LEVELS OF FLUORIDE IN WIDELY USED TRADITIONAL ETHIOPIAN SPICES

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ABSTRACT: The fluoride ion levels in the most widely used traditional Ethiopian spices were determined after alkali fusion extraction using a fluoride ion selective electrode. The levels of fluoride (mg F/kg dry weight) in the spices were: coriander seeds: 2.14±0.04, basil leaves: 2.53±0.05, ginger rhizome: 2.81±0.13, red chili pepper: 3.1±0.17, fenugreek seeds: 3.18±0.03, garlic granules: 3.27±0.13, turmeric rhizome: 3.29±0.07, nutmeg seeds: 4.51±0.12, cinnamon bark: 5.13±0.23, black cumin seeds: 8.14±0.15, and thyme leaves: 8.57±0.11. In general, the fluoride concentrations found in Ethiopian spices were comparable with the spices from other countries and the fluoride intake from spices for adults in Ethiopia contain only a relatively small amount of fluoride (0.082 mg/day) compared to the currently recommended daily fluoride intake (3 mg/day for a 60 kg adult based on 0.05 mg/kg body weight/day).

Keywords: Ethiopia; Fluoride; Food; Spices.

INTRODUCTION

The fluoride ion (F) is not considered to be essential for human growth and development,[1] including for the development of healthy teeth and bones, and the intake of fluoride at high levels (above 6 mg/day) for a long time causes dental, skeletal, and non-skeletal fluorosis.[2,3] It can also cause health problems affecting soft tissues like aorta, thyroid, lungs, kidneys, heart, pancreas, and brain[4] as well as birth defects, genetic damage, and cancer.[5] Dental fluorosis is a common health problem in many parts of the world including Ethiopia. Since the fluoride level of ground water in the Rift Valley areas is high, the risk of fluorosis in Ethiopia is mainly observed in the Rift Valley areas and is spread over several regional states.[6]

The total exposure to fluoride depends on the contributions from various sources, such as drinking water, water-based beverages, foods, food supplements, toothpaste, and air.[2,7,8] There are several studies in the literature on the dietary intake of fluoride from water, beverages and foods.[9-18] All vegetation contains some fluoride, which is absorbed from soil and water.[19] As a result, spices cultivated through the world may contain significant levels of fluoride and the fluoride content of spicy human diets contributes to the total fluoride intake. The delightful flavour and pungency of spices make them indispensable in the food industry to flavour, improve the taste, and increase the appeal of their products. Spices impart aroma, colour, and taste to food preparations and sometimes mask undesirable odours. In addition, they are reputed to possess several medicinal and pharmacological properties and hence find a position in the preparation of a number of medicines.[20]

Ethiopia is amongst the largest consumers of spices in Africa. The major use of spices in the country is in the preparation of a highly spiced stew known as “wot”

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which, together with "injera", is consumed by a large proportion of the population everyday as their main food. In addition, spices are also used by the numerous ethnic groups in the country to flavor bread, meat, soups, and various vegetables, and as medicines and perfumes.[21] Several different types of spices are used in Ethiopia including are red chili pepper (*Capsicum annum*), garlic tuber (*Allium sativum*), cinnamon bark (*Cinnamomum zeylanicum*), turmeric rhizome (*Curcuma longa*), ginger rhizome (*Zingiber officinale* Roscoe), fenugreek seeds (*Trigonella foenum graecum*), coriander seeds (*Coriandrum sativum* L.), nutmeg seeds (*Myristica fragrans* Houtt.), black cumin seeds (*Cuminum cyminum* Linn.), thyme leaves *(Thymus vulgaris*), and basil leaves (*Ocimum basilicum*).

Red chili pepper belongs to the family of Nightshade and is the most widely consumed spice throughout the world due to its intense flavor and pungency. Red pepper is the leading spice crop in Ethiopia and it is locally consumed in various food preparations, particularly for giving flavor and color to local stews. Red chili pepper is cultivated in different parts of Ethiopia.[22]

Garlic belongs to the family *Alliaceae* and it has grown as a spice and used for flavouring local dishes.[23] It is most widely cultivated in Ethiopia.[22] Most of the time, it is used in cooked form with various foods since it enhances flavor due to its pungency as well as adding nutritional benefit. The garlic tuber is the part of garlic plant which is used most often for cooking and medicinal purposes.[24]

