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Original Article

The fluoride content of select brewed and microwave-brewed black teas in the United States

Pamela Ruth Pehrsson *, Kristine Y. Patterson, Charles R. Perry

Nutrient Data Laboratory, ARS, Room 208A, Bldg. 005, 10300 Baltimore Avenue, USDA, MD 20705 USA

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ABSTRACT

Fluoride (F) intake is recognized to be important for dental health. Tea leaves are known F accumulators and brewed tea as well as the water used for brewing may contribute significantly to individual intake. The USDA's Nutrient Data Laboratory determined the F content of brewed and microwaved teas using geographically matched tap water samples. Two brands of top-selling regular and one of decaffeinated teabags were purchased in 36 locations and brewed either by steeping in boiled water or with microwave heating followed by steeping. The mean F content for caffeinated regular brewed tea was 373 ± 49 μg/100 g (n = 63) and for decaffeinated tea was 270 ± 46 μg/100 g (n = 34). The overall mean for F in microwaved regular tea was lower than regular brew (364 ± 40 μg/100 g vs. 322 ± 30 μg/100 g (n = 36)). In all cases, prepared tea using water from the Midwest had the highest F-values. The mean F content of the brewed teas was 3–4 times higher than the national mean of the tap water, analyzed separately (71 ± 33 μg/100 g). These data are the first nationally representative F-values for brewed teas, and will provide valuable information to the dental and medical research communities in assessment of fluoride intake and impact on dental health.

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1. Introduction

Fluoride (F) intake is recognized to be important to dental health; inadequate intake is associated with a higher incidence of dental caries, while an excessive intake is associated with enamel and possibly skeletal fluorosis (Institute of Medicine (IOM, 1997)). The Adequate intake (AI) from all sources for adult males is 4 mg/day, 3 mg/day for adult females, and 1–2 mg/day for children, established by the IOM as part of the Dietary Reference Intakes process (IOM, 1997). Dietary fluoride in the U.S. comes primarily from fluoridated water; commercially prepared foods and seafood, brewed and ready-to-drink teas, and oral health products also contribute to fluoride intake (ADA, 2005).

Tea consumption has increased over three-fold in the past two decades, primarily from ready-to-drink teas but also from brewed black teas (Simrany, The Tea Association, personal communication, 2007). According to the National Health and Nutrition Examination Survey (NHANES, 2003), Americans consume on average about one cup (240 mL) of brewed tea per day. Tea trees (*Camellia sinensis*), a perennial shrub, are a naturally rich source or accumulator of F, especially in the leaves; the plant absorbs F through passive diffusion from the usually acidic soil in which it grows (ATSDR, 2003; Ruan and Wong, 2001). Tea leaves can accumulate fluoride to concentrations in excess of 10 mg/100 g dry weight (Wei et al., 1989; Cremer and Büttner., 1970). Since much of the F released during brewing is available to the consumer, consumption of tea may contribute significantly to individual F intake (WHO, 1984). Historically, tea was grown in natural soil; however, many tea growers now use phosphate fertilizers, which may further increase the F content of the tea (ATSDR, 2003). After long-term use of these fertilizers the F concentration of the soil may be significantly increased (Loganathan et al., 2001). Since tea is grown in China, India, Sri Lanka, Japan, Africa and other areas around the world and is then imported to the U.S., use of these fertilizers may be difficult to determine.

In addition to F released from tea leaves, the contribution of F from the brewing water to the brewed tea can be significant, especially in areas where the F content of a fluoridated municipal water supply (or well water naturally high in fluoride) is well above the recommended fluoridation target of 1 ppm (the standard fluoridation practice for U.S. public water supplies). Imprecise fluoridation practices, brew time, caffeine content, the amount and F concentration of the dried tea used in the infusion, and the use of home water filtration systems that may remove F in the household water (Malde et al., 2006; Haman and Bottcher, 1986) also contribute to F content variability in teas. Literature values from

* Corresponding author. Tel.: +1 301 504 0693; fax: +1 301 504 0692.
E-mail address: pamela.pehrsson@ars.usda.gov (P.R. Pehrsson).

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studies that did not necessarily account for these considerations show brewed tea contains about 1–6 ppm F (Cao et al., 2006; Fung et al., 2003; Wei et al., 1989; Cremer and Büttner., 1970). Chandrajith and Abeypala (2007) analyzed tea brewed with deionized water and found no differences due to the region in which the tea was grown (all within Sri Lanka) or tea grade; the F concentration ranged from 0.3 to 1.7 ppm. Chan and Koh (1996) observed that decaffeinated tea infusions using distilled-deionized water and retail tea purchased in Texas were twice as high in F as were caffeinated infusions (3.2 ppm vs. 1.5 ppm); they attributed the increase to naturally high-fluoride mineral water used during the decaffeination process.

