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Original Article

# Risk assessment of fluoride daily intake from preference beverage

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**KEYWORDS**
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**Abstract** *Background/purpose:* Tea, coffee and alcohol beverages are called preference beverage and are drunk habitual and in large quantities. Therefore, there is a high possibility that a health risk is caused by the contained components, and risk assessment of intake is essential. However, the risk assessment of fluoride intake from preference beverages has not been sufficiently performed.
*Materials and methods:* This study estimated the daily fluoride intake from preference beverages by measuring the fluoride concentration of infusion liquid and measuring the total fluoride content by the microdiffusion method. In addition, Hazard Quotient (HQ) was calculated for children and adults to assess the risk of fluoride intake.
*Results:* As a result of this study, tea was the highest in all sample species, the infusion of tea was 1.06–6.68 mg/L and the total fluoride content of tea was 47.05–291.98 mg/kg. Green tea showed the next highest value, 0.26–4.09 mg/L, 21.91–83.68 mg/kg. Herbal tea fluoride levels were 0.07–0.17 mg/L and 0.05–1.90 mg/kg, Unique tea was 0.03–0.60 mg/L and 0.03–32.37 mg/kg, Coffee was 0.03–0.15 mg/L and 0.04–0.64 mg/kg.
*Conclusion:* The HQ values calculated from the average daily fluoride intake (DFI) of preference beverages were all within the safe range. Some products made from *Camellia sinensis*, such as tea and green tea, had 1.66 mg/day as DFI at maximum, and the Child's HQ exceeded 1. These results suggest that habitual consumption of some products requires risk management of dental fluorosis.


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## Introduction

Tea is drunk daily around the world, and the type and manufacturing method of tea reflects the climate and culture of each region and are highly valuable in terms of culture. Contains useful substances such as polyphenols, caffeine, and antioxidants, so it is popular with health-conscious people.[1,2] Coffee contains 40–60 mg of caffeine per 100 ml, and has an arousal effect and an increase in basal metabolism. In Japan, it is a preference beverage that is frequently drunk, and domestic consumption has been increasing year by year.[3–5] Alcohol is also a preference beverage for daily consumption and is a major source of exogenous ethyl alcohol in the blood.[6] Alcoholic beverages can be consumed only by adults over the age of 20 in Japan, and alcohol consumption per one adult is reported to be 80.5L per year.[7] Alcohol has been used in Shinto rituals and other events since the early era of Japanese history and has an important meaning as one of the communication tools that reflects locality and culture.[7] Tea, coffee and alcohol beverages are called preference beverage and are drunk habitual and in large quantities. Therefore, there is a high possibility that a health risk is caused by the contained components, and risk assessment of intake is essential. However, the risk assessment of fluoride intake from preference beverages has not been sufficiently performed. Low-dose fluoride enhances skeletal tissue and tooth acid resistance and is beneficial to tooth and bone health.[8] However, excessive chronic fluoride intake can cause dental and skeletal fluorosis.[9] Fluoride in the soil easily migrates to tea trees and accumulates mostly in tea leaves. Therefore, it has been reported that tea leaves contain a large amount of fluoride.[10–16] 40–90% of the F contained in tea leaves is water soluble and is a major source of fluoride intake from beverages.[17] Children under 15 years old are reported to be taking 0.075 L/day and adults over 35 years old are taking 0.75 L/day of tea in the world.[16] In Japan, green tea is traditionally consumed and the intake is reported to be higher than in Europe.[18] The health hazards of fluorosis have been reported in traditional tea beverage production areas around the world, and there are reports in Japan.[9] The limit for fluoride in drinking water is 1.5 mg/L, and the appropriate fluoride intake from all sources including water, beverages and meals is set at 0.05 mg/kg/day.[19] However, there are reports that fluoride intake from food and drink often exceeds the limit in areas where tea intake is high.[20] Coffee contains 0.10–0.58 mg/L of fluoride in the infusion solution and 0.15–0.56 mg/L in instant pack.[21] There are very few publications on the amount of fluoride in alcoholic beverages, but Polish studies reported that 0.093 mg/L of fluoride is contained in 10–20% alcohol and 0.056 mg/L in 20–40%.[22] There are also reports that beer brewed from water sources with high fluoride ion concentrations can contribute significantly to daily fluoride intake.[23] Measuring the daily fluoride intake from preference beverages such as tea, coffee and alcohol is necessary to assess the health risks associated with fluoride intake.

However, the amount of fluoride ingested from preference beverages has not been sufficiently evaluated. Therefore, there is no column of fluoride ion which is a biologically essential mineral in the standard tables of food composition in Japan. Daily Reference Intakes (DRI) in the US, Adequate Intake (AI) of fluoride is determined by age group, 1–3 years 0.7 mg/day, 4–8 years 1.0 mg/day, 9–13 years 2.0 mg/day, 14–18 years 3.0 mg/day, males over 19 years old 4.0 mg/day, females 3.0 mg/day, breastfeeding pregnant women (14–50 years old) 3.0 mg/day. The upper limit intake level (tolerable upper intake level, UL) is set to 1.3 mg/day for 1–3 years old, 2.2 mg/day for 4–8 years old, 10 mg/day for ages 8 and over, and 10 mg/day for pregnant women, and warning that insufficient fluoride intake increases the risk of dental caries.

This study aimed to estimate the daily fluoride intake from preference beverages by measuring the fluoride concentration of infusion liquid and measuring the total fluoride content by the microdiffusion method. In addition, Hazard Quotient (HQ) was calculated for children and adults to assess the risk of fluoride intake.

