ORIGINAL ARTICLE

Level of Fluoride in Well and Spring Waters of Jimma Zone, South West Ethiopia

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Abstract

In this study, the level of fluoride in well and spring waters of selected woreda (district) of Jimma zone, Oromia Regional State, Southwest Ethiopia was investigated. Water samples were collected from Jimma, Serbo, Seka Chekorsa and Agaro towns in July, 2016. The collected water samples were stored at 4 °C until analysis. The fluoride content of water samples was determined using fluoride ion selective electrode (FISE). External calibration curve was employed in the range of 0.10–30 mg/L of fluoride standard solutions. The resulting calibration curve was exhibited good linearity with coefficient of determination (R^2) 0.999. Satisfactory recoveries ranged from 79–120%, with standard deviation lower than 4.89 were obtained, indicating the validity of the employed method. Concentrations of fluoride in Jimma town well and spring waters were ranged from 0.19–17.73 mg/L and 0.18–0.25 mg/L, respectively. Whereas, the fluoride content of Serbo, Seka Chekorsa and Agaro well waters were ranging from 0.21–0.23 mg/L. The highest fluoride content (17.73 mg/L) was observed in well water of Jimma University. Except well water of Jimma University, the obtained fluoride content of all the studied water samples were below the WHO guideline (<1.5 mg/L) for drinking water. One way ANOVA test at $p < 0.05$ demonstrated the absence of significant difference in the fluoride content among the well waters of Serbo, Seka Chekorsa and Agaro. There was also no significant difference in the fluoride contents of spring waters collected from different area of Jimma town: Bacho Bore, Bossa Addis and Kochi.

Keywords*: Fluoride, Ion Selective Electrode, Spring and Well Waters*

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INTRODUCTION

Water is one of the essential natural gifts for all forms of life and is available in abundance, covering approximately three-fourth of the surface of the earth. The quality of water is one of the most important criteria that determine its usefulness for a specific need such as irrigation, recreation, drinking and so on. Primarily, large proportion of people in developing countries lack access to safe drinking water (UNICEF, 1995). Water can be contaminated by fluoride, heavy metals and organic waste residues. Fluoride is persistent and non-degradable that accumulates in soil, plants, wild life and in humans. Fluoride might enter into water ecosystems via geological processes as well as from industrial waste discharges. High fluoride concentrations are primarily associated with volcanic and fumarolic activities, which usually add fluoride to the groundwater (Fawell et al., 2006). Minerals that contain fluorides such as fluorite (CaF_2), apatite (Ca_5 (Cl,F,OH)$(PO_4)_3$), cryolite (Na_3AlF_6) are also other potential sources of fluoride in water (Nair et al., 1984).

Geographical belts of high fluoride concentrations in groundwater extend from Syria through Jordan, Egypt, Libya, Algeria, Morocco and the Rift Valley of East Africa through Ethiopia, Kenya and Sudan. Another belt stretches from Turkey through Iraq, Iran and Afghanistan to India, northern Thailand and China (WHO & EU, 2006). The East African Rift Valley, which passes through Ethiopia, is geomorphologically active volcanic activities. As a result, high amount of fluoride that could be released from volcanic rocks, such as the young basalt contains high concentrations of fluoride and fluorapatite (Ghiglieri et al., 2012). Moreover, the Rift Valley region is characterized by high hydrothermal activity that accelerates the solubility of fluoride, favoring the development of endemic fluorosis (Alemu et al., 2015).

