Kuwait J. Sci. Eng. **37** (2A) pp. 75-86, 2010

Determination of fluoride levels in soft drinks, fruit juices and milk consumed by the population in Kuwait using an ion-selective electrode

M.E. BHATTI, A. AL-RASHDAN, A. S. BOOTA, Z. AL-RASHID AND S. AL-RUWAIH.

Central Analytical Laboratory, Kuwait Institute for Scientific Research,
P.O. Box 24885, Safat, Kuwait.
Email: mejaz@kisr.edu.kw

ABSTRACT

Fluoride is an important anion present in various environmental, chemical and food samples. It is widely used in various branches of industry, and some fluoride compounds are formed as by-products in certain processes. Excessive amounts of fluoride in different compounds can enter the human body by means of polluted air, water and the food chain. Determination of fluoride in 86 food samples was performed by using an ion-selective electrode, as this method is simple, rapid and reliable. Fluoride concentrations were determined in the most-consumed fruit juices, soft drinks and milk samples among the population of Kuwait. A total of 15 soft drink samples available in the Kuwaiti markets were analyzed. The fluoride contents ranged from 0.05 - 0.15mg/L, with an average of 0.06 mg/L. From the producer KDD, 16 fruit juice samples were selected from the markets and analyzed for fluoride content. The fluoride level ranged from 0.05 - 0.20 mg/L, within an average of 0.08 mg/L. The 32 other main brands of fruit juices available in the supermarkets were analyzed for fluoride content and found to be in the range of 0.03 - 0.15 mg/L, within an average of 0.05 mg/L. The total 23 samples from different brands of milk were selected and analyzed, finding fluoride levels in the range of 0.02 - 1.20 mg/L with an average of 0.08 mg/L. Fluoride concentration in all soft drinks, fruit juices and milk samples were within the safe level.

Keywords: Fluoride, ion-selective electrode, dental, caries, cancer, drinks.

INTRODUCTION

Fluoride is an important anion present in various environmental, chemical and food samples. A small amount of fluoride is vital to humans 0.05 to 2.0mg/L), but it is toxic for human in larger amounts (Serife, 2004). Fluoride is widely used in various branches of industry, and some fluoride compounds are formed as by-products in certain processes (Serife, 2004). Excessive amounts of fluoride in different compounds can enter the human body by means of polluted air, water and the food chain. Fluoridated toothpaste and water are additional sources of fluoride. Fluoridated water is good for the teeth and bones, but if it is more than the required level, it can be lethal to health (0.2 to 0.35g F/kg body weight) (Serife, 2004). Fluoride can weaken bone and increase the risk of fractures for the people exposed to 4mg/L in their drinking water over their life time (Carton, 2006). A small amount of fluoride is beneficial in the prevention of dental caries, whereas fluorosis is caused by an elevated intake of fluoride over prolonged periods of time. Skeletal fluorosis and dental fluorosis are the two main problems resulting from high intake of fluoride over the needed level. In dental fluorosis, the structural integrity of enamel is affected, and small pits are left as the enamel breaks away. Skeletal fluorosis is the accumulation of fluoride in the skeletal tissues, and has been associated with pathological bone formation.

Systemically ingested fluoride disturbs thyroid hormone metabolism, leading to a delay in eruption of the teeth and their subsequent exposure to the delay-producing environment (Spittle, 2006). Recent studies showed a link of fluoride with effects on the brain. It is found that too much fluoride is associated with low performance on intelligence tests (Wang *et al.,* 2007).

In addition to bones and teeth, the kidney is a prime target organ for fluoride toxicity. Because fluoride is filtered from the blood by the kidneys and excreted in the urine, the quality of kidney function is very important and may be closely related to differences in susceptibility to fluoride toxicity. Greater accumulations of fluoride occur in people who have impaired renal function and thus can be more seriously affected. It is also seems likely that fluoride accumulation induces increased kidney damage (Hosokawa *et al*., 2010).

