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Short communication

Fluoride content of municipal water in the United States: what percentage is fluoridated?

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Abstract

Recently, a study was conducted to look at the trace element content of municipal waters sampled around the United States. This was a collaborative project defined by representatives of the Nutrient Data Laboratory and the Food Composition Laboratory of the United States Department of Agriculture. As part of the study, the fluoride content of nationally representative water samples was measured since water is one of the principal sources of fluoride in the US diet. This study served as a pilot for planning a more extensive future nationwide sampling. Samples were analyzed using a separate reference electrode and ion selective electrode with TISAB II buffer. The calibration range was from 0.5 to 3.0 ppm. Accuracy of the method was validated by analyzing NIST SRM 2671a Fluoride in Freeze-Dried Urine. Results from the municipal water study clearly showed that the distribution of fluoride in US municipal waters is bi-modal. That is to say, because of the fact that ambient levels are typically extremely low, the concentration of fluoride is basically quantized. Either the water is fluoridated and contains approximately 1 ppm of fluoride or it is not fluoridated with undetectable fluoride concentration. The distribution of these data make it difficult to assign a meaningful average value useful to health professionals and consumers to assess fluoride intake. This study revealed that approximately 40% of the water samples from this nationwide sampling were fluoridated with a mean concentration of $1.01 \pm 0.15\,\mu g/ml$.

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1. Introduction

The nutritional impact of fluoride is multifaceted. Ingestion of fluoride prior to the eruption of teeth has a cariostatic effect (Chow, 1990). The risk of dental caries is reduced due to the uptake of fluoride by enamel cystallites and formation of fluorhydroxyapatite which resists acid solubilization. The post-eruptive protective effect is attributed to the reduced acid production by plaque bacteria and an increased rate of enamel remineralization (Hamilton, 1990; Marquis, 1995). As such, the dental community generally favors the ingestion of fluoride to promote good dental health (Beltrán-Aguilar, Griffon, & Lockwood, 2002).

Orally ingested fluoride is readily absorbed from the gastrointestinal tract. Only high concentrations of calcium and other cations can prove problematic because they may bind with fluoride to form insoluble, poorly absorbed compounds. Nearly all of the fluoride in the body is found in calcified tissues and any elimination is done through the kidneys. Excessive fluoride intake can lead to fluorosis of both teeth and bones. Children between the ages of 2–3 are at most risk of suffering from cosmetic fluorosis (Evans & Darvell, 1995) but some evidence suggests that enamel fluorosis in primary teeth may be the result of infants ingesting formula reconstituted with fluoridated water (Larsen, Senderovitz, Kirkegaard, Poulsen, & Fejerskov, 1988). Skeletal fluorosis is the result of ingesting elevated levels of fluoride for extended periods of time. Prior to the onset of obvious symptoms, individuals will have slight increases in bone mass and bone concentrations which are 2–5 times higher than normal (Eble, Deaton, Wilson, & Bawden, 1992). The first obvious symptoms include pain and stiffness in joints and osteosclerosis and this may develop into stages 2 and 3, which may be crippling due to calcification of ligaments, osteoporosis of long bones, and neurological defects due to hypercalcification (Food and Nutrition Board, 1997). Crippling skeletal fluorosis is rare in the United States even in areas where the water supplies contained up to 20 ppm of fluoride.

Because of the conflicting health impacts associated with fluoride ingestion, the issue of fluoridating water is still debated (Takahashi, Akiniwa, & Narita 2001). Some people believe that fluoridation of water is only beneficial to children and argue against fluoridation of public water supplies. Others lobby for fluoridation of public water supplies arguing that there is little harm to fluoridation since literature reports show that fluoride intakes of less than 10 μg/day showed no adverse effects (National Research Council, 1993). Several authors agree with Karqagas, Baron, Barrett, and Jacobsen (1996) who noted that exposure to fluoride over time is more important than determining the instantaneous level being ingested when conducting epidemiology studies and trying to understand fluoride related health effects. The Water Fluoridation Reporting System developed by the Centers for Disease Control combines data from CDC and EPA and is the most comprehensive national fluoride database (CDC, 2002). Data included there are provided by several sources and do not necessarily facilitate a direct comparison of fluoride levels in a variety of water samples using the same analytical methodology and extensive quality control procedures. The CDC database is available on-line (http://www.cdc.gov/nohss/FSMain.htm) along with significant general fluoridation information.