## Materials and methods

### Collection of tea and alcoholic samples

In this study, tea, green tea, herbal tea, unique tea, coffee and alcoholic beverages were selected as samples for preference beverages. Randomly selected preference beverages prepared as samples are available in the general market in Tokyo. Products were sold at several supermarkets in Tokyo during April 2017 to July 2019. Total 31 tea samples and 41 alcoholic samples were collected from 15 markets, including 5 tea, 7 herbal tea, 5 green tea, 8 unique tea, 6 coffee and 41 alcoholic beverages. Table 1 shows information of each samples, including product name, brand and company, tea species and origin country. Alcoholic beverages are classified into the four categories according to the classification of the Japan Tax Bureau (Distilled liquor, Brewed liquor, Sparkling liquor and Mixed liquor).[7]

### Modified microdiffusion method with a Teflon vessel

In the case of tea leaves, a solution was prepared by extracting 1.0 g of tea leaves with 100 ml of distilled water at 80 °C for 5 min.[20] After cooling to 25 °C, infusion samples were used for measuring. In the case of a tea pack, 1.0 g of the contents was taken out and extraction was performed in the same method. The alcohol drinks were used for measurement after leaving carbon dioxide and leaving it to 25 °C. Liquid samples were analyzed using fluoride ion-selective electrode. Solid samples were homogenized using Centrifugal Mill (Ikeda Rika corp., Tokyo, Japan) before the microdiffusion process. All liquid samples were analyzed for pH value by a Standard ToupH electrode (9615S-10D, HORIBA, Japan) and a pH meter (D-73S, HORIBA, Japan).

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**Table 1** Fluoride content in infusion liquid and solid materials of tea and coffee.

| No. | Product name | Tea species | Brand, Company | Origin country | Shape | Infusion (mg/L) | Solid (mg/kg) | pH of infusion |
|---|---|---|---|---|---|---|---|---|
| 1 | Blacktea without pesticide | Tea | Hishiwa en, Yamanashi, Japan | India | Bag | 2.3503 ± 0.0603 | 183.4267 ± 3.5796 | 5.40 |
| 2 | Black tea | Tea | Nitto kocha, Tokyo, Japan | India | Bag | 1.7303 ± 0.0058 | 55.9467 ± 8.7641 | 5.50 |
| 3 | 100% pure ceylon tea | Tea | Donki jyote, Tokyo, Japan | Sri Lanka | Bag | 1.7370 ± 0.0572 | 52.2800 ± 2.3847 | 5.33 |
| 4 | Lipton | Tea | Lipton, Tokyo, Japan | India | Bag | 6.6803 ± 0.0471 | 291.9800 ± 12.9614 | 5.37 |
| 5 | Uba tea | Tea | big-a, Tokyo, Japan | Sri Lanka | Bag | 1.0570 ± 0.0374 | 47.0467 ± 7.6268 | 5.47 |
| 6 | chamomile | Herbal tea | celestial, Boulder, Colorado, USA | USA | Bag | 0.1450 ± 0.0026 | 0.1970 ± 0.0262 | 7.17 |
| 7 | peppermint | Herbal tea | celestial, Boulder, Colorado, USA | USA | Bag | 0.1707 ± 0.0091 | 1.0717 ± 0.5896 | 6.58 |
| 8 | Rare Ginger | Herbal tea | Newby, London, England | India | Bag | 0.0702 ± 0.0070 | 1.9013 ± 1.4630 | 6.62 |
| 9 | Hotta original ginger with honey | Herbal tea | Hotta, Bangkok, Thailand | Thai | Bag | 0.0790 ± 0.0108 | 0.1793 ± 0.0492 | 6.79 |
| 10 | Chapong thai tea number one | Herbal tea | Number one, Bangkok, Thailand | Thai | Bag | 0.0783 ± 0.0035 | 0.0523 ± 0.0045 | 6.43 |
| 11 | Lemongrass cut | Herbal tea | Ohtsuya, Bangkok, Thailand | Thai | Bag | 0.1390 ± 0.0141 | 0.2041 ± 0.0796 | 5.79 |
| 12 | tropical fruits herb tea | Herbal tea | celestial, Boulder, Colorado, USA | USA | Bag | 0.1463 ± 0.0140 | 0.3907 ± 0.0900 | 3.17 |
| 13 | Oui ocha ryokucha | Green tea | Itoen, Tokyo, Japan | Japan | Bag | 0.2557 ± 0.0542 | 83.6800 ± 18.3515 | 6.10 |
| 14 | Fukamushi senncha | Green tea | United supermarket, Tokyo, Japan | Japan | Leaf | 3.0270 ± 0.7813 | 22.7467 ± 1.6214 | 6.25 |
| 15 | Tokujyou mushi ryokucha 700 | Green tea | Itoen, Tokyo, Japan | Japan | Leaf | 4.0870 ± 0.1112 | 30.1133 ± 0.9877 | 5.96 |
| 16 | Yuuki maccha iri ryokucha | Green tea | Hishiwa en, Yamanashi, Japan | Japan | Leaf | 2.1770 ± 0.0047 | 30.3800 ± 6.6833 | 6.18 |
| 17 | Maccha iri senncha yabukita blend | Green tea | Harada seicha, Shizuoka, Japan | Japan | Leaf | 3.5570 ± 0.0573 | 21.9133 ± 7.8138 | 6.02 |
| 18 | Kaori kaoru mugi cha | Unique tea | Itoen, Tokyo, Japan | Australia | Bag | 0.0266 ± 0.0064 | 0.0256 ± 0.0117 | 7.36 |
| 19 | Alisang woolong tea | Unique tea | Tian, SanFrancisco,California,USA | Taiwan | Bag | 0.6043 ± 0.0232 | 32.3667 ± 3.7277 | 6.74 |
| 20 | Healthy rooibos tea | Unique tea | Itoen, Tokyo, Japan | South africa | Bag | 0.1260 ± 0.0044 | 0.1970 ± 0.0262 | 6.34 |
| 21 | Tamanegino kawa cha | Unique tea | Orihiro brandyu, Gnnma, Japan | Japan | Bag | 0.4360 ± 0.4242 | 1.0717 ± 0.5896 | 5.19 |
| 22 | Mate cha for pot tea | Unique tea | Kokutairou, Shizuoka, Japan | Brazil | Bag | 0.0833 ± 0.0020 | 0.4327 ± 0.0253 | 6.62 |
| 23 | 100%dattann soba cha | Unique tea | Itoen, Tokyo, Japan | China | Bag | 0.0792 ± 0.0022 | 0.0355 ± 0.0035 | 6.64 |
| 24 | Kokusan hatomugi cha | Unique tea | Itoen, Tokyo, Japan | Japan | Bag | 0.0815 ± 0.0016 | 0.0717 ± 0.0070 | 6.62 |
| 25 | Ranong tea | Unique tea | Mulberry, Bangkok, Thailand | Thai | Bag | 0.0783 ± 0.0024 | 7.8867 ± 0.9471 | 6.99 |
| 26 | Mild caldi | Coffee | Camel coffee, Tokyo, Japan | Brazill | Beans | 0.1463 ± 0.0029 | 0.3133 ± 0.0266 | 5.69 |
| 27 | italiano | Coffee | Camel coffee, Tokyo, Japan | Brazill | Beans | 0.1303 ± 0.0367 | 0.0623 ± 0.0582 | 6.18 |
| 28 | Mandheling | Coffee | Japan hills coffee, Tokyo, Japan | Indonesia | Bag | 0.0347 ± 0.0026 | 0.0430 ± 0.0290 | 5.75 |
| 29 | tanzania | Coffee | Japan hills coffee, Tokyo, Japan | Tanzania | Bag | 0.0277 ± 0.0230 | 0.0400 ± 0.0260 | 5.28 |
| 30 | Guatemala | Coffee | Japan hills coffee, Tokyo, Japan | Guatemala | Bag | 0.0259 ± 0.0029 | 0.0550 ± 0.0300 | 5.61 |
| 31 | colombia supremo | Coffee | Japan hills coffee, Tokyo, Japan | colombia | Bag | 0.0299 ± 0.0028 | 0.6360 ± 0.8090 | 5.43 |