In recent years, because of increasing consumption of soft drinks and fruit juices, fluoride content in these products has become an issue of growing concern. High levels of fluoride in fruit juices are usually due to the fluoride in water used in their preparation. (Stannard & Shim, 1991). Recent reviews of studies have reported that the prevalence of dental fluorosis has increased significantly in both fluoridated and non-fluoridated areas in the world (Heilman *et al*., 1999). This increase can be explained by the fact that in addition to water, significant levels of fluoride are now found in different products, including beverage and juices, as well as in non-dietary sources such as fluoride supplements. Excessive total fluoride intake from all sources during tooth development is the main cause of fluorosis (Mulder *et al*., 2002). Children are consuming more carbonated soft drinks, milk and juice drinks, and less water. A considerable number of infants are weaned early and artificially fed before they are a month old, reflecting a decline in breast-feeding. Thus, the concentration of fluoride in cow's milk and infant formula plays an important role in the supply of fluoride to infants. Although cow's milk is reported to have low fluoride concentrations, some studies related wide variations in fluoride concentrations in dairy milk types (Buzalaf *et al.*, 2006).

Considerable research has been conducted on the level of fluoride in fruit juices, soft drinks and milk in the world. Scientists are monitoring and studying the level of fluoride in water, soft drinks, fruits and milk to understanding its effects on health (Heilman *et al.*, 1999), however such a study has not previously been conducted in Kuwait.

Thus, the aim of the present study was to evaluate the fluoride concentrations in whole, low fat and skimmed milk with different flavors, along with soft drinks and fruit juices commercially available in Kuwait.

MATERIALS AND METHODS

Sampling

The most commonly consumed brands of soft drinks, fruit juices and milk samples were purchased from supermarkets (City Center and the Sultan Center) in the state of Kuwait. Eighty-six samples of juices, soft drinks and milk were purchased and divided into groups as follows:

15 samples of soft drinks from all brands.

16 samples of fruit juices from Kuwait Danish Dairy Co. (KDD).

32 samples of fruit juices from other major manufacturers.

23 samples of fresh full cream milk, low fat milk, and skimmed milk. (long-life full cream milk, low fat milk, skimmed milk and flavored milk).

The fluoride analyses were performed using an Orion model (940) Expandable Ion Analyzer meter with a combination fluoride electrode (Orion ISE 940900). Throughout the experiment, analytical reagent-grade chemicals were used, as well as distilled water. A set of fluoride standards ranging from 0.02 to 2.00 mg/L fluoride were prepared by using a serial dilution from a 1000 mg/L sodium fluoride (NaF) stock solution from (Orion # 940907). TISAB III (940911), which is commercially available from Orion, was used. TISAB III regulates the ionic strength of the samples to adjust the pH to between 5 and 7;

TISAB III also avoids interference by polyvalent cat ions, which are able to make complex and free fluoride concentrations in the solution, and to precipitate with fluoride. In this experiment single known standard addition method was used as follows:

A 50ml sample and 2.5ml of TISAB III were added in the beaker. Stirring was allowed until the reading was stabilized and displayed for standard addition. One ml of 100 mg/L standard solution of fluoride was added. Again the contents of the beaker were stirred, and until a stable reading was displayed. A final reading was displayed on the screen and recorded, giving the researchers the fluoride concentration present in the sample.

QUALITY CONTROL AND RECOVERY ASSURANCE

All samples for the study were analyzed in triplicate. For confirmation of the presence of fluoride contents in each sample, one ml of 100 mg/L fluoride standard was spiked, and the percentage recovery of fluoride in each sample was calculated. Reproducibility and accuracy of the results were ensured by triplicate determination and spiking the known standard in the sample. The boxes of juices and soft drinks were stored in the refrigerators and opened on the day of analysis. Each 50 ml sample was used for fluoride analysis. Fluoride determination was carried out by using an ion-selective electrode. A set of fluoride standards ranging from 0.02 to 2.00 mg/L fluoride were prepared by using a serial dilution (0.02, 0.05, 0.10, 0.5, 1.0, 1.5 and 2.0) from a 1000 mg/L sodium fluoride (NaF) stock solution (Orion # 940907). The calibration curve and performance of the electrode were checked.

