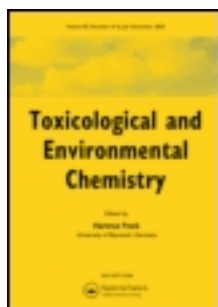


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Toxicological & Environmental Chemistry

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/gtec20>

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Published online: 27 Aug 2013.

To cite this article: Meseret Dessalegne & Feleke Zewge , Toxicological & Environmental Chemistry (2013): Daily dietary fluoride intake in rural villages of the Ethiopian Rift Valley, Toxicological & Environmental Chemistry, DOI: 10.1080/02772248.2013.827685

To link to this article: <http://dx.doi.org/10.1080/02772248.2013.827685>

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Daily dietary fluoride intake in rural villages of the Ethiopian Rift Valley

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(Received 16 December 2012; final version received 14 July 2013)

Dental and skeletal fluorosis is widespread in the Ethiopian Rift Valley region. Drinking water has been considered the main reason for the development of fluorosis, but dietary intake may also be a contributor in areas with high concentration of fluoride in water, soil, and biota. The purpose of this study is to assess the total daily dietary fluoride intake by adults in a rural part of the Ethiopian Rift Valley. The food, beverage, and water samples were collected from selected households of three neighboring villages with similar dietary pattern, but with different fluoride content in their water sources. Village A uses water with 1.0 mg L^{-1} fluoride, village B uses water with 3.0 mg L^{-1} fluoride, and village C uses water with 11.5 mg L^{-1} fluoride both for food preparation and for drinking. The level of fluoride was determined in all food ingredients, in the prepared food, beverages, and in the water used for food preparation and drinking. Recipe and food frequency questionnaires were used to gather household food preparation and consumption patterns. An alkali fusion method was used for digestion of food samples and for subsequent determination of fluoride with ion-selective electrode. The daily fluoride intake varied depending on its concentration in the water used for cooking and drinking. In households using water with 1 mg L^{-1} , 3 mg L^{-1} , and 11.5 mg L^{-1} fluoride, the total personal intake was found to be 10.5, 16.6, and 35.3 mg d^{-1} , respectively. Contribution of the water to the daily fluoride intake was 33%, 58%, and 86%, respectively. Even in households using water containing fluoride at a concentration of 1 mg L^{-1} , the daily intake was higher than the recommended safe intake of $1.5\text{--}4.0 \text{ mg d}^{-1}$ for adults, which indicates that the fluoride intake through food may cause health risks. Minimizing the fluoride concentration in water to the lowest possible level will greatly reduce the daily intake. The form of fluorine (organic or inorganic) in the food items and the associated health risk factors need further investigation.

Keywords: fluoride; daily intake; health risks; Ethiopia

Introduction

Fluoride is regarded as an essential element, primarily because of its benefits to dental health and its suggested role in maintaining the integrity of bone. To a certain extent, fluoride ingestion is useful for bone and teeth development, but excessive ingestion causes a disease known as fluorosis. As for several of the essential elements, there is a suggested range of safe and adequate intake. The guideline for drinking water recommends only 1.5 mg L^{-1} as a safe limit of fluoride in drinking water for human consumption assuming 2 L d^{-1} water consumption (WHO 1993). According to National Research Council (NRC 1989), the recommended total daily intake from various sources is in the range of $1.5\text{--}4.0 \text{ mg d}^{-1}$ for adults and less for children.

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In Ethiopia, fluoride contamination in groundwater has drawn the attention of the scientific community. Most of the studies, however, have so far focused on the investigation of the distribution and concentration of fluoride in groundwater, and severity of the problems to human health, particularly dental conditions. For example, Tekle Haimanot et al. (2006) studied the geographic distribution of fluoride in surface and groundwater in Ethiopia with a focus on the Rift Valley; Birkeland et al. (2005) assessed the severity of dental caries among children with different fluoride exposure. Other studies tackling similar issues include Wondwossen et al. (2004, 2006), Reimann et al. (2003), Chernet, Travi, and Valles (2001), and Kloos and Tekle Haimanot (1999).

Fluoride poisoning and the biological response leading to ill effects depend on the concentration of fluoride in drinking water and food; low calcium and high alkalinity of drinking water, age of the individual, and duration of intake may aggravate the disease. According to the studies carried out on fluoride and fluorosis in Ethiopia, a significant portion of the population of the Rift Valley region is exposed to high fluoride and likely to develop various forms of fluorosis. The Afar, Oromia, and Southern Nations Nationalities Peoples Region regional states are areas with high fluoride concentration in groundwater. The population estimated to live in the high-fluoride areas is about 14 million.

