



## Fluoride Content in Asian Produced Green Teas

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### ABSTRACT

**Methods:** We brewed 2.5 g each of 4 types of green tea (from China, South Korea, Japan and Sri Lanka) using deionized water heated to boiling. Water was cooled to 71°C, then tea was steeped for 2 minutes. This process was repeated 20 times, thus providing 20 samples for each tea type. In addition, 20 control samples of deionized water were brewed to the same specifications. Samples were analyzed using a fluoride probe, and statistical power was calculated.

**Results:** The tea samples from different countries varied in the amount of fluoride they contained. The Chinese sample contained the most fluoride, while the Japanese sample contained the least. Means and interquartile ranges of fluoride concentration were calculated for each sample: Sri Lanka 3.58 ppm (0.1425), Chinese 6.83 ppm (0.140), South Korean 5.36 ppm (0.0975), Japanese 1.88 ppm (0.1375) and control 0.33 ppm (0.0078).

**Conclusions:** The origin of tea and environmental factors, such as pollution, groundwater, air and the soil in which it was grown appear to directly affect the amount of fluoride that accumulates in the plants. Considering the fluoride content revealed by this study, green tea consumption habits should be taken into account when prescribing adjunct fluoride therapy as part of a preventative program for patient care.

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Next to water, tea is the world's most commonly consumed beverage.<sup>1</sup> Among the varieties of tea, green tea is the second most popular choice in the world.<sup>1</sup> In Canada, green tea ranks as the third most popular type of tea, after herbal and black tea, according to a study by Ipsos-Reid for the Tea Association of Canada.<sup>2</sup> While 56% of Canadians drink tea for pure enjoyment and relaxation, 41% of those polled felt that tea's purported health benefits were a good reason to drink tea. Green tea is a recognized source of antioxidants, being an excellent source of catechins, especially the polyphenol epigallocatechin-3-gallate (EGCG).<sup>3</sup> Catechins in green tea have been demonstrated to help lower cholesterol<sup>3</sup> as well as to reduce the risks of ischemic related diseases<sup>4</sup> and glaucoma.<sup>5</sup> In addition, research shows that green tea catechins may be effective in preventing various neoplasms, including gastrointestinal, breast, liver, lung and prostate cancer.<sup>6-8</sup>

Systematic reviews have shown that consumption of large amounts of green tea (> 4 cups/day) might be protective against early-stage breast cancer<sup>7</sup> and liver cancer.<sup>8,9</sup> However, green tea consumption was not beneficial in the later stages of breast cancer.<sup>7</sup> Meta-analyses have indicated that greater consumption of green tea can reduce the risk of liver cancer.<sup>8,9</sup> This association was most significant when participants were drinking 4 cups of green tea a day.<sup>9</sup>

All teas, regardless of whether they are classified as black, green or white, originate from the same tea plant, *Camellia sinensis*.<sup>1,10</sup> The method of bruising or manipulating the leaves, which leads to varying degrees of oxidation, is the primary determinant of whether teas become black, green or white.<sup>10</sup> Black tea is made by allowing the leaves to fully oxidize by drying and, thus, turning black, after which they are roasted.<sup>10</sup> In making green tea, the oxidation process is bypassed, allowing the leaves to remain green; the leaves are heat treated rapidly by pan frying, baking or steaming, then dried. Tea that is the least oxidized and the least processed becomes white tea.<sup>10</sup>

There are two major varieties of *Camellia sinensis*. *Camellia sinensis sinensis* is grown mainly to produce white and green teas, while *Camellia sinensis assamica* is grown to make the stronger black teas. Environmental factors, such as weather, harvest time and the soil that the plants grow in can also affect the flavour of tea.

Green tea is packed with nutrients.<sup>1,10</sup> In addition, it contains important antioxidants called catechin polyphenols, which contribute to its many health benefits. There are several derivatives of catechins in green tea: epicatechin, epigallocatechin, epicatechin gallate and EGCG. Of these, EGCG is the most potent and is believed to improve health in multiple ways, including lowering cardiovascular and cancer risks.<sup>10</sup> Green tea has a very high EGCG content, as well as vitamins B and C.

