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Total Diet Study 2014–2016: Assessment of dietary exposure to fluoride in adults and children in Ireland

TOTAL DIET STUDY 2014–2016: ASSESSMENT OF DIETARY EXPOSURE TO FLUORIDE IN ADULTS AND CHILDREN IN IRELAND

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EXECUTIVE SUMMARY

This report presents the findings of a total diet study (TDS) on fluoride intake carried out in Ireland during 2014–2016 by the Food Safety Authority of Ireland (FSAI). The objective was to evaluate possible risk, if any, to the health of children aged 1–12 years and adults resident in Ireland arising from exposure to fluoride from foods and beverages, including fluoridated water.

The estimates of fluoride intake in this study are for foods and beverages only and do not include possible fluoride contributions from non-dietary sources, particularly through swallowing of fluoridated toothpaste, which can make a significant contribution in young children, but much less so in older children and adults.

The most commonly consumed foods in Ireland, based on nationally representative food consumption data, were analysed for fluoride, and dietary exposure to fluoride was then estimated using the food consumption data and the level of fluoride present in each food. These estimates were based on fluoride content of food and water in regions with fluoridated public water supplies. The food consumption data used in the study were derived from the National Adult Nutrition Survey (NANS) for age 18 years and over (n=1,500) during 2008–2010, the National Children's Food Survey (NCFS) for age 5–12 years (n=594) during 2003–2004, and the National Pre-school Nutrition Survey (NPNS) for age 1–4 years (n=500) during 2010–2011.

Black tea, which can contain naturally high levels of fluoride, was found to contain by far the highest fluoride concentrations determined in this study, ranging from 1.7 mg/kg to 8.3 mg/kg in tea infusion. The exposure estimates derived here are based on the mean fluoride concentration in 10 individual brands of black tea (4.8 mg/kg in tea infusion), to reflect long-term exposure. With the exception of fish and fishery products (ranging from 0.04 to 5.8 mg/kg) and one nut sample (nuts and seeds ranging from 0.05 to 1.3 mg/kg), all other foods showed fluoride concentrations below 1 mg/kg. The average water fluoride concentration for fluoridated public water supplies in Ireland was 0.65 mg/L.

Mean fluoride exposures in preschool children (1–4 years of age) and children (5–12 years of age) (0.023 and 0.017 mg/kg bw/day, respectively) were much lower than in adults (0.040 mg/kg bw/day), largely due to the much higher consumption of black tea in adults. Adults are predominantly exposed to fluoride via consumption of black tea, which constitutes 76% of their total fluoride exposure, with tap water contributing 12%. Tap water was the main contributor to fluoride intake in children (49% in preschool children aged 1–4 years, 33% in children aged 5–12 years), while tea was a significant contributor (29%) in children aged 5–12 years.

The mean intakes of fluoride from foods and beverages were below the adequate intake (AI) of 0.05 mg/kg bw/day established by the European Food Safety Authority (EFSA) for caries protection, for all age groups, and particularly for children.

The exposure estimates obtained in the study were compared with tolerable upper intake levels (ULs) for fluoride established by the EFSA of 0.10 mg/kg bw/day for children aged 1–8 years related to moderate dental fluorosis and 0.12 mg/kg bw/day for children aged ≥9 years and adults, including pregnant and lactating women, related to adverse effects on bone.

For children aged 1–8 years, the probability of exceeding the UL (0.10 mg/kg bw/day) through fluoride intake from foods and beverages is very low, with higher percentile intakes of fluoride at P95 and P97.5 of 0.047 and 0.057 mg/kg bw/day, respectively, in children aged 1–4 years and 0.038 and 0.046 mg/kg bw/day, respectively, in children aged 5–8 years. This indicates that there is no appreciable risk of moderate dental fluorosis in children aged 1–8 years arising from fluoride intake from foods and beverages. While fluoride intakes from non-dietary sources were not assessed in the study, even where published estimates of average fluoride intake from toothpaste in Ireland were included (0.01–0.02 mg/kg bw/day for 1.5 to 3.5-year-old children), this would not result in total fluoride intake exceeding the UL, even in the highest consumers of fluoride from foods and beverages. However, it is recognised that misuse of toothpaste may result in higher intakes of fluoride in a small proportion of young children. The low rates of moderate dental fluorosis in permanent dentition in children in regions with fluoridated water in Ireland indicate that total fluoride intake is not excessive at the critical period of development of permanent teeth in early childhood (about age 15–30 months).

For children aged 9–12 years, the probability of exceeding the UL (0.12 mg/kg bw/day) through fluoride intake from foods and beverages is very low, with P95 and P97.5 fluoride intake of 0.042 and 0.056 mg/kg bw/day, respectively. This indicates that there is no appreciable risk of adverse effects on bone in children arising from fluoride intake from foods and beverages.

For adults, the probability of exceeding the UL (0.12 mg/kg bw/day) is low, with P95 and P97.5 fluoride intake of 0.094 and 0.111 mg/kg bw/day, respectively. Fluoride intake exceeded the UL in a small proportion (1.6%) of adults. This indicates that the risk of adverse effects on bone in the adult population arising from fluoride intake from foods and beverages is low.

Although not estimated in this study, it is considered that the probability of teenagers exceeding the UL for fluoride is very low, due to the much lower consumption of black tea in teenagers than in adults.

Based on the results of this study, the FSAI Scientific Committee concluded that there is currently no scientific basis for concerns about the safety of children and adults in Ireland from exposure to fluoride from foods and beverages.

ABBREVIATIONS

ADI	acceptable daily intake
AI	adequate intake
Boxplot	Boxplot is a summary plot of a dataset, graphically depicting the median, quartiles, and extreme values. The box represents the interquartile (IQ) range which contains the middle 50% of the data. The whiskers are lines that extend from the upper and lower edge of the box to the highest and lowest values which are no greater than 1.5 times the IQ range. Outliers are cases with values between 1.5 and 3 times the IQ range, i.e. beyond the whiskers. Extremes are cases with values more than three times the IQ range.

bw	body weight
CD	conductivity detector
DAFM	Department of Agriculture, Food and the Marine
EC	European Community
EFSA	European Food Safety Authority
EPA	Environmental Protection Agency
EU	European Union
FAO	Food and Agriculture Organization (FAO)
FSAI	Food Safety Authority of Ireland
HBGV	health-based guidance value
HSE	Health Service Executive
IC	ion chromatography
ISE	ion selective electrode
INFID	Irish National Food Ingredients Database (INFID)
IUNA	Irish Universities Nutrition Alliance (IUNA)

ABBREVIATIONS CONTINUED

kg	kilogram
LB	lowerbound (<LOD=0)
LOD	limit of detection
LOQ	limit of quantitation
mg	milligram
MI	Marine Institute
min	minute
mL	millilitre
µg	microgram
n	number of samples
n/a	not applicable
NANS	National Adult Nutrition Survey
NCFS	National Children's Food Survey
NPNS	National Pre-school Nutrition Survey
P95	95th percentile of a distribution
P97.5	97.5th percentile of a distribution
RfD	reference dose
SCHER	Scientific Committee on Health and Environmental Risks
sec	second
SFPA	Sea-Fisheries Protection Authority
SPSS	Statistical Package for the Social Sciences (statistics software package)
TDI	tolerable daily intake
TDS	total diet study
UB	upperbound (<LOD=LOD)
UL	tolerable upper intake level/upper limit
WHO	World Health Organization

1   INTRODUCTION

The Food Safety Authority of Ireland (FSAI) has a statutory responsibility to ensure the safety of food consumed, distributed, produced and sold on the Irish market. In this respect, the FSAI coordinates the collation of food safety surveillance information from laboratories operated by its official agencies, the Health Service Executive (HSE), the Department of Agriculture, Food and the Marine (DAFM), the Sea-Fisheries Protection Authority (SFPA), the Marine Institute (MI), and the local authorities. The FSAI also conducts targeted food safety surveillance in areas where potential food safety issues have been identified and/or on food contaminants for which there are currently no testing facilities in Ireland.

This report provides the results of a total diet study (TDS) on fluoride carried out during 2014–2016. It describes the health hazards, health-based guidance values (HBGVs) as established by the European Food Safety Authority (EFSA) and other international bodies, and evaluates exposure of the population resident in Ireland (adults >18 years of age, children 5–12 years of age and preschool children 1–4 years of age) via diet and fluoridated drinking water. Finally, it concludes on possible risk, if any, to the health of the adult and child populations resident in Ireland arising from exposure to fluoride from food and fluoridated water supplies.

Fluorine accounts for 0.06–0.09% of the Earth's crust and exists in the form of fluorides in a number of minerals, such as fluorspar, cryolite and fluorapatite. Most of the fluoride in rocks and soils is firmly bound and not biologically available. In some parts of the world, there can be significant environmental pollution coming from unprotected mines, industrial emissions, coal burning, fertilisers and pesticides (WHO, 1994).

Fluorides are ubiquitous in air, water and the lithosphere, and all vegetation contains some fluoride, which is absorbed from soil and water (WHO, 2017; SCHER, 2011). The availability of fluoride from soil depends on the solubility of the compound, the acidity of the soil, and the presence of water (WHO, 2017).

Human exposure to fluoride can vary considerably, depending on the levels of fluoride found in drinking water and on individual dietary and oral hygiene habits and practices (SCHER, 2011; WHO, 2017). Virtually all foodstuffs contain at least some traces of fluorine, and some foods are known to contain relatively high fluoride concentrations; examples include tea (WHO, 2017) and seafood (WHO, 2002). In addition, the fluoride content of foods and beverages is affected by its concentration in water used during preparation and processing (EFSA, 2005).

In some countries, fluoride may be added to table salt or drinking water in order to provide protection against dental caries (tooth decay) (EFSA, 2005). In Ireland, in accordance with the Health (Fluoridation of Water Supplies) Act, 1960 (Government of Ireland, 1960) and S.I. No. 42/2007 – Fluoridation of Water Supplies Regulations 2007 (Government of Ireland, 2007), public water supplies are required to be fluoridated. Since 2007, the level of fluoride in public water supplies in the Republic of Ireland has been set at between 0.6 and 0.8 mg/L.

Total diet studies

A TDS is considered a good complement to existing food monitoring or surveillance programmes to estimate population dietary exposure to beneficial and harmful chemical substances across the entire diet. The World Health Organization (WHO), the lead United Nations agency for health, supports the undertaking of TDSs as one of the most cost-effective means for ensuring that people are not exposed to unsafe levels of toxic chemicals through food (WHO, 2005). At the beginning of 2010, the EFSA formed a working group of experts on TDSs, aimed at reviewing TDSs worldwide with a particular emphasis on activities in Europe; subsequently, the EFSA developed a guidance document for a harmonised approach to TDSs in collaboration with the WHO (WHO/FAO/EFSA, 2011).

A comparison of the actual dietary intake of chemicals present in food, estimated via a TDS, with their corresponding HBGVs (such as the acceptable daily intake (ADI), tolerable daily intake (TDI) or tolerable upper intake level (UL)) gives a realistic estimate of exposure for risk assessment purposes. TDS results can be indicators of contamination of food from the environment and can be used to assess the effectiveness of specific risk management measures to control the levels of such chemicals in food. A TDS may also be used to provide an estimate of intake of food additives present in the diet, such as sweeteners or preservatives, or of intake of key nutrients such as vitamins.

The overall aim of a TDS is to provide an estimate of exposure of a given population to contaminants or other food chemicals of interest. It aims to estimate exposure of the general population and does not represent unusual exposure scenarios, e.g. above-average exposure to chemicals due to environmental contamination in specific geographical regions.