Cinnamon is the inner bark of cinnamomum plant having a buff or dark reddish color and belongs to the family of *Lauraceae*. It is called kerefa in Ethiopia and grows well in almost all soil types from the semi-dry to the wet zones, especially in Mizan-Tepi (South west of Ethiopia).[25] Cinnamon is commonly used in the cooking of sweets, vegetables (carrots, winter squash and sweet potatoes), for making hot drinks and beverages (white wine and vodka), and for its aroma, flavor, and taste. The flavor of cinnamon is due to an aromatic essential oil and its pungent taste and scent come from cinnamic aldehyde or the cinnamaldehyde component.[26]

Turmeric is a ground spice belonging to the family of *Zingiberaceae* (ginger). It is yellow-orange colored root and is commonly used as herb and spice all over the world. Turmeric is widely grown in the Southern part of Ethiopia.[25] Turmeric is extensively used as a spice for food coloring and flavoring purposes. Due to its active ingredient, curcumin, turmeric has been also used in traditional medicine as a household remedy for various diseases.[27]

Ginger is the underground stem or rhizome of the plant *Zingiber officinale* Roscoe and belongs to the family *Zingiberaceae*. Ginger rhizome is typically consumed as a fresh paste or dried powder, and is used as a food preservative in the preparation of syrup and candy. Due to its volatile oil components (zingerone, zingerols, and shogaols), ginger has been used for culinary purposes and for the treatment of various diseases.[28,29] At present, ginger is been cultivated in many parts of Ethiopia.[22,28] It is an important spice which is used in every Ethiopian kitchen for the preparation of pepper powder, stew, bread, etc. It is also used in traditional medicine for the treatment of flu, stomach-ache, and headache.[30]

Fenugreek seeds are one of the well known spices in human food which is cultivated worldwide as a semiarid crop. Fenugreek belongs to the family of *Fabaceae* and an extract of its soluble dietary fibre content is able to moderate the metabolism of glucose in the digestive tract, stimulate the appetite, and modify food texture. Fenugreek seeds are also used for medicinal purposes.[31] Fenugreek seeds are used in Ethiopia as a curry for their aroma and flavor, as a natural herbal medicine, and sometimes as an ingredient in the production of clarified butter.[32]

Coriander, commonly called dinbilal in Ethiopia, is an annual spice and aromatic herb that belongs to the family of *Umbelliferae/Apiaceae*. The leaves of coriander are used for flavoring soups and other foods. Since ancient times, coriander has been used to flavor foods and beverages, especially gin, and also medically to treat various diseases, particularly as a carminative.[33] Ethiopia has been the origin of genetic diversity in coriander.[34] The spice is used locally for flavoring purposes in the preparation of red pepper powder, bread, and sauces. Coriander is boiled in water and drunk on an empty stomach to treat stomach-ache.[33]

Nutmeg, which is called korerima in Ethiopia, is a shiny brown seed. It is usually sold enclosed in the dried fruit and when nutmeg fruit is crushed, it has a distinctive sweet smell and a delicious mild spicy taste. It belongs to the family of *Myristicaceae*. Nutmeg is well known in Ethiopia and Eritrea as an important and essential ingredient of many traditional Ethiopian spice mixtures. It is also used to flavor coffee as well as a traditional medicine in tonics and carminatives.[35]

Black cumin seeds are one of the aromatic seeds within the *Apiaceae* family. It is used in foods, fragrances, and medical preparations. Black cumin seeds are also used as an antispasmodic, carminative, and an appetite stimulating agent. Cumin is regularly used as a flavoring agent in a number of ethnic cuisines. It is used to season many dishes, as it draws out their natural sweetness. Cumin has also been used on meat and salsa, in addition to other common seasonings, to give extra flavor.[36]

Thyme belongs to the family of *Lamiaceae*. Thyme leaf is considered among the most valuable plants around the world and beyond its common culinary application, it is used cosmetically and medicinally. Due to its phenolic constituents (i.e., thymol and carvacrol), thyme has anti-bacterial and anti-oxidant activities.[37] Traditionally, it is edible as a remedy for whooping cough and other types of coughs, diarrhea, and digestive disorders.[38] Ethiopian thyme is a good source of iron and is widely used in Ethiopian cuisine, such as in tea, as a medicinal remedy to improve respiratory function and in many sauces including pasta sauce.[39]