Drinking water is usually the main source of fluoride intake, especially in areas where fluoride concentration is high in groundwater and/or surface water (Jeffery et al., 1998; Redda et al., 2006; Edmunds & Smedley, 2013). For instance, the water supplies in the Ethiopian Rift Valley regions are mainly from well water with depths of 10 to 100 m (Redda et al., 1987). Besides, foods, beverages and/or indoor air pollution due to the combustion of coal may also contribute to the total fluoride intake per day (Nielsen & Dahi, 2002). Perhaps, the presence of low concentration of fluoride (about 1.0 mg/L) in food and drinking water is useful to prevent dental caries or tooth decay (Fung et al., 1999; Shomar et al., 2004). However, exposure to high concentration of fluoride might cause severe diseases on teeth (dental fluorosis) and on bones (skeletal fluorosis). Prolonged intake of water with fluoride content of 3–6 mg/L also causes skeletal fluorosis, if these concentrations are exceeded, crippling skeletal fluorosis occur. Normally, the degree of fluorosis depends on the amount of exposure up to the age of 8 to 10 years. Due to isomorphic substitution, fluoride mainly gets deposited in joints of the knee, neck, pelvis and shoulder and makes it difficult to move or walk (WHO, 2002; Fawell et al., 2006). Fluorosis is an irreversible process and has no medication. The only remedy of its prevention is to keep the fluoride intake level below the safe limits (Raju et al., 2009; Teutli et al., 2012). The study conducted in Ethiopia indicated that the mean prevalence of the Rift Valley community was 83.2%. About 8.5 million people leaving in the Rift Valley region of

Ethiopia are exposed to high levels of naturally occurring fluoride (Rango et al., 2014). In China alone, more than 10 million people are estimated to suffer from skeletal fluorosis (Raju et al., 2009). Long-term exposure to high concentration of fluoride could also cause change of the DNA-structure, paralysis of volition, cancer, etc. (Nielsen & Dahi, 2002). Therefore, WHO has set the maximum permissible level of fluoride in drinking water to be 1.5 mg/L (WHO, 1984).

Groundwater in the Rift Valley region of Ethiopia contains high concentration of fluoride which severely affected the health of the community living in the area (Tamiru, 2000; Reimann et al., 2003; Tesfaye, 2004). Peoples living in this region are susceptible to severe dental fluorosis due to their exposure to high concentration of fluoride containing drinking water (Gikunju et al., 2002), raw vegetables and juices (Njenga et al., 2005), as well as milk (Kahama et al., 1997). Olsson (1979) reported that the high prevalence of dental fluorosis (99 %) was examined in 6‒7 years old children of Wonji and Hawassa in the Ethiopian Rift Valley region. The fluoride concentration in Wonji and Hawassa was 12.4 and 3.5 mg/L, respectively (Alemu, 2015). Redda et al. (2006) also found dental fluorosis in more than 80% of children in the Rift Valley region of Ethiopia. Other study conducted in two villages in the Rift Valley region also revealed that about 92–100% prevalence of dental fluorosis (Wondwossen et al., 2004). The presence of significant amount of fluoride in groundwater has also reported from other areas of Ethiopia such as in Oromia region (e.g., Jimma and Arsi Negele town) (FMOWR, 1997), Somalia region (e.g. Deghabur, Kebri Dehar, Jerer Valley, Hargele and Warder) (Yoseph, 2007), Tigay region (eg. Mekele town), Southern, Nations, Nationalities and Peoples' Region, SNNPR (e.g. Soddo, and Hawassa towns) (Reimann et al., 2003; Tesfaye, 2004).

The fluoride content of water, particularly, well water varied depending on many factors, like the geological, chemical and physical characteristics of the aquifer; porosity of soil and rocks, temperature and the depth of the wells (Maheshwari, 2006). In any geological environment, fluoride is a normal constituent of well waters (Edmunds & Smedley, 1996). In general, Ethiopian water contains high concentration of fluoride, ranging from 1.5–200 mg/L (Reimann et al., 2003). Some preliminary works on groundwater of Jimma town have shown the presence of significant amount of fluoride. But, there was no previous detailed study on the fluoride contents of well and spring waters of Jimma town and the surrounding woreda towns. Therefore, the objective of this study was to investigate the fluoride contents of well and spring waters of Jimma, Serbo, Seka Chorkosa, and Agaro towns. The variability of fluoride contents of the studied water samples were also statistically validated by employing t-test and one way ANOVA.

