.4	94	68.6	137	100
May cause injuries in teeth	34	37.8	56	62.2	90	100
May cause injuries to bones	6	4.5	128	95.5	134	100
Dental fluorosis among women attending clinic in Arusha	63	45.3	76	54.7	139	100

Absence/presence of dental fluorosis. As shown in Table 1, a total of 54.7 % of the women had no experience with dental fluorosis when examined in daylight with a dental mirror.

**Perceived personal risk** (susceptibility) with regard to oral health hazards. As shown in Table 2, the respondents perceived their personal risk or susceptibility to be close to but below **TABLE 2.** Means, standard deviations and range of reported amount of information about tooth decay from several sources and reported trust in these informational sources.

	Ν	Mean	SD	Range
Source of information				
Dental auxiliary	101	3.3	1.1	1-4
MCH aids	91	3.5	.99	1-4
Magazines/newspapers	88	3.4	.89	1-4
Radio/television	95	3.5	.88	1-4
Trust in information				
MCH aids	93	2.8	1.3	1-4
Dental auxiliaries	102	2.9	1.2	1-4
Magazines/newspapers	95	3.2	1.0	1-4
Radio/television	98	3.3	.99	1-4

average regarding tooth decay, gum disease, loss of teeth, children with pitted, coloured and fractured teeth and injuries in the bones.

Table 3 and 4 show descriptive statistics for the predictors of perceived susceptibility of having server tooth decay in terms of mean (median) standard deviations and range. As a whole there appeared to be minor differences in the amount of information received from newspapers and magazines, radio or television, dental auxiliaries and MCH aids as illustrated in Table 3. However, there is a slight indication in the data that the women received most information from dental auxiliaries, the source they trusted most.

**Correlates of perceived personal risk of having server tooth decay (bivariate analyses).** According to Table 5, significant positive associations in terms of Pearson's correlation coefficients emerged between perceived personal risk of having tooth decay and amount of information from and trust in health workers and media.

**TABLE 3.** One sample t- statistics for perceived personal risk (perceived susceptibility) of having tooth decay, gum disease, children with pitted, coloured and fractured teeth, loss of teeth and injuries in bones.(scale: very likely=+2, neither likely nor unlikely = 0, very unlikely = -2.). Significant levels refer to t-tests of the hypothesis that the mean is different from zero.

	Ν	Mean	SD	Т	Sig (2-tailed)
Tooth decay	139	13	1.01	-1.5	.282
Gum disease	138	30	.96	-3.60	.000
Pitted/discoloured/fractured teeth	148	55	.99	-6.6	.000
Loose all teeth	149	79	1.2	-8.3	.000
Injuries in bones	145	77	.80	-11.0	.000

**TABLE 4.** Descriptive statistics in terms of mean, (median\*), standard deviation (SD) and range of the predictors of perceived risk of having server tooth decay in bivariate and multivariate analyses.

	Mean	SD	Range
Sum score-information from health workers	6.9	1.4	2-8
Sum score-information from media	7.0	1.1	4-8
Sum score-trust in health workers	6.0	1.8	2-8
Sum score-trust in media	6.7	1.5	2-8
Level of information about caries*	2.0	0.7	1-3
Sum score-personal experience with symptoms	11.0	1.2	8-12
Sum score-intake of sugary products	7.2	1.3	3-9

**TABLE 5.** Factors associated with perceived likelihood (personal risk) of having tooth decay among young women in Arusha. (Bivariate analyses, Pearson's correlation =r).

/	
Predictors	Perceived personal risk/tooth decay (criterion)
Source of information	
Health workers/sum index	.45 **
Media/sum index	.32 **
Trust in sources	
Health workers/sum index	.47 **
Media/sum index	.40 **
Knowledge about caries	.16 n.s
Past experience	
Intake of sugary products/sum index	.19 *
Caries related symptoms/sum index	.25 **
* p<0.05, **p<0.000	
n.s not statistically significant	

Furthermore, there was a positive bivariate relationship between perceived risk and personal experience with symptoms related to tooth decay. Hence, the more information from health workers and media and the more trust in these informational sources, the higher the level of risk appraisal. In addition, these women take past experience into account when judging their personal susceptibility. The more frequent experience with symptoms related to tooth decay and intake of sugary products, the higher their reported level of perceived risk. This supports our expectation and is

186

consistent with findings in other health related domains. Level of knowledge, on the other hand, did not seem to be of significance for these women's perceived susceptibility of having tooth decay.