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## Determination of fluoride concentration

Liquid samples were analyzed for $F^-$ concentration by a fluoride combination ion selective electrode (9609BNWP, Thermo Science, USA) and an ion meter (930A, Thermo Science, USA). The standard fluoride concentrations were 10, 1.0, and 0.1 ppm were prepared. TISAB III (Total ionic strength adjustment buffer III solution, Sigma–Aldrich Co. LLC., Tokyo, Japan) was added to each sample at the ratio of 10 to 1. Fluoride concentrations of samples were calculated by comparison with a standard curve using 3 standards ranging.

## Modified microdiffusion method with a teflon vessel

For the microdiffusion method, We used modified microdiffusion method with a Teflon vessel system, previously described by Hinoide et al., in 1992. We have evaluated this method in previous studies.[24] The Teflon vessel system is composed of two parts, an inner cylinder and an outer cylinder, and is sealed by a screw type lid and has high airtightness. The outer cylinder is filled with the sample and the diffusion solution, and the inner cylinder is set with the trapping solution. It is designed to have low fluoride loss to carry out all reactions in a closed system. In addition, Teflon has heat resistance and chemical resistance, and can be used repeatedly. Five hundred milligrams of homogenized samples and 4 ml of hexamethyldisilazane (HMDS)-saturated 5M $HClO_4$ (Perchloric Acid, 7601-90-3, Wako, Tokyo, Japan) as the diffusion solution was poured into the outer cylinder. One milliliter of 0.1M NaOH (Sodium Hydroxide, 1310-73-2, Wako, Tokyo, Japan) was poured into the inner cylinder as the trapping solution for fluoride. The apparatus was placed at 60 °C water bath (TM-1A, ASONE, Tokyo, Japan) for 1 h. After the reaction, 0.1 ml of the TISAB III was poured into the inner cylinder. Fluoride concentration in trapping solution was measured using a fluoride ion-selective electrode. For the control, no sample vessel was prepared and subjected to the same process.

## Estimation of daily fluoride consumption and risk assessment of fluoride intake

Estimated daily fluoride intake (DFI) and fluoride intake risk due to the consumption of preference beverages were calculated from the following formulas (1) and (2) according to the method of USEPA (1992).[25]

$$CDI = C \times DI / BW \quad (1)$$

CDI: Chronic daily intake (mg/kg/day)
C: Fluoride concentration (mg/L or mg/kg)
DI: Average daily intake rate of preference beverage (L/day)
BW: Body weight (kg)
DFI: Daily fluoride intake (mg/day) = C × DI.