Despite the benefits of green tea, there are concerns that it might contain toxic amounts of harmful elements, such as aluminum,

lead and fluoride.<sup>11</sup> The presence of such toxins is a result of their potential uptake from contaminated soil and subsequent deposition and concentration in the stems, flowers and leaves of *Camellia sinensis*. *Camellia sinensis* is a natural "bio-accumulator" of fluoride, which is released when tea is brewed.

Fluoride is the negatively charged ion of fluorine and, in some contexts, it can be quite reactive.<sup>12</sup> In terms of its charge and size, fluoride resembles a hydroxide ion, but is less basic<sup>13</sup> and has a distinct reactivity that is solvent dependent.<sup>14</sup> The highly electronegative nature of fluoride allows it to integrate into bone and tooth hydroxyapatite crystals, converting them into stronger fluorapatite crystals. Fluorapatite crystals are larger, form quicker than hydroxyapatite and render tooth enamel more resistant to acid dissolution.

Naturally occurring fluoride is found in the earth's surface, where it exists in soil, water and air in the form of calcium fluoride. In some parts of the world, the level of naturally occurring fluoride is high, while in other areas fluoride is deemed to be deficient.

Where water supplies and diets are fluoride deficient, synthetic fluoride may be added systemically, for example in community water fluoridation. Fluoridation of water is considered by the Centers for Disease Control and Prevention to be "one of the 10 great public health achievements of the 20th century" and has been credited with an overall 18–40% reduction in caries.<sup>15</sup>

Ontario, Canada, was among the first jurisdictions in the world to introduce community water fluoridation as a public health initiative to reduce dental caries,<sup>16</sup> with the city of Brantford taking the lead in 1945.

We now know that there are multiple unanticipated sources of fluoride intake, including topical dentifrices, oral rinses, gels and varnishes. In addition, almost all foods and beverages contain certain amounts of fluoride. In 2007, a Canadian expert panel recommended that the amount of systemic fluoride added to water should not exceed 0.7 ppm to prevent deleterious effects on the human body.<sup>17</sup> For perspective, 1 mg of fluoride in 1 L of water is approximately equivalent to 1 ppm. In April 2015, the United States Department of Health and Human Services also lowered the recommended water fluoridation level to 0.7 mg of fluoride per litre of water.

A common question is, how much fluoride is too much? Fluoride toxicity is rare. For an acute fluoride overdose to cause death in a 70-kg man, a 1-time ingestion of 5–10 g of sodium fluoride is required.<sup>18</sup> Smaller acute exposures can cause mild to serious symptoms, most of which are related to gastrointestinal discomfort. These include vomiting, abdominal pain, diarrhea and nausea.<sup>19</sup>

Chronic toxicity can also develop after ≥ 10 years of ingesting large amounts of fluoride.<sup>18</sup> Skeletal fluorosis, a health condition

associated with long-term high fluoride exposure, can lead to brittle and painful bones.<sup>18</sup> The severity and extent of development of skeletal fluorosis depend on the amount and duration of fluoride intake. The mild form of skeletal fluorosis is called osteosclerosis. Advanced skeletal fluorosis can be crippling; however, this is considered a rare condition in Canada and the United States.

Dental fluorosis is more common and is caused by ingesting fluoride during enamel formation. Children,  $\leq 8$  years of age, can develop fluorosis while their permanent teeth are still forming. The mildest form of dental fluorosis can appear as random white flecks or lacy white patterns across the enamel surface. According to the Canada Health Measures Survey, 16% of children may have very mild or mild fluorosis.<sup>20</sup> Surveyed cases of severe dental fluorosis were too minimal to report.<sup>20</sup>

More severe dental fluorosis cases include brownish discolouration and pitting of the enamel. Severity depends on how much, how long and when fluoride was consumed.<sup>19</sup> Studies have shown that as the extent of water fluoridation increases, dental caries levels in children decline; but as fluoridation increases, dental fluorosis also rises.<sup>21</sup>

Previous studies have investigated the amount of fluoride released during the brewing of black tea. In 2013, Chan et al.<sup>22</sup> found that just 1 L of economy-range, bagged black tea provided a significantly greater amount of fluoride than the recommended adult intake of 4 mg/day. They also found that the amount of fluoride released by tea varied significantly by type (loose leaf tea vs. economy bagged tea), with bagged tea releasing the most fluoride. Furthermore, different steeping and brewing methods and times will affect the amount of fluoride released.