The fluoride research in the past decades suggests that fluoride concentrations in water below 1 mg L^{-1} are beneficial in the prevention of dental caries or tooth decay, but above 1.5 mg L^{-1} increase the severity of the incurable chronic disease “fluorosis”. The optimal level of fluoride intake is not known with certainty. A level of $0.05\text{--}0.07 \text{ mg kg}^{-1}$ is often thought of as “optimal” dietary fluoride intake in children; however, even lower levels of intake have been associated with dental fluorosis (Levy 1994). It cannot be assumed that because a person resides in a locality where excess fluoride is not present in drinking water, he or she is receiving low levels of fluoride as humans are exposed to fluoride from other sources or from water at locations other than home (e.g. childcare centers, schools, workplaces) (Levy et al. 1998, 2000).

Kloos and Tekle Haimanot (1999) reported that water is epidemiologically the most important source of fluoride (75–90% of daily intake) in most areas. Others indicate that considerable exposure risk is also associated with the consumption of fish bones, canned meat, vegetables, grains and other staples, local salt, drinks (especially tea), and air (WHO 1984; Smet 1990; Van Palenstein et al. 1995; Malde et al. 1997). In some African and Asian communities, intake of fluoride from food has been found to be higher than from water (Malde et al. 1997).

Virtually all foodstuffs and beverages including water contain at least trace amounts of fluoride (WHO 2002). Fluoride enters in human food-and-beverage chain in increasing amounts through the consumption of tea, wheat, spinach, cabbage, carrots, and other food items (Susheela 2003; Lakdawala and Punekar 1973).

Tea plants accumulate and store fluoride, absorbing it selectively from the air and soil (Cao et al. 2003; Zerabruk, Chandravanshi, and Zewge 2010). Tea plants are found having high fluoride uptake and 97% of it gets accumulated in leaves (Shu et al. 2003; Zerabruk, Chandravanshi, and Zewge 2010). The fluoride content of tea leaves is about 1000 times the soluble fluoride content of soil and 2–7 times the total fluoride content in soil (Fung, Zhang, and Wong 1999). Some studies showed that instant tea in distilled water has a fluoride concentration of 3.3 mg L^{-1} (Michael et al. 2005). This can be a causative factor of dental fluorosis (Cao et al. 1997; Cao, Zhao, and Liu 2000; Cao et al. 2003). Furthermore, brick tea prepared in China, which is fermented and then compressed into bricks, has a fluoride content of $590\text{--}780 \text{ mg kg}^{-1}$ (Sha and Zheng 1994). High fluoride-containing toothpastes are also a major source of fluoride intake (Levy et al. 2000).

Levels of daily exposure to fluoride depend mainly on the geographical area. In The Netherlands, the total daily intake was calculated to be 1.4–6.0 mg of fluoride (Slooff 1988). Food seems to be the source of 80–85% of fluoride intake; intake from drinking water is 0.03–0.68 mg d⁻¹ and from toothpaste 0.2–0.3 mg d⁻¹. For children, total intake via food and water is decreased because of lower consumption. Intake of food and water relative to the body weight is higher and can be further increased by the swallowing of toothpaste or fluoride tablets (Slooff 1988).

In Ethiopia, very few studies conducted on fluoride intake of children (6–59 months) from water and beverages indicated that fluoride intake is far above the maximum permissible level (Malde et al. 2003). To strengthen the mitigation and prevention of fluorosis, it is important to understand the level of fluoride from other sources than water, exposure level on specific age groups, potential health and economic impacts, dietary pattern, and nutritional status of the endemic communities.

The purpose of this study was to estimate the average daily fluoride intake of adult person based on most commonly consumed food and beverages including drinking water in selected villages in the Ethiopian Rift Valley. The contribution of food ingredients and water used for cooking to the fluoride concentration of prepared food and beverages was estimated in selected households of Dugda woreda.

Materials and methods

Sampling sites

Three sampling sites were selected based on the source of water they use for cooking and drinking purposes. These include three households using defluoridated water from community defluoridation (village A), three households using untreated water (village B), and three households using untreated water from a wind mill (village C). The geographical locations of the water points of villages A and B is N 080 05.762' E 0380 44.736' and that of village C is N 080 04.909' E 0380 45.134'. The villages are found in Dugda woreda, East Shewa zone of Oromia regional state where the observed risk of fluorosis is extremely high.

Food and beverage samples

Most commonly consumed food and beverage items in the area were selected based on food frequency questionnaires. Then the food ingredients and the water used for cooking were separately sampled and analyzed. This was used to calculate the contribution of each ingredient to the fluoride content of prepared foods. Six food items, namely enjera, homemade bread (kitta), kale stew, potato stew, shiro stew, fish stew, and beverages including tea, coffee, and the water used for drinking and cooking were selected. The selection of the foods and beverages considered the most consumed items as well as those which may be consumed rarely but expected to contribute high fluoride.

In addition, food ingredients used for the preparation of the selected food and beverages were collected. These include cereals, vegetables, legume powder (locally named shiro), spices, raw fish, coffee powder, tea leaves, sugar, and salt.

Sampling procedure

The selected nine households were interviewed for their willingness to respond to questionnaires and to conduct the sampling procedures for five consecutive days. All the nine

households were willing and a brief orientation on preparation of food was given to women who cook and be available during all the five days of sampling.