The adequate intake (AI) for adults for fluoride is 0.05 μg/kg/day while the tolerable upper intake level (UL) is 0.1 μg/kg/day (Food and Nutrition Board, 1997). Although drinking water is a primary source of fluoride in the diet, other sources related to dental care can play a major role, particularly for children. Levy, Kohout, Kiritsy, Heilman, and Wefel (1995) reported that in Iowa

City, approximately 25% of infants less than 9 months old were given fluoride supplements. As such, Levy and Muchow (1992) have found that supplements are often times prescribed at incorrect dosages, particularly in areas where water is fluoridated. Likewise, significant amounts of fluoride are ingested by swallowing toothpaste, fluoride rinses and mouthwash products (Food and Nutrition Board, 1997). This has led to recommendations from several groups to limit fluoride intake from non-dietary sources.

The US Public Health Service made recommendations for public water fluoridation in the United States more than 3 decades ago (US Public Health Service, 1962). Recommended fluoridation levels range from 0.7 to 1.2 ppm depending on the average daily temperature based on the fact that increased consumption is expected in warmer climates. Recent studies of water consumption (Heller, Woosung, Burt, & Eklund, 1999) suggest there may be no need for recommending different fluoridation levels based on mean maximum temperature and suggests further investigation.

This paper provides data reflecting the fluoride content for municipal waters from 24 different locations in a total of 15–16 cities from 10 states nationwide during three different seasons. Regional and seasonal variability is discussed and the appropriateness of establishing a "representative national average" fluoride content for water is discussed.

2. Methodology

2.1. Sample collection

Pehrsson, Haytowitz, Holden, Perry, and Beckler (2000) in the Nutrient Data Laboratory of USDA designed the sampling plan. This plan was originally developed for the National Food and Nutrient Analysis Program (NFNAP), which was an interagency agreement between the National Institutes of Health and the US Department of Agriculture. The sampling plan is a national probability-based plan developed to sample and analyze Key Foods for which existing data were either absent or of poor quality or where concentration levels were expected to be different than historical data based on changes in the formulation of the foods or changes in the analytical methodology. As a result, data collected under the NFNAP design for a specific food item are geographically and statistically representative of the actual food consumed by the US population. It was determined that self-weighting NFNAP data allow for accurate and representative mean estimates of nutrient profiles in generically described or brand-specific products and would, therefore, be suitable to replace historical data in NDL databases. A problem arises, however, with fluoride data because of the bi-modal distribution of fluoride in the US water supply, making mean concentrations of limited usefulness.

Water samples were collected from 24 locations nationwide during three phases (February, July and November 1999). Based on existing data related to the NFNAP sampling plan, the pick up locations were expected to provide nationally representative samples. The cities selected for sample pick-up correspond to the 24 Consolidated Metropolitan Statistical Areas (CMSAs) determined in the NFNAP sampling plan. Two water samples collected from each location were used for this study.

Acid cleaned, 125 ml low-density polyethylene bottles were used for water sample collection for both trace metal (Miller-Ihli, 2001) and fluoride analyses. The bottles were tested using acidified water and no detectable fluoride blanks were measured.

Instructions sent out to collection agents instructed that the tap water be run for 5 min at medium velocity prior to sample collection. In addition, a questionnaire was included asking participants to provide any specific information about the water system they were accessing including: type of pipes, municipal or well water, knowledge about fluoridation, etc. All samples returned for analysis were from municipal water sources.

