

ORIGINAL ARTICLE

Public-health risks from tea drinking: Fluoride exposure

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Abstract

Aims: Due to new evidence on fluoride neurotoxicity during early life, this study examined maternal exposure to fluoride through tea consumption in a low-fluoride region and measured fluoride releases from commercially available teas (tea bags and loose teas) to determine the need to limit fluoride exposure. **Methods:** Maternal urine fluoride (MUF) concentrations were measured in spot urine samples ($N=118$) from first-trimester pregnant women and in prepared tea infusions made with deionised water from 33 brand teas and 57 loose-tea products, as determined by the direct method of using a fluoride-selective electrode. **Results:** The fluoride concentration in the local drinking water supplies ranged from 0.10 to 0.18 mg/L, and the creatinine-adjusted MUF ranged from 0.09 to 1.57 mg/L. Seventeen per cent of the women were daily tea drinkers, and their MUFs were higher than those with no consumption ($p=0.002$). The fluoride concentration from tea bags ranged from 0.34 to 2.67 mg/L, while loose teas showed 0.72–4.50 mg/L (black), 0.56–1.58 mg/L (oolong), 1.28–1.50 mg/L (green), and 0.33–1.17 mg/L (white tea). **Conclusions: Fluoride exposure among pregnant women increases with tea consumption, with likely risks of developmental neurotoxicity to their children. As the fluoride release from tea varies widely, the fluoride concentration should be indicated on tea packages in order to allow consumers to make informed decisions on minimising their fluoride exposure.**

Keywords: Maternal urine fluoride, drinking water, neurotoxicity, prenatal exposure, tea

Introduction

Fluoride is considered beneficial in the prevention of dental caries, especially when applied topically in the oral cavity [1], but recent epidemiological studies suggest that fluoride is a potential developmental neurotoxicant [2]. By now, more than 40 cross-sectional studies have reported cognitive deficits in children living in communities with elevated, sometimes substantially elevated, fluoride concentrations in drinking water [3]. So far, this evidence has been considered inconclusive, and the World Health Organization's (WHO) recommended maximum fluoride concentration in drinking water of 1.5 mg/L has remained unchanged since 1986 [4].

New evidence from prospective studies call for a reassessment, as they provide substantial evidence that fluoride is a developmental neurotoxicant at prevalent exposures. Two studies were carried out in Mexico and measured the maternal urine fluoride (MUF) concentration during pregnancy. The first explored the association with scores on the Bayley Scales among 65 children evaluated at age 3–15 months [5]. The fluoride exposure during the first and second trimesters was associated with significantly lower Bayley scores after adjustment for covariates. The second relied on the ELEMENT birth cohort study that provided McCarthy Scale scores at the age of four years in 287 children and IQ by an abbreviated Wechsler scale at age 6–12 years in 211 children. An increase in

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MUF by 1 mg/L was associated with losses of 6.3 and 5.0 points on the two scores [6].

At slightly lower fluoride exposure levels in the Canadian MIREC cohort, intellectual abilities were assessed using a Wechsler scale in 512 children at three to four years of age. An increase in MUF by 1 mg/L was associated with a statistically significant loss in IQ of 4.5 points in boys, though not in girls. An increase of 1 mg/L in water fluoride was associated with an IQ loss of 5.3 points for both boys and girls. The daily fluoride intake was estimated, assuming a fluoride content of 0.52 mg in one cup of black tea, and an increased intake by 1 mg was found to be associated with an IQ loss of 3.7 points, again for both boys and girls [7]. In an extension of this study, the authors assessed the possible impact of fluoride exposure from reconstituted formula in fluoridated and non-fluoridated communities [8]. After adjustment for prenatal fluoride exposure and other covariates, each increase by 0.5 mg/L in the water fluoride concentration was associated with a decrease by 8.8 IQ points in the children who had been formula-fed in the first six months of life.

These prospective studies from North America relied on individual exposure indicators and focused on prenatal and early postnatal exposure known as a key window of neurological vulnerability [9]. Previous studies are less informative and suffer from important weaknesses [3]. Still, the fluoride-associated IQ losses are in accordance with the difference of almost 7 IQ points between exposed groups and controls in a meta-analysis of 27 cross-sectional studies [10].