Basil, called besobla in Ethiopia, is a popular herb that belongs to the family of *Lamiaceae* (mint). It is an annual spice herb. Basil is cultivated for use as a spice or flavoring agent and its aromatic leaves are used, fresh or dried, to add flavor to soups, tomato dishes, fish, poultry, vinegar, vegetables, salads, powdered beef, sausages candy, gelatins, and ice cream.[40] It is also used in dairy products to flavor yoghurt and cheese, to flavor tea, and to make berbere.[39]

The objective of the present study was to quantify the levels of fluoride in the spices most commonly used, cultivated, and consumed in Ethiopia. Since all Ethiopian foods are prepared with spices and there are no literature reports on the fluoride levels in the spices cultivated and consumed in Ethiopia, this study undertook to determine the exposure of humans in Ethiopia to fluoride from the commonly used spices.

MATERIALS AND METHODS

Sample collection: The spice samples in this study were collected from the Merkato and Shola spice markets in Addis Ababa, Ethiopia. The sellers in both spice markets had for sale different types of spices from cultivation areas in the rural parts of Ethiopia and all types of spices were available. It was therefore easy to get the representative samples of the selected spices for this study. All the selected spices (red chili pepper, garlic tuber, cinnamon bark, turmeric rhizome, ginger rhizome, fenugreek seeds, coriander seeds, nutmeg seeds, black cumin seeds, thyme leaves, and basil leaves) were collected from the Merkato and Shola spice markets on December 10 and 15, 2014, respectively. About 0.25 kg of each spice was bought from eight different spice shops (four shops from each of the two markets). A total of 88 spice samples (eight samples for each eleven spices) were collected and brought to the laboratory. The collected samples for each spice type were then mixed together into polyethylene plastic bags to get homogenized or representative spice samples weighing 2 kg for each spice.

Sample pre-treatment: The spice samples were washed first with tap water and was then with de-ionized water to reduce contamination. After washing, the samples were chopped into small sizes and dried in an oven (Digitheat, J.P. Selecta, Spain) at 70ºC to a constant weight. The dried samples were then ground using an electric grinder (Geepas Electric Coffee Grinder, Mainland, China) in the laboratory and sieved through a 1 mm sieve. The powder was stored in clean, dry, tightly closed plastic bottles until the digestion.

Instrumentation: A pH/ISE meter (Orion model, EA 940 Expandable Ion Analyzer, USA) equipped with a combination fluoride selective electrode (Orion Model 96-09, USA) was used for the determination of the fluoride ion in the standards and spice samples. The pH was measured with a pH/ION meter (HANNA instruments HI 9025, Malaysia) using a pH glass electrode. A muffle furnace (Audiotronics, Wagtech International Ltd., UK) was used to ash samples in nickel crucibles (50 mL).

Chemicals and reagents: The reagents used in this study were all of analytical grade. De-ionized water which is chemically pure, with a conductivity of 1.5 µs/cm, was used throughout the experiment. Nitric acid (69% HNO_3, Research Lab Fine Chem Industries, Mumbai, India) was used for cleaning purpose and sodium fluoride (99%, Analar, NaF, BDH Chemicals Ltd, England) was used to prepare the standard solutions. The pH standard buffers (pH of 4, 7, and 10) were used for the pH calibration. Sodium chloride (Fisher Scientific UK), glacial acetic acid

(100%, Sigma-Aldrich Laborchemikalien, Germany), trisodium citrate (BDH Laboratory Supplies Poole, England), and EDTA (Scharlau Chemie S.A., Barcelona, Spain) were used to prepare the total ionic strength adjustment buffer (TISAB II) solution. Sodium hydroxide (Scharlau Chemie S.A., Sentmenat, Spain) solution was used to dissolve the spice samples homogeneously before alkali fusion and also used to adjust the pH of the TISAB solution to a pH of 5.3. Hydrochloric acid (36%, Fisher Scientific UK Limited) was used for the neutralization of the dissolved fusion cake.