In carrying out a TDS at national level, the most commonly consumed foods in the country are selected, based on food consumption data, and the respective predominant food preparation methods are determined. The selected foods representative of the typical diet consumed by the population over a given period of time are collected from defined surveillance regions and consist of a number of subsamples per sample (e.g. a bread sample could contain 10 different subsamples of the particular type of bread selected). The food samples are prepared following the most commonly used kitchen preparation practices, to represent foods as they would be consumed, and are subsequently analysed for the chemicals of interest. The chemical concentration data are then combined with consumption data to calculate the exposure estimates for the population to the chemicals from the selected foods. A TDS explicitly takes into account the kitchen preparation of foods to assess the levels of chemicals in foods as consumed, as these may change during preparation and cooking.

2   METHODOLOGY

Exposure estimates for fluoride were calculated by combining food consumption data with fluoride concentration data for representative foods as prepared for consumption, e.g. boiled, grilled or baked. The concentration of fluoride in fluoridated water is known to fluctuate; therefore, an average fluoride concentration for Irish tap water was derived from an extensive external dataset. Fluoride monitoring data on the country-wide public water supply, in accordance with S.I. No. 42/2007 – Fluoridation of Water Supplies Regulations 2007, for the years 2015–2016 were obtained from the HSE. These data were subsequently used to derive the average fluoride concentration in tap water from public supplies in Ireland (i.e. 0.65 mg/L) and this value was used in the exposure assessment.

Planning and coordination of this project as well as sampling of the foods of interest was undertaken by FSAI staff; food preparation of the samples was undertaken under contract by Fera Science Ltd, UK and analysed at the Fluoride Laboratory, Centre for Oral Health Research, School of Dental Sciences, Newcastle University, UK.

The general approach for selection of foods and food preparation methods was developed as part of the first TDS (FSAI, 2011).

2.1   Food consumption data

Population groups included in this study were children aged 1–12 years and adults aged 18 years and over, but did not include infants aged under 1 year or teenagers aged 13–17 years. The food consumption data used in this TDS were derived from the National Adult Nutrition Survey (NANS) (IUNA, 2011), the National Children's Food Survey (NCFS) (IUNA, 2005) and the National Pre-school Nutrition Survey (NPNS) (IUNA, 2012b). These surveys investigated habitual food and beverage consumption in a representative sample (n=1,500) of adults aged 18 years and over in the Republic of Ireland during 2008–2010 (four-day dietary record), in 594 children aged 5–12 years in the Republic of Ireland during 2003–2004 (seven-day dietary record) and in 500 preschool children aged 1–4 years in the Republic of Ireland during 2010–2011 (four-day dietary record). The extensive electronic databases that were compiled from these surveys were used to obtain information on food consumption and food preparation habits.

2.2   Selection of foods for analysis

The choice of foods for this TDS was based on the list developed for the first TDS (FSAI, 2011), but was expanded and modified, based on information available from more recent food consumption surveys, in particular brand information. In addition, sample choice was influenced by the analyte under investigation, fluoride. The complete food list is shown in Table 1.

2.3   Shopping list/food sampling

For each individual food group, typically 10 samples were purchased and combined into a composite sample (see Appendix I), with the exception of tea (see Section 2.6), where 10 samples were purchased and analysed individually. The selection of brands was based on interrogation of the brand information in the food consumption databases. The quantity of each foodstuff purchased was as required for the analysis of fluoride in that foodstuff. This latter information was obtained from the laboratories contracted to carry out the food preparation and the analysis. Sampling of the foods was conducted by the FSAI in spring of 2014 and a total of 216 samples (comprising 1,861 subsamples) were sent for preparation and analysis to the contract laboratory. Food samples were mainly purchased from major retailers located in Dublin. Fluoridated tap water used in the preparation (e.g. boiling) of selected food samples was sourced from the public water supply in Dublin and supplied to the laboratory undertaking the food preparation.

Table 1: Foods analysed for fluoride

Food category	Food groups
CEREALS AND CEREAL PRODUCTS	White flour; Wholemeal flour; Cornflour; White bread/rolls; Granary/wholegrain breads; Brown bread and rolls; Plain frozen pastry; Plain biscuits; Chocolate biscuits; Other biscuits; Cakes; Other cakes, buns and pastries; Noodles (unflavoured); Pasta (cooked); Pasta (dry); Rice (cooked); Rice (dry); Couscous; Cornflakes; Bran flakes; Wheat-type cereals; Muesli; Oat flakes; Rice-type cereals; Rice cakes
DAIRY, DAIRY PRODUCTS AND DAIRY ANALOGUES	Whole milk; Low-fat, skimmed and fortified milk; Cream; Cheese (hard); Cheese (continental style); Cheese (soft and semi-soft); Yogurts; Fromage frais; Custard; Butter; Dairy and non-dairy spreads; Vanilla ice cream; Other ice creams; Soya milk; Coconut milk/cream; Other non-dairy milk (e.g. rice milk)
EGGS	Eggs (fried)
MEAT AND MEAT PRODUCTS	Pork; Ham; Pork sausage; Bacon rashers; Beef; Beef mince; Beefburger; Chicken; Turkey; Lamb; Offal (kidney); Offal (liver); Pudding, black; Pudding, white; Liver pâté (pork/chicken/duck)
FISH AND FISH PRODUCTS	Cod; White fish (other than cod); Oily fish other than salmon; Salmon; Tinned salmon; Tinned tuna; Tinned fish (excluding salmon and tuna); Mussels; Prawns; Crab; Fish fingers
VEGETABLES AND VEGETABLE PRODUCTS	Potatoes without skin (boiled); Potatoes without skin (raw); Potatoes (baby) with skin (microwaved); Chips; Garlic; Onion; Tomatoes; Canned tomatoes; Tomatoes (canned/concentrate); Peppers; Cucumber; Mushrooms; Canned sweetcorn; Carrots (boiled); Carrots (raw); Celery; Peas (boiled); Peas (raw); Canned peas; Green beans (boiled); Green beans (raw); Baked beans; Legumes (excluding peas) (boiled); Legumes (excluding peas) (raw); Canned legumes (excluding peas); Cabbage (raw); Cabbage (boiled); Broccoli (boiled); Broccoli (raw); Cauliflower (boiled); Cauliflower (raw); Root vegetables (excluding carrots) (boiled); Root vegetables (excluding carrots) (raw); Stir-fry vegetables; Lettuce; Spinach and other leaves; Avocado; Pumpkin/Squash; Seaweed (Irish moss, Kombu, Nori) (dry); Aubergine/Courgette; Oriental vegetables (water chestnuts/bamboo shoots)
FRUIT AND FRUIT PRODUCTS	Apples; Citrus fruit; Bananas; Grapes; Pears; Peaches and nectarines; Kiwis; Plums; Berries; Other fruit; Canned fruit; Melons; Smoothies; Olives; Dried raisins/sultanas/currants; Apricots, prunes, and other dried fruit; Figs and dates; Glacé cherries and mixed peel
HERBS AND SPICES	Herbs; Spices
NUTS AND SEEDS	Peanuts (roasted salted); Seeds; Tree nuts
FATS AND OILS (EXCLUDING DAIRY)	Animal fat; Olive oil; Vegetable oil
SOUPS, SAUCES AND CONDIMENTS	Soup (fresh) (Tetra Paks and tubs); Soups (canned); Soups (dried packet); Stock cubes, concentrated savoury spreads and essences; Tomato sauce; Mayonnaise; Gravy; Cook-in sauces (other); Cook-in sauces (tomato-based); Dry sauces; Soy sauce; Mustard; Other sauces and condiments; Salad dressings; Pickled vegetables; Pickles, chutneys and relish
SAVOURY SNACKS	Crisps; Other savoury snacks
SUGAR AND PRESERVES	Sugar; Marmalade; Jam; Honey; Sugar syrups; Jelly

Food category	Food groups
CONFECTIONERY	Chocolate confectionery; Non-chocolate confectionery; Plain chocolate; Cocoa powder; Drinking chocolate powder; Chocolate/nut spreads
BEVERAGES	Lager; Stout; Wine (white/red/rosé); Spirits; Cider; Carbonated soft drinks; Soft drinks (wine substitutes); Sports drinks; Squashes; Apple juice (fresh); Orange juice (fresh); Apple juice (from concentrate); Orange juice (from concentrate); Fruit nectars; Cranberry juice; Multivitamin juices; Other fruit juices; Tea; Herbal tea; Instant coffee; Percolated coffee; Bottled water; Tap water*
OTHER	Vinegar; Water used for food preparation (e.g. boiling)
FOOD FOR INFANTS AND YOUNG CHILDREN	Infant formula (powder); Infant formula (dairy alternatives) (powder); Growing-up milk (powder); Infant-specific beverages; Infant food (meat-based); Infant food (fish-based); Infant food (fruit-based); Infant food (vegetable-based); Infant food (cereal-based); Infant food (dairy-based); Baby snacks
COMPOSITE FOOD	Pizza; Rice pudding

* Calculated value, derived from 2015–2016 HSE monitoring data on fluoride in drinking water from public supplies in Ireland

2.4   Food preparation

Where appropriate, foods were prepared ready for consumption before analysis by the laboratory. Information on the most commonly used kitchen preparation practices was obtained as part of the analysis of the food consumption database for development of the food list. A description of the methods of food preparation employed can be found in Appendix I.

Uptake of fluoridated water used in the preparation of food was of particular interest in this study. Therefore, foods known to absorb water during cooking (e.g. rice, pasta, noodles, and certain vegetables) were boiled in fluoridated water.

For tea and percolated coffee, deionised water was used in the preparation in order to determine the fluoride concentration in the brewed beverage attributable to the tea leaves or roasted coffee beans, respectively. Contribution to exposure from fluoridated tap water in the preparation of these beverages was accounted for separately (see Section 2.6).

2.5   Fluoride analysis

Analysis of fluoride in complex samples, such as the food and beverage samples in the TDS, posed a number of challenges related to the different forms (inorganic, organic, complexed) in which fluoride may occur and to the possibility of gains/losses due to contamination from extraneous sources of fluoride, incomplete release of fluoride from the sample matrix, and volatility of fluoride at high temperatures (Campbell, 1987). Methods for extraction of fluoride from food and beverage samples include ashing, acid digestion, fusion with alkali, and microdiffusion, while methods for measurement of fluoride in sample extracts include colorimetry, ion chromatography, gas chromatography, and potentiometry – ion selective electrode (ISE) (Ponikvar *et al.*, 2007; Rocha *et al.*, 2013). However, by far the most commonly used sample extraction procedures are either ashing/alkali fusion or microdiffusion, and fluoride determination with an ISE (Taves, 1983; Malde *et al.*, 2001; Martínez-Mier *et al.*, 2011).

In preliminary investigations for this study, the two sample extraction methods of ashing/alkali fusion and microdiffusion, both coupled with determination by ISE, were compared using different food samples. However, the limit of detection (LOD) for the ashing/alkali fusion method, at between 2 and 10 mg/kg, was unsuitable for most samples in the study, which contained fluoride at levels of <1 mg/kg. In contrast, the microdiffusion method had an adequate limit of determination, at 0.005 mg/kg, rendering it the most suitable method for the study.

Following selection of the microdiffusion method for the study, the ongoing performance of the method was evaluated through recovery studies and repeat analyses. The 216 samples in the TDS were distributed across 18 categories of foods and beverages, and a total of 71 recovery samples, comprising 1–10 samples per category depending on the size and variation in sample type within the category, were analysed. Mean recovery of fluoride ranged from between 80% and 119% across most of the food categories; in the case of a number of samples from two of the food categories, i.e. 'Soups, sauces and condiments' and 'Beverages', very low recoveries were determined and particular steps had to be taken in dealing with the affected samples (n=8) in the exposure study (see Section 2.6). As part of the quality control of the sample analyses, repeat analyses were carried out on 43 samples, representative of the various sample categories, and, for these samples, the mean value obtained from both analyses was used for the exposure study.