The samples of each food ingredient and cooking water were placed in plastic bags and plastic bottles labeled with identification number, date, and type of the sample. After taking the samples, the weight of each ingredient including the cooking water was measured and recorded on recipe questionnaires. After the food was prepared, the weight was measured and recorded. 50–100 g sample of the prepared food was taken after the weight was measured into polyethylene plastic sample bottles. All the ingredients, prepared food and beverages were kept under a deep freezer until analysis.

Sample preparation

Homogenized samples of 0.5 g and 1 g depending on the expected fluoride content were measured directly into 40-mL nickel crucibles (Baoji Xinnuo New Metal Materials Co. Ltd., Shaanxi, China). The samples were covered with 5 mL of 8 mol L⁻¹ sodium hydroxide solution (extra pure, Scharlau Chemie S.A., Sentmenat, Spain). The sample and sodium hydroxide solution were slowly shaken to make the mixture as homogenous as possible.

The crucibles were placed on a hot plate for evaporation to dryness, covered and introduced into a muffle furnace for combustion. The temperature was set at 200 °C for approximately 16 h in an oven and was increased to 525 °C for 3 h in the muffle furnace. The crucibles were then cooled to room temperature and 10–15 mL deionized water was added to the crucibles and kept on a hot plate in order to aid the dissolution of the fusion cake. After 2 h, the sample solutions were transferred to 50-mL plastic beakers (Biologix Plastics Co. Ltd., Jiangsu, China).

The sample solutions were neutralized using concentrated and then diluted hydrochloric acid. Concentrated hydrochloric acid (37%, Sigma-Aldrich Chemie, Steinheim, Germany) was added dropwise until the pH decreased from 12.0–13.0 to 8.0–8.5 then to pH of 7.2–7.5 by adding dilute hydrochloric acid. The sample solutions were then diluted to 50 mL with deionized water (Malde, Bjorvatn, and Julshamn 2001).

Fluoride measurement

A pH/ion-selective electrode meter (model EA940, Orion, Beverly, MA, USA) equipped with a combination fluoride-selective electrode (model 96-09, Orion) was employed for the determination of fluoride in the samples and standard solutions. The pH was measured with a pH/ion meter (HI 9025, Hanna Instruments, Selangor, Malaysia) using a pH glass electrode.

The reagents used in the analysis were all of analytical grade. Fluoride stock was prepared from anhydrous sodium fluoride (99.0%, BDH Chemicals, Poole, Dorset, England) and standard solutions of the required concentration were prepared by appropriate dilution from the fluoride stock solution with deionized water. Glacial acetic acid (Techno Pharmchem, New Delhi, India), sodium chloride (Oxford Laboratory, Mumbai, India), trisodium citrate (BDH Chemicals), ethylene diamine tetra acetic acid (EDTA) (Scharlau Chemie S.A., Barcelona, Spain), and sodium hydroxide were used to prepare the total ionic strength adjustment buffer (TISAB II). The TISAB II was prepared by dissolving 58.5 g sodium chloride, 57.0 mL glacial acetic acid, 7 g of trisodium citrate, and 2 g EDTA in 500 mL distilled water. The pH of this solution was adjusted to 5.0–5.5 with 5 mol L⁻¹ sodium hydroxide and finally diluted to 1000 mL in a volumetric flask with deionized water.

The reagent also includes concentrated hydrochloric acid for neutralization after alkali fusion and dissolution of food samples and buffer solutions for pH calibration. Fluoride was determined by taking 5 mL of the sample and 5 mL of TISAB to maintain the pH in the range of 5.2–5.4.

Reagent blanks were prepared together with the samples and brought through the whole procedure for blank determination. The standard solutions and the sample solutions were analyzed with pre-calibrated fluoride ion-selective electrode (Malde, Bjorvatn, and Julshamn 2001). The beverages tea and coffee including water were analyzed directly without digestion.

Data quality assurance

All the prepared samples were ashed and measured in duplicate and mean results were recorded. For the quality control, recovery study was done by spiking known concentration of standards to some food samples such as enjera, bread, and fish, and percentage recovery was calculated. The result of the recovery analysis was in the range of 85–115%, which shows a good reproducibility of the method.

One certified reference material (CRM) for fluoride in vegetation, (Timothy high, 2695) from National Institute of Standards and Technology, USA was measured together with the samples following the same procedure. The certified value and the fluoride measured at National Institute of Nutrition and Seafood Research are $277 \pm 27 \text{ mg kg}^{-1}$ and $277 \pm 34 \text{ mg kg}^{-1}$, respectively. The average value ($n = 7$) measured during this study is $282 \pm 19 \text{ mg kg}^{-1}$.