TISAB was prepared by dissolving 58 g of sodium chloride, 57.0 mL of glacial acetic acid, 7 g of trisodium citrate, and 2 g of EDTA in 500 mL of de-ionized water into a 1000 mL beaker and the pH was adjusted to 5.3 with 5 M sodium hydroxide. The solution was then transferred to a 1000 mL volumetric flask and diluted to the mark with de-ionized water.[41]

Fluoride determination in the spice samples: The determination of fluoride in the spices using a fluoride ion selective electrode was done by slightly modifying the reported method.[9] Nine reagent blanks, containing 5 mL of 8 M sodium hydroxide solution were also fused by the same procedure as samples for blank determination. The calibration standard solutions of 0.05, 0.5, 1, 5, and 10 mg/L were prepared by serial dilution from the 1000 mg/L fluoride stock solution. An aliquot of 5 mL of each of standard solution was poured into a 50 mL plastic beaker separately and 5 mL of TISAB II solution was mixed with it to calibrate the fluoride ion selective ion electrode. An aliquot of 5 mL of de-ionized water and 5 mL of TISAB were poured into another 50 mL plastic beaker as a standard blank solution for the blank correction. The electrode potentials of these standard and blank solutions were then measured by stirring the solutions constantly at room temperature with the fluoride ion selective electrode. The calibration curve of five series points was constructed as electrode potentials versus log of standard fluoride concentrations. The slope of the calibration curve was –58.5 mV per decade of concentration change which showed a good precision of measurement.

An aliquot of 5 mL of each sample solution was poured into a 50 mL plastic beaker and 5 mL of TISAB II mixed with it. Similarly 5 mL of fused blank solution and 5 mL of TISAB II were mixed into 50 mL plastic beakers. The electrode potentials of these standard and blank solutions were then measured by stirring the solutions constantly at room temperature with the fluoride ion selective electrode. All analyses were made in triplicate at room temperature.

Method validation for fluoride determination: In this study, validation of fluoride measuring method was checked by performing recovery tests as well as evaluating the precision of the triplicate determinations for all the spice samples. A satisfactory recovery tells us that there are no interfering agents and that the fluoride added has been detected. For the recovery study, the spiked samples were prepared by adding a known concentration of fluoride from a 10 mg/L fluoride standard solution. Samples of 0.5 g of red chili pepper, garlic granules, cinnamon bark, turmeric rhizome, ginger rhizome, fenugreek seeds, coriander seeds, nutmeg seeds, black cumin seeds, thyme leaves, and basil leaves were spiked separately with 154.5, 163.5, 128.5, 82.3, 140.5, 79.5, 214, 225.5, 407, 214.3, and 253 μL of 10 mg/L fluoride standard solution, respectively. The fluoride levels of the unspiked and spiked spice samples were analyzed in triplicate in similar conditions and the percent recoveries were calculated.[42]

Statistical analysis: All measurements were made in triplicate and the results were recorded as mean±standard deviation (SD). The variation between the mean fluoride levels in spice samples was tested. Analysis of variance (ANOVA) was used to check the for the presence of a significant difference at the 95% confidence level ($p<0.05$) between the mean fluoride levels in the spice samples. It indicates whether the source of variation in the fluoride levels was random error during the sampling, measurement, or treatment of the samples or due to heterogeneity among the samples. One way ANOVA was used to compare whether there was a significant difference between the mean fluoride levels of selected spice samples.

RESULTS AND DISCUSSION

Recovery results of fluoride determination: The recovery test was done for all the samples since all the samples had different matrices and the percentage recoveries were calculated by spiking known amounts of fluoride into the 0.5 g spice samples (Table 1).

Table 1. Recovery test results of fluoride determination in Ethiopian spices
(Values are mean ± SD, n = 3)

Spice type	F^- in unspiked sample (mg/kg)	Amount of F^- added to sample (mg/kg)	F^- in spiked sample (mg/kg)	% Recovery
Red chili pepper	3.10 ± 0.07	3.10	6.34 ± 0.1	105 ± 5
Garlic granules	3.27 ± 0.13	3.27	6.25 ± 0.63	97 ± 7
Cinnamon bark	5.13 ± 0.23	2.57	7.66 ± 0.31	105 ± 8
Turmeric rhizome	3.29 ± 0.07	1.65	4.98 ± 0.14	106 ± 7
Ginger rhizome	2.81 ± 0.13	2.81	5.64 ± 0.59	96 ± 6
Fenugreek seeds	3.18 ± 0.13	1.59	4.83 ± 0.07	103 ± 6
Coriander seeds	2.14 ± 0.04	4.28	6.55 ± 0.03	103 ± 3
Nutmeg seeds	4.51 ± 0.12	4.51	9.5 ± 0.5	98 ± 4
Black cumin seeds	8.14 ± 0.15	8.14	15.9 ± 0.16	96 ± 3
Thyme leaves	8.57 ± 0.11	4.29	12.6 ± 1.2	98 ± 8
Basil leaves	2.53 ± 0.05	5.06	7.74 ± 0.18	103 ± 3