The recovery study gave information about possible interference with fluoride from other analytes in the sample, which were aluminum and iron. The amount of fluoride added should correspond to the level normally present in the sample material. With a ion-selective electrode method, the reproducibility and recovery were reliable and consistent throughout the study, indicating its suitability as a reliable method for fluoride determination in juices, soft drinks and milk samples.

RESULTS AND DISCUSSION

In different parts of the world, scientists are monitoring and studying the level of fluoride in water, soft drinks, fruit juices, vegetables and dairy products to understanding its effects on health (Heilman *et al*., 1999).

Observation and evaluation of populations taking fluoride and facing the risk of mild chronic fluoride intoxication are insufficient. The target organs of chronic fluoride intoxication are not only the teeth and the skeleton, but also the liver, kidney, and the nervous and reproductive systems (Guan, 1986).

A total of 15 soft drink samples available in the Kuwaiti markets were collected and analyzed. The fluoride contents ranged from 0.05 to 0.16mg/L (SD ± 0.001 to 0.01), with an average of 0.06 mg/L. Reproducibility and accuracy were determined by spiking the samples with 1 mg/L of fluoride standard. The recovery percentage in spiked samples ranged from 98 to 108% with an average of 106 % (RSD ± 1.51 to 9.19). The results are presented in Table 1, while average concentrations and the recovery percentage are shown in Fig. 1 and Fig. 2, respectively.

Table 1. Fluoride concentration in soft drinks

Sample	**Conc.± S.D. (mg/L) (n = 3)**	**Recovery ± RSD (%)**
Pepsi	0.05 ± 0.003	106 ± 6.002
7-up	0.06 ± 0.005	108 ±7.979
Sprite	0.06 ± 0.001	100 ± 2.925
Mountain Dew	0.05 ± 0.002	107 ± 4.852
Coca Cola	0.06 ± 0.001	106 ± 2.053
Lipton Iced Tea	0.10 ± 0.01	104 ± 4.969
Mirinda Orange	0.06 ± 0.005	102 ± 2.132
Fanta Blackcurrent	0.06 ± 0.005	102 ± 8.379
Fanta Strawberry	0.05 ± 0.001	105 ± 2.965
Fanta Orange	0.06 ± 0.004	102 ± 1.797
Coca Cola Light	0.06 ± 0.004	100 ± 6.292
Shani	0.04 ± 0.004	102 ± 9.196
Soda water	0.06 ± 0.001	98 ± 1.505
Lemon	0.07 ± 0.002	106 ± 3.251
Sunkist Orange	0.07 ± 0.007	106 ± 8.979

From Kuwait Danish Dairy (KDD), 16 fruit juice samples were selected from the markets and analyzed for fluoride content. The fluoride level ranged from

0.05 to 0.20 mg/L (SD ± from 0.001 to 0.009), within an average of 0.08 mg/L. Reproducibility and accuracy were determined by spiking the samples with 1mg/ L of fluoride standard. The recovery percentage in spiked samples ranged from 96 - 110 % an average of 100% (RSD ± 0.80 to 12.69). The results are presented in Table 2, and average concentrations and the recovery percentage are shown in Fig. 1 and Fig. 2, respectively.