Given that thresholds for toxicity cannot be observed, regulatory agencies often use so-called benchmark dose calculations to establish safe exposure limits [11], where a loss of 1 IQ point is considered an adverse effect [12]. Relying on the results from the two major studies [6,7] calculations show that a protective limit for both urine fluoride and water fluoride should be <0.2 mg/L [3,13], that is, much below the WHO recommendation. Due to this new insight, a renewed assessment seems appropriate in regard to common sources of fluoride intake.

In Scandinavia, fluoride concentrations in community drinking water must be monitored regularly, and in Denmark, results commonly range from very low to 1.5 mg/L [14]. In regard to bottled waters, any concentration above the 1.5 mg/L limit must be indicated on the label. Most products on the Danish market show concentrations <0.2 mg/L, although a Swedish brand has a concentration almost twice the 1.5 mg/L limit [15]. While fluoride in some other countries is added to table salt, known sources of fluoride intake otherwise include dental products (toothpaste, mouth rinses), fluoride supplements

(pharmaceuticals or fluoride tablets) and some minor dietary sources [4,16].

Among beverages, tea has the highest potential for increasing daily fluoride intake [17], as the tea plant, *Camellia sinensis*, accumulates fluoride that is released into tea infusions [18]. Tea bags available in Ireland were found to release fluoride concentrations of 1.6–6.1 mg/L (ppm) [17], and those in the UK, 3.60–7.96 mg/L [19]. Apart from certain sources of drinking water and some brands of bottled water, tea therefore appears to represent a major source of fluoride exposure. High fluoride exposure through the consumption of tea, particularly by vulnerable populations (e.g. pregnant women and infants) may negatively affect early cognitive development. However, no published data are available from Scandinavia whether populations in low-fluoride regions may be exposed to adverse effects due to tea consumption. The present study therefore examines the MUF concentration among Danish pregnant women and its association with tea drinking, and estimates the fluoride intakes from commercially available teas (tea bags and loose teas) in Denmark. This information provides documentation on the relative importance of fluoride intake from community water and different types of tea on the market as a possible basis for guidance of consumers, especially vulnerable populations.

Methods

Maternal urine collection

Maternal spot urine samples were collected from 118 first-trimester pregnant women aged 22–43 years ($M_{\text{age}}=29.5$ years), who attended their first ultrasound scan at an antenatal clinic in Esbjerg, Denmark. The population was drawn from an area with low fluoride exposure in drinking water at the residence (<0.2 mg/L). The fluoride concentrations in drinking water at home were obtained from the Geological Survey of Denmark and Greenland. Information on the consumption of drinking water, bottled water and black and green tea was obtained by a self-administered questionnaire. In connection with the urine sampling, a questionnaire asked about beverage consumption and other fluoride sources during the past four weeks. Women indicating the habit of swallowing toothpaste were excluded. Samples were labelled, and 0.2 g of EDTA was added to a 10 mL aliquot of the urine sample prior to storage (–80°C).

Tea sampling

Popular international tea brands, such as Pickwick, Lipton and Twinings, together account for >60% of

Table I. Tea consumption data and maternal urine fluoride (MUF) concentration.*

| At least one cup | Number of women | <i>M</i> (<i>SD</i>), mg/L | 25th–75th percentiles | Total range |
|------------------|-----------------|------------------------------|-----------------------|-------------|
| Daily | 20 | 0.59 (0.30) | 0.40–0.66 | 0.20–1.57 |
| Weekly | 21 | 0.40 (0.14) | 0.29–0.48 | 0.17–0.67 |
| Monthly | 13 | 0.41 (0.16) | 0.32–0.48 | 0.17–0.68 |
| None | 63 | 0.40 (0.20) | 0.23–0.49 | 0.09–0.91 |

* $p=0.009$ by Kruskal–Wallis analysis of variance and $p=0.002$ by Dunn's test comparing MUF at no tea consumption and at daily consumption.

the Danish tea market [20], and we obtained a total of 33 different brands of tea bags in common use, as well as 57 popular types of loose tea, from supermarkets, retail outlets and tea stores in the cities of Odense and Copenhagen, Denmark. The samples were classified on the basis of information provided by the tea merchant or as revealed on the tea bag boxes. The loose-leaf products represented black, green, oolong and white teas from a variety of countries. None of them were decaffeinated.