Green tea is sourced from a number of countries with different environmental standards. Although research shows the amount of fluoride released from black tea, similar studies have not been conducted for green tea. Because of the popularity of green tea, we sought to quantify fluoride release from whole loose-leaf organic green tea from various parts of Asia, where green tea is mainly produced. We aimed to compare teas from China, Japan, South Korea and Sri Lanka to determine which contained lower fluoride content for safer consumption.

## Materials and Methods

A convenience sample size of 4 types of green tea (Chinese, Japanese, South Korean and Sri Lankan) was chosen and brewed according to the following protocol. Deionized water was used to ensure a purified water source.

For each tea sample, 2.5 g of loose tea was measured into a stainless-steel mesh tea ball. Deionized water was boiled and 80 mL was poured into a fresh disposable paper coffee cup, as Asian green tea is traditionally served in small portions. Water temperature was monitored with a digital thermometer and when it reached 71°C, the tea ball was placed in the cup and steeping was allowed to begin. Steeping time was measured using a chronograph with a digital readout.

Each sample was steeped for 2 minutes. Harney and Sons Master Tea Growers ([harney.com/pages/green-teas](http://harney.com/pages/green-teas)), an award-winning company internationally known for quality teas and education of how to handle tea, recommends a brewing time of 1–3 minutes for green tea; thus, a standard of 2 minutes was chosen for all the teas. After 2 minutes, the tea ball was removed from the cup, and the brewed tea was analyzed using a fluoride probe (Thermo Scientific Orion DualStar, Thermo Fisher Scientific, Waltham, Mass., USA). Each tea type was brewed 20 times, resulting in a total of 80 samples. As a control, the same process was carried out using a clean, empty tea ball of the same type as the experimental tea ball. All reusable articles (thermometer, tea ball) were cleaned with deionized water between experimental runs.

Statistical power was calculated using nQuery Advisor (v. 7.0, GraphPad Software, San Diego, Calif., USA). The Kolmogorov-Smirnov test was used to assess the statistical assumption of normality. Because of the presence of non-normally distributed data, the Kruskal-Wallis test was used to compare the groups in terms of their fluoride concentrations.

Medians and interquartile ranges (IQRs) were reported as descriptive statistics. IQR is the difference between the 75th percentile and the 25th percentile, with a higher IQR indicating greater variability. Following a significant Kruskal-Wallis test, Mann-Whitney U tests with the Bonferroni correction were employed as pairwise comparisons to determine which sample pairs exhibited statistically significant differences. SPSS v. 22 was used in the analysis.

## Results

We calculated median fluoride concentration and IQR for each tea type (**Table 1**): Sri Lanka 3.58 ppm (0.1425), Chinese 6.83 ppm (0.140), South Korean 5.36 ppm (0.0975), Japanese 1.88 ppm (0.1375) and Control 0.33 ppm (0.0078).

Based on a Kruskal-Wallis test, we found a significant difference in fluoride concentration between tea types ( $H = 95.06$ ,

df = 4,  $p < 0.001$ ). The Chinese samples contained the highest concentration, while the Japanese samples had the lowest. A Mann-Whitney  $U$  test with the Bonferroni correction revealed significant differences between the Chinese samples and both

the Sri Lankan and Japanese samples, and a significant difference between the South Korean and Japanese samples. All tea types differed significantly from the control (Table 1).

**Table 1:** Differences in median fluoride concentration and interquartile range among 4 types of green tea, measured in 20 samples of each.