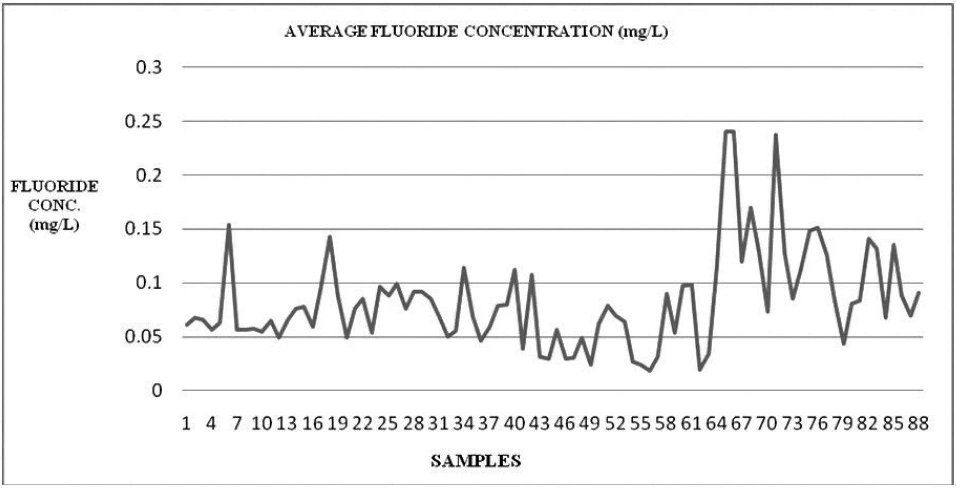

Figure .1 Average fluoride concentration

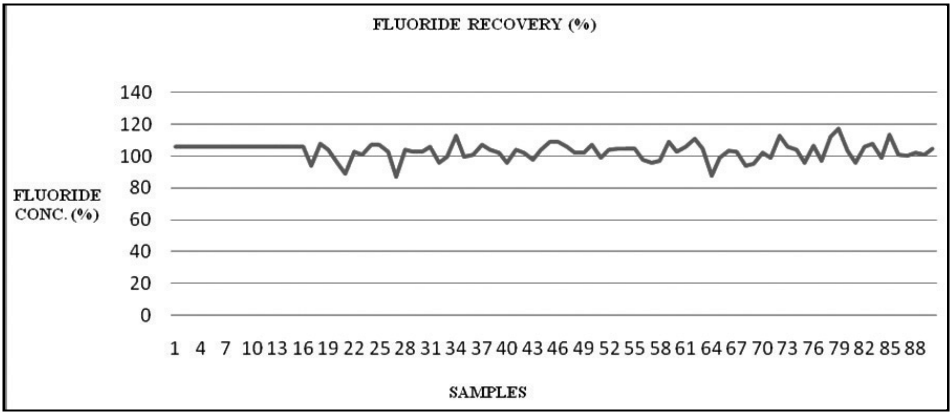

Figure. 2 Percent fluoride recovery

Table 2. Fluoride concentration in Kuwait Danish Dairy (KDD) fruit juices

Sample	Conc.± S.D. (mg/L) (n = 3)	Recovery ± RSD (%)
Mango	0.05 ± 0.009	94 ± 3.691
Apple	0.09 ± 0.001	108 ±1.661
Grape (Red)	0.02 ± 0.002	104 ±1.732
Grape (White)	0.08 ± 0.000	96 ±0.939
Guava	0.04 ± 0.001	89 ±2.734
Peach	0.07 ± 0.005	103 ±7.048
Cherry	0.08 ± 0.002	101 ±2.882
Tomato	0.05 ± 0.004	107 ±9.046
Orange	0.09 ± 0.007	107 ±7.756
Grapefruit	0.06 ± 0.000	103 ±0.807
Pineapple	0.09 ± 0.006	85 ±6.965
Lemonade	0.07 ± 0.007	104 ±2.827
Carrot	0.09 ± 0.006	103 ±7.073
Cocktail	0.09 ± 0.002	103 ±2.455
Passion fruit	0.08 ± 0.002	106 ±3.35
Apricot	0.06 ± 0.008	96 ±12.69

The other 32 popular brands of fruit juices available in the supermarket were also analyzed for fluoride content and found to vary in the range 0.03 - 0.15 mg/L, within an average of 0.05 mg/L (S.D. ± from 0.001 to 0.009). Reproducibility and accuracy were determined by spiking the samples with 1mg/L of fluoride standard. The recovery percentage in spiked samples ranged from 88 - 113% with an average of 99.8% (RSD ± 0.51 to 10.30). The results are presented in Table 3, and average concentration and recovery percentages are shown in Fig. 1 and Fig. 2, respectively.