CDI means Chronic daily intake (mg/kg/day) and C means Fluoride concentration from preference beverages (mg/L or mg/kg). DI means Average daily intake rate, which based on National Health and Nutrition Survey (2018) conducted by Ministry of Health, Labor and Welfare in Japan. The DI for tea was set to 0.2485 L/day, the coffee was set to 0.0658 L/day, and the alcoholic beverage was set to 0.1002 L/day. BW is body weight (kg). According to USEPA (1992), the default weights for children and adults were set to 20 kg and 70 kg, respectively. The multiplication of C and DI means the estimated daily fluoride intake (DFI) (mg/day).

Hazard Quotient (HQ) was calculated using the following formula (2) (USEPA, 1999).[26]

$$HQ = CDI / RfD \quad (2)$$

HQ: Hazard Quotient.
RfD: Reference dose.

Reference dose (RfD) is an estimate of daily exposure that is not expected to be a significant risk of adverse effects throughout life. The RfD of fluoride is 0.06 mg/kg/day, which includes both 0.05 mg/kg/day due to fluoride intake via beverage and 0.01 mg/kg/day due to fluoride intake via meals. When the HQ is greater than 1, the estimated potential fluoride exposure exceeds the RfD and a risk of fluorosis may be posed.

## Statistical analysis

All data are expressed as means of three replicates with standard deviation. The statistical analysis among the sample was performed by one-way analysis of variance (ANOVA), and differences were considered significant at $p < 0.05$. The Bonferroni test was used for post-hoc comparisons when significance was determined by analysis of variance ($p < 0.05$). All statistical analysis were performed with Origin 2019b (version 9.6.5.169, LightStone, USA)

# Results

## Fluoride content in infusion liquid and solid materials of tea and coffee

In this study, we categorized tea species as follows; tea, green tea, herbal tea, unique tea. We carried analysis of fluoride concentration in both total concentrations and its infusion. The tea infusion showed fluoride ion concentrations of 1.06–6.68 mg/L, and the total fluoride content of tea leaves was 47.05–291.98 mg/kg, the highest fluoride content of all sample species (Table 1).

The pH of the infusion was 5.33–5.50, and all of the five samples showed weak acidity. In this study, infusion was performed with the same extraction time and temperature as drinking, but the elution ratio calculated from the fluoride content in tea leaves was low at 2.15%. Herbal tea infusion had a fluoride ion concentration of 0.07–0.17 mg/L and total fluoride content of 0.05–1.90 mg/kg. Most of infusion sample pH were near neutral, however there was also a highly acidic sample showing pH 3.17. The elution rate of fluoride into the infusion was as high as 20.74%. The concentration of fluoride ion in the infusion of green tea was the highest value after tea, 0.26–4.09 mg/L and the amount of fluoride contained in tea leaves was

21.91–83.68 mg/kg. The infusion rate of fluoride was low at 6.94%, and the pH of the infusion was slightly acidic at 5.96–6.25. Unique tea showed a fluoride ion concentration of 0.03–0.60 mg/L in the infusion and a fluoride content of 0.03–32.37 mg/kg in the tea leaves. The pH of the extract was 5.19–7.36, ranging from slightly acidic to neutral. In the case of Coffee, the infusion showed a fluoride ion concentration of 0.03–0.15 mg/L, and the fluoride content of the coffee bean powder was 0.04–0.64 mg/kg. The extraction rate of fluoride was 34.34%, the highest among the sample species. The pH of the coffee infusion was 5.28–6.18 and was slightly acidic (Table 1).

### Fluoride content in alcoholic beverages

Distilled liquor includes alcoholic beverages such as shochu, whiskey and brandy, has a higher alcohol concentration of 12.0–55.0%. The fluoride ion concentration of distilled liquor were 0.01–2.15 mg/L, pH were 2.92–8.95, and the individual differences by product were large (Table 2). The vodka that showed the highest fluoride ion concentration in all alcoholic beverage samples in this study was included in distilled liquor. The sample of brewed liquor contained sake and wine and showed an alcohol concentration of 0–16%, and the fluoride ion concentration was 0.03–0.46 mg/L. The pH of the brewed liquor was 3.09–4.40 and was acidic. Sparkling liquor, classified as Beer or Sparkling drink, had a low alcohol content of 0–5.4% and a fluoride ion concentration of 0.05–0.10 mg/L. Mix liquor showed the lowest fluoride ion concentration among alcoholic species and was stable at a low value of 0.04–0.07 mg/L. Both Sparkling liquor and Mix liquor had low pH and acidity of pH 3.40–4.26 (Table 2).

### Risk assessment of fluoride daily intake from preference beverage

The CDI of adult calculated according to the method of USEPA (1992) was highest at tea at 0.0096 mg/kg/day and lowest at coffee at 0.0001 mg/kg/day (Table 3). Green tea showed 0.0093 mg/kg/day, and herbal tea and unique tea were 0.0004 mg/kg/day and 0.0007 mg/kg/day, respectively. The CDI of child showed the highest 0.0337 mg/kg/day for tea and the lowest 0.0005 mg/kg/day for coffee (Table 3). Green tea showed 0.0326 mg/kg/day, the same high value as tea, and herbal tea and unique tea were 0.0015 mg/kg/day and 0.0024 mg/kg/day respectively, which were the same as adult data. In all species, CDI did not exceed 0.06 mg/kg/day of RfD. The child CDI in tea was 0.0337 mg/kg/day, which was highest CDI in all species, was about half of RfD. The CDI in alcoholic beverages was generally much lower than tea and coffee, the highest at distilled liquor, 0.0003 mg/kg/day, followed by brewed liquor at 0.0002 mg/kg/day, and sparkling liquor and Mixed liquor at 0.0001 mg/kg/day. The CDI range of alcoholic beverages were very low, which were about 1/200-1/600 of RfD.