To evaluate the performance of the method, a comparison of measured values of standard reference material with that of certified value was made according to the procedure in Application Note 1 of the European Reference Materials ERM (Linsinger 2005). This statistically evaluates the difference between the measured values in a standard reference material and its certified value based on the expanded uncertainty of the difference between measured concentration value (C_m) and the certified value C_{CRM} ,

$$\Delta_m = |C_m - C_{\text{CRM}}|. \quad (1)$$

The combined uncertainty of the difference Δ_m is u_Δ , calculated from the uncertainty of the certified value and the uncertainty of the measurement result according to

$$u_\Delta = \sqrt{u_m^2 + u_{\text{CRM}}^2}. \quad (2)$$

The standard uncertainty u_m of the measured fluoride in the CRM was estimated from the measured values of seven CRM ashed samples in different days $\left(u_m = \frac{\sigma}{\sqrt{7}}\right)$. The standard uncertainty of the CRM was calculated from its expanded uncertainty ($U_{\text{CRM}} \div 2$).

The expanded uncertainty at 95% confidence level (coverage factor $k = 2$) is thus,

$$U_\Delta = 2u_\Delta. \quad (3)$$

The difference between the concentration in the CRM and the measured value Δ_m is compared with U_Δ : If $\Delta_m \leq U_\Delta$ (i.e. $\Delta_m/U_\Delta \leq 1$), there would be no significant difference between the measurement result and the certified value at 95% confidence level.

The performance of fluoride measurement in the matrix CRM, Δ_m compared with U_Δ shows

$$\begin{aligned}\Delta_m &= 5 \\ U_\Delta &= 30.7.\end{aligned}$$

$\Delta_m \leq U_\Delta$ (i.e. $\Delta_m/U_\Delta \leq 1$), which implies that there is no significant difference between the measurement result and the certified value at 95% confidence level. This implies a good performance of the method.

Results and discussion

Fluoride in food ingredients

The fluoride content of the food ingredients is shown in [Table 1](#). The ingredients used for cooking different food items were different from one food item to the other and from household to household though some ingredients such as onion, salt, for preparation of most stews and cereals such as maize, teff, and wheat for preparation of enjera and bread were commonly used in the households. Food ingredients that were repeatedly used in one household were collected once except cooking water. The water used for cooking was collected every day of sampling in each of the nine households. The standard deviations of some of the measurements were higher because the samples were collected from each household where the sources may differ.

Table 1. Fluoride concentration of food ingredients and water ($N = 9$).

Ingredients	Mean F ⁻ (mg kg ⁻¹)	SD
Tea leaves	625	12
Salt	27.7	5.6
Berberbe	18.8	6.4
Red teff flour	15.1	6.9
Maize flour	12.2	5.1
Tumeric (erd)	10.2	4.0
Shiro powder	9.8	2.8
Wheat flour	6.8	0.3
Potato	5.6	1.7
Green pepper	4.9	3.1
Tomato	4.7	2.7
Garlic	4.1	–
Raw fish	4.0	1.3
Kale	3.9	2.0
Onion	2.1	0.3
Coffee	1.8	0.4
Sugar	0.3	0.01
Water source in village A	1.15	0.02
Water source in village B	3.19	0.02
Water source in village C	11.6	0.1

Since food ingredients were collected from households, all the cereals were in the form of powders. Legume powder (locally named shiro) can be prepared from different legumes such as peas, beans, chickpea, and a mixture of those. All the households that participated in this study were buying the powder from a nearby market and did not have exact information on the specific legumes used for the preparation of shiro powder.

Most commonly used spices in the selected households were red chilli powder (locally named berbere) and turmeric, *Curcuma longa* (locally named erd). The households use the two spices interchangeably when preparing potato stew and shiro stew.

Fish was mostly not consumed in the selected households. However, fish was found to accumulate high fluoride concentration in different studies, it was selected to be included in this study. The fish samples collected from all the nine households were from the nearby Lake Ziway. Malde et al. (1997) measured a fluoride concentration of 4.4 mg kg⁻¹ of dry weight in *Oreochromis niloticus* species (large size) found in Lake Ziway.

Sugar, salt, coffee, and tea leaves were not produced or grown around the sampling area. Salt was found to have high fluoride concentration because it is produced in Afdera, which is a part of the Rift Valley region in Ethiopia. Tea leaves are known to accumulate high fluoride concentration. The households used a brand called “Good Morning Tea” to prepare tea. It was found to have a fluoride concentration of 624.5 ± 12 mg kg⁻¹. Zerabruk, Chandravanshi, and Zewge (2010) calculated the fluoride concentration of nine Ethiopian black tea leaves in the range of 248 mg kg⁻¹ in “Almeta tea” to 682 mg kg⁻¹ in “Abay tea”.

Selected food and beverage items were prepared using different water sources containing different fluoride levels. These include defluoridated water from community defluoridation (with bone char) in Weyo Gabriel (village A), untreated water from Meskan village in Weyo Gabriel (village B), and untreated water from Gura village (village C).

As shown in Table 1, the fluoride content of food ingredients is variable. In addition, similar ingredients from different households showed different fluoride content as the sources of the ingredients available in the market are different.