Preparation of tea infusions

To determine the fluoride release, we prepared tea infusions using deionised water (without detectable amounts of fluoride). Duplicate samples of 2 g loose tea or randomly selected tea bags from each box were used to prepare duplicate infusions of each type of tea. The weight of tea in the bags was determined using an AT261 Delta Range scale (Mettler, Toledo, OH). Because variations were observed in the weight of different types of tea bags, and in order to facilitate comparisons with loose-tea results, the fluoride release is reported both as amount and concentration per tea bag, as well as the results adjusted to 2 g tea. In agreement with previous studies [17,21], we used a brewing time of five minutes and aimed at a ratio of 1% w/v (using one tea bag or 2 g loose tea) in 200 mL boiled deionised Milli-Q-treated water.

Analysis of MUF and tea fluoride

The total fluoride concentration in the tea infusions was determined by a potentiometric measurement with an Orion™ Ion Selective Electrode (ISE 9609 BNWP; Thermo Fisher Scientific, Waltham, MA) coupled to a Model 15 pH-meter from Denver Instruments (Sartorius, Goettingen, Germany) in accordance with a previously described method [22]. Calibration curves were made with Fluoride Standard 0.1 mol/L NaF (certified by NIST SRM) from Thermo Fisher Scientific prepared from dilutions with deionised water. Due to a lack of linearity in the low-concentration range, two separate calibration curves were made: one for the 0.0–0.2 mg/L interval

and a linear curve covering 0.2–10 mg/L. The calibration curves were plotted and fitted to the mV readings from the electrode versus the log fluoride concentration. All samples were diluted prior to the analysis (1:1) with total ionic strength adjusted buffer (TISAB II) solution, as recommended by the manufacturer. The accuracy of the method was controlled during each batch of samples analysed by the use of fluoride solution CRM 0.52 ± 0.02 mg/L (Merck, Darmstadt, Germany). The limit of determination was 0.02 mg/L. The average imprecision of the method was <5.1%.

As the spot urines may have been affected by different degrees of dilution, the mean MUF concentrations were adjusted for the creatinine concentration using the following equation: $MUF_{CR} = (MUF/MUC) \times MUC_{average}$, where MUF_{CR} is the creatinine-adjusted fluoride concentration (in mg/L), MUF is the measured fluoride concentration, MUC is the individual creatinine value and $MUC_{average}$ is the average creatinine concentration of available samples [6]. We used the non-parametric Kruskal–Wallis test and Dunn's multiple comparison test due to distributions deviating from normality.

Results

In the study area in Western Jutland, the fluoride concentrations in the community drinking water vary from 0.10 to 0.18 mg/L, with a mean of 0.12 mg/L. The MUF values adjusted for creatinine ranged between 0.09 and 1.57 mg/L ($M=0.42$ mg/L). The MUF results across categories of tea consumption are shown in Table I. Increased MUF is observed among women with a daily consumption compared to no tea consumption.

The fluoride extracted from the teas varied across types, and the results are therefore classified into commercial tea bags, with loose teas further categorised into black, oolong, green and white. The 33 brands of tea bags were mainly labelled as black teas, although without identifying the origin (Table II). The weight of the tea in each tea bag varied from 1.35 to 2.31 g, and results adjusted to 2 g are also provided for comparison purposes. The fluoride concentrations ranged from 0.29 to 3.95 mg/L, and thus the fluoride intake

Table II. Fluoride content in popular tea bags on the Danish market.