MATERIALS AND METHODS

Instrumentation

Hanna, pH 211 microprocessor-based pH/mv/°C Bench Meter and glass pH electrode were obtained from Hanna instruments, Inc (Póvoa de Varzim, Portugal). Fluoride ion selective electrode (FISE)-HI4010, and silver-silver chloride, HI5315 Reference Electrode were also purchased form Hanna instruments, (Woonsocket, Rhode Island, USA). Poly vinyl chloride (PVC) plastic bottles were used for sample collection.

Chemicals and reagents

All chemicals and reagents used in the analysis were analytical grade. Anhydrous sodium fluoride (NaF, 99.0%) and Glacial acetic acid were obtained from BDH Chemicals Ltd (Poole, England). Sodium chloride (NaCl), sodium hydroxide (NaOH) and disodium salt of ethylene diaminetetraacetic acid (EDTA) were purchased from Fisher Chemicals (Leicestershire, England). Nitric acid (HNO_3, 69–72%), was obtained from Fine Chemical Industries (Mumbai, India). Hydrochloric acid was obtained from Wardle Chemicals Ltd (Birmingham, England). Distilled water was used for preparation of standard solutions and rinsing glassware's and sampling bottles.

Preparation of solutions

Total ionic strength adjustment buffer (TISAB) solution was prepared by dissolving 58 g NaCl and 4 g the sodium salt of EDTA in a sufficient amount of distilled water, then 57 mL glacial acetic acid was added and the resulting solution pH was adjusted to 5.3 ± 0.1 using10 M NaOH. Finally, distilled water was added to obtain the final volume of 1000 mL.

A stock solution containing 1000 mg/L fluoride was prepared in polystyrene volumetric flask by dissolving 2.21 g NaF (dried in oven at 105 °C for 1 hour) in 1000 mL distilled water. Then, a series of standard solutions comprising seven concentration levels, ranging from 0.1–30 mg/L were prepared by serial dilution of the stock solution and used for construction of calibration curve.

Fluoride content determinations of the standard and/or real samples were then performed using FISE as follows: first, equal volumes of the sample or standard and TISAB were placed in a 100 mL plastic beaker. Then, after stirring the mixture using magnetic stirrer, the FISE was immersed into the solution to measure the concentration of the fluoride ion. In all cases, prior to quantitative determinations of fluoride content of the samples, the FISE was calibrated over a concentration range of interest.

Sampling

The study was conducted in four towns of Jimma zone including Jimma town (the capital of the zone) and three woreda towns: Serbo, Seka Chokorsa, and Agaro, which are located in Oromia Regional State, in Southwest of Ethiopia. Six well and four spring water samples were collected from different localities (Kebeles) of Jimma town. Well water samples were collected from Medianalem Sefer (MSW), Bacho-Bore (BBW), Furustale Sefer (FSW), Kochi Sefer (KSW), Bosa–Addis (BAW) and Jimma University Main Campus, Kole Ber (JUKBW) and spring water samples were obtained from Bocho-Bore (BBS), Kochi-Sefer (KSS), Bosa-Addis (BAS) and Furstale Sefer (FSS). Moreover, six well water samples, i.e., two samples from different localities of each woreda town: Serbo, Seka Chokorsa, and Agaro were collected. Details of the Global Position System (GPS) of the specific sampling sites of well and spring waters are presented in Table 1.

Table 1. Specific sampling sites of the well and spring water samples

Samples	Sampling Sites	Altitude (m)	North-direction	East- direction
Jimma town well water	JUKBW	1716	N-07^{0}40'42.3'	E-036^{0}51'3.72"
	BBW	1712	N-07^{0}41'8.44'	E- 036^{0}51'3.22"
	BAW	1821	N-07^{0}41.155'	E-036^{0}50'3.95"
	KSW	1724	N-07^{0}41'2.31'	E-036^{0}50'57.31"
	FSW	1730	N-07^{0}40.016'	E-036^{0}51.083'
	MSW	1729	N-07^{0}40.321'	E-036^{0}51'4.22"
Jimma town spring water	BBS	1722	N-07^{0}39.210'	E-036^{0}49'6.25"
	KSS	1728	N-07^{0}41'2.31'	E-036^{0}50'57.31"
	BAS	1709	N-07^{0}41'8.44'	E- 036^{0}51'3.22"
	FSS	1730	N-07^{0}40.016'	E-036^{0}51.083'
Woreda towns well water	AGW1 & AGW2	1614	N- 07^{0}51.034'	E-036^{0}35'11.5"
	SKW1 & SKW2	1838	N-07^{0}36.300'	E-036^{0}44'356"
	SEW1 & SW2	2660	N-07^{0}35.800'	E-036^{0}46'3.14"