The contribution of the ingredients to the prepared dishes depends on the fluoride content of the ingredients and the amount that is used during food preparation. The experimental results also showed that some ingredients appear to have high fluoride concentration but their contribution is very less as they are used in small quantity. This is true for salt (27.7 mg kg⁻¹), tea leaves (625 mg kg⁻¹), cereals, and vegetables depending on the amount used by each household.

Fluoride content of prepared food and beverages

The fluoride content of prepared food and beverages is shown in Table 2. Fluoride in the prepared food items varied depending on factors such as the type of ingredients used, the amount of ingredients added, and fluoride concentration of the water used for cooking.

Despite the fact that the recipe for preparing similar food items differs from one household to another, it was possible to evaluate the influence of fluoride in the water used for cooking. As the fluoride concentration in the water used increases, the fluoride content of the prepared food and beverages also increases. Other studies also reported similar results (Dabeka et al. 1982).

Table 2. Fluoride content of prepared food and beverages ($N = 3$).

Items	Fluoride concentrations in the water used for cooking		
	A (1 mg L ⁻¹)	B (3 mg L ⁻¹)	C (11 mg L ⁻¹)
Enjera	9.7	11.9	16.9
Bread	11.9	16.0	18.1
Shiro stew	2.5	3.7	10.9
Potato stew	4.0	6.0	9.0
Kale stew	7.2	8.8	16.1
Fish stew	2.6	4.9	7.41
Tea	3.7	6.8	13.3
Coffee	1.2	2.8	11.3

Estimation of daily fluoride intake

Daily intake of fluoride was estimated for each household using three different water sources (Equation (4)),

$$\text{i.e. } X_T = \sum_{j=1}^{10} x_j, \quad (4)$$

where X_T is total daily intake (mg person⁻¹ d⁻¹),

x_j is daily intake through food or beverage j ,

j_1, j_2, \dots, j_{10} is prepared food and beverage items including drinking water,

$$\text{where } x_j = \text{consumption of } j^* c_j,$$

where consumption of j is the daily consumption of j (kg person⁻¹ d⁻¹), c_j is the concentration of fluoride in food or beverage j (mg kg⁻¹).

The fluoride intake from different prepared foods and beverages including drinking water was calculated to assess the contribution of each food and beverage item to the total intake (Equation (5)),

$$\text{Consumption of } j = \frac{(\text{Amount of } j \text{ consumed (kg)} \times \text{frequency of consumption in a week})}{\text{No. of days in a week}}. \quad (5)$$

The average amount of each food consumed by an adult person was estimated from the results of recipe questionnaires by taking the average of all the nine households. The information of frequency of the food and beverages to be consumed within a week was obtained from the field observation and results of the questionnaires. For assessing the influence of the water used for drinking and the cooking process itself, total daily intake was calculated from estimated results of food and beverages before cooking (Equation (6)),

$$\% \text{ contribution to total intake} = \left(\frac{(X_T - X_T^*)}{X_T} \right) \times 100, \quad (6)$$

where X_T is total daily fluoride intake (mg d⁻¹) and X_T^* is total daily fluoride intake calculated with zero F⁻ in cooking and drinking water.

Similarly, the influence of the water fluoride content is calculated (Equation (7)),

$$\% \text{ contribution to } j = \left(\frac{(x_j - x_j^*)}{x_j} \right) \times 100, \quad (7)$$

where x_j is daily intake through food or beverage j , x_j^* is daily intake through j calculated with zero fluoride in cooking water, and j_1, j_2, \dots, j_9 are different types of food and beverages.

Daily fluoride intake

The total daily fluoride intake as shown in Tables 3–5 is the sum of daily intake through different food items and beverages. From the food frequency questionnaire, the amount of enjera that is consumed is very high (eightfold) as compared to the consumption of tea. Consequently, the daily fluoride intake per person from enjera (village A) is 2.9 mg, while it is only 0.5 mg from tea. But when we compare the contribution of the water it is higher in tea (18%) than in enjera (12%) as the actual water content of tea is much higher than that of enjera.

The fluoride intake through food and beverages prepared with defluoridated water is shown in Table 3. In these households, the intake of fluoride from the food was much higher than the fluoride from the beverages indicating that the fluoride concentration of the ingredients also played a vital role on the total intake, in addition to the fluoride concentration in the water.

Malde, Zerihun, and Bjorvatn (2004) studied the total daily fluoride intake of children living in two villages A and K (with fluoride concentration in the water about 2 mg L⁻¹ and 14 mg L⁻¹, respectively), in Wonji Shoa sugar estate, a rural part of the Ethiopian Rift Valley. According to this study, the daily fluoride intake of children in village A was derived 63% from food, while children in the high-fluoride village K consumed most of the fluoride through beverages (60%).

As shown in Table 3, the calculated daily intake of fluoride from enjera is 4.63 mg person⁻¹d⁻¹, whereas it is 2.61 mg person⁻¹d⁻¹ from bread. On the other hand, the

Table 3. Fluoride intake by an adult using defluoridated water.

