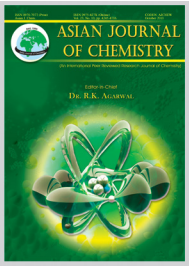
Asian Journal of Chemistry; Vol. 24, No. 7 (2012), 3225-3227

ASIAN JOURNAL OF CHEMISTRY

www.asianjournalofchemistry.co.in



Determination of Fluoride in Drinking Water and Black Tea by Ion Selective Electrode Using Standard-Addition Method

MARYAM KARAMI NOGOURANI[1], S.M. TALEBI[2,*] and M. EBRAHIMIAN[1]

[1]Department of Pediatric Dentistry, Dental Faculty, Islamic Azad University, Khorasgan Branch, Isfahan, Iran
[2]Department of Chemistry, the University of Isfahan, Isfahan, Iran

*Corresponding author: E-mail: smtalebi@yahoo.com

(*Received*: 30 August 2011; *Accepted*: 24 February 2012) AJC-11107

The fluoride content of drinking water and popular black tea in Iran was determined by ion selective electrode and standard addition method. A recovery test indicated that the method is quite reliable to determine the trace fluoride in sample solutions. The concentration of fluoride in drinking water in different mega cities was in the range of 0.12-0.31 µg/L. The concentration of fluoride in popular black tea extracts was in the range of 2.54-3.73 but the fluoride content in tea extract is depended to the infusion time.

Key Words: Determination of fluoride ion, Ion selective electrode, Fluoride in drinking waters.

INTRODUCTION

Fluoride is known as the most reactive non-elemental and electronegative element. It never occurs in nature in its elemental state. It combines with all elements except oxygen and noble gases to form fluoride. Fluoride is a natural component of earth crust. Fluoride is an important anion present in various environmental, pharmaceutical and food samples[1,2]. Orally ingested fluoride can easily be absorbed from gastrointestinal tract. The risk of dental caries is reduced due to uptake of fluoride, because fluoride interacts with hydroxyapatite and forming fluorohydroxyapatite which is more resistant to erosion by plaque acid. Although fluoride has many benefits, it is also toxic for human being and plants in larger amounts[3]. Excessive fluoride intake can cause fluorsis of both teeth and bones as a result intake of suitable amount of fluoride is of great importance. The adequate intake of fluoride for adults[4] is 0.05 µg Kg^{-1} per day. Drinking water is a source of fluoride in human diet. In some countries fluorinated water is a source for intake of adequate fluoride. United State Public Health Service has recommended fluorination levels 0.7-1.2 mg L^{-1} [5]. Although fluoride has many benefits, it is also toxic in larger amounts. For adults the lethal dosage is 0.2-0.35 g F^- per Kg body weight. Appreciable amounts of fluoride in the form of different compounds can enter the human body by food chain and drinks and water[6,7].

There are many different methods for determination of fluoride concentration include spectrometric method[8], ion selective electrode[9] and ion chromatography[10], among them ion selective electrode is most popular because of its simplicity, accuracy and precision. Fluoride selective electrode is very sensitive and it works in a wide range of temperature from 0 to 50 ºC. For potentiometric analysis of fluoride ion, in commercial usage, there are mostly used electrodes with homogenic membrane made from fluoride lantana (LaF_3), which was first suggested by Frant and Ross[11]. Today the potentiometric determination of fluoride using an ISE reference electrode pair is performed routinely in water analysis laboratories throughout the world.

The standard addition method has been used for determination of fluoride ion concentration. In this method, the electrode potential is measured for the sample solution and for the sample solution plus a small amount of a standard solution of the analyte. Essentially, we measure responses for the sample and calculating the concentration of analyte in sample would be possible. The standard method is better than measuring a calibration curve for standard solutions prepared in distilled water. With the method of standard additions, the analyte response is measured in the complex sample matrix.

In this work, the fluoride content of drinking water and tea infusions were determined by ion selective electrode and standard addition and the effect of water fluoride concentration on extracting of fluoride from tea was also investigated.

EXPERIMENTAL

Fluoride stock solution (1000 mg/mL) was prepared from sodium fluoride and stored in polyethylene containers.

Total ionic strength adjustment buffer (TISAB) solution contains 58.5 g of sodium chloride, 57 mL of glacial acetic

acid, 61.5 g of sodium acetate and 5 g of *trans*-1,2 cyclohexanediamino *N,N,N',N'* tetra acetic acid (CDTA), which were dissolved in Milli-Q water and diluted to 500 mL. pH of the solution was adjusted to 5.5 with 6 M solution of sodium hydroxide and then diluted to 1000 mL with Milli-Q water.

The working range fluoride standard solutions were made from the stock standard solution by sequential dilution.

Collection of water samples: Water samples were collected from the water supply system of different mega cities in Iran. Each time 500 mL of water was collected from each sampling point and transferred into the laboratory for analysis. Two water samples were collected from each location. The sampling sites were chosen across the country and were expected to provide nationally representative of Iranian drinking water.

Four more popular black tea brands were chosen and the fluoride content was determined after infusion and extraction of fluoride.

Extraction and analysis of fluoride: 2 g of each black tea sample was infused in about 70 mL of boiling Milli-Q water and the mixture was left in a water bath at 80 ºC for different periods of times from 5 to 15 min. After the period of infusion the mixture was filtered into a 100 mL volumetric flask and its volume was adjusted to 100 mL with Milli-Q water.

Determination of fluoride concentration: A 10 mL of sample solution from each 100 mL solution was chosen and 5 mL of total ionic strength adjustment buffer solution was added and used for the determination of fluoride concentration by fluoride ion selective electrode. The determination of fluoride ion was carried out using a double junction electrode Ag/AgCl and a fluoride ion selective electrode (Metrohm, Switzerland).