The samples were collected using PVC bottles previously cleaned by soaking in 10 % HNO_3 for 24 h. Then, after washing using non-ionic detergent, it was rinsed with distilled water prior to use. Before sampling, well and spring waters were allowed to dewater for 10 minutes to get the real sample from the depth of the site. During sampling, sampling bottles and their caps were repeatedly rinsed with the water to be sampled. Afterwards, the collected samples were stored in refrigerator at 4 ^{o}C until analysis time.

Method validation

Calibration curve linearity

For quantitative determination of fluoride in the collected water samples, external calibration curve was employed. A series of seven fluoride standard concentrations were prepared in distilled water and were then used for construction of the calibration curve. Determination of fluoride content in the real samples was then performed using the linear equation of the calibration curve.

Determination of limits of detection and quantification

Limit of detection (LOD) is the minimum concentration of an analyte that can be measured and reported with 99% confidence that the analyte concentration is greater than zero. Whereas, limit of quantitation (LOQ) which is also called limit of determinations is the lowest concentration of the analyte that can be measured in the sample matrix at an acceptable level of precision and accuracy. In this study, LOD and LOQ were determined using blank samples which were prepared by mixing equal volume of TISAB and distilled water. Their values are then obtained using equation 1 (Muriphy & Riley, 1962).

$$\mathrm{LOD} = \mathrm{X_b} + 3\mathrm{S_b} \ \& \ \mathrm{LOQ} = \mathrm{X_b} + 10\mathrm{S_b} \quad (1)$$

Where, X_b and S_b are mean concentration and standard deviation of the blank samples, respectively.

Precision and recovery studies

Precision study was carried out in terms relative standard deviation (%RSD) of the replicate analysis of both the blank and spiked real samples (Muriphy & Riley, 1962). Similarly, recovery (%R) was also studied by spiking the real water samples and the equation 2 is used for %R calculation.

$$\%R = \frac{C_{sp} - C_{unsp}}{C_{an}} \times 100 \qquad (2)$$

Where, %R, C_{sp} C_{unsp} and C_{an} are recovery, concentration of the analyte in the spiked sample, concentration of the analyte in unspiked sample and concentration of the spiked analyte, respectively.

Statistical analysis

The obtained data were statistically evaluated using t-test, and one-way ANOVA. Statistical tests are usually employed to differentiate whether the studied samples contain either equal or different concentrations of fluoride. Accordingly, the tests were performed at ($p < 0.05$) confidence interval (Miller et al., 1984).

RESULTS AND DISCUSSION

Construction of the calibration curve

Quantitative determination of the target analyte was performed using FISE based on an external calibration curve. A calibration curve was constructed using a series of seven standard concentrations of the analyte prepared in distilled water. The curve was constructed based on potential as the function of concentration of the target analyte. Figure 1 shows the constructed calibration curve. As can be seen, the obtained calibration curve has good linearity with coefficient of determination (R^2) of 0.999. The concentrations of the analyte in the studied water samples were then determined using the linear equation of the curve. The LOD and LOQ of the method were also determined using blank sample (equal volume of distilled water and TISAB), which was free from the analyte of interest. Accordingly, the LOD and LOQ, which was obtained based on the determination of seven blank samples (n = 7), were 0.07 mg/L and 0.095 mg/L, respectively.