**Predictors of perceived risk of having tooth decay (multivariate analyses).** The predictors of perceived risk (likelihood) of having tooth decay shown in Table 5 were not independent, and much of the variance apparently explained by the independent variables is redundant. Multiple regression calculations in which all variables were entered simultaneously into the prediction equation were carried out in order to determine which variables make unique contributions to the prediction of perceived vulnerability, how much variance can be predicted by these variables and whether the variance is greater than that expected by chance alone. Stepwise regression analyses which is more vulnerable to chance associations was then used to determine the most parsimonious set of predictors. Only independent variables significantly related to the dependent variable in the bivariate analyses were entered into the multivariate analyses.

**TABLE 6.** Summary statistics and results from stepwise regression analyses for the prediction of perceived personal risk of having tooth decay in a sample of Tanzanian women. Standardised regression coefficients beta.

Prediction model	Beta	Sig.
Age (control variable)	.13	.168
Information from health workers	.32	.010
Trust in information from health workers	.26	.073
Information from media	15	.300
Trust in information from media	.10	.547
Experience with symptoms related to tooth decay	.068	.468
Sugar related behaviour	.056	.574
R square = .35, F(7,78)=6.1 p<0.000).		

Table 6, presents the results of the multivariate stepwise regression analyses in terms of summary statistics for the sample. This table presents the standardised regression coefficients, betas, which is the correlation between each independent and the dependent variable after controlling for all other independent variables in the model. With all predictors entered the full regression model was significant, R square change = .35, F(7,78)=6.1 p<0.000), i.e. 35% of the variance in perceived likelihood of having tooth decay was explained by the model. Table 6 shows that when all variables were in the equation, only one of the individual predictors, amount of information from health workers, contributed significantly, while trust in information from health workers approached significance. Hence, for these women, the more information received from health workers and the more trust in information from these particular sources the higher risk appraisal for future oral health problems in terms of tooth decay.

#### Discussion

**Predictors of perceived susceptibility in terms of indirect and direct experience with oral health hazards.** It might be argued that changing risk perception is a goal of oral health education, if perceived risk is indeed a bridge to oral health behaviour change. In order to change risk perception the relationship between perceived risk and level of information, sources of information and trust of these sources should be established. An interesting aspect of these data was how sources of information, levels of trust, past experience with oral health problems and previous risk behaviour might influence women's perceived susceptibility. In this present study it is focused upon how these variables may influence perceived personal risk of having server tooth decay.

The results from bivariate analyses indicate that the mother's perceived susceptibility of having tooth decay is based on the amount of information they receive from health workers and media, trust in these sources as well as direct experience with individual risk factors. In contrast, women's level of knowledge did not seem to be that important in arriving at personal risk judgements. From one point of view, the present data clearly support a common-sense premise of health education confirmed in previous studies;<sup>14</sup> that peoples perception of susceptibility is influenced by exposure to information about such risks. Hence, amount of information from health workers and trust in these informational sources were significantly associated with perceived susceptibility of having tooth decay both in bivariate and multivariate analyses. On the other hand, the insignificant role played by women's level of knowledge as compared to the significance of their personal risk experience, points to the notion that direct experience are superior to information transmitted via other people and the media, in influencing perceived personal risk.<sup>15</sup>

The data in table 5 and 6 might have implications for preventive oral health programs. Since previous experience with disease and risk behavior seems to constitute important influences of perceived personal risk, providers of oral health education need to go beyond the plain provision of general oral health information. Rather, they must try to make certain that the recipients actually apply information to themselves and form personal risk perceptions that reflect their own standing on different risk factors. In this regard, it has been suggested that an understanding of the powerful but complex effect of direct personal experience may shed light on the determinants of self protective behavior.<sup>11</sup> In other words, when people are exposed to information that have some of the ingredients of direct experience such exposure may feel "just as if it happened to me", and be as effective in influencing personal risk perceptions. In fact, an intervention derived from these line of reasoning proved successful in increasing the use of automobile seat belts.<sup>16</sup>

Nevertheless, the present results underscore the important role of health workers as providers of oral health information. Women attending MCH clinics reported to receive approximately as much information from health workers as from other sources in terms of radio/television and newspapers/magazines. However, they trusted the information from experts sources the most, such as dental auxiliaries and MCH aids. One implication for health education is that health workers besides of providing information in clinical settings also should place information through other sources such as magazines, newspapers, radio and television. The need to tie popular sources of information to the most trusted ones seems apparent.