The HQ of tea and coffee in adult showed the same tendency as CDI, tea showing the highest HQ 0.1604 and Coffee showing the lowest HQ 0.0022 (Table 3). In the HQ of child, tea had the highest value of 0.5614 and coffee had the lowest value of 0.0077 (Table 3). All varieties had HQ $< 1$, and tea HQ was significantly higher than herbal tea, unique tea, coffee HQ ($p < 0.05$) (Fig. 1).

The HQ of alcoholic beverages showed the same tendency as CDI, with the highest being 0.0055 for distilled liquor, 0.0032 for brewed liquor, 0.0018 for sparkling liquor, and 0.0013 for mixed liquor (Table 4). All species of alcoholic beverages were HQ $< 1$, and there was no significant difference between each sample type ($p > 0.05$) (Fig. 2).

## Discussion

In this study, tea and green tea have a high fluoride ion concentration and the total amount of fluoride in tea leaves, and herbal tea, unique tea, and coffee have low fluoride content among the preference beverages (Table 1). This result was almost consistent with the previous study reports that the tea extract had a fluoride content of 0.70–6.01 mg/L and 0.57–3.72 mg/L of tea from Turkey, Sri Lanka, Kenya, and India.[27,28] Moreover, the total fluoride content of tea leaves measured by the microdiffusion method was generally consistent with the previous report of 35–289 mg/kg.[13,29] Tea and green tea are both made from the flowering plant *Camellia sinensis*, which is tea tree and an evergreen tree belonging to the theaceae family, cultivated in temperate areas with high annual rainfall. In the tea production and processing steps, tea is in the process of fermentation and green tea is non-fermented of *C. sinensis*. On the other hand, herbal tea and unique tea are mostly not made from *C. sinensis*. Among unique tea species, only No.20 oolong which was made by partially fermented *C. sinensis*, showed 0.6043 mg/L of high fluoride ion level in infusion, and the total fluoride content of tea leaves was 32.3667 mg/kg (Table 1).

In previous studies, the fluoride ion concentration of herbal tea infusions was as low as 0.02–0.04 mg/L, which was similar to our data 0.0702–0.1707 mg/L.[20,28] It has been reported that more than 96% of the total fluoride content of tea leaves is soluble in tea infusion.[30] The maximum infusion rate of fluoride in this experiment was 34.34% of coffee, but a considerable amount of fluoride may be released into the solution depending on the extraction time and conditions. This suggests that there is a risk of increasing the amount of fluoride ingested under the extraction conditions. In the case of a preference beverage with infusion, measurement of the total fluoride content of tea leaves as well as the fluoride ion concentration of the infusion is important for risk assessment.

Tea contains caffeine and vitamins, and has antioxidative and awakening effects, and blood cholesterol lowering effects. There are also reports of prevention of dental caries against *Streptococcus mutans* by tea catechins.[31,32] In recent years, domestic consumption of tea has also increased due to heightened health awareness. According to a questionnaire survey in Japan, It has been reported that average daily intake of drinking water is 1486 ml, of which 45% is the intake of water and tea, 35% is coffee and soft drinks and 17% is alcohol.[33] Although coffee is the next most consumed tea, both the fluoride concentration of the infusion and the total fluoride content of the coffee beans themselves are very small (Table 1).

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**Table 2** Fluoride Content in Alcoholic beverage.