International and national research on the F contribution of brewed tea and research correlating dental caries and fluorosis with intake (Chan and Koh, 1996; Malde et al., 2006; Cao et al., 2006; Chandrajith and Abeypala, 2007; Hallanger Johnson et al., 2007; IOM, 1997) suggests regionally and nationally representative data on commonly consumed teas would be highly useful in the U.S. and elsewhere around the globe. In previously published research, tea was prepared with either distilled or deionized water (Chan and Koh, 1996; Malde et al., 2006; Cao et al., 2006; Chandrajith and Abeypala, 2007). In order to best represent tea as consumed across the U.S., this study analyzed black tea (the most common type consumed nationally) purchased across the country and municipal water from the same general region as the point of purchase and preparation, simulating tea as prepared for consumption. Fluoride from the brew water, the tea leaves, different brands, the influence of the brewing method, and caffeine content were examined; resulting data were incorporated in the USDA National fluoride Database maintained by the Nutrient Data Laboratory (NDL), Beltsville Human Nutrition Center, Beltsville, MD (USDA, 2005). While F-values are typically reported in parts per million based on its presence in the water supply, where 1 ppm is equivalent to 100 μg/100 g water, the results for this study are reported on a weight/100-g basis, consistent with consumer understanding of consumption amounts and the USDA National Nutrient Database (USDA, 2010).

2. Materials and methods

2.1. Sampling frame development and water collection

The sampling and collection of tap water used to brew the tea in this study is described in detail in Pehrsson et al. (2006). Tap or municipal water was used (as opposed to deionized water) and matched to dry tea purchase locations to best simulate the tea as consumed. Briefly, the water sampling method involved: serpentine ordering of the U.S. population by census region, division and county; dividing the population into 72 equal population-size zones and randomly selecting one county per zone and two residences per county. Los Angeles County, CA was selected twice in the first stage because of its size; therefore, the sample includes only 71 unique counties (see Fig. 1). The mathematical approach to the sample frame development is described in greater detail in Perry et al. (2002).

Participants at each of the two residences in each county were recruited by phone to provide two tap water samples from their homes. Samples were collected in high-density polyethylene (HDPE) bottles, 3–4 months apart, for a total of 288 samples and sent to Virginia Polytechnic Institute and State University (VPI), Blacksburg, VA for processing. These 288 water samples were combined to give a total of 36 water samples by compositing all the water from two counties, paired by geographic proximity (the exception being Maryland and Kansas), both sites in each county and both pickups. These water composites were stored at −60 °C until they were used to make the brewed tea. One of the four sites in each set of two counties was randomly chosen for the purchase of dry tea from a retail outlet (Table 1).

2.2. Dry tea samples

Two highly consumed and nationally available brands of caffeinated black tea (brands A and B) and a decaffeinated product (brand B) were procured as boxes of teabags. These brands were selected based on national market share data; however, retail outlets were selected locally or within counties according to the