In spite of the endemic nature of dental fluorosis due to high fluoride drinking water in Arusha, approximately half of the women investigated in this study showed no visible signs of fluorosis when examined in daylight with a dental mirror. This might reflect that many inhabitants of Arusha have moved to this town as adults. Consequently, they might have grown up in parts of the country where the Fluoride content of drinking water is below the critical value. Unfortunately, there was no information on whether the participants of this study were life time residents of Arusha or not.

**Methodological considerations.** In spite of restriction in the range of the measures utilised in this study, the correlation with perceived risk was strong and the significant regression coefficients quite large. It should be noted, however, that the small sample size constrains the confidence in the observed relationships. This means that some measures which did not satisfy the criterion for significance but which may be true predictors of perceived risk have been ignored and some measures determined as salient might prove unstable or unreliable in repeated samples. As such the findings of this present study should be interpreted with caution. Apparently, further investigations are needed.

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# CRITICAL SUSTAINABILITY PARAMETERS IN DEFLUORIDATION OF DRINKING WATER

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SUMMARY: Experiences from household and community defluoridation projects have been collected. They are presented in the form of critical parameters that need to be considered for the success of household defluoridation projects. Parameters are classified in three groups. Motivation of households seems to be critical since fluorosis is not always considered as the main problem of concern and improvements are not always visible for a number of years. Appropriate and cheap technique is always a must in poor villages. Finally the organisation of supporting functions that may include quality control, technical and motivating support, as well as general educational initiatives.

**Key words:** Defluoridation; Water supply projects; Critical parameters; Appropriate technology; Sustainability criteria.

#### INTRODUCTION

Defluoridation of drinking water in third world countries has generally been unsuccessful in spite of many attempts to implement projects aiming at provision of safe drinking water in fluorotic areas. Furthermore, no defluoridation initiatives have been taken in many areas where fluorosis is prevalent, probably due to lack of knowledge of appropriate technologies or lack of means to provide all the project components associated with defluoridation. The difficulties to achieve sustainable results are illustrated by the fact that very few projects are actually practising defluoridation at present.

A review has been initiated by the Danish International Development Agency, Danida, in order to assess practical defluoridation experiences in selected areas and to identify critical parameters in defluoridation projects. This paper presents the preliminary findings by the reviewer, based on study visits to defluoridation projects in India, Sri Lanka and Tanzania, interviews with a number of researchers and practitioners in the field of defluoridation and experiences expressed in project reports and scientific papers.

It seems practical to classify the critical parameters into three groups, each of which is absolutely essential to consider in any defluoridation project:

- Motivation of users.
- Appropriate and economic technique.
- Proper organisational setup.

Basically, defluoridation can be introduced at two organisational levels; as *household defluoridation*, carried out by the single households for their own consumption, and as *community defluoridation*, carried out for the public in a village, town, sub-village etc. Experiences with other forms like institutional defluoridation (schools, health centres etc.) or private defluoridators shared by several households are presently limited. The specific parameters are in the following grouped in household defluoridation and community defluoridation.

## HOUSEHOLD DEFLUORIDATION

Defluoridation of drinking water has been implemented in different numbers of households. The Nalgonda technique, i.e. flocculation with alum and lime, has been experienced in India and Tanzania. Adsorption in activated alumina columns has been experienced in India and in crushed brick columns in Sri Lanka. Adsorption in bone char columns has been experienced in Thailand, Sri Lanka and Tanzania.

**Motivation.** Motivation of users to actually procure and use an available defluoridation technique is absolutely essential in household defluoridation. Any of the selected techniques requires attendance and an extra workload and payment either daily in case of the Nalgonda technique or (as an example) tri-monthly in the case of the column adsorption methods. To carry out defluoridation on top of the other burdens often faced in the regarded countries requires that the "water manager" in the household, almost always the woman, is properly motivated to do so.

First of all it requires awareness about the advantages of defluoridation, i.e. the possibility of reducing the skeletal and non-skeletal fluorosis among the family members. Many villages have only been exposed to fluoride during few years and the examples of fluorosis are scarce. The awareness has been induced through an awareness camp, i.e. a public meeting in the village where fluoride's health hazards, sources of fluoride, the fluoride situation in the village and fluorosis prevention both through defluoridation and changes in dietary habits is introduced. Use of living examples of fluorosis victims in the village has been mentioned as effective in raising the understanding of the seriousness of the matter.