| No. | Species | Category | Product name | Company | Country | Mean ± S.D. (mg/L) | pH | Alcohol degree (%) |
|---|---|---|---|---|---|---|---|---|
| 1 | Distilled liquor | Shochu | Kikunotsuyu | Kikunotsuyusyuzou, Okinawa,Japan | Japan | 0.0570 ± 0.012 | 8.95 | 12.0 |
| 2 | Distilled liquor | Shochu | Mugikon | Shinozaki, Fukuoka,Japan | Japan | 0.2014 ± 0.016 | 6.37 | 25.0 |
| 3 | Distilled liquor | Shochu | Iichiko Mugi 25 | Sanwa, Ooita, Japan | Japan | 0.0861 ± 0.015 | 8.12 | 25.0 |
| 4 | Distilled liquor | Shochu | Iichiko Mugi 20 | Sanwa, Ooita, Japan | Japan | 0.0772 ± 0.001 | 8.20 | 20.0 |
| 5 | Distilled liquor | Shochu | SugoMugi | Oenon Holdings, Tokyo,Japan | Japan | 0.1657 ± 0.004 | 8.08 | 12.0 |
| 6 | Distilled liquor | Shochu | Sugoimo | Oenon Holdings, Tokyo,Japan | Japan | 0.1491 ± 0.020 | 8.07 | 12.0 |
| 7 | Distilled liquor | Shochu | Kurokirishima | Kirishimasyuzou, Miyazaki,Japan | Japan | 0.0717 ± 0.014 | 4.79 | 25.0 |
| 8 | Distilled liquor | Shochu | Akaobisugi | Inouesyuzou, Miyazaki,Japan | Japan | 0.0085 ± 0.004 | 4.99 | 25.0 |
| 9 | Distilled liquor | Shochu | Kyogetsu | Suntory, Osaka,Japan | Japan | 0.3551 ± 0.015 | 2.92 | 16.0 |
| 10 | Distilled liquor | Shochu | Sousou | Ooumisyuzou, Kagoshima,Japan | Japan | 0.5537 ± 0.040 | 7.77 | 25.0 |
| 11 | Distilled liquor | Shochu | Akamaou | Inouesyuzou, Miyazaki,Japan | Japan | 0.0117 ± 0.001 | 4.73 | 25.0 |
| 12 | Distilled liquor | Whiskey | Glenfiddich | Suntory, Osaka,Japan | England | 0.0773 ± 0.009 | 4.38 | 40.0 |
| 13 | Distilled liquor | Whiskey | Custom | Suntory, Osaka,Japan | Japan | 0.1103 ± 0.032 | 4.72 | 33.0 |
| 14 | Distilled liquor | Rum | BACARDI Gold | BACARDI Japan, Tokyo,Japan | Puerto Rico | 0.0843 ± 0.004 | 3.91 | 40.0 |
| 15 | Distilled liquor | vodka | Stolichnaya | Stoli Group USA,NewYork,USA | Russia | 2.1491 ± 0.150 | 7.50 | 40.0 |
| 16 | Distilled liquor | Gin | BEEFEATER | Suntory, Osaka,Japan | England | 0.0814 ± 0.015 | 6.82 | 40.0 |
| 17 | Distilled liquor | Liquor | MALIBU | Suntory, Osaka,Japan | England | 0.0233 ± 0.005 | 5.01 | 21.0 |
| 18 | Distilled liquor | Liquor | ABSENTE55 | Marukai Corp, Osaka,Japan | France | 0.5731 ± 0.067 | 8.89 | 55.0 |
| 19 | Distilled liquor | Liquor | Midori | Suntory, Osaka,Japan | USA | 0.0604 ± 0.010 | 4.85 | 20.0 |
| 20 | Distilled liquor | Liquor | DISARONNO | Suntory, Osaka,Japan | Italy | 0.0559 ± 0.006 | 5.06 | 28.0 |
| 21 | Distilled liquor | Whiskey | TORYS Classic | Suntory, Osaka,Japan | Japan | 0.0590 ± 0.018 | 5.32 | 37.0 |
| 22 | Distilled liquor | Whiskey | JimbeamHONEY | Suntory, Osaka,Japan | USA | 0.2094 ± 0.004 | 4.81 | 35.0 |
| 23 | Distilled liquor | Brandy | CakeMagicBrandy | Suntory, Osaka,Japan | Japan | 0.0442 ± 0.008 | 4.82 | 39.0 |
| 24 | Brewed liquor | Sake | Amazake | Marukome, Nagano,Japan | Japan | 0.0330 ± 0.006 | 4.40 | 0.0 |
| 25 | Brewed liquor | Sake | Kome Amazake | Marukome, Nagano,Japan | Japan | 0.0320 ± 0.007 | 4.40 | 0.0 |
| 26 | Brewed liquor | Wine | Vintense Merlot | Stassen, Mittelrhein,Germany | Belgium | 0.0364 ± 0.003 | 3.63 | 0.0 |
| 27 | Brewed liquor | Wine | Fort du Mirail | Fort du Mirail, Bordeaux,French | France | 0.1877 ± 0.002 | 3.29 | 10.0 |
| 28 | Brewed liquor | Wine | Shoukousyu | Suntory, Osaka,Japan | China | 0.4554 ± 0.072 | 4.20 | 16.0 |
| 29 | Brewed liquor | Sake | Onigoroshi | Kiyosuzakura Brewery, Aichi,Japan | Japan | 0.1994 ± 0.005 | 4.33 | 13.0 |
| 30 | Brewed liquor | Sake | Kaminarisanndai | Kyouhimesyuzou, Kyoto,Japan | Japan | 0.0931 ± 0.003 | 4.25 | 15.0 |
| 31 | Brewed liquor | Sake | Umesyu | Choya umeshu, Osaka,Japan | Japan | 0.0273 ± 0.003 | 3.09 | 14.0 |
| 32 | Sparkling liquor | Sake | Szune | Ichinokura, Miyagi,Japan | Japan | 0.0827 ± 0.009 | 3.40 | 5.4 |
| 33 | Sparkling liquor | Beer | Mugi 50%off | Norlake Int.,Kanagawa, Japan | Korea | 0.0645 ± 0.026 | 4.05 | 4.0 |
| 34 | Sparkling liquor | Beer | Mugi soukai | Norlake Int.,Kanagawa, Japan | Korea | 0.0453 ± 0.001 | 4.25 | 5.0 |
| 35 | Sparkling liquor | Beer | Asahi Healtystyle | Asahi, Tokyo,Japan | Japan | 0.1044 ± 0.007 | 3.94 | 0.0 |
| 36 | Sparkling liquor | Beer | Ichibanshibori | KIRIN, Tokyo,Japan | Japan | 0.0781 ± 0.001 | 4.26 | 5.0 |
| 37 | Mixed liquor | Whiskey | TORYS Highball | Suntory, Osaka,Japan | Japan | 0.0421 ± 0.022 | 3.50 | 7.0 |
| 38 | Mixed liquor | Whiskey | TAKARAHighball | Takarasyuzou, Kyoto,Japan | Japan | 0.0540 ± 0.007 | 3.49 | 7.0 |
| 39 | Mixed liquor | Beer | Hoppy (Black) | Hoppy Beverage, Tokyo,Japan | Japan | 0.0720 ± 0.002 | 3.98 | 0.8 |
| 40 | Mixed liquor | Beer | Hoppy | Hoppy Beverage, Tokyo,Japan | Japan | 0.0547 ± 0.003 | 4.02 | 0.8 |
| 41 | Mixed liquor | Liquor | Cidre sweet | Kikusui, Nagano,Japan | Japan | 0.0401 ± 0.001 | 3.64 | 3.0 |