Tea type	Median fluoride concentration, ppm	Interquartile range
Control	0.33	0.0078
Sri Lanka	3.58	0.1425
China	6.83	0.140
South Korea	5.36	0.0975
Japan	1.88	0.1375

*The following tea types were significantly different at the  $p < 0.001$  level: control versus Sri Lanka, control versus China, control versus South Korea, Sri Lanka versus China, China versus Japan, and South Korea versus Japan. The following types were significantly different at the  $p = 0.029$  level: control versus Japan, Sri Lanka versus South Korea, Sri Lanka versus Japan and China versus South Korea.*

## Discussion

The amount of time taken to brew green tea affects the amount of fluoride released, but different types of green tea require slightly different brewing times to fully release their flavours. We consulted the recommendations of Harney and Sons Master Tea Growers (harney.com/pages/green-teas) for brewing and steeping directions. They recommend that to fully experience the complex sweet, fragrance and flavour of green tea, the water boiling point should be ~160 °F, which equates to the ~ 71 °C used in this study. According to Harney and Sons, different green teas have varying steeping times, but the average steeping time is two (2) minutes. If brewing temperature is above 71 °C and tea is steeped longer than the recommended time, the tea may taste bitter. Increased steeping time will also result in a tea infusion that has increased fluoride content.<sup>22</sup>

Tea quality will also influence fluoride content. The highest grades of tea consist of the bud and the youngest leaves of the tea plant, usually those closest to the bud.<sup>23</sup> Thus, loose leaf and younger leaf teas are higher in quality and more expensive. Less expensive teas include older leaves, woody stems and branches. The least expensive teas are often made up of leftover bits of tea leaves and stems, called “tea dust,” packaged in teabags. Studies have demonstrated that inexpensive, lower quality teas contain much more fluoride than higher quality loose-leaf teas.<sup>22</sup>

Bagged teas were not considered in this research, on the understanding that these could contain lower quality tea parts, which might affect fluoride levels. Instead, we sought pure (non-blended) loose-leaf organic tea samples. Organic cultivation methods do not use synthetic pesticides, some of which contain fluoride and would affect the collection results.

Nevertheless, the organic Chinese tea samples revealed high levels of fluoride. Organic South Korean tea samples contained the second highest level of fluoride, followed by Sri Lankan tea samples. The Japanese organic tea samples contained the least amount of fluoride.

Geographic and environmental factors may account for the results in this study. As mentioned in the introduction, natural fluoride exists at various concentrations in different parts of the world. Fluoride is found in soil and in fresh, salt and rainwater. The concentration of fluoride in rainwater is influenced by fluoride-containing particulates that may be released into the atmosphere from various pollutant sources.<sup>24</sup>

As a natural bio-accumulator, tea plants take up fluoride from the soil, water and air. Plants transport fluoride via the xylematic system resulting in much of the fluoride ending up in the tea leaves.<sup>25</sup>

In China, the concentration of fluoride in groundwater is naturally high, with levels up to 8 mg/L in some parts of the country.<sup>26</sup> In South Korea, 1 study demonstrated fluoride concentrations of 4.4mg/L in bedrock groundwater.<sup>27</sup> In Sri Lanka, there is a significant difference in fluoride levels between wet and dry areas<sup>28</sup>: fluoride concentrations of > 8.0 mg/L were found in dry zones, while wet zones were found to contain < 0.8mg/L of fluoride. Sri Lankan tea is produced in many parts of the wet zone.<sup>29</sup> According to the World Health Organization, the fluoride content of groundwater in Japan varies from 0.01 mg/L to 7.8 mg/L, but fluoride was not found in a quantifiable amount in soil.<sup>26</sup> Fluoride levels in Canada's natural groundwater are low compared to those of China, South Korea, Sri Lanka and Japan; most are < 0.05 mg/L.<sup>26</sup>