| Tea brand | Mean weight of tea per bag, g | Fluoride released per tea bag | | Fluoride released from 2 g | |
|------------------------------------|-------------------------------|-------------------------------|--------------------------------|----------------------------|--------------------------------|
| | | Amount, mg | Concentration in one cup, mg/L | Amount, mg | Concentration in one cup, mg/L |
| White temple | 2.31 | 0.07 | 0.34 | 0.06 | 0.29 |
| Mandela | 1.92 | 0.12 | 0.60 | 0.13 | 0.63 |
| Tea2you Organic Earl Grey | 1.88 | 0.18 | 0.91 | 0.19 | 0.97 |
| Fredsted Organic English Breakfast | 1.35 | 0.18 | 0.91 | 0.27 | 1.35 |
| Steuarts Ceylon | 2.01 | 0.20 | 1.01 | 0.20 | 1.00 |
| leveis organic Earl Grey | 1.56 | 0.22 | 1.12 | 0.29 | 1.44 |
| Lipton Green Tea Orient | 1.99 | 0.22 | 1.12 | 0.23 | 1.13 |
| Ronnefeldt EB | 1.50 | 0.22 | 1.12 | 0.3 | 1.49 |
| Twinings Pure Green | 1.94 | 0.23 | 1.15 | 0.24 | 1.19 |
| Tea2you Organic English Breakfast | 1.85 | 0.23 | 1.15 | 0.25 | 1.24 |
| Princip Earl Grey | 1.89 | 0.23 | 1.16 | 0.25 | 1.23 |
| Continental Darjeeling | 2.08 | 0.23 | 1.15 | 0.22 | 1.11 |
| Fredsted English Earl Grey | 1.75 | 0.24 | 1.18 | 0.27 | 1.34 |
| Lipton Green Tea Citrus | 2.02 | 0.26 | 1.29 | 0.26 | 1.28 |
| vores Earl Grey | 1.91 | 0.27 | 1.35 | 0.28 | 1.41 |
| Budget Earl Grey | 1.54 | 0.28 | 1.41 | 0.37 | 1.83 |
| Øgo organic Earl Grey | 1.64 | 0.29 | 1.46 | 0.36 | 1.78 |
| Clipper Organic Earl Grey | 2.07 | 0.30 | 1.50 | 0.29 | 1.45 |
| Pickwick Earl Grey | 2.07 | 0.32 | 1.58 | 0.31 | 1.53 |
| Bigelow Green | 1.65 | 0.34 | 1.71 | 0.41 | 2.07 |
| Westminister English Breakfast | 1.75 | 0.36 | 1.79 | 0.41 | 2.04 |
| Lipton Russian Earl Grey | 2.04 | 0.38 | 1.91 | 0.37 | 1.87 |
| Twinings Earl Grey | 1.97 | 0.40 | 2.02 | 0.41 | 2.05 |
| Lipton Chai – Thé Noir | 1.94 | 0.42 | 2.11 | 0.44 | 2.18 |
| AC Perch Breakfast | 3.00 | 0.43 | 2.15 | 0.29 | 1.43 |
| Twinings English Breakfast | 2.03 | 0.46 | 2.32 | 0.46 | 2.29 |
| Lipton Yellow Label | 2.00 | 0.50 | 2.52 | 0.50 | 2.52 |
| Lipton English Breakfast | 1.97 | 0.52 | 2.61 | 0.53 | 2.65 |
| Medova | 2.02 | 0.53 | 2.67 | 0.53 | 2.64 |
| Lord Nelson Green Tea | 1.75 | 0.55 | 2.73 | 0.62 | 3.12 |
| Lord Nelson English Breakfast | 1.75 | 0.56 | 2.79 | 0.64 | 3.19 |
| Mebmer | 1.75 | 0.59 | 2.97 | 0.68 | 3.39 |
| Veer Breakfast Black | 1.96 | 0.77 | 3.87 | 0.79 | 3.95 |

from one cup of tea prepared from a tea bag ranged between 0.06 and 0.79 mg. Without taking into consideration any fluoride content of community water, 10 teas analysed (45% of the products examined) exceeded the WHO's current drinking water guideline (1.5 mg/L). While the results reflect random samples, they may be associated with a substantial degree of variability, as the weight of the tea in each bag may not be uniform, the origin of tea in the tea bags is not declared and may change and the product may well vary with time and between different markets.