The sample analyses were carried out during 2015–2016. Details on methods of analysis and analytical sensitivity are provided in Appendix II, and a summary of analytical results is listed in Appendix III.

2.6   Exposure assessment

Fluoride exposure estimates were calculated by combining food consumption data with the fluoride concentration data generated in this study and, for particular samples, with the average water fluoride concentration determined from monitoring data provided by the HSE.

The food consumption databases are structured using the McCance and Widdowson Food Code System (Holland *et al.,* 1988, 1989, 1991, 1992a, 1992b, 1993; Chan *et al.,* 1994, 1995, 1996; FSA 2002; MAFF 1998) and foods were classified, based on this system. To enable the exposure assessment undertaken in this TDS, the food consumption databases were restructured and recoded, thereby splitting composite foods, e.g. dishes, into ingredients and regrouping them accordingly (see Appendix IV for more detailed information).

Since uptake of fluoridated water used in the preparation of food was of particular interest in this study, water absorbed during cooking was accounted for either by:

- Cooking of the food (e.g. broccoli, rice, pasta) in fluoridated water, or

- Including the food and water portion separately (e.g. a paella recipe might provide ingredient proportions for raw rice and water used in the recipe). Water loss during cooking was accounted for through the application of weight loss factors. The average water fluoride concentration (0.65 mg/L) determined in the dataset provided by the HSE was used in these cases.

Consequently, fluoride contribution from tap water is divided across several food categories, such as tea and percolated coffee prepared with tap water, infant formula prepared with tap water, cereals (rice, noodles, pasta) boiled in tap water, and tap water in composite foods not further disaggregated (e.g. ready-to-eat soups, sauces, etc.). The food group 'tap water' therefore comprises tap water consumed as such, and tap water used in the preparation of composite beverages (e.g. squashes) and dishes (e.g. beef stew), where the composite beverage or dish was disaggregated into ingredients.The databases were recategorised into 216 food groups in accordance with the food list determined for this TDS. The relevant food groups were then matched with the associated fluoride concentration levels determined in the food samples contained in the food list, with the exception of tap water. Here, the average water fluoride concentration was determined from a dataset provided by the HSE, covering all fluoridated water supplies in Ireland for the years 2015–2016. Concentration data for those food groups not on the food list were extrapolated from comparable foods for which data were available, e.g. the concentrations obtained in prawns were used to estimate fluoride exposure from comparable shellfish, such as lobster and shrimp, for which no fluoride concentration data were available.

The recoded food consumption data and fluoride concentration data were combined using the probabilistic web-based 'Creme' software (Creme Food® 3.6). For the purpose of this TDS, a semi-probabilistic approach was used, i.e. the fluoride concentration determined in each food group was

combined with population food intake distribution data. Specifically, exposure was calculated, based on individual food consumption, averaged over the total consumption survey period. Exposure to fluoride was calculated by multiplying the fluoride concentration for each food by the respective consumption amount in grams per day and per kilogram of body weight (kg bw) separately for each individual in the database. Exposure from each food was subsequently added to derive an individual total exposure per day. Finally, these exposure estimates were averaged over the number of consumption survey days and normalised for individual bw, resulting in an individual average exposure per day per kg bw for the survey period (see Appendix V for the exposure function used). This was done for all individuals in the consumption surveys, resulting in distributions of individual average exposure per survey. Based on these distributions, the mean, 95th percentile (P95), and 97.5th percentile (P97.5) exposures were calculated per survey for the total population.

Results are expressed as average intake and above-average intake (P95, P97.5) of fluoride, together with average intake per kg bw and above-average intake per kg bw for the total population to reflect average and above-average consumers. Per kg bw calculations take into account the distribution of weights of individuals within the survey population and are included for direct comparison with the reference values, which are expressed on a per kg bw basis.

Fluoride concentration data determined for each food group were combined with the matching food group in the food consumption survey, with the exception of:

a) **Tea:** Ten individual black tea samples were analysed as part of the TDS to capture the fluctuation in fluoride concentration known to occur naturally in this food group. For exposure calculation purposes, the average concentration of the 10 individual samples was used. As the samples were prepared using deionised water, for exposure purposes, the average fluoride concentration found in tap water (2015–2016 HSE data) was combined with the average tea concentration to reflect exposure to fluoride from tea as consumed (i.e. tea prepared with tap water).

b) **Herbal tea:** As the sample was prepared using deionised water, for exposure purposes, the average fluoride concentration found in tap water (2015–2016 HSE data) was combined with the fluoride concentration determined in the tea sample to reflect exposure to fluoride from herbal tea as consumed (i.e. tea prepared with tap water).

c) **Percolated coffee:** The measured fluoride concentration for this food group did not conform with the required quality criteria (i.e. analyte recovery in the sample was insufficient); therefore, only the average fluoride concentration found in the tap water (2015–2016 HSE data) component of percolated coffee (i.e. coffee percolated with water) was included in the exposure assessment.

d) **Instant coffee;[1] tomato sauce; mustard; salad dressings; pickles, chutneys and relish; and other sauces and condiments:** For these food groups, the measured fluoride concentration results did not conform with the required quality criteria (i.e. analyte recovery in the samples was insufficient); therefore, these results were excluded from the exposure assessment.

e) **Infant formula (powder); infant formula (dairy alternatives) (powder); and growing-up milk (powder):** As these food groups were analysed as sold (i.e. powder), for exposure assessment purposes, the average fluoride concentration found in tap water (2015–2016 HSE data) was combined with the fluoride concentration measured in the powder sample to reflect actual consumption (i.e. formula prepared with tap water) in accordance with manufacturers' instructions (e.g. 13% powder, 87% water).

To estimate potential additional contribution from those samples excluded from the assessment (i.e. (c) and (d) above), fluoride concentration data determined in other surveys (Warren *et al.*, 1996; USDA, 2005) were used as a substitute (see Substitution Model, Appendix VI).

2.7   Mathematical treatment of analytical results

Each fluoride value determined for each food sample is the calculated average for replicate (n=2 or 3) fluoride measurements carried out on the sample.

Fluoride was not determined at measurable levels in any of the replicates for approximately 13% of samples. For these samples, fluoride content was assumed to be present at the LOD of the method, i.e. 0.005 mg/kg, which is a conservative approach.

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[1] Unlike percolated coffee, the water component of the instant coffee beverage as consumed is accounted for separately in the food consumption surveys; therefore, it did not need to be considered here.

3   RESULTS

3.1   Concentration data

TDS samples

In total, 216 samples distributed across 18 categories of foods and beverages were analysed as part of this study.

Table 2 and Figure 1 provide an overview of the analytical concentration of fluoride determined across the 18 food categories, which were further divided into subcategories. Analytical results which did not meet the required quality criteria (see Section 2.6), and results for samples not used in the exposure assessment (e.g. fluoride in dry tea leaf, water used in cooking), are not shown.

Table 2: Fluoride concentration in mg/kg determined in Total Diet Study samples*

Food category	Food subcategory	*n*	Sub-*n* per *n*	Mean**	Min	Max
CEREALS AND CEREAL PRODUCTS	Flours, etc.	4	10	0.258	0.041	0.691
	Breads and rolls	3	10	0.343	0.284	0.44
	Breakfast cereals	6	10	0.062	0.006	0.085
	Fine bakery ware	7	10	0.155	0.101	0.209
	Pasta and noodles (boiled)	2	10	0.619	0.579	0.658
	Pasta and noodles (raw)	2	10	0.166	0.139	0.193
	Rice (boiled)	1	10	0.7	0.7	0.7
	Rice (raw)	1	10	0.065	0.065	0.065
	Total	*26*	*260*	*0.225*	*0.006*	*0.7*
DAIRY, DAIRY PRODUCTS AND DAIRY ANALOGUES	Milks	2	10	0.009	0.008	0.009
	Creams	1	10	0.006	0.006	0.006
	Yogurts	2	10	0.038	0.029	0.046
	Cheeses	3	10	0.024	0.007	0.054
	Butter	1	10	0.006	0.006	0.006
	Other non-dairy milk (e.g. rice milk)	3	10	0.253	0.213	0.324
	Other dairy products	3	10	0.055	0.017	0.119
	Fat/dairy spreads (excluding butter)	1	10	0.005	0.005	0.005
	Total	*16*	*160*	*0.069*	*0.005*	*0.324*
EGGS	Eggs (fried)	1	10	0.007	0.007	0.007
	Total	*1*	*10*	*0.007*	*0.007*	*0.007*
MEAT AND MEAT PRODUCTS	Beef	3	10	0.047	0.005	0.11
	Lamb	1	10	0.057	0.057	0.057
	Pork	4	10	0.084	0.005	0.157
	Poultry	2	10	0.012	0.005	0.018
	Offal and derived products	5	10	0.106	0.005	0.202
	Total	*15*	*150*	*0.073*	*0.005*	*0.202*

Food category	Food subcategory	*n*	Sub-*n* per *n*	Mean**	Min	Max
FISH AND FISH PRODUCTS	White fish	3	10	0.956	0.159	2.175
	Oily fish	5	10	2.371	0.036	5.8
	Shellfish	3	10	0.949	0.474	1.659
	Total	*11*	*110*	*1.597*	*0.036*	*5.8*
VEGETABLES AND VEGETABLE PRODUCTS	Allium	2	10	0.005	0.005	0.005
	Brassica (boiled)	3	10	0.629	0.401	0.958
	Brassica (raw)	3	10	0.025	0.005	0.05
	Fruiting vegetables	7	10	0.059	0.005	0.263
	Leafy vegetables	2	10	0.058	0.037	0.078
	Stem vegetables	1	10	0.038	0.038	0.038
	Root vegetables (boiled)	2	10	0.512	0.374	0.65
	Root vegetables (raw)	2	10	0.006	0.005	0.007
	Tuber vegetables (boiled)	1	10	0.382	0.382	0.382
	Tuber vegetables (raw)	3	10	0.067	0.018	0.139
	Legumes (boiled)	3	10	0.524	0.244	0.948
	Legumes (raw)	6	10	0.205	0.01	0.469
	Mushrooms	1	10	0.008	0.008	0.008
	Other vegetables	4	10	0.116	0.021	0.235
	Seaweed	1	10	0.806	0.806	0.806
	Total	*41*	*410*	*0.201*	*0.005*	*0.958*
FRUIT AND FRUIT PRODUCTS	Banana	1	10	0.005	0.005	0.005
	Exotic/tropical fruit	1	10	0.005	0.005	0.005
	Berries	1	10	0.008	0.008	0.008
	Citrus fruit	1	10	0.005	0.005	0.005
	Pome fruit	2	10	0.006	0.005	0.006
	Stone fruit	3	10	0.005	0.005	0.006
	Other fruit	4	10	0.058	0.007	0.141
	Fruit preserves	2	10	0.042	0.033	0.05
	Drupes	1	10	0.456	0.456	0.456
	Dried fruit	4	10	0.197	0.064	0.413
	Total	*20*	*200*	*0.08*	*0.005*	*0.456*
HERBS AND SPICES	Herbs	1	10	0.437	0.437	0.437
	Spices	1	10	0.325	0.325	0.325
	Total	*2*	*20*	*0.381*	*0.325*	*0.437*
NUTS AND SEEDS	Nuts	2	10	0.655	0.053	1.257
	Seeds	1	10	0.045	0.045	0.045
	Total	*3*	*20*	*0.452*	*0.045*	*1.257*
FATS AND OILS (EXCLUDING DAIRY)	Fats and oils	3	10	0.005	0.005	0.005
	Total	*3*	*30*	*0.005*	*0.005*	*0.005*