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**Table 3** Estimated fluoride daily Intake and hazard quotient from tea and coffee.

| Species | Fluoride | | | | CDI (mg/kg/day) | | | Hazard Quotient (HQ) | |
|---|---|---|---|---|---|---|---|---|---|
| | Mean (mg/L) | Range (mg/L) | Average daily intake (L/day) | Daily Fluoride intake (mg/day) | Adult | Child | RfD (mg/Kg/day) | Adult | Child |
| Tea | 2.7110 | 1.0570–6.6803 | 0.2485 | 0.6737 | 0.0096 | 0.0337 | 0.06 | 0.1604 | 0.5614 |
| Herbal tea | 0.1184 | 0.0702–0.1707 | 0.2485 | 0.0294 | 0.0004 | 0.0015 | 0.06 | 0.0070 | 0.0245 |
| Green tea | 2.6207 | 0.2557–4.0870 | 0.2485 | 0.6513 | 0.0093 | 0.0326 | 0.06 | 0.1551 | 0.5427 |
| Unique tea | 0.1894 | 0.0266–0.6043 | 0.2485 | 0.0471 | 0.0007 | 0.0024 | 0.06 | 0.0112 | 0.0392 |
| Coffee | 0.0658 | 0.0277–0.1463 | 0.1400 | 0.0092 | 0.0001 | 0.0005 | 0.06 | 0.0022 | 0.0077 |

Therefore, it is considered that the intake of fluoride is small even if an amount close to the total fluoride is eluted in the infusion solution depending on the infusion conditions. Alcoholic beverages are considered to have a low fluoride intake because the fluoride ion concentration is low overall, except for some samples (Table 2).

In this study, we examined whether a product with a high fluoride content could be inferred from the features such as the type and origin of the product. However, the differences among products were large, and the amount of fluoride was different even in the same production area and origin (Table 1). The difference in fluoride concentration in the same production area may be due to the fact that the fluoride concentration of tea leaves is affected by the soil fluoride concentration of the growing environment.[10] A rough estimation of the amount of fluoride is possible based on the type of material and tea, such as whether *C. sinensis* is used or not. However, the measurement of the product itself by the microdiffusion method is considered to be the most accurate and effective for creating basic data and risk assessment of fluoride intake. In the case of children, it has been reported that the amount of fluoride uptake in the body is higher than that in adults, and strict monitoring of the amount of fluoride ingested is necessary for application. Children under the age of 8 are susceptible to dental fluorosis during the dental calcification process.[8] On the other hand, in the case of adults, the amount of fluoride accumulated in the body by metabolism is small, and the upper limit of the tolerable amount is increased by increasing body weight, so the safety level with respect to the amount of fluoride taken from preference beverages is higher than that of children. Non-alcoholic beverages are often drunk by children, and it is necessary to accumulate basic data on tea and coffee consumed by children.

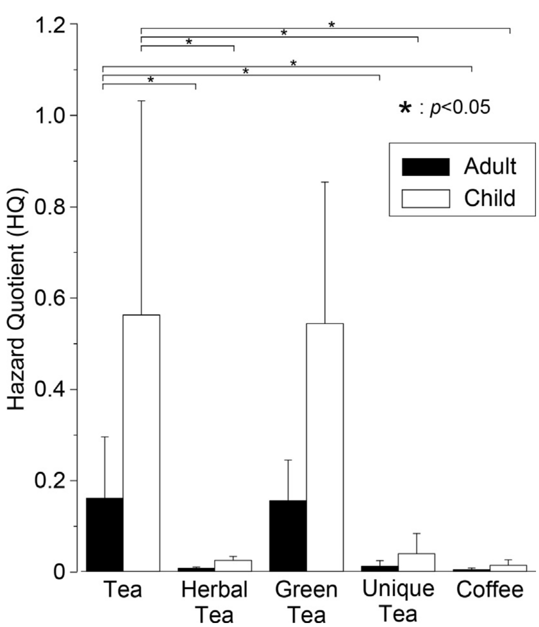


**Figure 1** Hazard quotient of each tea spices and coffee beverages. All values are presented as mean ± SD of replicates per following; 5 tea, 7 herbal tea, 5 green tea, 8 unique tea and 6 coffee. Black bars show Adult HQ. White bar shows Child HQ. The statistical analysis among the sample was performed by one-way analysis of variance (ANOVA), and differences were considered significant at $p < 0.05$. The Bonferroni test was used for post-hoc comparisons when significance was determined by analysis of variance ($p < 0.05$).

To monitor the amount of fluoride intake in humans, there is a method of directly calculate the fluoride intake from food and drink, indirectly blood fluoride concentration, bone fluoride concentration, urine fluoride concentration.[34] Since indirect methods are metabolically affected, direct methods are desirable for accurate estimation of total fluoride intake. Therefore, in this study, the amount of fluoride in preference beverages was measured by a direct method, and CDI and HQ were calculated (Tables 3 and 4). Our results showed the CDI of all tea species and coffee is 0.0001–0.0096 mg/kg/day for adult and 0.0005–0.0337 mg/kg/day for child, which were significantly lower than RfD (Table 3). Moreover, HQ of all tea species and coffee was 0.0022–0.1604 for adult and 0.0077–0.5614 for child, which was lower than 1 (Table 3). It was confirmed that the intake of fluoride from four types of tea and coffee was within the safe range for both adults and children. Alcohol showed that distilled liquor tended to have a higher fluoride content than other liquors, but there was no significant difference (Fig. 2). It was considered that the large amount of fluoride in distilled liquor is because concentration of fluoride occurred during the alcoholic distillation process during production. Alcohol showed smaller CDI and HQ values than tea and coffee. Alcohol consumption is limited to adults with a large body weight. There is almost no risk of fluoride overdose in the average daily intake range (Table 4). However, alcoholic beverages showed acidic pH in 33 out of 41 items, and in 21 items were below the critical enamel pH of 4.5 (Table 2). These results