Awareness camps seem to be appropriate for teaching of fluorosis subjects to a large group in the first instance. It has however been mentioned that sometimes villagers tend to forget the knowledge with time.<sup>1</sup> A large number of villagers are often illiterate and more used to learn by experience than by teaching. Special attention has to be paid to the procurement of understandable handouts and perhaps also certain forms of reminder sessions or continuous information.

It is not always enough to be aware of the consequences to actually act accordingly. Like the smokers smoke, fully aware of the high risk of lung cancer, some people will not defluoridate the water even though they have means to do so and they have their own children as living examples of dental fluorosis victims. This irrational behaviour, whether attributed to lack of interest, ignorance or laziness, seems to be a general human character that must be realised.

A commitment to the investment done when the household has paid for the defluoridator has been mentioned as a driving force to carry out defluoridation. The feeling of ownership is generally considered as a positive factor in operation and maintenance.

One parameter that seems to hinder peoples motivation is the lack of viable results, since fluoride cannot be sensed and reduction in fluorosis prevalence are usually not seen the first many years. The Unicef supported project in Rajasthan has had success in making a health survey before introduction of defluoridation and a re-examination of fluorosis victims three months after defluoridation start. The victims felt a relief in both skeletal and non-skeletal symptoms. Furthermore an increase in the ability to make certain movements was measured.<sup>2</sup> The understanding of the positive benefits when the collective results were presented to the community has been judged to be

one of the most important motivating factor in the project. It may for this reason be much easier to motivate people to carry out household defluoridation in areas with high fluoride concentrations ( $\geq$ 4-5 mg/L) than in areas with lower fluoride concentrations (1.5-3 mg/L).

Factors that seem to enhance the motivation to some extent are other improvements in water quality, experiences like the brick defluoridation seems to cool the water and bone char may give a better taste. Among other factors that "motivates" is perhaps a continuous attention of the defluoridation project manager, combined with a commitment (like a signed contract) to carry out defluoridation for a number of years.

**Appropriate and economic technique.** Because of the difficulties in motivating people for defluoridation it is extremely important to choose a technique that is cheap and which requires only minor workload. At the same time it should be easy to learn and remember, even for illiterate people. Obviously it should not impart any adverse taste to the treated water and the maintenance and repair should be easy and affordable.

For the Nalgonda technique and other possible methods that need a daily operation it is important that the operation period is limited to a minimum. It has been experienced that it is very difficult for a housewife to find 15-20 undisturbed minutes every day to carry out defluoridation.<sup>1</sup> Probably 5 minutes for defluoridation should be considered as a maximum. This is also sufficient for the Nalgonda technique.<sup>3</sup>

For the column treatment methods the daily operation is normally negligible. The hurdle for the proper operation/maintenance is the regeneration or exchange of media, normally carried out every few months. This is either connected with a relatively heavy workload (crushing of bricks) or a relatively high investment in regeneration or new media. The cost may not seem high on a monthly basis, but when 3 months media has to be paid at one time it may seem overwhelming for a poor household thus pushing the payment till next month - and yet another month - etc. To reduce "irrational excuses" replacements and regeneration should be arranged as cheap and simple as possible.

All the mentioned defluoridation methods can be appropriate as such for household defluoridation, but proper designs and materials have to be selected. Users have expressed complaints of heavy manual workloads both in daily defluoridation practise and during exchange of media. This may, to a certain extent, hinder the success of the project. Simple procedures / easy operation and durable materials that are easy to maintain and repair, will be important if not crucial for the success of household defluoridation, especially where the motivation is not very high.

Safety of handling of chemicals for defluoridation and regeneration should be sought. Wrong dosage of e.g. alum in the Nalgonda technique may result in inappropriate pH in the treated water. Thereby fluoride removal will be less, but it is more crucial that the taste may become offensive and thereby demoralises people from using the method. Poor taste together with rumours about side effects of the treatment can be effective in discouraging people from drinking the water.<sup>1</sup> Variation in qualities of e.g. alum and lime may result in different fluoride removals and water qualities. Special attention has therefore to be paid to the fact that these chemicals are commercially available in different qualities.