Table III shows the fluoride releases from loose teas according to the country of origin. The fluoride concentrations showed a higher maximum (4.50 mg/L) and average (1.63 mg/L) for black teas, somewhat lower levels in oolong (average 1.17 mg/L) and green (average 1.30 mg/L) teas, while white teas showed the lowest fluoride results (average 0.54 mg/L). The black teas revealed large differences between countries of origin, with teas from Nepal

having the lowest and Kenyan teas the highest fluoride release. Fluoride in teas from Sri Lanka, India and China varied between different types of teas.

Based on the most recent evidence, the benchmark dose level (for IQ loss) is approximately 0.2 mg/day [3], while the NOAEL has been previously identified at 2.5 mg/kg/day [23]. Based on these limits, Table IV shows that tea intake by small children may readily result in excessive fluoride intake, even from teas with the lowest fluoride concentrations. Daily consumption of five cups of Chinese or Sri Lankan teas or two cups of Kenyan tea by adults would easily exceed the acceptable daily intake (ADI), and the benchmark dose level (BMDL) may be easily exceeded from just one cup of tea.

Discussion

Despite low levels of fluoride in the drinking water, elevated MUFs were found in samples from the

Table III. Fluoride content in teas originating from various regions.

| Type of tea | Country | Number of teas analysed | Fluoride released from 2 g tea | | |
|-------------|------------------------|-------------------------|--------------------------------|--------------------------------|-----------|
| | | | Amount, mg | Concentration in one cup, mg/L | |
| | | | | <i>M</i> | <i>SD</i> |
| Black tea | Nepal | 5 | 0.14 | 0.72 | 0.30 |
| | Vietnam | 1 | 0.17 | 0.86 | |
| | Darjeeling First flush | 5 | 0.09 | 0.49 | 0.30 |
| | Darjeeling, India | 4 | 0.24 | 1.20 | 0.56 |
| | Assam, India | 5 | 0.28 | 1.40 | 0.40 |
| | Sri Lanka | 6 | 0.35 | 1.73 | 0.90 |
| | China | 4 | 0.36 | 1.82 | 0.87 |
| | Kenya | 3 | 0.90 | 4.50 | 0.72 |
| Oolong tea | Nepal | 1 | 0.11 | 0.56 | |
| | India | 1 | 0.16 | 0.81 | |
| | Taiwan | 2 | 0.29 | 1.46 | 0.32 |
| | China | 1 | 0.31 | 1.58 | |
| Green tea | Nepal | 1 | 0.26 | 1.28 | |
| | China | 4 | 0.22 | 1.08 | 0.53 |
| | Japan | 9 | 0.28 | 1.38 | 0.42 |
| | India | 1 | 0.30 | 1.50 | |
| White tea | China | 3 | 0.07 | 0.33 | 0.03 |
| | Japan | 1 | 0.23 | 1.17 | |

Table IV. Estimated daily fluoride intake from tea (% of exposure limit for children and adults).

| Exposure limit | Age group | Daily exposure to fluoride from tea drinking | | |
|----------------------|---------------------|--|-------------------|------------------|
| | | 1 cup (200 mL) | 2.5 cups (500 mL) | 5 cups (1000 mL) |
| | | 0.14–0.90 mg/day | 0.35–2.25 mg/day | 0.70–4.50 mg/day |
| (% ADI) ^a | Children (<3 years) | 40–255% | 99–635% | 198–1270% |
| | Adults (>18 years) | 9–66% | 24–150% | 50–300% |
| % BMDL ^b | Adults (>18 years) | 70–450% | 175–1125% | 350–2250% |

^aAcceptable daily intake (ADI) of 0.35 mg/day (children); 1.5 mg/day (adults) derived from the NOAEL.²³

^bBenchmark dose level (BMDL) of 0.2 mg/day (adults).³

newly pregnant women. The 17% of the women who were daily tea drinkers had the highest average MUF concentrations that greatly exceeded the BMDL. Tea consumption therefore seems to represent a major dietary source of fluoride intake.

Worldwide, tea is the most popular beverage consumed, although it is not as commonly consumed in Scandinavian countries as it is in, for example, Ireland and the UK [17,24]. Data from Statistics Denmark show that tea consumption increased by 47% between 2016 and 2018 [25], perhaps in part because tea bags are highly convenient and because of the availability of greater varieties. In a recent survey of Danish women aged 20–30 years, 23.7% drank more than five cups of tea every day, that is, a daily consumption of about 1 L tea, and 32% had a daily consumption of 400–800 mL [26]. These consumption levels will surely exceed the safe exposures and may therefore constitute a public-health hazard.