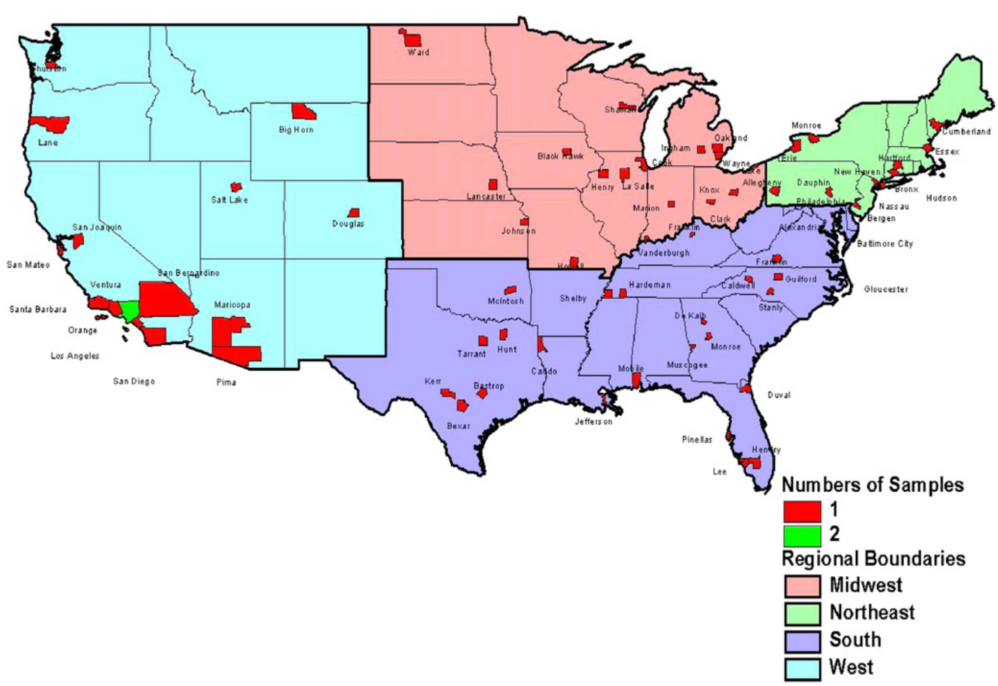

Fig. 1. Fluoride sampling by regions and counties.

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Table 1
Examples[a] of sampling locations: brew wattaber[b] and dry tea.

Source of water for brewing				Source of tea
State	County-water pickup	Site	Season	County—dry tea pickup
OK	McIntosh	55A	Winter	Tarrant
			Spring	
		55B	Winter	
			Spring	
TX	Tarrant	66A	Winter	
			Spring	
		66B	Winter	
			Spring	
UT	Salt Lake	67A	Winter	Salt Lake
			Spring	
		67B	Winter	
			Spring	
WA	Thurston	70A	Winter	
			Spring	
		70B	Winter	
			Spring	

[a] These are examples of 2 of the 36 composited water samples and the matching locations for dry tea samples.
[b] Water samples combined across two counties, two sites per county and two seasons per site for a total of 8 samples to make up one brew water composite.

annual sales volume of the outlets. All three levels – location, store, and brand name – were sampled probability-proportional-to-size (PPS; level denominators were population density, sales volume, and market share, respectively); that is, all level members had a chance for selection and the chance of the level individual being selected was consistent with its contribution to the level total.

2.3. Tea brewing

2.3.1. Tea brewing procedures

The day before brewing, frozen water samples were removed at the end of the day and placed on the counter at room temperature to thaw, unopened, overnight for 16–20 h; post-thaw water temperatures ranged from 19 to 24 °C. One tea bag, randomly selected from the location-matched box of teabags, was used for each cup of tea prepared; the paper tab was removed and the dry bag was weighed but the string remained attached for handling convenience. Tea samples were brewed in cups, using boiling water ("cup brewing") and as recommended by the tea industry, or in the microwave, according to typical consumer practices. All data (dry and wet tea bag weights, temperature after brewing and when sub-sampled, and sample description information) were recorded for each prepared sample. Excess moisture from the tea bag upon removal was extracted using a plastic spoon; a clean/separate spoon was used for each sample. The tea bag was wrapped with string and gently squeezed; only the string and spoon contacted the teabag. The tea was allowed to cool, uncovered, from 29 to 31 °C for safety in handling; ice baths were used in some cases to speed cooling. The tea was stirred immediately before sampling using either a glass rod or plastic spoon. Samples were aliquotted using a pipette into 15 mL polypropylene screw-cap test tubes (Sarstedt #60540; Sci-Mart, Inc., St. Louis, MO) for fluoride analysis and into 50 mL bottles for mineral analysis (to be reported separately).

2.3.2. Cup brewing

First, 180 ± 0.5 g of specified water was transferred to a 250 mL Erlenmeyer flask by pouring water from the bottle, then using a disposable pipette to adjust the weight. The water was heated in the flask to boiling on a flat surface stove, then boiled 30 s and removed from stove (resulting in 6 oz (178 g (mL) ± 2%) considering vaporization losses). The temperature of the water (°C) was measured after boiling for 30 s. The water was poured over the tea bag into a 400 mL laboratory beaker and steeped for exactly 4 min, within the tea industry's recommended range for brew time of 3–5 min.

2.3.3. Microwave brewing

The specified bottle of composited water was shaken for 30 s and 180 ± 0.5 g transferred to a 400 mL beaker. Tea bags were placed in the beaker, using a plastic spoon to push the tea bag under the water. The microwave oven (1200 W as specified by manufacturer) was used at full power; beakers (containing water and tea bag) were heated for 1 min. Tea bags then remained in the beaker and steeped for exactly 30 s. The steep time was short enough to prevent tea bag bursting (which often occurred at 1.5–2 min in pilot testing). Temperatures (°C) at end of 1.5 min microwave brewing period (1 min in microwave plus 30 s steep) were recorded.