The percentage recoveries were in the range of 96–106% which confirms that the analytical method for the total fluoride measurement used in the study was reliable (Table 1). The method detection limit, which is defined as the least concentration of the analyte that can be detected within a predetermined level of confidence and is equal to three times the standard deviation of the blank, was 0.04 mg/kg. The percentage relative standard deviations (% RSD) were <5% which showed that the method of analysis was precise.

Level of fluoride in the spice samples: The spices selected for the study were found to contain significant amounts of fluoride (mg/kg dry wt) with a range of 2.14 (coriander seeds) to 8.57 (thyme leaves, Table 2). The decreasing order for the fluoride levels in the spices was thyme leaves > black cumin seeds > cinnamon bark > nutmeg seeds> turmeric rhizome > garlic granules > fenugreek seeds> red hot chili pepper > ginger rhizome > basil leaves > coriander seeds. Such variations in the fluoride levels amongst the spice samples may be due to variations in the spice type, soil pH, the agrochemicals (fertilizers, pesticides, and herbicides) used during cultivation, and the mineral content (Al, Ca, Fe, and F^-) of the soil, the irrigation water, and the atmosphere.

Table 2. Mean concentration of fluoride in spices

(Values are mean ± SD, mg/kg dry weight basis, n = 9)

Spice type	F^- level (mg/kg)	Spice type	F^- level (mg/kg)
Red chili pepper	$3.10 \pm 0.07^{*,\dagger}$	Coriander seeds	$2.14 \pm 0.04^{*}$
Garlic granules	$3.27 \pm 0.13^{*,\dagger,\ddagger}$	Nutmeg seeds	$4.51 \pm 0.12^{*}$
Cinnamon bark	$5.13 \pm 0.23^{*}$	Black cumin seeds	$8.14 \pm 0.15^{*}$
Turmeric rhizome	$3.29 \pm 0.07^{*,\ddagger}$	Thyme leaves	$8.57 \pm 0.11^{*}$
Ginger rhizome	$2.81 \pm 0.13^{*}$	Basil leaves	$2.53 \pm 0.05^{*}$
Fenugreek seeds	$3.18 \pm 0.13^{*,\dagger,\ddagger}$		

Comparing any two spices, *p<0.05 except for those also marked † for whom p>0.05 and for those also marked ‡ for whom p>0.05.

The ANOVA results showed that statistically significant differences (p<0.05) at 95% confidence level existed between the mean fluoride levels (mg/kg dry wt) of the selected spices except for the mean fluoride levels between red chili pepper, fenugreek seeds, and garlic granules which were not significantly different (p>0.05, Table 2). The same was true for turmeric rhizome, garlic granules and fenugreek seeds (Table 2). The variation of the fluoride levels in the samples may be due to heterogeneity of the samples, mineral variations in the soil, irrigation water, and the atmosphere, and variations in the agrochemicals used during cultivation, such as fertilizers, pesticides, and herbicides. On the other hand, the cause of non-significant different mean fluoride levels in the mentioned spice sample groups may be due to the presence of similarities in climatic conditions, soil type, and irrigation water.

Daily intake of fluoride by an adult person through spices: The total fluoride intake (TFI) is the summation of the daily fluoride intake from the entire food sources. The daily intake of fluoride (DIF) from a particular food was calculated by multiplying the fluoride concentration in the particular food item by the total quantity of that particular item consumed per person per day. The total fluoride intake was calculated using the following equations:

$$\text{TFI} = \sum(\text{DIF})$$

and

$$\text{DIF} = \text{FC} \times \text{QF}$$

where TFI = total fluoride intake, DIF = daily intake of fluoride, FC = fluoride concentration in the food source, and QF = quantity of the particular food item taken per person per day.

The quantity of a particular food item taken per person per day was calculated by multiplying the amount of food item taken in a day by the frequency of consumption in a week and divided by the number of days in a week (7).

The daily intake of fluoride from spices was calculated based on the spices consumed regularly and did not include other spices which were infrequently consumed. The suggested average amounts of particular spices consumed by adults and the predicted daily intake of fluoride from these are shown in Table 3.