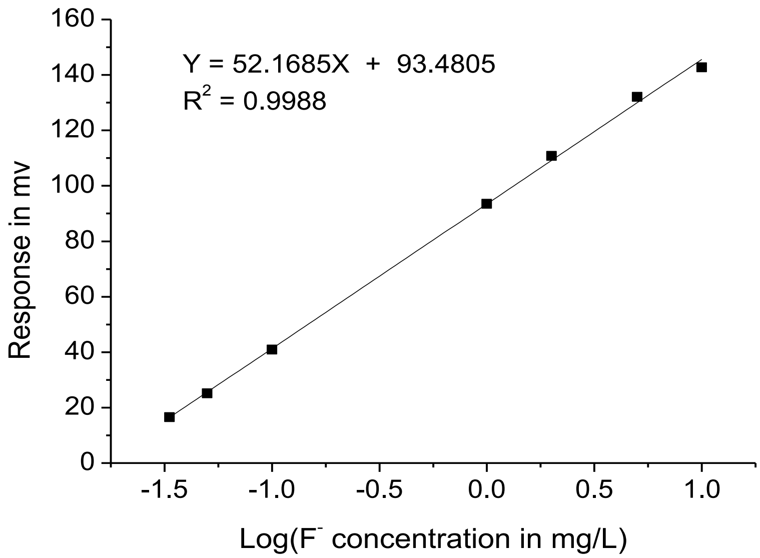

Figure 1. Calibration curve for the determination of fluoride in well and spring water samples

Recovery studies

The accuracy of the analytical method was evaluated by analyzing the spiked well and spring water samples with 0.4 mg/L of fluoride standard solution. The recoveries were calculated using equation 2 and they are found to be in the range of 79–120 %. The observed recoveries are generally in the acceptable range which indicates the validity of the employed method. The obtained recoveries are presented in Table 2.

Table 2. Recoveries (%R ± SD, n = 4) of fluoride in well and spring water samples

Sample	**%R ± SD**	**Sample**	**%R ± SD**	**Sample**	**%R ± SD**
BBS	94 ± 4.89	KSW	83 ± 1.55	SEW2	120 ± 0.62
BAS	109 ± 2.14	MSW	97 ± 0.92	SKW	116 ± 2.45
FSS	117 ± 1.23	AGW	82 ± 0.61	SKW2	79 ± 1.57
KSS	117 ± 0.61	AGW2	79 ± 1.05	FSW	93 ± 0.31
BBW	86 ± 2.14	SEW	111 ± 2.14	JUKBW	98 ± 0.93

SD: standard deviation

Determination of fluoride in the well and spring water samples

The fluoride content of water samples was determined by analyzing duplicate samples of each water sample collected from different localities. The observed fluoride concentrations exhibited variability with sampling location, particularly in well and spring waters of Jimma town. Table 3 demonstrates the fluoride concentrations of the studied well and spring water samples.

Table 3. Concentrations of fluoride (Mean ± SD, in mg/L) in well and spring water samples

Jimma town Well water	F^- (mg/L) Mean ± SD	Woreda towns Well water	F^- (mg/L) Mean ± SD	Jimma town Spring water	F^- (mg/L) Mean ± SD
KSW	0.20 ± 0.003	SEW1	0.21 ± 0.005	BBS	0.18 ± 0.009
BBW	0.19 ± 0.004	SEW2	0.21 ± 0.001	BAS	0.19 ± 0.004
MSW	0.17 ± 0.004	SCW1	0.23 ± 0.006	FSS	0.25 ± 0.003
FSW	1.06 ± 0.003	SCW2	0.21 ± 0.003	KSS	0.19 ± 0.001
JUKBW	17.73 ± 0.003	AGW1	0.22 ± 0.002		
BAW	0.24b ± 0.005	AGW2	0.22 ± 0.002		

SD: Standard deviation

As can be observed from Table 3, the highest fluoride concentration was observed in well-water of JUKBW (17.73 ± 0.003 mg/L) and the lowest were observed in well-water of MSW (0.17 ± 0.004 mg/L). Except JUKBW, the obtained fluoride content of all water samples were below the permissible limits set by WHO for drinking water (Alemu et al., 2015). One way ANOVA test at $p < 0.05$ demonstrated the absence of significant difference in the fluoride content of the well water samples collected from the three woreda towns (Serbo, Seka Chekorsa and Agaro). Similarly, there was no significant difference in the fluoride concentrations of Bacho Bore, Bossa Addis and Kochi spring waters. The high fluoride concentration observed in JUKBW might be primarily associated to low calcium concentrations, which restrict the precipitation of fluoride as fluorite (CaF_2) (Apambire et al., 1997).