| Food category	Food subcategory	*n*	Sub-*n* per *n*	Mean**	Min	Max
SOUPS, SAUCES AND CONDIMENTS	Sauces	5	10	0.245	0.014	0.455
	Soups	3	10	0.261	0.109	0.56
	Gravies and stock cubes	2	10	0.247	0.129	0.365
	Total	*10*	*100*	*0.25*	*0.014*	*0.56*
SAVOURY SNACKS	Potato snacks	1	10	0.191	0.191	0.191
	Mixed or other snacks	1	10	0.237	0.237	0.237
	Total	*2*	*20*	*0.214*	*0.191*	*0.237*
SUGAR AND PRESERVES	Sugars and molasses	2	10	0.029	0.005	0.052
	Sugar confectionery	1	10	0.137	0.137	0.137
	Honey	1	10	0.605	0.605	0.605
	Jellies	1	5	0.641	0.641	0.641
	Total	*5*	*45*	*0.288*	*0.005*	*0.641*
CONFECTIONERY	Confectionery	4	10	0.02	0.005	0.034
	Drinking powder	1	4	0.163	0.163	0.163
	Total	*5*	*44*	*0.048*	*0.005*	*0.163*
BEVERAGES	Non-alcoholic beverages	12	10	0.241	0.01	0.475
	Alcoholic beverages	5	10	0.13	0.033	0.181
	Tea	10	10	4.755	1.711	8.295
	Tea (herbal)	1	10	1.649	1.649	1.649
	Bottled water	2	10	0.423	0.196	0.65
	Tap water***			0.65		
	Total	*30*	*300*	*1.788*	*0.01*	*8.295*
OTHER	Vinegar	1	10	0.005	0.005	0.005
	Total	*1*	*10*	*0.005*	*0.005*	*0.005*
FOOD FOR INFANTS AND YOUNG CHILDREN	Infant formula	3	5–10	0.241	0.081	0.438
	Infant formula (dairy alternatives)	8	6–10	0.169	0.116	0.23
	Total	*11*	*101*	*0.189*	*0.081*	*0.438*
COMPOSITE FOOD	Composite food	2	10	0.13	0.052	0.208
	Total	*2*	*20*	*0.13*	*0.052*	*0.208*

* Not shown are samples that failed the analytical quality control criteria, i.e. Tomato sauce; Other sauces and condiments; Mustard; Salad dressings; Pickled vegetables; Pickles, chutneys and relish; Instant coffee; Percolated coffee. Also excluded are results for the water used in cooking, dry tea leaf (aggregate), an aggregate tea sample and aggregate tea samples containing milk, which were not used in the exposure assessment.

n = number of samples; sub-*n* per *n* = number of subsamples for each sample.

** Results are rounded to three decimal places.

*** Calculated value, derived from 2015–2016 HSE monitoring data on fluoride in drinking water from public supplies in Ireland.

Figure 1: Fluoride concentration (mg/kg) boxplot distribution in food categories/groups included in the Total Diet Study
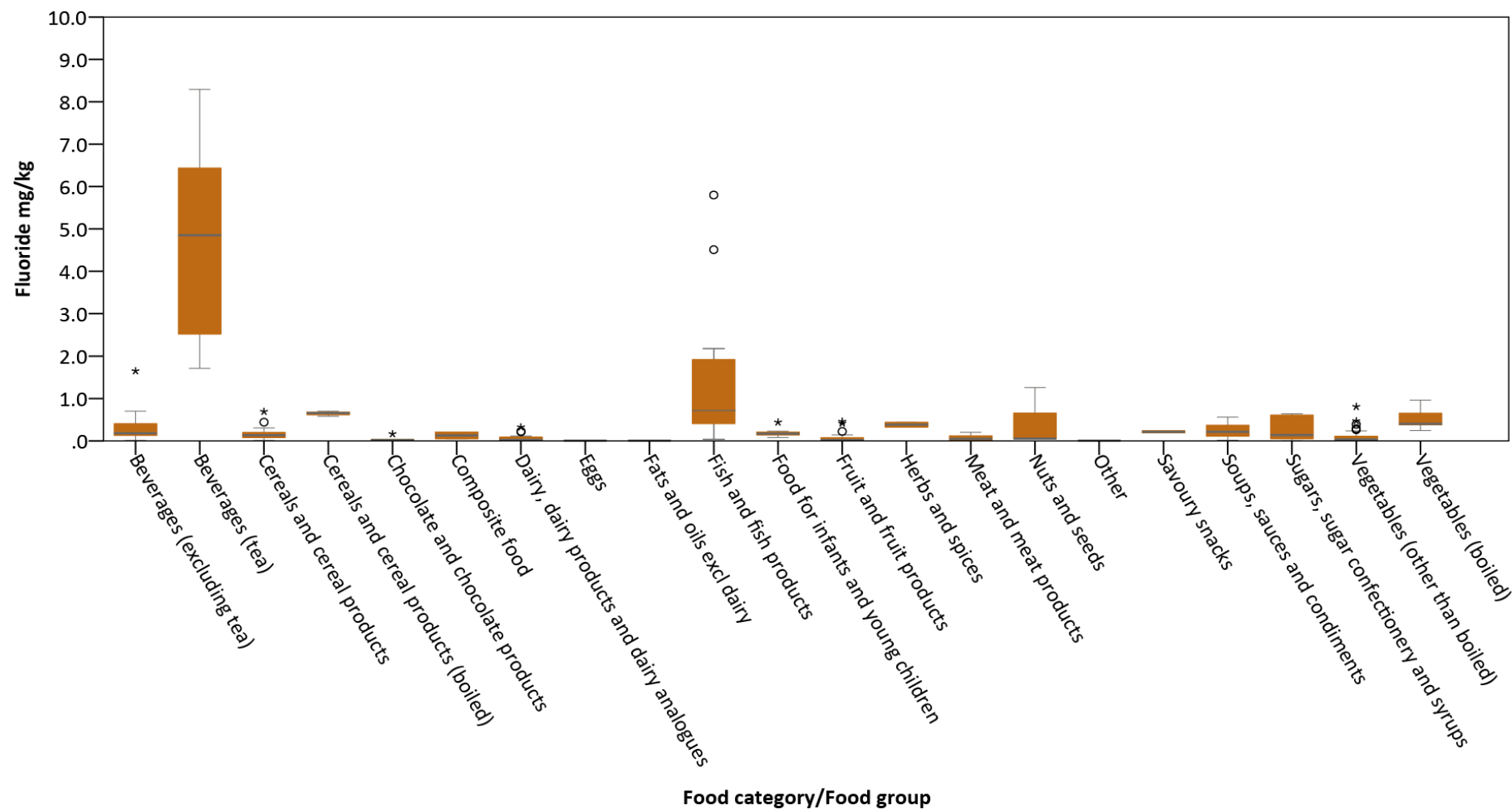
As can be seen in Figure 1, by far the highest fluoride concentrations were observed in the 10 tea samples, which ranged from 1.711 mg/kg to 8.295 mg/kg. For exposure assessment purposes, the mean of the concentration determined in the 10 individual tea samples (4.755 mg/kg) was used to reflect long-term chronic exposure. The mean value takes into account the wide variation in fluoride concentration between brands and the large fluctuation in fluoride content of tea brands over time, arising from changing sources of tea used for blending (Chan *et al.*, 2013).

The concentrations in the 10 tea samples analysed in this study were found to be in line with results determined in other studies on black tea samples carried out in the UK and Ireland (see Table 3). Common to these studies is a large range of fluoride concentration in black tea (leaves and infusions). This variation in fluoride content can be attributed to plant variety, plant part, age of plant tissue and growing site conditions (Ruan and Wong, 2001; Kalayci and Somer, 2003; Chan *et al.*, 2013; Cai *et al.*, 2016) and will also be influenced by tea leaf processing and blending methods (Cai *et al.*, 2016), tea preparation practices and conditions (Gulati *et al.*, 1993; Chan *et al.*, 2013; Maleki *et al.*, 2016; Ruxton and Bond, 2014) and, possibly, by the analytical methodology employed in determining fluoride concentration in the tea samples (see Section 2.5).

Three aggregate tea samples, to which 10, 30 and 50 mL respectively of milk had been added, were also analysed as part of this study and compared with the aggregate sample prepared without addition of milk (data not shown) to examine any potential decrease in fluoride availability due to complexing of fluoride with milk components (e.g. calcium). No consistent pattern of change in fluoride concentration could be observed; this is in line with findings reported by Gulati *et al.* (1993), who reported no difference in levels of fluoride in tea prepared with or without milk.

Table 3: Fluoride concentration in black tea reported in the literature (UK and Ireland)

Product	Test material	n	Country*	Strength of infusion	Water used in infusion	Infusion time	Method of analysis	Mean (mg/kg)	Range (mg/kg)	Reference
Pure blends	Dry leaves	38	UK	n/a	n/a	n/a	Ashing/IC-CD	132	103–200	Chan *et al.*, 2013
Oolong/Pu'er	Dry leaves	38	UK	n/a	n/a	n/a	Ashing/IC-CD	216	174–268	Chan *et al.*, 2013
Black blends	Dry leaves	38	UK	n/a	n/a	n/a	Ashing/IC-CD	266	130–382	Chan *et al.*, 2013
Economy blends	Dry leaves	38	UK	n/a	n/a	n/a	Ashing/IC-CD	578	330–839	Chan *et al.*, 2013
Blended black tea	Dry leaves	28	UK	n/a	n/a	n/a	Ashing/ISE	1082	540–1700	Ruxton and Bond, 2015
Decaffeinated black	Dry leaves	10	UK	n/a	n/a	n/a	Ashing/ISE	1200	750–1800	Ruxton and Bond, 2015
Speciality	Dry leaves	11	UK	n/a	n/a	n/a	Ashing/ISE	769	25–1300	Ruxton and Bond, 2015
Black tea	*Dry leaves*	*1*	*IRL*	*n/a*	*n/a*	*n/a*	*Microdiffusion/ISE*	*454*	*n/a*	*This study*
Pure blends	Infusion	4	UK	2 g tea in 100 mL	Deionised water	2 min	ISE	1.63	0.71–2.5	Chan *et al.*, 2013
Pure blends	Infusion	4	UK	2 g tea in 100 mL	Deionised water	10 min	ISE	1.99	1.09–3.07	Chan *et al.*, 2013
Pure blends	Infusion	4	UK	2 g tea in 100 mL	Deionised water	30 min	ISE	2.08	1.19–3.33	Chan *et al.*, 2013
Oolong/Pu'er	Infusion	6	UK	2 g tea in 100 mL	Deionised water	2 min	ISE	0.81	0.43–1.43	Chan *et al.*, 2013
Oolong/Pu'er	Infusion	6	UK	2 g tea in 100 mL	Deionised water	10 min	ISE	1.47	0.89–2.27	Chan *et al.*, 2013
Oolong/Pu'er	Infusion	6	UK	2 g tea in 100 mL	Deionised water	30 min	ISE	1.65	1.16–2.57	Chan *et al.*, 2013
Black blends	Infusion	7	UK	2 g tea in 100 mL	Deionised water	2 min	ISE	3.28	0.76–4.98	Chan *et al.*, 2013
Black blends	Infusion	7	UK	2 g tea in 100 mL	Deionised water	10 min	ISE	3.74	0.63–5.26	Chan *et al.*, 2013
Black blends	Infusion	7	UK	2 g tea in 100 mL	Deionised water	30 min	ISE	4.10	0.76–6.45	Chan *et al.*, 2013
Economy blends	Infusion	15	UK	2 g tea in 100 mL	Deionised water	2 min	ISE	5.86	3.60–7.96	Chan *et al.*, 2013
Economy blends	Infusion	15	UK	2 g tea in 100 mL	Deionised water	10 min	ISE	6.53	3.70–8.37	Chan *et al.*, 2013
Economy blends	Infusion	15	UK	2 g tea in 100 mL	Deionised water	30 min	ISE	6.98	3.90–8.85	Chan *et al.*, 2013
Speciality	Infusion	11	UK	2 g tea in 100 mL	Deionised water	40 sec	ISE	3.06	1.00–3.06	Ruxton and Bond, 2015
Blended black tea	Infusion	28	UK	3x (1 teabag in 240 mL water)	Deionised water	40 sec	ISE	4.91	2.14–9.33	Ruxton and Bond, 2015
Decaffeinated black	Infusion	10	UK	3x (1 teabag in 240 mL water)	Deionised water	40 sec	ISE	6.64	2.91–8.25	Ruxton and Bond, 2015
Black tea	Infusion	54	IRL	1 teabag in 200 mL water	Deionised water	5 min	ISE	2.60	0.80–5.50	Waugh *et al.*, 2016
Black tea	Infusion	54	IRL	2x (1 teabag in 200 mL water)	Fluoridated water (0.7 mg/L)	5 min	ISE	3.30	1.60–6.10	Waugh *et al.*, 2016
Black tea	*Infusion*	*10*	*IRL*	*1 teabag in 250 mL water*	*Deionised water*	*3 min*	*Microdiffusion/ISE*	*4.76*	*1.71–8.26*	*This study*

n = number of samples; * Country where survey was carried out; n/a = not applicable; min = minutes; sec = seconds; IC = ion chromatography; CD = conductivity detector; ISE = ion selective electrode; Pure blends are from specific regions only (e.g. Ceylon Tea)

With the exception of fish and fish products (ranging from 0.036 to 5.8 mg/kg) and one nut sample (nuts and seeds ranging from 0.053 to 1.257 mg/kg), all remaining foods showed fluoride concentrations below 1 mg/kg.