The percentage of the recommended daily intake of fluoride (% RDIF) from a particular spice was calculated from the following equation:

$$\%\ \text{RDIF} = \frac{\text{DIF}}{\text{RDIF}} \times 100$$

where DIF = daily intake of fluoride
and % RDIF = percentage of the recommended daily intake of fluoride.

Although it was found that fluoride was not an essential nutrient, and therefore no average requirement for the performance of essential physiological functions could be defined, the European Food Safety Authority Panel on Dietetic Products, Nutrition, and Allergies (NDA) considered that the setting of an Adequate Intake (AI) was appropriate because of the beneficial effects of dietary fluoride on the prevention of dental caries based on epidemiological studies, performed before the 1970s, showing an inverse relationship between the fluoride concentration of water and caries prevalence.[43] They recommended a daily intake of fluoride (RDIF) from all sources for both children and adults, including pregnant and lactating women of 0.05 mg/kg body weight.[43] For pregnant and lactating women, the adequate fluoride intake is based on their body weight before pregnancy and lactation.[43] According to the European Food Safety Authority and the National Academy of Sciences Institute of Medicine, the recommended daily intake of fluoride (RDIF) for adults having 60–70 kg body weight is usually within the range of 3–4 mg/kg.[43,44] The value of a daily intake of 0.05 mg/kg body weight has been extensively used as a reference to estimate the risk of fluorosis from spices or the relative contribution of fluoride intake from the spices selected for the present study.

In the present study, the daily intake of fluoride was estimated for an adult consuming the amount of a particular spice suggested in Table 3 together with the percentage of the recommended intake of fluoride from the spice for an adult of 60 kg body weight, based on an AI of 0.05 mg F/kg body weight per day (Table 3).

Table 3. Daily intake of fluoride for adults from different spices

Parameter	Spice type					Total
	Red chili pepper	Garlic granules*	Cinnamon bark	Ginger rhizome*	Turmeric rhizome	
Average amount of the spice consumed by an adult on the days when spices are eaten (g)	20	2.1	1.5	1.83	4.5	
Frequency of consumption per week	7	6	5	4	4	
Average daily amount of spice consumed (weekly consumption ÷ 7, g/day)	20	1.8	1.07	1.05	2.57	
Concentration of fluoride in the spice (mg/kg dry wt)	3.1	3.27	5.13	2.81	3.29	
Fluoride intake (mg/day)	0.06	0.006	0.005	0.003	0.008	0.082
Relative daily intake of fluoride (%)†	2	0.2	0.18	0.1	0.28	2.76

*An adult normally consumes 15 g fresh garlic granules and 10 g fresh ginger rhizome which corresponds to 2.1 g and 1.83 g dry weight, respectively. The moisture content of fresh garlic granules and ginger rhizome were found to be 84.6% and 81.7%, respectively. †Based on an adequate intake (AI) of 3 mg/day (0.05 mg F/kg body weight per day for a 60 kg adult).

The daily intake of fluoride from spices for an adult of 60 kg weight ranged from 0.003 mg/day (ginger rhizome) to 0.06 mg/day (red chili pepper). The percentage of the daily AI for fluoride for a 60 kg adult, based on an AI of 0.05 mg F/kg body weight per day, from the individual spices were: red chili pepper: 2%, cinnamon bark: 0.18%, turmeric rhizome: 0.28%, ginger rhizome: 0.1%, and garlic granules: 0.2%. These five spices, if used daily, would provide 2.76% of the recommended AI for fluoride level (3 mg/day for adults, Table 3).

Comparison of fluoride levels of this study with literature values: Several studies have been made of the fluoride levels in spices cultivated in different countries (Table 4).