2.4. Analytical methodology

The University of Iowa (UI) College of Dentistry analyzed the brewed tea and the original water samples for fluoride. The results for the water analysis have been reported previously (Pehrsson et al., 2006). The general methodology and equipment used for the fluoride measurements has been described in detail by Heilman et al. (2006). Briefly, the fluoride concentration in the samples was determined using a fluoride ion-specific electrode and five F standards to set the instrument calibration curve.

2.5. Quality control

Brewing blanks were prepared by following the protocol for cup brewing (2 blanks) or microwave brewing (1 blank) using samples of distilled, de-ionized (DDI) water that had been stored frozen in bottles. The only difference from the protocol was that no tea bag was used. For all segments of the project, labware was acid cleaned. Each day two control samples were prepared using water from an in-house water control composite and a tea bag from a control tea that was a locally procured box of caffeinated tea matching one of the two brands used in the study using the same protocols as previously described.

A freeze-dried urine reference material, SRM 2671a (National Institute of Standards and Technology, Gaithersburg, MD) certified for fluoride, was used as the in-house control. During routine analysis of tea samples, blinded quality control samples which were aliquots of F control solutions produced at VPI, were shipped with test samples at a rate of one high-concentration control sample or one low-concentration control sample for every 16 test samples. In addition, 10% of the samples were analyzed for F in duplicate.

2.6. Data analysis

SAS statistical software (Littell et al., 1996) was used to analyze fluoride values in the tea samples. A covariate model analysis using F in the brewing water as the random covariate and region, caffeine status, and brew type as fixed variables was performed ($p < 0.05$); water F level was used as a random covariate because of the known variability (Pehrsson et al., 2006). Predictive models based on fixed effects were developed to allow prediction of tea F content based on a given fixed effect (brew type, region, or caffeine status) and the F content of water used in the brew.

3. Results and discussion

3.1. Water analysis

The composites of water used to brew the tea ranged from 31.5 μg F/100 g (Pair 4, 4 New York sites, 2 fluoridated, 2 pickups each) to 129.5 μg F/100 g (Pair 7, 4 Pennsylvania sites, all

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Table 2
Fluoride content in brewed black teas, national averages.

Tea type	N[a]	Total F[b] (μg/100 g)	% F[c] from tea	% F[c] from water
Cup brewed[d]	97	337 ± 69	78.9	21.1
Microwave brewed[e]	36	322 ± 30	78.0	22.0
With caffeine[f]	99	354 ± 50	79.9	20.1
With caffeine, cup brewed[g]	63	373 ± 49	81.0	19.0
Decaffeinated, cup brewed[h]	34[h]	270 ± 46	73.7	26.3

[a] Number of sample analyses.
[b] Mean ± s.d.
[c] Calculated using 71 μg/100 g = average national fluoride content of drinking water (Pehrsson et al., 2006).
[d] Two brands, one brand includes regular and decaffeinated types.
[e] One brand.
[f] Two brands, one brand includes cup and microwave brewed.
[g] Two brands, cup brew only.
[h] One brand, decaffeinated tea not found in some locations so number is less than 36.

fluoridated, 2 pickups each). The brew water contributed significantly to the final F concentration of the prepared tea ($p < 0.05$) contributing up to one-fourth of the F in the prepared tea (Tables 2 and 3). Fluoride content of the prepared tea varied significantly ($p < 0.05$; slope = 0.6735, a linear effect) among the 36 location-specific water samples and was considered the most influential of the covariates.

3.2. Analytical quality control results

The mean value obtained for the certified NIST freeze-dried urine was 56 μg/100 g F (0.56 mg/L), which compared well to the certified value of 55 μg/100 g (with an uncertainty range of 52–58 μg/100 g). The high level control mean was 210 μg/100 g (with an expected range of 200–250 μg/100 g) and the low-level control mean was 40 μg/100 g (with an expected range of 40–50 μg/100 g). All of the brewing process blanks were below the detection limits for F (2 μg/100 g). The tea control samples that were run daily with the test samples had a mean value of 390 μg/100 g) (%rsd = 7.6) for the cup brewing and 350 μg/100 g (%rsd = 3.0) for the microwave brewing. The difference between duplicates on average was ±1.3%.