In general, a higher level of fluoride could be observed in samples that were boiled in fluoridated water (e.g. rice, noodles and certain vegetables) compared to their raw counterparts (see Figure 1 and Appendix III). The fluoride concentration determined in the cooking water was 0.88 mg/kg. This value is above the target range for fluoride in fluoridated water in Ireland (0.6–0.8 mg/L; see Section 3.2). Therefore, samples cooked in this water may contain somewhat higher fluoride levels than would be expected on average.

Public water supply

The concentration of fluoride in tap water is known to fluctuate; therefore, an average fluoride concentration was derived from an extensive external dataset, representative of the fluoridated public water supply in Ireland.

Country-wide information on fluoride concentrations determined in the public water supply (fluoridated schemes only) for the years 2015–2016 was provided by the HSE. This information was used to derive the average fluoride concentration in fluoridated tap water in Ireland and was used in the exposure assessment, as described in detail in Section 2.6.

This calculated value of 0.65 mg/L provides a representative estimate of long-term average exposure to fluoride from fluoridated water.

3.2   Dietary exposure

Dietary exposure to fluoride was calculated as described in Sections 2.6 and 2.7. Table 4 provides results of total fluoride exposure from all food groups, for adults, for children and for preschool children resident in the Republic of Ireland.

Table 4: Total mean, P95 and P97.5 dietary exposure to fluoride in mg/day and mg/kg bw/day in adults, children and preschool children estimated in the Irish Total Diet Study

Population group (age in years)	Total population					
	mg/day			mg/kg bw/day		
	Mean	P95	P97.5	Mean	P95	P97.5
Adults (18+)	2.982	6.623	7.949	0.040	0.094	0.111
Children (5–12 years)	0.534	1.302	1.597	0.017	0.039	0.052
Preschool children (1–4 years)	0.342	0.648	0.744	0.023	0.047	0.057

The mean dietary exposure to fluoride ranged from 0.017 to 0.040 mg/kg bw/day across the population groups covered in this study. Exposures at P95 and P97.5 ranged from 0.039–0.094 mg/kg bw/day and 0.052–0.111 mg/kg bw/day, respectively.

3.3   Sources of dietary exposure in Ireland

Figure 2 provides an overview of major dietary contributors to fluoride exposure. Only food categories contributing more than 8% are labelled. As can be seen in Figure 2, for all population groups covered in this TDS, the food category 'Beverages' is found to be the major contributor, contributing 94%, 76% and 63% to total exposure in adults, children aged 5–12 years, and preschool children aged 1–4 years, respectively.

Further subanalysis at the food group level (see Table 1 for food categories and food groups) is displayed in Figure 3, showing the top contributors to total dietary exposure in all population groups covered in this TDS. Food groups individually contributing to <5% were summed into the group 'Other food groups'.

Adults are predominantly exposed to fluoride via consumption of black tea, which constitutes 76% of their total fluoride exposure, with tap water contributing 12%. For children aged 5–12 years, major contributors to exposure are tap water (33%) and black tea (29%). Preschool children aged 1–4 years are mainly exposed via consumption of tap water (49%), and to a lesser degree via consumption of growing-up milk (9%) and black tea (5%). For both children aged 5–12 years and preschool children aged 1–4 years, a host of other food groups, which individually contribute to less than 5%, make up about one-third of the exposure to fluoride.

Figure 2: Percentage contribution of food categories to total dietary fluoride exposure in adults, children and preschool children
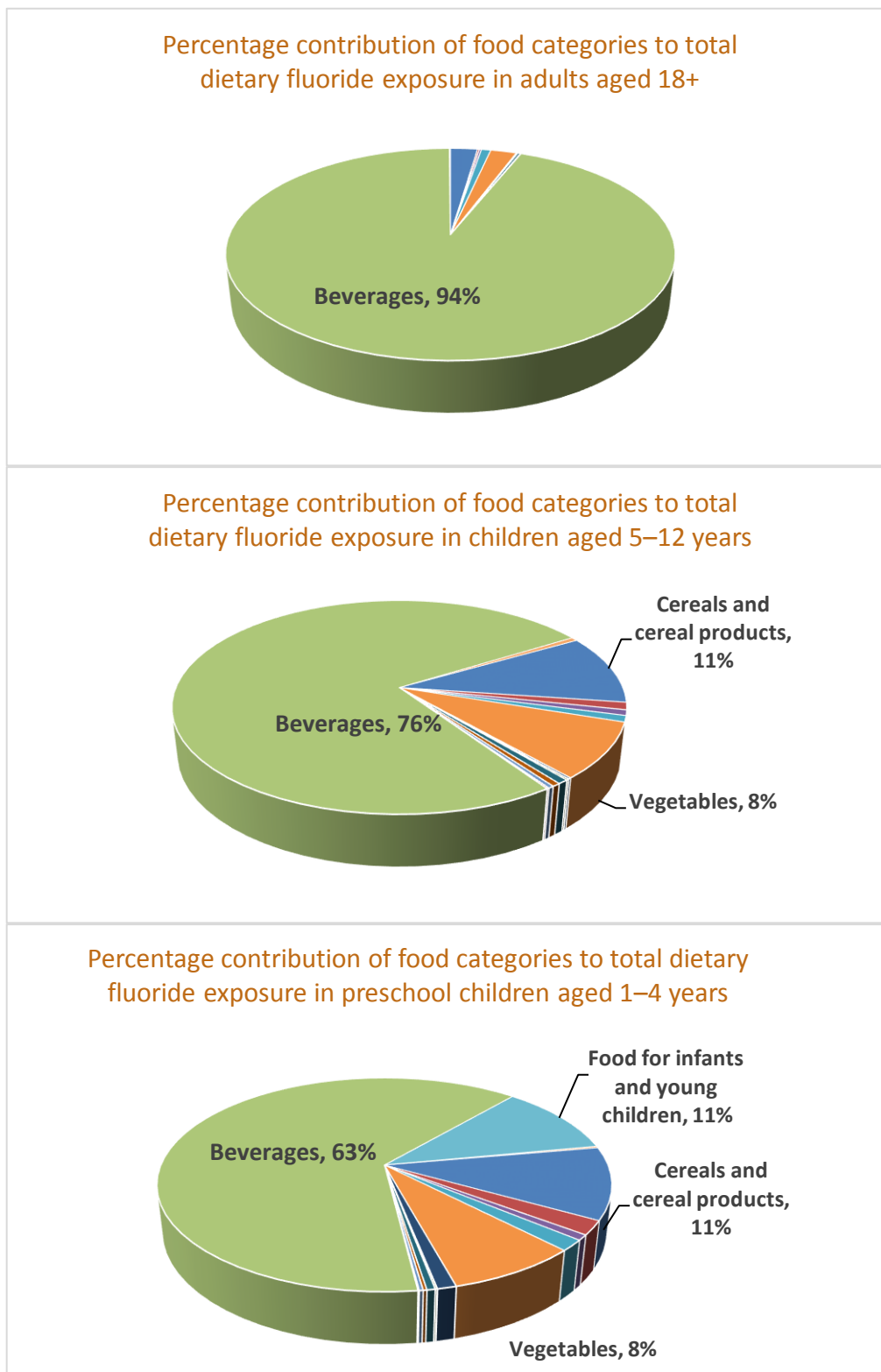

Percentage contribution of food categories to total dietary fluoride exposure in adults aged 18+

Beverages, 94%

Percentage contribution of food categories to total dietary fluoride exposure in children aged 5–12 years

Cereals and cereal products, 11%

Beverages, 76%

Vegetables, 8%

Percentage contribution of food categories to total dietary fluoride exposure in preschool children aged 1–4 years

Food for infants and young children, 11%

Beverages, 63%

Cereals and cereal products, 11%

Vegetables, 8%

Figure 3: Percentage contribution of individual food groups to total dietary fluoride exposure in adults, children and preschool children

Percentage contribution of food groups to total dietary fluoride exposure in adults aged 18+

Tap water 12%
Other food groups 12%
Tea 76%

Percentage contribution of food groups to total dietary fluoride exposure in children aged 5–12

Tea 29%
Other food groups 38%
Tap water 33%

Percentage contribution of food groups to total dietary fluoride exposure in preschool children aged 1–4