There is at the moment not much experience with self-financing of defluoridation. The most obvious cost recovery method for household defluoridation would be self-payment of (at least) operation and maintenance costs. Obviously they should be kept at a level that people will pay. To meet the poor people's ability to pay, chemicals can be subsidised if there is economic basis to do so. That may also prevent people from buying commercial poor quality chemicals. The defluoridators should also be cheap enough for people to meet the costs. Otherwise there should be a public funding to provide or to subsidise the defluoridators.

It is experienced in various projects that defluoridators are in most cases too expensive to sell for the majority of the population. If no subsidies are available, an option like installation a tap in a bucket already at use in the household would in many cases be the only affordable solution. Moreover it has to be realised, that even methods which are known to be cheap, demand steady extra expenditure for water and the cost of chemicals, whether paid monthly or biannually, may be a significant factor of discouragement for many people to continue treating the water. More experience is needed with respect to people's willingness to pay, but there is a good reason to keep costs to a minimum.

**Proper organisational set-up.** Any defluoridation method would sooner or later need supply of chemicals for dosage or for regeneration of media and a reliable supply of spare parts and spare media. It is obvious that these needs can be covered either through arrangement with the private retailers or through the water authorities. The system of choice will depend on the local conditions. It is also obvious in case activated alumina or bone char is used, certain arrangements have to be made in order to ensure proper regeneration/supply of media.

It has been experienced that when introducing a technique utilising breakable parts, it is necessary to set up units to carry out minor repair of the households defluoridators.<sup>1</sup> That is because the households neither have the skills nor the simple tools to carry out the repair on their own. In some cases, the households do not give priority to buy even cheap spare parts or to spend the necessary time to do the repair. Easy access to technical support (both as adviser and manual support), e.g. a man in every village that has got specific training, may be needed to keep as many defluoridators as possible in operation.

In this connection it has also been experienced that people, after some time of operation, tend to forget procedures and/or to loose interest in the defluoridation. It may be necessary to ensure a continuous follow up of awareness or motivation among the households for an extended period of time. Collaboration with community development NGO's has been suggested and is being tried out in some projects. In the project using crushed bricks in Sri Lanka it has been decided to follow the implementation for at least five years. Such a follow up may be required in order for people to observe better oral health among children and to ensure the sustainability of the implementations. Organisation of the provision of qualitative test kits for fluoride measurements based on the alizarin method may be useful for the households to gain assurance about the efficiency of treatment (motivation).<sup>5</sup> However still this method needs more documentation about its usability in practise as an on/off method.

The preceding applies for the local organisation necessary for maintaining household defluoridation. The superior water authority needs to play an active role in at least the control and monitoring functions.

A certain control of, ultimately, the quality of the treated water in the households is necessary. Frequent sampling of treated water for analysis can be used to point out problems in operation. The water authorities should carry this out. Other kinds of water quality assurance will include control of quality of chemicals and control that these are actually used by the households. Finally the responsible authorities should control the procedures and guidelines given for defluoridation. The Nalgonda technique represents a special problem that should be addressed, because the quality of the alum and lime is critical and so is the dosage of chemicals, that ultimately needs to be determined for every single water source (at different times of the year) and every single bucket size.

It may be crucial for a large scale implementation that some functionaries at all levels of the water supply and health authorities are aware of the fluorosis problem and the possible means to solve it. Higher levels of administration will only be able to advice the lower executing levels about the strategies if they themselves are aware of the fluoride problem and its implications. This would require a general educational program in the administration.

Finally the water authorities should be the institution that collects the experiences gained in the particular geographical and cultural environment since no technique will be applicable the same way in all environments. Together with the health authorities they have an important role to play in surveying the health (fluorosis) status as a result of defluoridation programmes.

## **COMMUNITY PLANT DEFLUORIDATION**

Many practitioners as an intermediate solution see household defluoridation until a more permanent low-fluoride water supply has been established. In fluorotic areas the permanent water supply may well be based on defluoridation of water in a community water treatment plant. The reason why community plants are not always introduced in the first place is higher costs of construction, higher costs of chemicals, especially if all domestic water should be treated and distributed. Moreover because of problems in maintaining and paying the O & M costs of a public water treatment plant in areas where the public management is poor. The following is partly based on village defluoridation experiences gained in India..<sup>4</sup>

**Motivation.** It is presumably easier to motivate people to walk a little longer to fetch their drinking water than to do the additional work connected with household defluoridation. They will however need to be informed about fluorosis and the installed defluoridator. Information meetings by the time of implementation are therefore necessary. Plants should also be marked clearly with a message like "water fit for drinking and cooking".