2.2. Sample preparation and analysis

Fluoride determinations were obtained with a Cole Parmer (Vernon Hills, IL, USA) ion selective electrode with a separate reference electrode. Readings were made on a Denver Instruments Model 250 pH/ISE/Conductivity Meter (Arvada, CO). Samples and standards were prepared for analysis by diluting 15 ml of water sample with 15 ml of Total Ionic Strength Adjustment Buffer, TISAB II (Orion, Beverly, MA). Samples and standards were placed in 100 ml beakers and were continuously mixed using a magnetic stir bar during analysis. The calibration range was from 0.0 to 3.0 μg/ml. Triplicate readings were made in all instances and the final results were averaged.

2.3. Quality assurance/quality control performance

Accuracy of the method was validated by analyzing NIST SRM 2671a Fluoride in Freeze-Dried Urine (low concentration: certified value 0.55 ± 0.03 μg/ml). The concentration determined over the course of this study was 0.553 ± 0.005 μg/ml for $n = 9$ determinations (3 readings were averaged for each determination). In addition, an in-house control material, prepared to have a fluoride concentration of 0.85 μg/ml, was run throughout the study and the accuracy and precision were excellent (0.854 ± 0.006 μg/ml).

3. Results and discussion

Data were reviewed for all 24 locations for each of the 3 phases and these are summarized in Table 1. Our lower level for quantification (10 sigma) was determined to be 0.27 μg/ml (based on a 3 sigma detection limit of 0.081 μg/ml). Sigma is defined as the standard deviation of 10 blank measurements. When measurable values could not be determined, ‘‘nd’’ may be found in the table indicating that the fluoride content was not detectable ($<$ detection limit).

Although both hot and cold water samples were collected in phase 1, because there was no significant difference in the trace metal content between the samples, only cold water samples were collected for phases 2 and 3. As might be expected, phase 1 data show no evidence of significant fluoride concentration variability as a function of temperature. It is interesting to note that the fluoride level was undetectable at 12 of the 24 pickup locations in phase 1 (representing 8 different cities). If the mean concentration of only the quantitative values is determined, the mean fluoride concentration for phase 1 is 0.78 ± 0.28 μg/ml ($n = 12$). If we want to consider only the