Tap water 49%
Growing-up milk 9%
Tea 5%
Other food groups 37%
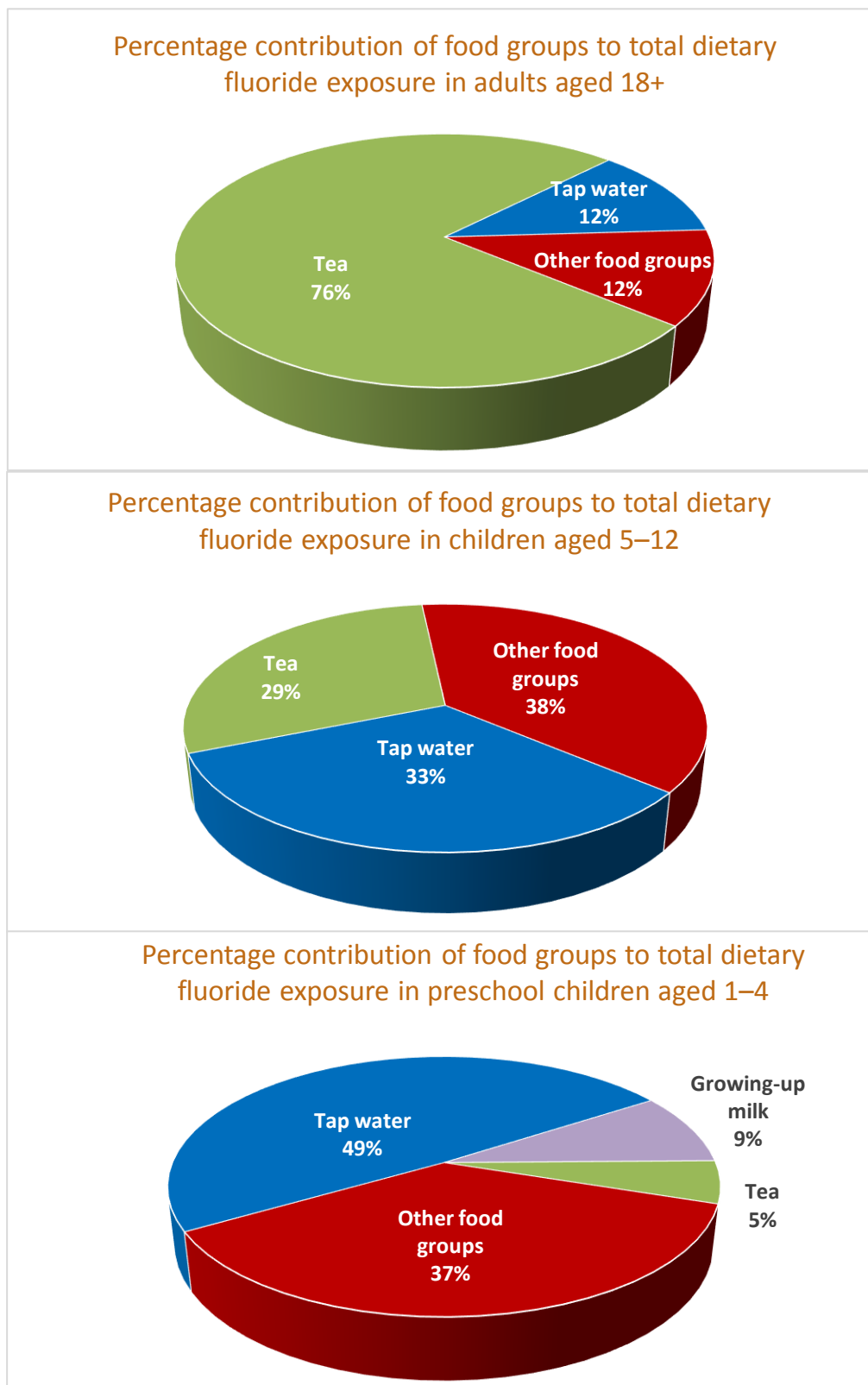
4. RISK ASSESSMENT

4.1 Health effects of fluoride

Fluoride is not essential for human growth and development. It is mainly associated with calcified tissue (teeth and bone) due to its high affinity for calcium (EFSA, 2005).

At low concentrations, it is beneficial in the prevention of dental caries (tooth decay). As a result, fluoridation of drinking water and the development of fluoride-containing oral care products (toothpastes and mouth rinses) and supplemented food and medical products have been employed since the early 20th century in a number of countries as a public health protective measure against tooth decay (SCHER, 2011; Government of Ireland, 2002; Sutton *et al.*, 2015).

The United States Institute of Medicine, in determining dietary reference intakes for fluoride, stated that the primary adverse effects associated with chronic excess fluoride intake were enamel (dental) fluorosis in children up to age 8 years and skeletal fluorosis in children from age 9 years upwards and in adults (Institute of Medicine, 1997).

Available data indicate a clear dose/response relationship between fluoride intake and both incidence and severity of dental fluorosis (EFSA, 2005). Excessive fluoride ingestion during the pre-eruptive formation and enamel maturation of teeth can cause fluorosis. This critical phase starts *in utero* and persists up to the formation of permanent teeth in preteens. Thus, enamel fluorosis may not become apparent until the final tooth eruption more than four to five years after exposure (Fejerskov *et al.*, 1996).

Systemically available fluoride can affect the ameloblasts, with effects on tooth structure at high levels. The incidence and severity of dental fluorosis is dose-dependent. At lower fluoride levels, the effects are on the mineralising matrix with a hypomineralised subsurface enamel covered by well-mineralised enamel (Bronckers *et al.*, 2009). This mild fluorosis is seen as mottling or striation on the tooth surface and is considered an aesthetically acceptable effect. On the other hand, moderate fluorosis is considered aesthetically objectionable and an unwanted effect. The occurrence of moderate enamel fluorosis was less than 5% in populations with fluoride intakes of 0.1 mg/kg bw/day (EFSA, 2005).

Skeletal fluorosis initially manifests as an increase in bone density on radiography, but with increasing fluoride incorporation into bone, pain and stiffness of joints can occur together with osteosclerosis and calcification of ligaments (McDonagh *et al.*, 2000). A fluoride intake of at least 15–20 mg/day via consumption of drinking water high in fluoride (>4 mg/L) for periods of 20 years can result in crippling skeletal fluorosis, a condition rarely found in non-tropical countries without occupational exposure to high airborne fluoride concentrations. Therapeutic studies with fluoride in postmenopausal osteoporosis suggest an increasing risk for skeletal fractures at or above fluoride intakes of 0.6 mg/kg bw/day (EFSA, 2005).

There is no consistent evidence of an association between the consumption of fluoridated drinking water and morbidity or mortality due to cancer, although this has been examined in a large number of epidemiological studies performed in many countries (McDonagh *et al.*, 2000; NHMRC, 2007; Jack *et al.*, 2016; Sutton *et al.*, 2015). With regard to other potential adverse health effects, there is insufficient good-quality evidence on any particular outcome to establish that levels of fluoride intake observed in Ireland can cause such effects (McDonagh *et al.*, 2000; NHMRC, 2007; SCHER, 2011; Jack *et al.*, 2016; Sutton *et al.*, 2015).

In 2011, the Scientific Committee on Health and Environmental Risks (SCHER) concluded that all risk assessments on fluoride carried out thus far pointed at a narrow margin between the recommended intakes for the prevention of dental caries and the recommended upper limits of exposure (SCHER, 2011).

4.2   Health-based guidance values

In 2013, the EFSA Panel on Dietetic Products, Nutrition and Allergies (NDA Panel), accounting for the beneficial effects of dietary fluoride on prevention of dental caries, set an adequate intake (AI) for fluoride from all sources, based on epidemiological studies showing an inverse relationship between the fluoride concentration of water and caries prevalence. The AI from all sources (including non-dietary sources) is 0.05 mg/kg bw/day for both children and adults, including pregnant and lactating women. For pregnant and lactating women, the AI is based on the body weight before pregnancy and lactation (EFSA, 2013).

In 2005, the NDA Panel (EFSA 2005) set tolerable upper intake levels (ULs) for fluoride for adults and children. Based on a prevalence of less than 5% of moderate dental fluorosis of permanent teeth in populations ingesting 0.08–0.12 mg/kg bw/day, the EFSA considered that the UL for fluoride is 0.10 mg/kg bw/day in children aged 1–8 years.

For children aged 9 years and older and for adults (including pregnant and lactating women), a UL of 0.12 mg/kg bw/day was set. This was based on an increased risk for non-vertebral bone fractures seen in studies with therapeutic oral administration of fluoride in amounts of 0.6 mg/kg bw/day in postmenopausal women over several years, incorporating an uncertainty (safety) factor of 5 (EFSA, 2005).

In 2010, the US Environmental Protection Agency's Health and Ecological Criteria Division, Office of Water (US EPA, 2010) developed an estimated reference dose (RfD) of 0.08 mg/kg bw/day for protection of 99.5% of the vulnerable population (i.e. children) against severe dental fluorosis and concluded that this value is also protective against fractures and skeletal effects in adults. The RfD is an estimate of the fluoride dose that will allow for a fluoride exposure adequate to protect against tooth decay for children and adults while protecting against severe dental fluorosis, clinical stage II skeletal fluorosis and skeletal fractures (US EPA, 2010).

The United States Institute of Medicine (IOM, 1997) set a UL of 0.1 mg/kg bw/day for preschool children and children up to 8 years of age, based on epidemiological research carried out in the 1930s and 1940s showing that chronic fluoride intake of less than 0.1 mg/kg bw/day by children at risk of enamel fluorosis was associated with a low prevalence of the milder form of dental fluorosis.

For adults and children above the age of 8 years, the UL was set at 10 mg/day, based on epidemiological studies on skeletal fluorosis in North America showing that risk of developing early signs of skeletal fluorosis is associated with a fluoride intake greater than 10 mg/day for 10 or more years (IOM, 1997). This value may be converted to 0.14 mg/kg bw/day for a 70 kg adult,[2] which is comparable to the UL set by the EFSA in 2005.

In 2007, a Health Canada expert panel recommended that the TDI of fluoride should be based mainly on estimated total fluoride intake from fluids and foods recorded in the 1940s – when children were exposed to no other major sources of fluoride and there was a low incidence (approximately 10%) of mild and very mild dental fluorosis. Adding all the relevant sources of exposure, the updated TDI was set at 0.105 mg/kg bw/day, which was the same as the one that had been proposed by the Advisory Review Panel in 1993, based on the absence of moderate dental fluorosis (Health Canada, 2010).

As detailed in the previous paragraphs, HBGVs have been set for fluoride in Europe, the US and Canada. Concerning threshold limits, the 2005 EFSA ULs correspond well to the ULs set by the IOM in 1997 and the TDI set by Health Canada in 2007. Concerning risk/benefit values, the EFSA AI of 0.05 mg/kg bw/day may be compared to the US EPA RfD of 0.08 mg/kg bw/day.

For the purposes of this report, the ULs set by the EFSA in 2005 were used, i.e. 0.1 mg/kg bw/day and 0.12 mg/kg bw/day for children (1–8 years) and for adults and children (≥9 years), respectively.

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[2] The EFSA Scientific Committee recommends that 70 kg be used when there is a need to apply a default body weight value for adults (EFSA, 2012).

4.3   Risk characterisation

Calculated fluoride exposure for all population groups covered in this study were compared against the ULs of 0.12 mg/kg bw/day and 0.1 mg/kg bw/day set by the EFSA (EFSA, 2005) for children (≥9 years) and adults, and for children (1–8 years), respectively. To allow for the calculated exposure for the population group of children aged 5–12 years to be compared with the relevant UL, this group has been divided into two population groups; namely, children aged 5–8 years and children aged 9–12 years.

Table 5 provides the results for total dietary exposure to fluoride expressed in mg/kg bw/day and as percentages of the EFSA UL.

Table 5: Percentage contribution of total dietary fluoride exposure estimated in the Irish
Total Diet Study to the ULs as set by the EFSA (EFSA, 2005)

| Population group (age, years) | Total population | | | | | |
| | mg/kg bw/day | | | % UL (0.1 mg/kg bw/day (1–8 years)) (0.12 mg/kg bw/day (9+ years)) | | |
	Mean	P95	P97.5	Mean	P95	P97.5
Adults (18+)	0.040	0.094	0.111	33	78	93
Children (9–12 years)	0.018	0.042	0.056	15	35	47
Children (5–8 years)	0.016	0.038	0.046	16	38	46
Preschool children (1–4 years)	0.023	0.047	0.057	23	47	57

As can be seen in Table 5, average exposure and exposure at the higher percentiles (P95, P97.5) are below the ULs set by the EFSA for all age groups. Fluoride exposure in adults, the highest exposed population, was much higher than in children, due to their relatively higher consumption of black tea. In addition, the mean intakes of fluoride were below the AI (0.05 mg/kg bw/day) for caries protection for all age groups, and particularly for children.

Figure 4 provides an overview of the exposure distribution for all population groups in relation to the applicable UL (0.1 mg/kg bw/day (1–8 years) and 0.12 mg/kg bw/day (≥9 years)) and AI (0.05 mg/kg bw/day) values set by the EFSA. None of the preschool children or children aged 5–12, and only a small percentage (1.6%) of the adult population, exceed the respective UL.