It is rather de-motivating for people if the plant is not functioning as expected. The plant can be non-functioning in periods because of power cuts or operational problems (technical breakdowns or lack of chemicals). It can also deliver water of poor aesthetic quality e.g. because of erroneous dosage of chemicals. Both cases can lead to rejection by the users and by-pass of the treatment plant.

If the water treatment is functioning, affordable and not too distant, lack of motivation should not be a problem. There are probably no experiences so far with payment for defluoridated water and how it effects the users' willingness to use defluoridated water. Payment will probably lead to a higher demand of service. **Appropriate and economic technique.** In relation to community operated plants, appropriate mainly means that it should be feasible to operate by someone in the village and not too difficult to procure breakable spare parts. Obviously it should not be situated too far away from the users, but it needs not be the easiest accessible source. If consumption is not controlled, people living close to the plant may use the treated water for their entire needs, leaving too little water for other households.

The dimensioning of the plant requires selection between three options:

Treatment of all water for domestic use including shower, cloth-washing etc. There will be no confusion about selection between different water sources, but both construction and running costs will be many times as high as required for drinking purposes.

Treatment of only required amount of water, e.g. 4-5 lpcd. This is the cheapest and most rational option. If all households shall be able to have their share of water it will require careful control of how much water every household gets, e.g. by payment for every single bucket.

Treatment of more water, e.g. 7-10 lpcd. This may be a pragmatic solution in order to ensure that everyone covers own need and still pays for it.

As any other water supply in third world villages, materials for construction should be locally available so it is possible to repair. The experience with many types of water treatment systems in small societies is that if they are not repaired/rehabilitated easily, they will soon be bypassed. In areas with frequent power cuts (or no electric power), operation should preferably be possible without electricity. Raw water for column systems should be supplied by gravity from a gravity scheme, a high level reservoir or even a hand-operated pump in the case of smaller plants. Batch systems like Nalgonda technique may in the simplest form be mixed by the power of the inflowing water, alternatively by manually operated paddles. Paddles and gearboxes are often corroding and breaking down.

Daily routine check of treated water quality on site should be ensured. It is important for the motivation that the taste and visible appearance are not offensive. In the case of adsorption in columns, where the effluent fluoride concentration increases with time, the test parameter will be the fluoride concentration, which should be measured frequently around the expected time of regeneration or exchange of media. This may be simply measured e.g. by the use of the zirconium-alizarin method<sup>5</sup>. In systems where fluoride is removed by dosage of chemicals other parameters may be more important. E.g. for the Nalgonda technique pH is the control parameter to be tested at every batch treated, using pH-paper strips.

When water is treated centrally in the village, the waste is also concentrated more than in household treatment. Care should be taken that waste medium, sludge or regeneration wastes are treated safely for the environment and the people and animals around the plant.

**Proper organisational set-up.** As for the household defluoridation some organisational issues specific for the single techniques (procurement of bone char, regeneration of activated alumina) have to be considered separately and will not be mentioned here.

Operation and maintenance is often problematic, not only because of technical constraints. Special training of a local caretaker is very critical, but when the caretaker leaves the village the knowledge of operation may get lost. Employing at least two trained caretakers alternating in operating the plant may prevent this. A full and detailed operation and maintenance manual should be available with the responsible body and the caretakers. It should also contain a detailed list of where to find all breakable parts.

The local caretaker performs the day to day operation and control of treated water quality. A control and supervision infrastructure should be established with the water authorities. This includes regular water sampling, bringing samples from the plants to the laboratory, processing of data, and regular evaluation of performance. Action should be taken to rectify poor performance of the plants. It will in most cases also involve training of employed in the water authorities.

Many problems can arise if responsibilities are not clear and attended from the planning phase. The local body in charge of defluoridation should be able to take over responsibility of operation and maintenance in terms of trained caretakers and operational funds (cost recovery system). Engineers who erect the plants should be made responsible for the proper functioning of the plants and provision of manuals. Water supply departments should approve the plant and be able to assist when larger operational problems occur.

Like for household defluoridation, higher levels of co-ordination will be necessary for the creation of institutional memory and development of area-specific knowledge. Engineering departments, health departments and other local organisations should work together in connection with erection of plants to maximise the effect and use of defluoridated water. Since defluoridation has still not proved to be sustainable in third world rural areas, building of co-ordinated institutional memory within the country will be determining for the country's possibilities to develop and sustain defluoridation in future.

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