Daily intake of F ⁻ (mg person ⁻¹) through	Food alone (0 mg L ⁻¹ in water)	Calculated from raw food (1 mg L ⁻¹)	Contribution of water (%)	Calculated from prepared food (1 mg L ⁻¹)
Bread	1.2	1.3	5	2.6
Enjera	2.6	2.9	12	4.6
Shiro stew	0.1	0.2	27	0.2
Potato stew	0.32	0.37	11	0.34
Kale stew	0.27	0.31	12	0.59
Fish stew	0.03	0.04	19	0.04
Total	4.56	5.11	11	8.37
Tea	0.37	0.45	18	0.17
Coffee	0.07	0.22	66	0.24
Drinking water	0.00	1.73	100	1.73
Total	0.45	2.40	81	2.14
Total daily intake	5.0	7.5	33	10.5

Table 4. Fluoride intake of an adult person using untreated water from village B.

Daily intake of F ⁻ (mg person ⁻¹) through	Food alone (0 mg L ⁻¹ in water)	Calculated from raw food (3 mg L ⁻¹)	Contribution of water (%)	Calculated from prepared food (3 mg L ⁻¹)
Bread	2.14	2.71	21	3.52
Enjera	2.76	4.22	35	5.68
Shiro stew	0.12	0.37	67	0.24
Potato stew	0.35	0.48	28	0.51
Kale stew	0.14	0.32	54	0.71
Fish stew	0.04	0.07	45	0.08
Total	5.6	8.2	32	10.7
Tea	0.26	0.59	85	0.41
Coffee	0.11	0.74	56	0.66
Drinking water	0.00	4.78	100	4.78
Total	0.37	6.12	94	5.85
Total daily intake	5.9	14.3	58	16.6

fluoride content of enjera and bread is 9.7 mg kg⁻¹ and 11.9 mg kg⁻¹, respectively (Table 2). The observed difference is due to the higher daily consumption of enjera as it is the staple food in those villages in particular and in Ethiopia in general.

People living in village B use water with fluoride concentration of about 3 mg L⁻¹. The fluoride intake of an adult person through the food and beverages prepared by using this water source is shown in Table 4.

As shown in the table, the fluoride intake through food and beverages is higher as compared with those households using defluoridated water.

The people in village C used untreated groundwater with fluoride content of about 11.6 mg L⁻¹ for preparing food, beverages, and for drinking purposes. As shown in Table 5, the results are much higher than the results found by using defluoridated water and untreated water from village B. Moreover, the fluoride intake from beverages is higher than that from the food. In a Kenyan study, Opinya et al. (1991) found a total daily fluoride intake of 14.5 mg (range 6–24 mg) in children aged 1–4 years living in an area with 9 mg L⁻¹ in the drinking water. Anasuya, Bapurao, and Paranjape (1996) studied a

Table 5. Fluoride intake of an adult person using the water source in village C.

Daily intake of F ⁻ (mg person ⁻¹) through	Food alone (0 mg L ⁻¹ in water)	Calculated from raw food (11 mg L ⁻¹)	Contribution of water (%)	Calculated from prepared food (11 mg L ⁻¹)
Bread	0.99	2.88	66	3.97
Enjera	2.71	7.26	63	8.1
Shiro stew	0.14	0.77	81	0.69
Potato stew	0.30	0.87	65	0.77
Kale stew	0.29	1.07	73	1.3
Fish stew	0.11	0.16	33	0.12
Total	4.54	13.0	65	15.0
Tea	0.28	1.48	81	1.67
Coffee	0.01	2.38	99.5	1.29
Drinking water	0.00	17.4	100	17.35
Total	0.30	21.2	99	20.3
Total daily intake	4.8	34.2	86	35.3

range of intakes based on consumed foodstuffs and local supplies of drinking water (normal and fluoride-endemic areas) by adults living in the rural areas of India. The study showed a daily fluoride intake of 0.84–4.69 mg d⁻¹ in normal and 3.40–27.1 mg d⁻¹ in fluoride-endemic areas.

Coffee was found to be consumed very frequently (seven days in a week) in the selected households. The percentage contribution of fluoride in the water used for the preparation of coffee was high in all the three water sources compared to the contribution of the coffee powder because coffee powder has low fluoride content (1.8 ± 1.4 mg kg⁻¹).

It is important to note that significant amount of fluoride from water is retained in the prepared food. In the households using water with fluoride concentration of 1, 3, and 11.5 mg L⁻¹, the daily fluoride intake was 10.5, 16.6, and 35.3 mg d⁻¹, respectively. This shows that a 11-fold increase in fluoride concentration of the water used for food preparation corresponds to a 3-fold increase in the daily fluoride intake. Malde, Zerihun, and Bjorvatn (2004) found a 7-fold increase in the fluoride concentration of the water used for food preparation, which resulted in the doubling of fluoride intake through food. In households using low-fluoride water (village A), the contribution of water was only about 33%, whereas the value was about 86% in the case of households using high-fluoride water (village C).