Figure 4: Histograms of total dietary exposure of adults, children and preschool children to fluoride (mg/kg bw/day) estimated in the Irish Total Diet Study, expressed as percentage population exposure in relation to the applicable EFSA UL and AI values
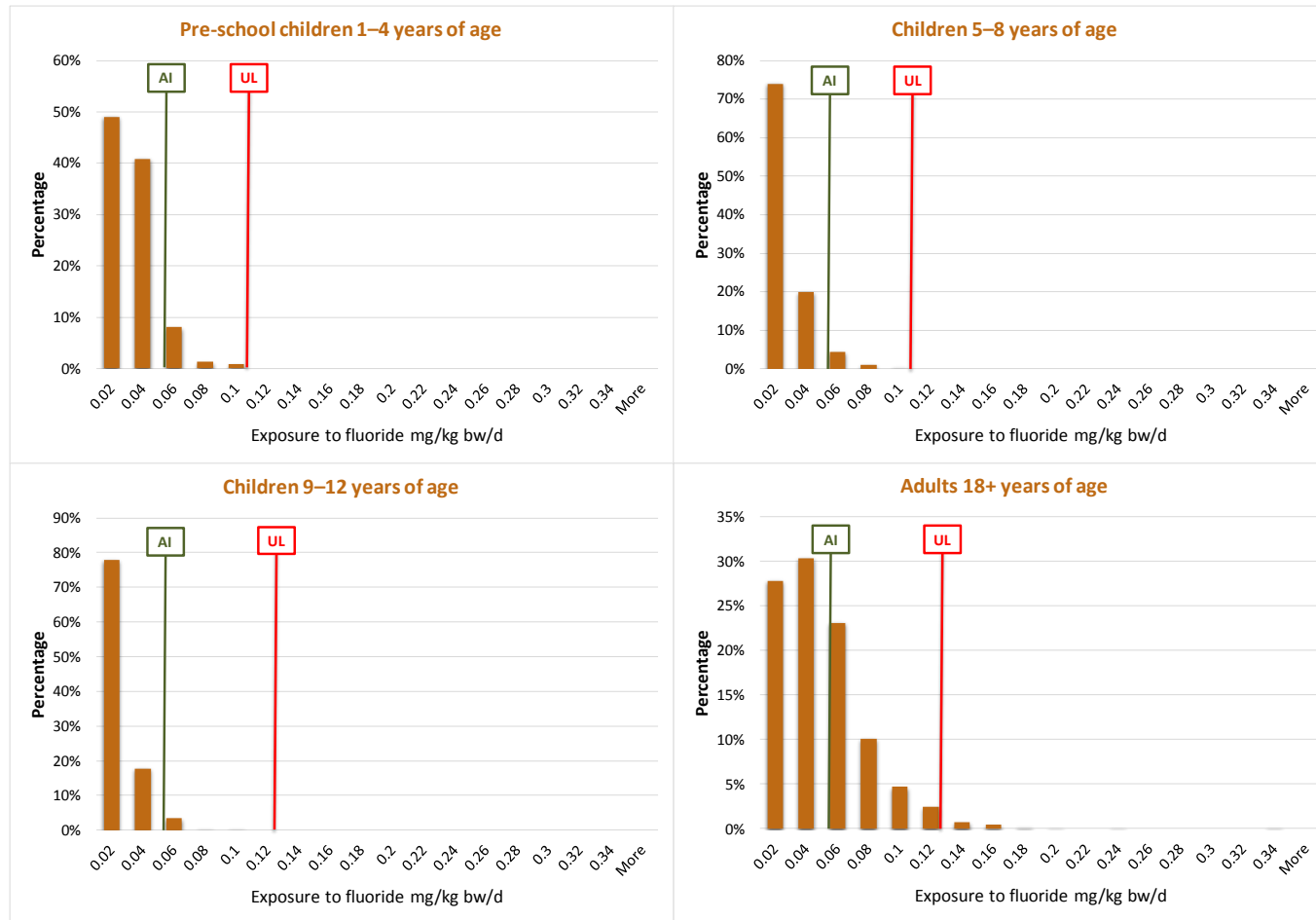
5.   DISCUSSION

In this study, estimates were made of long-term fluoride exposure in adults, children and preschool children resident in Ireland as a result of consumption of food and fluoridated tap water. The TDS approach was deemed most appropriate to derive such estimates, as it takes into account typical food preparation practices. Uncertainties associated with the approach chosen, having a potential influence on the exposure estimates, were identified and are provided in Appendix VII.

The estimates of fluoride intake in this study are for foods and beverages only and do not include possible fluoride contributions from non-dietary sources, particularly through swallowing of fluoridated toothpaste, which can make a significant contribution in young children, but much less so in older children and adults (Cochran *et al.*, 2004; Cressey *et al.* 2009).

Of particular interest in this study was the contribution of fluoride from tap water to overall fluoride exposure, and this contribution, as such, had to be accounted for by designing a study that would also account for absorption of fluoride during the cooking of food in fluoridated tap water. To meet this requirement, the standard format of the TDS typically employed in Ireland (i.e. use of deionised water in food preparation) was modified, and fluoridated tap water was used in the preparation of foodstuffs requiring boiling. Based on national monitoring data for 2015–2016, the average water fluoride concentration in Ireland was 0.65 mg/L. Since the cooking water used in the TDS had a fluoride concentration of 0.88 mg/L, the results obtained for the foods boiled in fluoridated water (e.g. rice, pasta, potatoes, broccoli, etc.) are likely to be an overestimation of the typical situation in Ireland. However, none of these foods were found to be a major contributor to dietary exposure and the effect on the overall exposure can therefore be considered small.

Due to the assumption that fluoridated water is used in all food preparation, the results of this study are conservative for people on a fluoridated public water supply (approximately 70% of the population). People obtaining their water from non-fluoridated supplies (approximately 30% of the population draws water from non-fluoridated public supplies or private water supplies) will likely have exposures below the estimates derived in this study.

Also adding to the conservatism of this study was use of the LOD value of 0.005 mg/kg for those foods showing no measurable content of fluoride. However, the overall impact of this on the calculated exposures is considered to be small, because all major dietary contributors had fluoride levels above the LOD of the analytical method.

Black tea was found to be the most important dietary contributor to fluoride intake in adults, and to a much lesser degree in children and preschool children. As discussed in Section 3.1, the fluoride concentration in black tea as consumed can vary greatly, and exposure will be influenced by several factors (e.g. tea preparation, tea origin, etc.). The exposure estimates derived here reflect the tea-brewing practices employed in the study and are based on the mean fluoride concentration derived from 10 individual brands of black tea. The mean fluoride concentration provides the best estimate of long-term intake, given wide variation in fluoride concentration between brands and the large

fluctuation in fluoride content of tea brands over time, arising from changing sources of tea used for blending (Chan *et al.* 2013). This mean concentration for black tea, used in the exposure assessment in this study, was within the range and close to the highest mean levels reported in similar studies on fluoride in black tea conducted in the UK and Ireland (Chan *et al.*, 2013; Ruxton and Bond, 2015; Waugh *et al.*, 2016).

In this study, it was also assumed that fluoride present in food is 100% bioavailable to the human body, which has been shown to not always be the case. This is because the extent of absorption is influenced by concomitant food intake, stomach acidity and the chemical form of fluoride (Cerklewski, 1997; Trautner and Einwag, 1989; Warneke and Setnikar, 1993; Ekstrand and Ehrnebo, 1979; Patz *et al.,* 1977; Shulman and Vallejo, 1990; Chan, 2014). However, the impact on the risk characterisation is considered negligible, since the studies on which the EFSA ULs are based (EFSA, 2005) made similar assumptions.

Fluoride exposures in children were much lower than in adults, largely due to the much higher consumption of black tea in adults. Adults are predominantly exposed to fluoride via consumption of black tea, which constitutes 76% of their total fluoride exposure, with tap water contributing 12%. Tap water was the main contributor to fluoride intake in children (49% in preschool children aged 1–4 years, 33% in children aged 5–12 years), while tea was a significant contributor (29%) in children aged 5–12 years.

The mean intakes of fluoride from foods and beverages were below the AI (0.05 mg/kg bw/day) for caries protection for all age groups, and particularly for children aged 1–12 years.

For all population groups included in this study, the estimated mean exposure to fluoride from foods and beverages at the mean and at higher percentiles (P95, P97.5) were below the ULs set by the EFSA.

For children aged 1–8 years, the probability of exceeding the UL (0.10 mg/kg bw/day) from fluoride intake from foods and beverages is very low, with P95 and P97.5 fluoride intake of 0.047 and 0.057 mg/kg bw/day, respectively, in children aged 1–4 years and 0.038 and 0.046 mg/kg bw/day, respectively, in children aged 5–8 years. This indicates that there is no appreciable risk of moderate dental fluorosis in children aged 1–8 years arising from fluoride intake from foods and beverages. While fluoride intakes from non-dietary sources were not assessed in this study, it has been estimated that the average contribution of toothpaste to fluoride intake is 0.01–0.02 mg/kg bw/day for 1.5–3.5-year-old children in Ireland, with over 90% having intakes <0.04 mg/kg bw/day (Cochran *et al.*, 2004), similar to estimates of 0.01–0.02 mg/kg bw/day for 1–3-year-old children in New Zealand (Cressey *et al*., 2009). Including such fluoride contributions of toothpaste would not result in total fluoride intake exceeding the UL of 0.10 mg/kg bw/day in 1–4-year-old children, even in the highest consumers of fluoride from foods and beverages. However, it is recognised that misuse of toothpaste may result in higher intakes of fluoride in a small proportion of young children (Cochran *et al.*, 2004). The low rates of moderate dental fluorosis in permanent dentition in children in regions with fluoridated water in Ireland (Whelton *et al.*, 2004) indicate that total fluoride intake is not excessive at the period of

development of permanent teeth in early childhood (about age 15–30 months) (Government of Ireland, 2002).

For children aged 9–12 years, the probability of exceeding the UL (0.12 mg/kg bw/day) from dietary fluoride intake is very low, with P95 and P97.5 fluoride intake of 0.042 and 0.056 mg/kg bw/day, respectively. This indicates that there is no appreciable risk of adverse effects on bone in children aged 9–12 years arising from fluoride intake from foods and beverages.

For adults, the probability of exceeding the UL (0.12mg/kg bw/day) is low, with 95% having intake of fluoride of less than 0.094 mg/kg bw/day and 97.5% having intake of less than 0.11 mg/kg bw/day. Fluoride intake exceeded the UL in a small proportion (1.6%) of adults. This indicates that the risk of adverse effects on bone in the adult population arising from fluoride intake from foods and beverages is low.

Although not estimated in this study, it is considered that the probability of teenagers exceeding the UL (0.12mg/kg bw/day) for fluoride is very low, due to the much lower consumption of tea in teenagers (mean 110 g/day, P95 499 g/day) (IUNA, 2006a) than in adults (mean 422 g/day, P95 1121 g/day) (IUNA, 2011). For high consumers (P95) of tea, the fluoride contribution from tea alone is 0.076 mg/kg bw/day in adults, but only 0.040 mg/kg bw/day in teenagers. Thus, assuming that tea is the predominant contributor to above-average fluoride intakes in teenagers (as in adults), the probability of fluoride exceeding the UL in teenagers is much lower than the (low) probability in adults.

Exposure of infants to fluoride from infant formula reconstituted with fluoridated water has previously been assessed by Anderson *et al.* (2004), who concluded that there is a very low risk of moderate dental fluorosis of the primary or permanent dentition in infants. The FSAI Scientific Committee recently concluded that, at levels of fluoride intake observed in women in this TDS study, exposure to fluoride during foetal development in Ireland is not excessive and does not give rise to moderate or severe fluorosis of primary teeth (FSAI, 2018).

Based on the results of this study, the FSAI Scientific Committee concluded that there is currently no scientific basis for concerns about the safety of children and adults in Ireland from exposure to fluoride from foods and beverages.