The method of variable volume standard addition was used to determine fluoride content of water samples. In this method the sample contains fluoride was mixed with total ionic strength adjustment buffer and its potential was measured. Then successive amount of a fluoride standard solution was added and the potential was measured after each addition. The following equations describe how the concentration of fluoride in unknown can be calculated.

The measured potential (E) can be represented by:

$$E = K + S \log C$$

where, K is a constant, S is the slope of the calibration curve, C is the analyte ion (F^-) concentration.

This equation can be rearranged to give

$$C = \frac{10^{E/S}}{10^{K/S}}$$

$$C = C_oV_o + C_{std}V_{std} / V_o + V_{std} = 10^{E/S}/10^{K/S}$$

The equation can be rearranged and converted to the following:

$$10^{E/S} (V_o + V_{std}) = 10^{K/S} C_0V_o + 10^{K/S} C_{std}V_{std}$$

A plot of $10^{E/S} (V_o + V_{std})$ *versus* $C_{std}V_{std}$ will represent a linear plot, in that the amount of fluoride in the sample solution before addition of the standard is equal to the negative amount of intercept (Y = 0). It can be written as follows:

$$\frac{-b}{m} = \frac{-10^{K/S}C_0V_0}{10^{K/S}} = -C_0V_0$$

Finally the concentration of fluoride (µg/L) can be calculated from the following equation.

$$C_{unk} = C_oV_o/V_{unk}$$

RESULTS AND DISCUSSION

Recovery test: In order to investigate the accuracy and precision of the determination method a series of samples were spiked with fluoride standard solution and the concentration of fluoride was determined in each sample. The results obtained from this investigation for four-replicate analysis (n = 4) are shown in Table-1 and indicate that the method is quite reliable for the determination of trace fluoride.

TABLE-1
RECOVERY TEST FOR DETERMINATION OF FLUORIDE IN DRINKING WATER BY ISE

Sample	F^- added (mg/L)	F^- found (mg/L)	Recovery (%)
1	-	0.18	-
	0.50	0.67	98.6
2	-	0.25	-
	0.50	O.74	98.7
3	-	0.17	-
	0.50	0.66	98.5
4	-	0.31	-
	0.50	0.80	99.0

Concentration of fluoride in drinking water: Drinking water samples were collected from seven Iranian mega cities and their fluoride content was determined by ion selective electrode and standard addition method. Table-2 shows the results obtained from this investigation.

TABLE-2
CONCENTRATION OF FLUORIDE IN DRINKING WATER

Location	Concentration (µg/L)	SD
Tehran	0.12	0.03
Isfahan	0.31	0.04
Shiraz	0.15	0.03
Booshehr	0.24	0.05
Rasht	0.24	0.04
Mashhad	0.16	0.03
Arak	0.17	0.04

Concentration of fluoride in black tea: The fluoride content of each black tea sample was extracted according to the procedure and at two different infusion times, 10 and 20 min. It was cleared that the more infusion time the more fluoride extracted. Table-3 shows the results of this investigation.

TABLE-3
EFFECT OF INFUSION TIME ON THE EXTRACTION OF FLUORIDE FROM TEA (µg/L)

Tea sample	Popularity	Infusion time (10 min)	Infusion time (20 min)
1	Very popular	2.60 ± 0.262	3.12 ± 0.301
2	Very popular	2.63 ± 0.183	3.12 ± 0.385
3	Popular	3.73 ± 0,112	4.17 ± 0.234
4	Very popular	2.54 ± 0.126	2.77 ± 0.138

Conclusion

The fluoride content of drinking water is considerably low in Iranian mega cities. Tea is a popular drink all around the world and especially in Iran. Tea is a popular drink in Iran and a source of receiving fluoride. Although Iranian adults can intake some of the demanded the fluoride through drinking tea but the children are faced to an apparent fluoride deficiency and its adverse health effects. The fluoridation of drinking water should be focused on for prevention of children dental cares.

REFERENCES

1. M.M. Bayon, A. Garrcis, R.J.I. Garcis-Alonso and A. Sanz-Medel, *Analyst*, **124**, 27 (1999).
2. S.M. Talebi, A.H. Abdollahi and M.D. Jelas-Haron, *Pollut. Res.*, **29**, 375 (2010).
3. M. Yuwonu, *Dental J.*, **38**, 91 (2005).
4. N.J. Miller-Ihli, P.R. Pehresson, R.L. Cutrifelli and J.M. Holden, *J. Food Comp. Anal.*, **16**, 621 (2003).
5. K.E. Heller, S. Woosung, J.A. Burt and S.A. Eklund, *J. Public Health Dentistry*, **59**, 3 (1999).
6. J.E. Johnson, A.E. Kearns, P.M. Doran, T.K. Khoo and R.A. Wermers, *Mayo Clin. Proc.*, **82**, 719 (2007).
7. P. Konieczka, B. Zygmunt and J. Namiesnik, *Toxicol. Environ. Pollut.*, **104**, 197 (1999).
8. B. Culik, *Anal. Chim. Acta*, **189**, 239 (1986).
9. M.B. Rajkovic and I.D. Novokovik, *J. Agric. Sci.*, **52**, 155 (2007).
10. Y. Michigami, Y. Kuroda, K. Ueda and Y. Yamamoto, *Anal. Chim. Acta,* **274**, 299 (1993).
11. M. Frant and J.W. Ross, *Science*, **154**, 1553 (1966).