3.3. Tea sample results

The F contents of brewed teas are summarized in Table 2, and are reported several ways. The F concentration (national means) in regular brewed tea (brands A and B) was 373 ± 49 μg/100 g (Table 2); decaffeinated tea (brand B) was 270 ± 46 μg/100 g; and microwaved tea (brand B) was 322 ± 30 μg/100 g. Regional differences were not seen for tea F when F in the water was considered: significant differences in tea F were shown across brew type and caffeine level ($p < 0.05$; Table 3). In all cases, tea from the Midwest had the highest F-values, consistent with the F content of the water. F content in traditional (cup brewed) tea was higher than microwaved tea; subtracting out the F from the water, F coming from tea in the traditional brew ranged from 29 to 58 μg/100 g (South-Midwest) more than from microwaved tea. F content from traditional brew regular tea was higher than from decaffeinated tea, possibly due to steep time or water temperature differences. The range in differences was 47–88 μg/100 g with the largest difference between teas in the Midwest and the lowest in the West. The F content of the brand B tea, excluding the F contributed by the water, either showed no difference or was slightly higher than brand A tea: 0–33 μg/100 g F, with the largest difference in the South and no difference in the Northwest and West samples. In all cases, most of the F was contributed by the tea leaves (range 74–85%, Table 3).

Using these data, predictive models for contribution of fluoride based on specific characteristics of the prepared tea were developed. For matched pairs (matched by location, excluding missing values), F variability was significantly affected by brew type (traditional 364 ± 40 μg/100 g vs. microwave 322 ± 30 μg/100 g, $p < 0.0001$) and caffeine level (regular 270 ± 44 μg/100 g vs. decaf 364 ± 40 μg/100 g, $p < 0.0001$). For matched pairs, no significant differences were observed for effect of region (p = 0.33–0.75) and brand of tea (p = 0.17). For the latter, the average amount of F coming from the water contributed most to the variability of F in the brewed tea. Missing values for brand A are a limitation of these results.

Given that a cup of brewed tea weighs 178 g (6 oz)–237 g (8 oz), as typically served in the U.S. (USDA, 2010), doubling the F concentrations of the samples presented in these tables should be considered when estimating F intake per serving from brewed tea. Using the results of this study, about 4–5 cups of brewed tea for women and about 6 cups for men would achieve the recommended daily intake for fluoride; this does not account for additional amounts provided by natural or intended fluoridation of drinking water higher than 1 ppm, fluoride consumed from drinking water and other foods and beverages, and the use of oral health products. Data from this study may be useful in developing tools for managing fluoride intake, which is correlated with prevalence of dental caries and, at higher intakes, with dental fluorosis.

Table 3
Fluoride in brewed black teas[a]: sources of variability.

Variable	No. of sites	Total F[b] (μg/100 g)	F from water[c] (μg/100 g)	F from tea[d] (μg/100 g)[b]
Region (all teas, cup brewing)				
Midwest	8	345 ± 65[1]	87 ± 26	258 ± 59 (75%)
Northeast	7	326 ± 56[1]	69 ± 46	256 ± 45 (78%)
South	13	342 ± 57[1]	78 ± 24	264 ± 58 (77%)
West	8	313 ± 65[1]	48 ± 26	265 ± 67 (85%)
Brand (cup brewing)[e]				
Brand A	27	385 ± 59[1]	73 ± 33	312 ± 31 (81%)
Brand B	27	367 ± 43[1]	73 ± 33	294 ± 31 (80%)
Brew (caffeinated, brand B)[e]				
Cup brewing	36	364 ± 40[1]	71 ± 33	293 ± 28 (80%)
Microwave	36	322 ± 30[2]	71 ± 33	251 ± 31 (78%)
Caffeine (cup brewing, brand B)[e]				
Decaffeinated	34	270 ± 46[1]	71 ± 34	199 ± 35 (74%)
Regular caffeinated	34	364 ± 40[2]	71 ± 34	294 ± 28 (81%)

[a] Mean ± S.D. Slight differences from mean and variability data in the USDA National Fluoride Database (USDA, 2005) may be attributed to the combining of composite data for this research.
[b] Differences in numerical superscripts within categories show significance at $p < 0.05$.
[c] Fluoride in water used for preparing tea.
[d] Contribution (%) from dry tea to total fluoride in prepared tea given in parentheses.
[e] Matched by location.

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4. Conclusions

Clearly, while the level of sampling of water and tea was extensive in this study, the variability in local water supplies within regions of the U.S. is wide, even within counties and because of alternate water sources (well water vs. municipal water supplies). However, on average, the dry tea contributes 3–4 times as much fluoride to the brewed tea as does the water. The fluoride provided by brewed tea may contribute significantly amounts of F, and should be considered when assessing total daily intake.

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