Table 1
Fluoride content of US municipal water samples

	Location	Feb 1999 Phase 1	Mean (1)	Jul 1999 Phase 2	Mean (2)	Nov 1999 Phase 3	Mean (3)	3-phase mean	S.D.
1	DeQueen, AR	nd, nd	nd	nd, nd	nd	nd, nd	nd	nd	nd
2	DeQueen, AR	nd, nd	nd	nd, nd	nd	nd, nd	nd	nd	nd
3	Costa Mesa, CA	0.72, 0.73	0.72	0.72, 0.73	0.72	0.36, 0.35	0.35	0.60	0.21
4	Anaheim, CA	0.40, 0.44	0.42	0.51, 0.52	0.51	0.39, 0.40	0.40	0.44	0.06
5	Niles, IL	0.92, 0.89	0.91	1.3, 1.3	1.3	1.0, 1.0	1.0	1.07	0.20
6	Niles, IL	0.93, 0.93	0.93	1.3, 1.3	1.3	1.0, 1.0	1.0	1.08	0.20
7	Springfield, MO	nd, nd	nd	1.6, 1.3	1.5	nd, nd	nd	0.50	—
8	Springfield, MO	nd, nd	nd	1.4, 1.3	1.4	nd, nd	nd	0.47	—
9	Mountainside, NJ	nd, nd	nd	nd, nd	nd	nd, nd	nd	nd	nd
10	City not identified	0.99, 1.10	1.05	1.3, 1.3	1.3	1.0, 1.1	1.0	1.12	0.16
11	Portland, OR	nd, nd	nd	nd, nd	nd	nd, nd	nd	nd	nd
12	Vancouver, WA	0.82, 0.85	0.84	0.95, 0.89	0.92	0.86, 0.88	0.87	0.88	0.04
13	Longview, WA	0.99, 1.0	1.0	1.1, 1.1	1.1	0.77, 0.81	0.79	0.96	0.16
14	Longview, WA	0.91, 1.0	0.95	1.1, 1.1	1.1	0.77, 0.78	0.78	0.94	0.16
15	Tarentum, PA	nd, nd	nd	nd, nd	nd	nd, nd	nd	nd	nd
16	Gibsonia, PA	nd, nd	nd	nd, nd	nd	nd, nd	nd	nd	nd
17	Oil City, PA	nd, nd	nd	nd, nd	nd	nd, nd	nd	nd	nd
18	Oil City, PA	nd, nd	nd	nd, nd	nd	nd, nd	nd	nd	nd
19	Nashville, TN	1.0, 0.89	0.94	1.1, 1.2	1.2	1.0, 1.0	1.0	1.05	0.14
20	Nashville, TN	0.98, 0.92	0.95	1.2, 1.2	1.2	1.0, 1.0	1.0	1.05	0.13
21	Houston, TX	0.33, 0.31	0.32	0.44, 0.46	0.45	0.45, 0.42	0.44	0.40	0.07
22	Houston, TX	0.26, 0.31	0.29	0.37, 0.42	0.40	0.44, 0.47	0.45	0.38	0.08
23	Gilchrist, TX	nd, nd	nd	nd, nd	nd	0.27, 0.26	nd	nd	nd
24	Gilchrist, TX	nd, nd	nd	nd, nd	nd	0.27, 0.23	nd	nd	nd
	# detectable values		**12**		**14**		**19**		
	Mean $\pm$ S.D.		0.78 ± 0.28		1.03 ± 0.37		0.55 ± 0.35	0.79	0.24
	# fluoridated (>0.70)		**9**		**11**		**8**		
	Mean $\pm$ S.D.		0.92 ± 0.09		1.18 ± 0.22		0.93 ± 0.10	1.01	0.15
	% fluoridated		37%		46%		33%		
	Total with zeros/nd		**24**		**24**		**24**		
	Mean $\pm$ S.D.		0.39 ± 0.44		0.60 ± 0.59		0.44 ± 0.39	0.48	0.11
	Variance		0.19		0.35		0.15		

fluoridated water samples and we use a threshold of 0.75 μg/ml, then the mean is $0.92 \pm 0.0.09$ μg/ml ($n = 8$). If the value of zero is substituted for non-quantitative values (nd), then the mean is 0.39 ± 0.44 μg/ml ($n = 24$). These data combined with the data for phases 2 and 3 highlight the difficulty in trying to compute a representative mean value for the fluoride content of water in the United States.

The fact that fluoridation of water is not regulated nationwide leads to a challenge for individuals interested in generating meaningful data for nationwide databases. Add to that the fact that the naturally occurring levels of fluorine in water is quite variable, albeit typically much lower than fluoridated levels, and it is hard to provide a meaningful estimate of the amount of fluoride ingested without having specific regional, municipal water data. Most often municipal water is "treated" and the general public might assume that this includes fluoridation. A quick survey taken in our lab revealed that fewer than 10% of our staff knew if they received fluoridated water in their home water supply and it is likely that the general public might be even less aware.

What percentage of the waters sampled were fluoridated? Using the CDC 0.7–1.2 μg/ml range, approximately 40% of the water samples collected over the course of the 3-phase study were fluoridated. The notable exceptions were the samples collected in Anaheim, CA and Houston, TX which had measurable levels which were one third to one half of the typical fluoridation level. Also, all of the locations which had levels which were not detectable can safely be designated as non-fluoridated water supplies. One location which was unusual was Costa Mesa, CA which had average levels of 0.72 μg/ml for phases 1 and 2 and in the third phase the average level was only 0.35 μg/ml. Overall, phase 3 provided significantly lower levels, and that is interesting, but no obvious explanation was identified. It is interesting to note that the seasonal variability (evaluated by looking at the 3-phase mean ± standard deviation) was less than 20% RSD for all samples which were identified as being fluoridated (based on a 0.70 μg/ml threshold). The largest variability was seen for the Costa Mesa, CA water sample discussed above. The smallest variability was seen for Vancouver, WA where fluoride levels over the three phases averaged 0.88 ± 0.04 μg/ml (RSD of 4.6%). The expected achievable seasonal reproducibility at a given location is dependent on many things including: the method used for fluoridation, mixing volume in the treatment plant, etc. More typical reproducibilities over the 10 month period represented by the 3 phases were 10–15% on average.