Fluoride releases above the water quality criterion (>1.5 mg/L) were observed in more than one out of three popular tea bags and in a similar proportion of black teas (loose) available on the Danish market. While the present study showed that a single tea bag could result in a fluoride intake of 0.06–0.79 mg, lower or higher releases have been reported in the same commercially tea bags available elsewhere [17,19,27]. As the method for brewing the tea was standardised, the considerable differences identified are likely related to production methods and origins. Thus, because the variety and grade, relative composition and particle size of teas likely affect the fluoride content [17], similarly named products marketed at different times and in different markets may well differ in fluoride releases.

The country of origin of the loose teas is clearly of importance for the fluoride content. However, the majority of tea-bag brands did not reveal the origin of

the tea, perhaps because teas from different regions are used over time. The quality of the tea product may play a role, as previous studies have reported higher fluoride contents in tea bags compared to loose tea leaves [19]. Past studies have found elevated fluoride concentrations in Kenyan teas compared to teas from other origins [17]. The present study did not include decaffeinated teas, but a previous study found that decaffeination does not remove fluoride and may even increase the amount of fluoride released during brewing [28].

Observational studies have concluded that tea consumption is beneficial for health [29], but few epidemiological studies have so far focused on the fluoride exposure from tea consumption, especially in regard to risks during pregnancy. In the MIREC study [7], the estimated fluoride exposure assumed an additional fluoride intake of 0.52 mg from one cup of black tea, which is in accordance with the average seen in the present study. However, the results in Table II show a lower fluoride content in green tea (0.28 mg) than the 0.39 mg used by the MIREC authors for calculation of the daily fluoride intake in Canada [7].

Because the tea infusions were made with fluoride-free water, the fluoride in the potable water must be added to the results [18]. Thus, depending on residence and the tea chosen, a daily fluoride intake from two cups of tea could easily exceed 1 mg. The consumption data therefore suggest that many women who often drink tea [26] are likely to exceed BMDL or ADI levels for fluoride.

In regard to adverse effects, dental fluorosis among children has been linked to increased consumption of tea [30], but fluorosis is not the critical effect of elevated fluoride exposure. Thus, the new evidence on developmental neurotoxicity requires greatly increased attention to the fluoride intake in pregnant women and likewise in small children [3,13]. Additional concerns relate to subjects with a high water intake, for example due to renal disease, increased physical activity or work in hot climates, and subjects with further sources of fluoride intake, such as fluoride-containing medications [17].

Notwithstanding the growing demand for certified tea over the past few years [31], there is no food safety standard for fluoride in tea entering the European Union. Thus, no quality certifications or labelling requirements have been imposed on imported teas, and all grades of tea are therefore freely available in the market [18].

Disclosing the fluoride content on tea packet labels may be a possibility to help the consumer consider the impact on fluoride exposure from different teas. The variability in fluoride releases from different

tea types suggests that production of low-fluoride tea may be an option and that regulatory authorities could consider imposing safety standards for the fluoride content in commercially available teas.

Conclusions

Daily consumption of tea is likely a major source of fluoride, as revealed by fluoride in maternal urine samples from tea drinkers, despite the low fluoride content in drinking water. Both tea bags and loose teas available in Denmark can result in tea infusions exceeding the present water-fluoride limit of 1.5 mg/L. Thus, fluoride intake from tea consumption could easily exceed safe limits, thereby resulting in a risk of cognitive deficits in the progeny, as shown in prospective studies. While community water must be monitored regularly for the fluoride concentration, there is no such regulation for tea products. Some types of teas release substantial amounts of fluoride, while certain types or origins of tea contain much less fluoride. Given the new insight on developmental fluoride neurotoxicity, tea should be considered as a major source of exposure, in addition to drinking water, and thus requires public-health attention in regard to prevention of excess fluoride exposure, for example through labelling requirements.

Declaration of conflicting interests

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