Nevertheless, both well and spring water samples collected from Frustale area have relatively higher fluoride concentrations than the rest except JUKBW, and also statistical analysis indicated the presence of the significant differences. This variation of fluoride from one source to another might be attributed to the irregular distribution of fluoride bearing minerals in the soils and composition of bedrocks, calcium-poor aquifers and anthropogenic sources for example fertilizers and pesticides as reported by Mwende (2000). Earlier reports also indicated that some Ethiopian well and spring waters contain high fluoride concentrations (Alemu et al., 2015). Table 4, presents some Ethiopian well and spring waters that contain fluoride concentration above WHO guideline (1.5 mg/L) for drinking water.

Table 4. Well and spring waters of Ethiopia that contains fluoride concentrations above WHO guideline (Alemu et al., 2015)

Region	Zone, wereda or another village	Source of Sample	Conc. F, in mg/L
Addis	Kotebe w2	well	3.35
Ababa	Yeka sub-city (Medeksu)	well	3.36
Afar	Hugub (Awash 7 kilo)	well	8.50
	Awash 7^{th}Camp	well	7.25
	Segento	well	10.30
	Metehara Suger Factory	well	16.25
Amhara	Dalecha D. birhan	well	5.53
	Hara Dembele	well	7.88
	Malima Biri	well	15.20
Somali	Afder, Bare town Kebele 01	well	12.12
	Afder, Bare	well	3.23
Oromia	Wonchi Gadam	Spring	7.75
	Arsi	Spring	8.50
	Wonji-Tabel	Spring	14.20
	Dugda woreda Tejitu	Well	8.67
	Dodo woreda, Jerme bora, East Shoa and Abo no 2	Well	10.00
	Gedema Kufa (Adama)	Well	26.25
	Dugda woreda Jido	Well	33.67
	Dugda woreda Urgi Mechefera	Well	36.00
	Meki Bole Elementary School	Well	40.00
SNNPR	Wondogenet	Spring	7.75
	Hawassa Hw-1	Spring	11.25
	Hawassa (Doga)	Well	13.32
	Hawassa 01(HD1)	Well	24.00
Tigray	Atsgede Tsimbla	Spring	6.52

The concentrations of fluoride obtained in well and spring water samples are relatively lower than the reported results from other parts of the country. High fluoride concentrations, above WHO guideline for drinking water, have been reported from various parts of Ethiopia. The highest fluoride level was reported from the Rift Valley region, which is the lowland area with the most recent volcanic activity in the country (Alemu et al., 2015). The results of the present study also indicated that the fluoride content in the groundwater of Jimma area (JUKBW) was comparable to that of the Rift Valley region.

CONCLUSION

In this study, fluoride concentrations of well water samples collected from Jimma town and its surrounding three woreda towns including Serbo, Seka Chekorsa and Agaro were investigated using FISE. In addition, the fluoride content of spring waters collected from different localities in

Jimma town was studied. Reliability of the utilized method was investigated using recovery studies, by spiking the sample with known concentration of fluoride and the observed recoveries in all cases were in the acceptable range, i.e., 79–120 %. The obtained fluoride contents of all the well and spring water samples were below the WHO guideline (1.5 mg/L), with the exception of the well water of JUKBW (17.73 mg/L). Thus, except JUKBW, the studied well and spring waters are safe in terms of fluoride content for drinking or other domestic uses. However, in order to obtain sufficient information about the fluoride content of well and spring waters of the area, further study should be conducted based on seasonal variations.

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