Table 3. Fluoride concentration in different brands of fruit juices

Sample	Conc. ± S.D. (mg/L) (n = 3)	Recovery ± RSD (%)
Mango ABC	0.05 ± 0.004	113 ± 9.067
Orange ABC	0.05 ± 0.004	100 ± 8.362
Sultan Apple	0.1 ± 0.002	101 ± 2.664
Cocktail Dana	0.07 ± 0.001	107 ± 2.654
Peach ABC	0.04 ± 0.004	104 ± 2.029
Cocktail Sunkist	0.05 ± 0.002	102 ± 3.559
Orange Sunkist	0.07 ± 0.001	96 ± 2.341
Pineapple Sunkist	0.07 ± 0.001	104 ± 2.113
Orange Al-Rabie	0.11 ± 0.006	102 ± 0.514
Orange Sultan	0.03 ± 0.001	98 ± 3.072
Apple Sultan	0.10 ± 0.009	104 ± 8.454
Cocktail Sultan	0.03 ± 0.002	109 ± 8.853
Orange Awal	0.03 ± 0.001	109 ± 3.442
Pineapple Awal	0.05 ± 0.003	106 ± 6.581
Mango Awal (children)	0.02 ± 0.002	102 ± 8.268
Orange Awal (children)	0.02 ± 0.002	102 ± 8.899
Mango Caesar (children)	0.04 ± 0.001	107 ± 2.393
Orange + Carrot Caesar	0.02 ± 0.002	99 ± 10.3
Orange Caesar	0.06 ± 0.001	104 ± 2.397
Orange Shereen	0.07 ± 0.002	105 ± 3.238
Peach Shereen	0.06 ± 0.001	105 ± 2.197
Guava Shereen	0.06 ± 0.001	105 ± 2.495
Orange Al-Marai	0.02 ± 0.005	98 ± 5.466
Mango Al-Marai	0.02 ± 0.005	96 ± 6.398
Guava Al-Marai	0.02 ± 0.001	97 ± 6.038
Mixed Berry Al-Marai	0.03 ± 0.001	109 ± 4.768
Berry Mix Al-Rabie	0.09 ± 0.005	103 ± 5.618
Chocolate Coconut Rabie	0.05 ± 0.003	106 ± 6.315
Pinapple + Coconut Rabie	0.09 ± 0.008	111 ± 8.656
Kiwi + Lime Rabie	0.09 ± 0.004	105 ± 4.31
Tomato Juice Caesar	0.02 ± 0.001	88 ± 9.044
Strawberry Nada	0.03 ± 0.002	99 ± 5.863

Table 4 represents all brands of cow's milk analyzed with the ion selective electrode. The cow milk selection includes full cream, low fat, skimmed and flavored milks. The manufacturers of different brands of milk are mentioned in the table, and the fluoride concentrations were determined for all brands of milk (expressed in mg/L). All full cream, low cream and skimmed as well as flavored milk had fluoride concentrations ranging from 0.04 to 0.24 mg/L (SD ± from 0.001 to 0.02). Reproducibility and accuracy were again determined by spiking the samples with 1 mg/L of fluoride standard. The recovery percentage in spiked samples ranged from 94 - 108% with an average of 99.8% (RSD ± 1.12 to 13.32). These fluoride levels were expected, because fluoride is poorly transported from plasma to milk, and the concentration of fluoride in milk remains low even when the intake of fluoride by the mammals is high (Buzalaf *et al.*, 2006). Among the different types of milk analyzed in this study, these products we selected may be the most consumed. Taking into account that enamel fluorosis may occur, following either an acute or chronic exposure to fluoride during tooth formation, dairy companies should consider providing information to the public about fluoride concentrations in their products.