Table 4. Comparison of the fluoride levels in spices reported in the literature with those found in the present study

Spice type	F^- concentration (mg/kg dry wt)	Origin	Reference
Green chili pepper	4.9	Ethiopia	14
Red chili pepper	8.13	China	3
Red chili pepper	7.82	Spain	3
Red chili pepper	3.1	Ethiopia	Present study
Garlic granules	8.73	Spain	3
Garlic granules	5	India	7
Garlic granules	3.27	Ethiopia	Present study
Cinnamon bark	6.95	Mexico	3
Cinnamon bark	5.13	Ethiopia	Present study
Turmeric rhizome	3.3	India	7
Turmeric rhizome	3.29	Ethiopia	Present study
Ginger rhizome	2	India	7
Ginger rhizome	2.81	Ethiopia	Present study
Fenugreek seeds	3.18	Ethiopia	Present study
Coriander seeds	3.97	India	45
Coriander seeds	2.3	India	7
Coriander seeds	2.14	Ethiopia	Present study
Nutmeg seeds	4.51	Ethiopia	Present study
Black cumin seeds	1.8	India	7
Black cumin seeds	8.14	Ethiopia	Present study
Thyme leaves	8.57	Ethiopia	Present study
Basil leaves	8.85	Egypt	3
Basil leaves	2.53	Ethiopia	Present study

Emanuel and Jiří[3] determined the fluoride levels (mg F/kg dry wt) in spices cultivated in four countries: Spanish red hot pepper: 7.82, Chinese chili pepper: 8.13, Egyptian basil leaves: 8.85, Spanish garlic granules: 8.73, and Mexican cinnamon bark: 6.95. These reported values are higher than the fluoride levels found in the present study (red chili pepper 3.1, basil leaves 2.53, garlic granules 3.27, and cinnamon bark 5.13). These variations in the fluoride levels may be due to variations in the combustion techniques and to differences in the levels of fluoride and other minerals, like Al, Ca, Mg, and Fe, in the soil, the irrigation water, and the atmosphere. Variations in the agrochemicals, like fertilizers, pesticides, and herbicides, used during cultivation may also cause differences in the fluoride content between similar spices cultivated in different countries or even between spices cultivated in the same country but in different places. In industrialized countries, fluoride-containing industrial dusts, mining activities, and coal burning may increase the fluoride levels of spices.[37]

In the other studies in Ethiopia, Dessalegne and Zewge found that the level of fluoride in green chili pepper was 4.9 mg/kg dry wt.[14] This reported value is higher than the fluoride level of red chili pepper (3.1 mg/kg dry wt) found in the present study. This variation of fluoride content may be due to differences in the geographical locations of the cultivation sites of the samples.

Manoj[7] found the fluoride levels (mg/kg dry wt) in spices cultivated in Orissa, India, were: garlic granules: 5, coriander seeds: 2.3, cumin seeds: 1.8, turmeric rhizome: 3.3, and ginger rhizome: 2.0. The fluoride levels reported by Manoj[7] for coriander seeds, ginger rhizome, and turmeric rhizome were comparable with the levels of fluoride found in the present study while the fluoride levels found in the present study were higher for cumin seeds (8.14) and lower for garlic granules (3.27) than the values reported by Manoj. In another Indian study, Khandare and Rao[45] found that the level of fluoride in coriander seeds (3.97) was somewhat higher than the value found in the present study (2.14). We did not find any reports in the literature for fluoride levels in fenugreek seeds, thyme leaves, or nutmeg seeds.

CONCLUSION

In the present study, the levels of fluoride in 11 of the most widely consumed traditional Ethiopian spices were determined. The levels of fluoride (mg/kg dry wt) in the spices were, in decreasing order: thyme leaves (8.57±0.11) > black cumin seeds (8.14±0.15) > cinnamon bark (5.13±0.23) > nutmeg seeds (4.51±0.12) > turmeric rhizome (3.29±0.07) > garlic granules (3.27±0.13) > fenugreek seeds (3.18±0.03) > red chili pepper (3.10±0.17) > ginger rhizome (2.81±0.13) > basil leaves (2.53±0.05) > coriander seeds (2.14±0.04). Analysis of variance showed that there were statistically significant differences ($p<0.05$) in the mean levels of fluoride among the spice samples apart from those in two groups: (i) red chili pepper, fenugreek seeds, and garlic granules; and (ii) turmeric rhizome, garlic granules, and fenugreek seeds. It was also found that the daily intake of fluoride from red chili pepper, cinnamon bark, turmeric rhizome, ginger rhizome, and garlic granules for an adult (60 kg body weight) who consumed all of these spices was 0.082 mg/day or 2.76% of the recommended daily fluoride intake of 3 mg fluoride, based on the guideline for an adequate intake of 0.05 mg F/kg body weight. This indicates that the spices consumed by adults in Ethiopia contain only a relatively small amount of fluoride compared to the currently recommended daily fluoride intake.

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