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**Table 4** Estimated fluoride daily Intake and hazard quotient from alcohol beverages.

| Species | Average (mg/L) | Range (mg/L) | Average daily intake (L/day) | Daily Fluoride intake (mg/day) | CDI (mg/Kg/day) | RfD (mg/Kg/day) | Hazard Quotient (HQ) |
|---|---|---|---|---|---|---|---|
| Distilled liquor | 0.2289 | 0.0085–2.1491 | 0.1002 | 0.0229 | 0.0003 | 0.06 | 0.0055 |
| Brewed liquor | 0.1330 | 0.0320–0.4554 | 0.1002 | 0.0133 | 0.0002 | 0.06 | 0.0032 |
| Sparkling liquor | 0.0750 | 0.0453–0.1044 | 0.1002 | 0.0075 | 0.0001 | 0.06 | 0.0018 |
| Mixed liquor | 0.0526 | 0.0401–0.0720 | 0.1002 | 0.0053 | 0.0001 | 0.06 | 0.0013 |

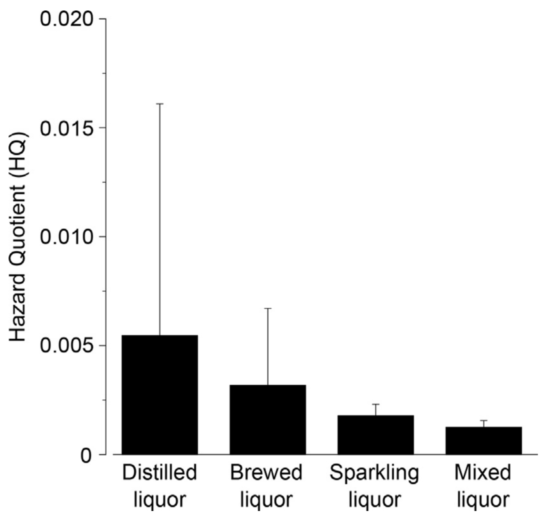


**Figure 2** Hazard quotient of each alcoholic beverages. All values are presented as mean ± SD of replicates per following; 23 distilled liquor, 8 brewed liquor, 5 Sparkling liquor, 5 Mixed liquor.

suggest that there is a risk of direct demineralization of the tooth due to frequent intake alcoholic beverages. According to data from a survey of 189 countries around the world, the world's annual alcohol consumption from 1990 to 2017 increased by approximately 70% from 20.99 billion liters to 35.676 billion liters. The number of drinkers increased from 45% to 47%. This trend will continue, and by 2030, the number of drinkers is expected to rise to 50% and lifetime non-drinkers to 40%.[35] Increasing trends in the pH and alcohol consumption of alcoholic beverages may require attention in terms of risk of erosion.

When measuring the total fluoride content of the direct method, the difference between products is large as described above. Some samples, such as Tea No.4 and Green Tea No.15, show extremely high fluoride ion concentrations than other products (Table 1). In the Tea No.4 sample, the CDI is 0.0237 mg/kg/day for adults and 0.0830 mg/kg/day for children, and these RfD exceeds 0.06 mg/kg/day in children. The HQ was 0.3953 for adults and 1.3834 for children, and the HQ for children exceeded 1 and was at risk. The green tea No. 15 sample had an adult CDI of 0.0145 and a child of 0.0508, and it exceeded 0.05 even if the child did not exceed the RfD of 0.06. HQ is 0.2418 for adults and 0.8463 for children, and HQ for children is close to 1. Previous studies have reported that the green tea CDI was higher than the other types, with CDI exceeding the limits in 5 out of 10 samples.[20] Tea infusion from Turkey, Sri Lanka, Kenya and India also have high fluoride concentrations and some products exceed CDI regulations.[27,28] In this study, HQ was estimated using the average value for each tea and alcoholic species. However, because individual differences among products are large, it is desirable to examine each product in order to conduct risk assessment more strictly.

The value of the RfD 0.06 mg/kg/day for fluoride, 0.05 mg/kg/day is the intake of fluoride via beverage, and 0.01 mg/kg/day is the dietary intake. This is a value calculated by taking the fluoride intake from water fluoridation into account, but in Japan, fluoride systemic application by water fluoridation is not performed. Therefore, it is considered that the upper limit of the amount of fluoride ingested from meals and preference beverages has a margin as compared to other countries. However, the intake of fluoride from food is not taken into account in this study, so the health risks due to the amount of fluoride taken from preference beverages cannot be completely eliminated. The HQ values calculated from the average DFI of preference beverages were all within the safe range. However, some products made from *C. sinensis*, such as tea and green tea, had 1.66 mg/day as DFI at maximum, and the Child's HQ exceeded 1. These results suggest that habitual consumption of some products requires risk management of dental fluorosis. There is a need to collect more fluoride intake data from preference beverages for proper caries prevention effects and risk management for dental fluorosis.

## Declaration of Competing Interest

The authors have no conflicts of interest relevant to this article.

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