Table 4. Fluoride concentration in different brands of milk samples

Sample	**Conc. ± S.D. (mg/L) (n = 3)**	**Recovery ± RSD (%)**
Al-Marai full cream	0.11 ± 0.003	104 ± 3.072
KDD full cream	0.24 ± 0.005	103 ± 2.406
KDD half cream	0.24 ± 0.007	94 ± 3.005
KdCow half cream	0.12 ± 0.01	95 ± 8.333
KdCow skimmed	0.17 ± 0.01	102 ± 5.882
ABC full cream	0.13 ± 0.01	99 ± 7.692
AWAL full cream	0.07 ± 0.005	113 ± 7.873
AWAL half cream	0.23 ± 0.024	106 ± 10.45
Saudia low fat	0.12 ± 0.005	104 ± 4.558
Saudia Junior-full cream	0.08 ± 0.006	96 ± 7.036
Saudia-banana	0.11 ± 0.002	107 ± 2.24
Saudia-strawberry	0.14 ± 0.010	97 ± 7.081
Saudia-chocolate	0.15 ± 0.011	113 ± 7.585
Glaxay-chocolate	0.12 ± 0.002	117 ± 1.587
KDD-vanilla	0.08 ± 0.005	104 ± 6.928
Al-Marai-banana	0.04 ± 0.005	96 ± 13.32

Cont. Table 4. Fluoride concentration in different brands of milk samples

Sample	Conc. ± S.D. (mg/L) (n = 3)	Recovery ± RSD (%)
Al-Marai-strawberry	0.08 ± 0.010	106 ±12.48
Al-Marai full cream	0.08 ± 0.005	108 ± 6.928
Al-Marai low fat	0.14 ± 0.010	99 ± 7.156
KdCow low fat	0.13 ± 0.010	114 ± 7.645
Safi -laban	0.06 ± 0.006	101 ± 9.432
Pepsi Cola	0.13 ± 0.001	100 ± 1.126
Sultan full cream	0.08 ± 0.006	102 ± 7.452
Sultan low cream	0.07 ± 0.007	101 ± 10.87
ABC chocolate	0.09 ± 0.004	105 ± 5.401

CONCLUSION

The fluoride concentrations in our study during the year 2007 were found to be in the range of 0.02 to 0.24mg/L., which are considered acceptable by World Health Organization (WHO) (0.02 to 2.0 mg/L). This indicates that fluoride content in soft drinks, fruit juices and milk available in the Kuwaiti markets are well in the acceptable range. However, it is suggested to form an advisory forum to regulate intake of this important chemical at different stages of human life.

REFERENCES

Buzalaf, M. A., Pessan, J. P., Fukushima, R., Dias A. & Rosa, H. M. 2006. Fluoride content of UHT milks commercially available in Bauru, Brazil. Journal of Applied Oral Science. **14** (1): 38-42.

Carton, R. J. 2006. Fluoride in drinking water. Fluoride **39** (3): 163-172.

Guan, Z. Z. 1986. Morphology of the brain of the offspring of rats with chronic fluorosis. (CH). Zhonghua Bing Li Xue Za Zhi. Chinese Journal of Pathology **15**: 297-299.

Heilman, J.R., Kirtisy, M.C., Levy, S.M. & Wafel, J.S. 1999. Assessing fluoride levels of soft drinks. Journal of the American Dental Association **130** (11): 1593-1599.

Hosokawa, M. & Asakawa, H. 2010. Deterioration of renal function in ICGN mice by fluoride in drinking water. Fluoride **43**(1): 31-44.