Environmental factors may also affect fluoride concentrations. China is the world's largest producer and consumer of coal, which is used to drive electrical power plants. Coal has a high fluoride content; when burned, it releases high fluoride-containing particulates into the air.<sup>30</sup> These pollutants float in the atmosphere and eventually leach into the soil or fall into bodies of water.<sup>30</sup> This leaching can impact farming environments, even the 28% of China's cultivatable land that is "certified organic."<sup>31</sup> In China, coal is also used in household heating, cooking and food drying. Indoor airborne fluoride emissions are inhaled; or are ingested once the fluoride particulates land on food and beverages, accounting for unanticipated and potentially harmful ambient sources of fluoride.<sup>30</sup>

South Korea is also largely dependent on fossil fuels for energy; coal accounts for 40% of electricity generation and is also a main pollutant.<sup>32</sup> Meanwhile, Japan relies on coal for a fourth of its electricity and is particularly dependent on fossil fuels after the Fukushima nuclear disaster of 2011.<sup>33</sup>

Cai et al.<sup>34</sup> demonstrated that the harvesting season affects fluoride content in a variety of teas made from the *Camellia sinensis* plant. There are typically 3 harvesting seasons for tea: spring, summer and late summer/fall. Leaves picked in early spring contained the least amount of fluoride, whereas those harvested in the last summer season contained the most.<sup>34</sup>

Finally, just as the cuisines of China and Japan differ, so too do the methods used to cultivate, harvest, oxidize and process green tea. This might also account for differences in fluoride content. Chinese teas are grown on more spreading bushes and the tea leaves are picked by hand, then quickly pan fried or lightly roasted. In contrast, because Japan's arable land is minuscule compared with China's, Japanese tea bushes are cultivated in compact rows and leaves are harvested by machine. Leaves are then quickly steam processed, allowing Japanese green tea to retain its vibrant green colour.

We used deionized water in this study. Most people brew tea with tap water, which may already be fluoridated. In addition, some people may live in areas containing much higher natural fluoride in the groundwater than in other areas of the country. Therefore, it is important to note that people may be ingesting much more fluoride while drinking tea than reported in this project.

## Conclusions and Implications

The origin of tea plants and the environment in which they grow appear to affect the amount of fluoride that accumulates in them. As future research, it would be prudent to study environmental factors in different parts of the arable land in the 4 Asian countries chosen as tea sources in this research, including the soil in which the tea plants are grown, the water used to irrigate them and the surrounding air. Consideration might also be given to studying how fermentation and processing methods (pan frying vs. steaming) affect the fluoride content of green tea.

Although we chose green teas that were labeled "organic," fluoride was found in the samples. This is quite possibly because the tea plants were irrigated with fluoridated water or the plants were subjected to atmospheric pollutants that contain fluoride.<sup>30</sup>

When deciding on adjunct fluoride therapy as part of a preventative program for patient care, the dental professional must consider the fluoride concentrations in fluoride-containing toothpastes, oral rinses, gels, varnishes and fluoride-releasing restorative materials. Dentists must also think about their patients' habits in terms of drinking fluoridated water as well as the fluoride content of foods and beverages. Although virtually all foods contain fluoride,<sup>26</sup> crustaceans, fish, grapes and tea contain the highest levels.<sup>35,36</sup>

In addition, conventionally grown fruits and vegetables will possibly contain more fluoride than organically sourced produce, and conventionally grown tea plants will possibly contain more fluoride than organically sourced ones. Fruit and vegetables grown in chemically laden soil can also be irrigated as well as sprayed with fluoride-containing pesticides and herbicides.<sup>36</sup> Different crops tend to absorb fluoride through their root system at various levels.

When a dental professional considers a person's daily fluoride intake, green tea consumption should be taken into account. This is especially prudent for habitual tea drinkers who drink multiple cups a day on a regular basis.

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**Disclaimer:** Although we hope that the green teas used in this study are representative of the country of origin, we appreciate that the suppliers may have provided a blend of teas from different tea farms in each country. Our intention is to provide a representative snapshot of what differences may be expected among green teas and how fluoride content may vary among countries.

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