Conclusions

An estimation of the dietary intake of fluoride in adults will help to evaluate the prevalence of skeletal fluorosis due to long-term total daily intake. The study showed that the daily fluoride intake of adults living in the selected villages was 10.5, 16.6, and 35.3 mg. A significant portion of the fluoride intake is from food items frequently used by the studied households. As the water fluoride content increases, the contribution of water to the daily fluoride intake becomes more pronounced. A 11-fold increase in the fluoride concentration of water resulted in a 3-fold increase in the daily intake of fluoride. Reducing the fluoride concentration of the water to the lowest possible level is recommended to minimize the total daily fluoride intake. Fluoride in food ingredients also played a role on the daily fluoride intake, especially in the households using less fluoride-containing water. As the actual absorption of ingested fluoride may depend on other factors such as the presence of nutrients like Ca and Mg, further study will be required on complete dietary composition. In addition, as the fluoride in food may exist in organic form, which may be less or more toxic, further study will be required by extracting with organic solvent and subsequent analysis.

Acknowledgments

The authors would like to thank the Department of Chemistry and Food Science Program of Addis Ababa University for providing the required laboratory facilities. Financial support from the Swiss Federal Institute of Aquatic Science and Technology is greatly appreciated. The author Meseret Dessalegn is thankful to DireDawa University for sponsoring her study.

References

- Anasuya, A., S. Bapurao, and P.K. Paranjape. 1996. "Fluoride and Silicon Intake in Normal and Endemic Fluorotic Areas." *Journal of Trace Elements in Medicine and Biology* 10: 149–155.

- Birkeland, J.M., Y.E. Ibrahim, I.A. Ghandour, and O. Haugejorden. 2005. "Severity of Dental Caries Among 12-Year-Old Sudanese Children With Different Fluoride Exposure." *Clinical Oral Investigations* 9: 46–51.
- Cao, J., X. Bai, Y. Zhao, D. Zhou, S. Fang, M. Jia, and J. Wu. 1997. "Brick Tea Consumption as the Cause of Dental Fluorosis Among Children From Mongol, Kazak and Yugu Populations in China." *Food and Chemical Toxicology* 35 (8): 827–833.
- Cao, J., Y. Zhao, and J. Liu. 2000. "Fluoride in the Environment and Brick-Tea Type Fluorosis in Tibet." *Journal of Fluorine Chemistry* 106: 93–97.
- Cao, J., Y. Zhao, J. Liu, R. Xirao, S. Danzeng, D. Daji, and Y. Yan. 2003. "Brick Tea Fluoride as a Main Source of Adult Fluorosis." *Food and Chemical Toxicology* 41 (4): 535–542.
- Chernet, T., Y. Travi, and V. Valles. 2001. "Mechanism of Degradation of the Quality of Natural Water in the Lakes Region of the Ethiopian Rift Valley." *Water Research* 35 (12): 2819–2832.
- Dabeke, R.W., A.D. McKenzie, H.B.S. Conacher, and B.S. Kirkpatrick. 1982. "Determination of Fluoride in Canadian Infant Foods and Calculation of Fluoride Intake by Infants." *Canadian Journal of Public Health* 73: 188–191.
- Fung, K.F., Z.Q. Zhang, and J.W.C. Wong. 1999. "Fluoride Contents in Tea and Soil From Tea Plantations and the Release of Fluoride Into Tea Liquor During Infusion." *Environmental Pollution* 104: 197–205.
- Kloos, H., and R. Tekle Haimanot. 1999. "Distribution of Fluoride and Fluorosis in Ethiopia and Prospects for Control." *Tropical Medicine and International Health* 4 (5): 355–364.
- Lakdawala, D.R., and B.D. Punekar. 1973. "Fluoride Content of Water and Commonly Consumed Foods in Bombay and a Study of the Dietary Fluoride Intake." *Indian Journal of Medical Research* 61: 1679–1687.
- Levy, S.M. 1994. "Review of Fluoride Exposures and Ingestion." *Community Dentistry and Oral Epidemiology* 22 (3): 173–180.
- Levy, S.M., J.A. McGrady, P. Bhuridej, J.J. Warren, J.R. Heilman, and J.S. Wefel. 2000. "Factors Affecting Dentifrice Use and Ingestion Among a Sample of U.S. Preschoolers." *Pediatric Dentistry* 22 (5): 389–394.
- Levy, S.M., M.C. Kiritsy, S.L. Slager, and J.J. Warren. 1998. "Patterns of Dietary Fluoride Supplement use During Infancy." *Journal of Public Health Dentistry* 58: 228–233.
- Linsinger, T. 2005. European Reference Materials: Application Note 1. Comparison of a Measurement Result With the Certified Value. http://www.bam.de/en/fachthemen/referenzmaterialien/referenzmaterialien_medien/erm_application_note_1_en.pdf.
- Malde, M.K., K. Bjorvatn, and K. Julshamn. 2001. "Determination of Fluoride in Food by the Use of Alkali Fusion and Fluoride Ion-Selective Electrode." *Food Chemistry* 73: 373–379.
- Malde, M.K., A. Maage, E. Macha, K. Julshamn, and K. Bjorvatn. 1997. "Fluoride Content in Selected Food Items From Five Areas in East Africa." *Journal of Food Composition and Analysis* 10: 233–245.
- Malde, M.K., L. Zerihun, and K. Bjorvatn. 2004. "Fluoride, Calcium and Magnesium Intake in Children Living in a High-Fluoride Area in Ethiopia: Intake Through Food." *International Journal of Paediatric Dentistry* 14: 167–174.
- Malde, M.K., L. Zerihun, K. Julshamn, and K. Bjorvatn. 2003. "Fluoride Intake in Children Living in a High-Fluoride Area in Ethiopia: Intake Through Beverages." *International Journal of Paediatric Dentistry* 13 (3): 27–34.
- Michael, P., M.D. Whyte, M.S. Kevan Essmyer, H. Francis, M.D. Gannon, R. William, and M.D. Reinus. 2005. "Skeletal Fluorosis and Instant Tea." *American Journal of Medicine* 118: 78–82.
- NRC (National Research Council). 1989. *Recommended Dietary Allowances*. 10th ed. Washington, DC: National Academy Press.
- Opinya, G.N., N. Bwibo, J. Valderhaug, J.M. Birkeland, and P. Lokken. 1991. "Intake of Fluoride and Excretion in Mothers' Milk in a High Fluoride (9 ppm) Area in Kenya." *European Journal of Clinical Nutrition* 45: 37–41.
- Reimann, C., K. Bjorvatn, B. Frengstad, Z. Melaku, R. Tekle Haimanot, and U. Siewers. 2003. "Drinking Water Quality in the Ethiopian Section of the East African Rift Valley I – Data and Health Aspects." *Science of the Total Environment* 311: 65–80.
- Sha, J.Q., and D.X. Zheng. 1994. "Study on the Fluorine Content in Fresh Leaves of Tea Plant Planted in Fujian Province." *Journal of Tea Science* 14: 37–42.
- Shu, W.S., Z.Q. Zhang, C.Y. Lan, and M.H. Wong. 2003. "Fluoride and Aluminum Concentrations of Tea Plants and Tea Products From Sichuan Province, PR China." *Chemosphere* 52: 1475–1482.