Mulder, J., Chatterjee, R. & Wolff, M.S. 2002. Fluoride levels in commercially available beverages. State University of New York at Stony Brook, Stony Brook, USA. San Diego International Association for Dental Research. The IADR/AADR/CADR 80th General Session. Poster Session (6-9 March).

Spittle, M.B. 2006. Fluoridation promotion by scientists in 2006. An example of "Tardive Photopsia". Fluoride **39** (3): 157-162.

Serife, T. 2004. Determination of fluoride in various samples and infusions using a fluoride selective electrode. Turkish Journal of Chemistry **28**: 203-211.

Stannard, J. G. & Shim, Y.S. 1991. Fluoride levels and fluoride contamination of fruit juices. Journal of Clinical Pedriatric Dentistry **16**: 38-48.

Wang, S.X. & Wang, Z.H. 2007. Arsenic and fluoride exposure in drinking Water: Children's IQ and growth in Shanyin County, Shanxi province. China Environmental Health Perspectives **115**(4): 643-647.

Submitted : *11/3/2009*
Revised : *29/9/2010*
Accepted : *6/10/2010*

تحليل معدلات مادة الفلورايد في المشروبات الرطبة، وعصير الفواكه وعينات الحليب المستهلكة من سكان الكويت، باستخدام إلكترود الأيونات الخاص

محمد إعجاز بهاتي ، أمل الرشدان ، أنور بوتا ، زكية رشيد و شعاع الرويح

المختبر التحليلي المركزي – معهد الكويت للأبحاث العلمية
ص.ب. 24885 – الصفاة – 13109 – الكويت

خلاصة

تعتبر مادة الفلورايد من الأيونات المهمة الموجودة في أنواع مختلفة من البيئة، والكيماويات وعينات الأطعمة. وتستخدم مادة الفلورايد في مجالات مختلفة من الصناعة، وبعض مواد الفلورايد تتكون نتيجة بعض الطرق والعمليات كناتج إضافي. وكمية كبيرة من الفلورايد قد تدخل جسم الإنسان بمركبات مختلفة عن طريق تلوث الهواء، الماء، وسلسلة الأطعمة. وتحديد كمية الفلورايد في 86 عينة من الأطعمة قد تم فحصها عن طريق استخدام إلكترود الأيونات الخاص. الطريقة التي استخدمت بسيطة، سريعة ويمكن الاعتماد عليها. وتراكيز الفلورايد حددت في كل من عصير الفواكه، المشروبات الرطبة، وعينات الحليب، وهذه المشروبات تستهلك عن طريق معظم سكان الكويت.

وقد تم تحليل 15 عينة من المشروبات الرطبة المتوافرة في السوق الكويتي. وكان تركيز مادة الفلورايد يتراوح ما بين 0.05–0.15 مليجرام/لتر ومعدل 0.06 مليجرام /لتر. ومن شركة المرطبات الكويتية الدنمركية (KDD)، 16 عينة من عصير الفواكه تم اختيارها من السوق وتحليلها لمعرفة مدى تواجد مادة الفلورايد. وكانت مادة الفلورايد تتراوح ما بين 0.05–0.2 مليجرام /لتر ومعدل 0.08 مليجرام/لتر. وكان معدل ما مجموعة 32 عينة من مختلف منتجات عصير الفواكه المتوافرة في السوق المحلي تتراوح ما بين 0.03–0.15 مليجرام /لتر ومعدل 0.05 مليجرام /لتر. وقد تم اختيار 23 عينة من مختلف المنتجات التجارية للحليب وتم تحليلها، وكان معدل مادة الفلورايد يتراوح ما بين 0.02–1.2 مليجرام/ لتر ومعدل 0.08 مليجرام/لتر. وكانت النتيجة من هذه التحاليل أن مادة الفلورايد المتواجدة في عصير الفواكه، والمشروبات الرطبة وعينات الحليب تتراوح في الحدود الآمنة المصرح فيها.