Although the data are very interesting, they demonstrate the challenge of trying to generate meaningful values for food composition databases. If only the quantitative values are considered, then the apparent mean concentration is elevated. If zeroes are substituted for values not detected (nd) then the apparent mean concentration is biased low and the standard deviation is artificially inflated. One thing that may be done is to calculate the mean value for all of the fluoridated samples (defined as those with fluoride levels > 0.70 μg/ml). If this is done, we see that the mean concentrations for phases 1–3 are: 0.92 ± 0.09, 1.18 ± 0.22, and 0.93 ± 0.10 μg/ml, respectively. The mean value for the fluoridated waters for the 3 phases combined is 1.01 ± 0.15 μg/ml which is in good agreement with the often-cited target value of 1 μg/ml.

Returning to the question of what values should be put into food composition databases, what is the harm in averaging everything together? What should be done with values below the detection limit? Should zero values or 0.5 × detection limit be substituted for not detected (nd)? If only detectable values are used, what is the significance of the mean being elevated? The fact that water is typically "artificially" fluoridated is the real issue. It is inappropriate to average all 24 values together and conclude that based on this 3-phase study representing 15–16 cities in 10 states, an individual is likely to ingest 0.48 ± 0.47 μg/ml fluoride by drinking municipal water. In fact, none of the water samples had that concentration. The bi-modal distribution of fluoride in water poses a unique challenge in how best to interpret, share and utilize the data. Based on the results from this study, it is most appropriate to conclude that there is a 40% probability of an individual ingesting fluoridated water with an average expected fluoride content of: 1.04 μg/ml and a 60% probability that they will ingest unfluoridated water with an average fluoride content less than the analytical detection limit of 0.081 μg/ml. Results from the CDC database

(CDC, 2002) for the 10 states studied here, indicate that, on average, 57% of the water supplies are fluoridated but the CDC database contains no specific information on the expected variability between cities within the states.

4. Conclusions

Future large-scale studies are needed to better characterize the large variability in the fluoride content in water samples in the United States. To ensure that optimal and safe levels of fluoride intake can be recommended to the US public, those foods which are the primary sources of fluoride to the diet should be monitored and their levels made public (Levy and Guha-Chowdhury, 1999). Because water consumption is so high and because municipal waters are very often used for the preparation of soft drinks, fruit drinks, and other beverages such as formula, coffee and tea, it is critical to have a meaningful estimate of the fluoride content of water so that predictions of fluoride intake can be more accurately made. Any food that absorbs water while cooking may be affected by the fluoride content of the water including rice, pasta, and vegetables. Tea has been reported to be the most popular beverage in the world (Fung, Zhang, Wong, & Wong, 1999) and while tea leaves have been reported to be a natural concentrator of fluoride, the water the tea is prepared in can be a significant source of fluoride as well and cannot be ignored.

Future experiments will focus on the fluoride content of national brands of soft drinks as well as bottled waters. In addition, a tea pilot study will be conducted looking at fluoride extraction from the tea leaves into the water as a function of water temperature and brewing time. Conventional brewing/steeping methods will be compared to microwave preparation of teas. Colleagues in the Nutrient Data Laboratory of USDA have undertaken a large-scale nationwide project concentrating on determining the fluoride content of water samples, tea samples, beverages and other foods which are potentially significant contributors of fluoride to the diet to better characterize the variability of this element in the food supply.

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