- Slooff, W. 1988. "Basis Document Fluoriden." *National Institute of Public Health and Environmental Protection*. Bilthoven, Netherlands (Report No. 758474005).
- Smet, J. 1990. "Fluoride in Drinking Water." In *Symposium on Fluorosis in Developing Countries: Causes, Effects and Possible Solutions*, edited by J.E. Frencken, 10–19. Leiden: NIPG-TNO.
- Susheela, A.K. 2003. *A Treatise on Fluorosis*. Revised 2nd ed. New Delhi: Fluorosis Research and Rural Development Foundation.
- Tekle Haimanot, R., Z. Melaku, H. Kloos, C. Reimann, W. Fantaye, L. Zerihun, and K. Bjorvatn. 2006. "The Geographic Distribution of Fluoride in Surface and Groundwater in Ethiopia With an Emphasis on the Rift Valley." *Science of the Total Environment* 367 (1): 182–190.
- Van Palenstein, H.W., E. Mkasabuni, H.J. Mjengera, and L. Mabelya. 1995. "Severe Fluorosis in Children Consuming Fluoride Containing Magadi." In *Proceedings of the First International Workshop on Fluorosis and Defluoridation of Water*, edited by E. Dahi and H. Bregnhøj, 15–19. Auckland: International Society for Fluoride Research.
- Wondwossen F., A.N. Astrom, K. Bjorvatn, and A. Bardsen. 2004. "The Relationship Between Dental Caries and Dental Fluorosis in Areas With Moderate-and High-Fluoride Drinking Water in Ethiopia." *Community Dentistry and Oral Epidemiology* 32 (5): 337–344.
- Wondwossen, F., A.N. Astrom, K. Bjorvatn, and A. Bardsen. 2006. "Sociodemographic and Behavioural Correlates of Severe Dental Fluorosis." *International Journal of Paediatric Dentistry* 16 (2): 95–103.
- WHO (World Health Organization). 1984. *Fluorine and Fluorides, Fluoride in Drinking-Water. Environmental Health Criteria 36*. Geneva: World Health Organization.
- WHO (World Health Organization). 1993. *Guidelines for Drinking-Water Quality. Recommendations*. 2nd ed. Vol. 1. Geneva: World Health Organization.
- WHO (World Health Organization). 2002. *Fluorides. Environmental Health Criteria Number 227*. Geneva: World Health Organization.
- Zerabruk, S., B.S. Chandravanshi, and F. Zewge. 2010. "Fluoride in Black and Green Tea (*Camellia Sinensis*) Infusions in Ethiopia: Measurement and Safety Evaluation." *Bulletin of The Chemical Society of Ethiopia* 24 (3): 327–338.