6.   CONCLUSIONS

This TDS has provided estimates of dietary exposures (mean, 95th and 97.5th percentile) of a representative population of adults (n=1,500, males and females), children aged 5–12 years (n=594) and preschool children aged 1–4 years (n=500), resident in Ireland, to fluoride present in the typical diet. These estimates are based on fluoride content of foods and beverages, including fluoridated water. They do not include possible fluoride contributions from non-dietary sources, particularly through swallowing of fluoridated toothpaste, which can make a significant contribution in young children, but much less so in older children and adults.

Black tea, which can contain naturally high levels of fluoride, was found to contain by far the highest fluoride concentrations determined in this study, ranging from 1.7 to 8.3 mg/kg in tea infusions. The exposure estimates derived here are based on the mean fluoride concentration in 10 individual brands of black tea (4.8 mg/kg in tea infusions) to reflect long-term exposure. With the exception of fish and fish products (ranging from 0.04 to 5.8 mg/kg) and one nut sample (nuts and seeds ranging from 0.05 to 1.3 mg/kg), all other foods showed fluoride concentrations below 1 mg/kg. The average water fluoride concentration for fluoridated public water supplies in Ireland was 0.65 mg/L.

Mean fluoride exposures in preschool children (1–4 years of age) and children (5–12 years of age) (0.023 and 0.017 mg/kg bw/day, respectively) were much lower than in adults (0.040 mg/kg bw/day), largely due to the much higher consumption of black tea in adults. Adults are predominantly exposed to fluoride via consumption of black tea, which constitutes 76% of their total fluoride exposure, with tap water contributing 12%. Tap water was the main contributor to fluoride intake in children (49% in preschool children aged 1–4 years, 33% in children aged 5–12 years), while tea was a significant contributor (29%) in children aged 5–12 years.

The mean intakes of fluoride from foods and beverages were below the AI of 0.05 mg/kg bw/day established by the EFSA for caries protection, for all age groups, and particularly for children.

These exposure estimates were compared with ULs for fluoride derived by the EFSA, enabling a conclusion to be made regarding risk to consumers.

For children aged 1–8 years, the probability of exceeding the UL (0.10 mg/kg bw/day) from fluoride intake from foods and beverages is very low. This indicates that there is no appreciable risk of moderate dental fluorosis in children aged 1–8 years of age arising from fluoride intake from foods and beverages. Where published estimates of average fluoride intake from toothpaste in Ireland were included (0.01–0.02 mg/kg bw/day for 1.5–3.5-year-old children), this would not result in total fluoride intake exceeding the UL, even in the highest consumers of fluoride from foods and beverages. However, it is recognised that misuse of toothpaste may result in higher intakes of fluoride in a small proportion of young children. For children aged 9–12 years, the probability of exceeding the UL (0.12 mg/kg bw/day) from fluoride intake from foods and beverages is very low. This indicates that there is no appreciable risk of adverse effects on bone in children aged 9–12 years of age arising from fluoride intake from foods and beverages.

For adults, the probability of exceeding the UL (0.12 mg/kg bw/day) is low and fluoride intake exceeded the UL in a small proportion (1.6%) of adults. This indicates that the risk of adverse effects on bone in adults arising from fluoride intake from foods and beverages is low.

Although not estimated in this study, it is considered that the probability of teenagers exceeding the UL for fluoride is very low, due to the much lower consumption of black tea in teenagers than in adults.

Based on the results of this study, the FSAI Scientific Committee concluded that there is currently no scientific basis for concerns about the safety of children and adults in Ireland from exposure to fluoride from foods and beverages.

7.   REFERENCES

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ACKNOWLEDGEMENTS

APPENDIX I FOOD PREPARATION

Table A1 Food preparation applied to TDS food groups

Food category	FSAI code	Food group	Number of subsamples per sample	Food preparation applied
CEREALS AND CEREAL PRODUCTS	1	White flour	10	–
	2	Wholemeal flour	10	–
	3	White bread/rolls	10	–
	4	Granary/wholegrain breads	10	–
	5	Brown bread and rolls	10	–
	6	Plain frozen pastry	10	Baked
	7	Plain biscuits	10	–
	8	Chocolate biscuits	10	–
	9	Other biscuits	10	–
	10	Cakes	10	–
	11	Other cakes, buns and pastries	10	–
	12	Noodles (unflavoured)	10	Boiled (in fluoridated tap water)
	12.1	Noodles (unflavoured) (dry)	10	–
	13	Pasta (cooked)	10	Boiled (in fluoridated tap water)
	14	Pasta (dry)	10	–
	15	Rice (cooked)	10	Boiled (in fluoridated tap water)
	16	Rice (dry)	10	–
	17	Couscous	10	Boiled (in fluoridated tap water)
	18	Cornflakes	10	–
	19	Bran flakes	10	–
	20	Wheat-type cereals	10	–
	21	Muesli	10	–
	22	Oat flakes	10	–
	23	Rice-type cereals	10	–
	24	Rice cakes	10	–
	25	Cornflour	10	–

Food category	FSAI code	Food group	Number of subsamples per sample	Food preparation applied
DAIRY, DAIRY PRODUCTS AND DAIRY ANALOGUES	26	Whole milk	10	–
	27	Low-fat, skimmed and fortified milk	10	–
	28	Cream	10	–
	29	Cheese (hard)	10	–
	30	Cheese (continental style)	10	–
	31	Cheese (soft and semi-soft)	10	–
	32	Yogurts	10	–
	33	Fromage frais	10	–
	34	Custard	10	–
	35	Vanilla ice cream	10	–
	36	Butter	10	–
	37	Dairy and non-dairy spreads	10	–
	38	Other ice creams	10	–
	40	Soya milk	10	–
	203	Coconut milk/Cream	10	–
	39	Other non-dairy milk (e.g. rice milk)	10	–
EGGS	41	Eggs (fried)	10	Dry-fried
MEAT AND MEAT PRODUCTS	106	Pork	10	Grilled
	107	Ham	10	–
	108	Pork sausage	10	Grilled
	109	Bacon rashers	10	Grilled
	110	Beef	10	Grilled
	111	Beef mince	10	Dry-fried
	112	Beefburger	10	Grilled
	113	Chicken	10	Roasted
	114	Turkey	10	Roasted
	115	Lamb	10	Roasted
	116	Offal (kidney)	10	Dry-fried
	117	Offal (liver)	10	Dry-fried
	118	Pudding, black	10	Dry-fried
	119	Pudding, white	10	Dry-fried
	120	Liver pâté (pork/chicken/duck)	10	–

Food category	FSAI code	Food group	Number of subsamples per sample	Food preparation applied
FISH AND FISH PRODUCTS	121	Cod	10	Baked
	122	White fish (other than cod)	10	Baked
	123	Oily fish other than salmon	10	Grilled
	124	Salmon	10	Grilled
	125	Tinned tuna	10	Drained
	126	Tinned fish (excluding salmon and tuna)	10	Drained
	127	Tinned salmon	10	Drained
	128	Mussels	10	Steamed (in fluoridated tap water)
	129	Prawns	10	Dry-fried
	130	Crab	10	–
	205	Fish fingers	10	Baked
VEGETABLES AND VEGETABLE PRODUCTS	42	Potatoes without skin (boiled)	10	Boiled (in fluoridated tap water)
	43	Potatoes without skin (raw)	10	–
	44	Potatoes (baby) with skin (microwaved)	10	Microwaved
	45	Chips	10	As purchased/baked (home fries)
	46	Garlic	10	–
	47	Onion	10	–
	48	Tomatoes	10	–
	49	Canned tomatoes	10	Juice included in analysis
	50	Tomatoes (canned/concentrate)	10	–
	51	Peppers	10	–
	52	Cucumber	10	–
	53	Mushrooms	10	Dry-fried
	54	Canned sweetcorn	10	Drained
	55	Carrots (boiled)	10	Boiled (in fluoridated tap water)
	56	Carrots (raw)	10	–
	57	Celery	10	–

Food category	FSAI code	Food group	Number of subsamples per sample	Food preparation applied
	58	Peas (boiled)	10	Boiled (in fluoridated tap water)
	59	Peas (raw)	10	–
	60	Canned peas	10	Drained
	61	Green beans (boiled)	10	Boiled (in fluoridated tap water)
	62	Green beans (raw)	10	–
	63	Baked beans	10	–
	65	Legumes (excluding peas)	10	Boiled (in fluoridated tap water)
	66	Legumes (excluding peas) (raw)	10	–
	67	Canned legumes (excluding peas)	10	Drained
	68	Cabbage (raw)	10	–
	69	Cabbage (boiled)	10	Boiled (in fluoridated tap water)
	70	Broccoli (boiled)	10	Boiled (in fluoridated tap water)
	71	Broccoli (raw)	10	–
	72	Cauliflower (boiled)	10	Boiled (in fluoridated tap water)
	73	Cauliflower (raw)	10	–
	74	Root vegetables (excluding carrots) (boiled)	10	Boiled (in fluoridated tap water)
	75	Root vegetables (excluding carrots) (raw)	10	–
	76	Stir-fry vegetables	10	Dry-fried
	77	Lettuce	10	–
	78	Spinach and other leaves	10	–
	79	Avocado	10	Inedible parts removed
	80	Pumpkin/Squash	10	Baked
	81	Seaweed (Irish moss, Kombu, Nori) (dry)	10	Soaked (in deionised water)

Food category	FSAI code	Food group	Number of subsamples per sample	Food preparation applied
	82	Aubergine/Courgette	10	Grilled
	83	Oriental vegetables (water chestnuts/bamboo shoots)	10	Dry-fried
FRUIT AND FRUIT PRODUCTS	84	Apples	10	–
	85	Citrus fruit	10	–
	86	Bananas	10	–
	87	Grapes	10	–
	88	Pears	10	–
	89	Peaches and nectarines	10	–
	90	Kiwis	10	Inedible parts removed
	91	Plums	10	–
	92	Berries	10	–
	93	Other fruit	10	Inedible parts removed
	94	Canned fruit	10	–
	95	Melons	10	Inedible parts removed
	64	Smoothies	10	–
	96	Olives	10	Drained; inedible parts removed
	97	Dried raisins/sultanas/currants	10	–
	98	Apricots, prunes, and other dried fruit	10	Inedible parts removed
	99	Figs and dates	10	Inedible parts removed
	100	Glacé cherries and mixed peel	10	Inedible parts removed
HERBS AND SPICES	104	Herbs	10	–
	105	Spices	10	–
NUTS AND SEEDS	101	Tree nuts	10	Inedible parts removed
	102	Peanuts (roasted salted)	10	–
	103	Seeds	10	–
FATS AND OILS (EXCLUDING DAIRY)	197	Olive oil	10	–
	198	Vegetable oil	10	–
	199	Animal fat	10	–

Food category	FSAI code	Food group	Number of subsamples per sample	Food preparation applied
SOUPS, SAUCES AND CONDIMENTS	132	Soup (fresh) (Tetra Paks and tubs)	10	–
	133	Soups (canned)	10	–
	134	Soups (dried packet)	10	–
	131	Stock cubes, concentrated savoury spreads and essences	10	–
	135	Tomato sauce	10	–
	136	Mayonnaise	10	–
	137	Gravy	10	–
	138	Cook-in sauces (other)	10	–
	139	Cook-in sauces (tomato-based)	10	–
	140	Dry sauces	10	–
	142	Soy sauce	10	–
	144	Mustard	10	–
	141	Other sauces and condiments	10	–
	145	Salad dressings	10	–
	146	Pickled vegetables	10	Drained
	147	Pickles, chutneys and relish	10	–
SAVOURY SNACKS	200	Crisps	10	–
	201	Other savoury snacks	10	–
SUGAR AND PRESERVES	154	Sugar	10	–
	155	Marmalade	10	–
	156	Jam	10	–
	157	Honey	10	–
	158	Sugar syrups	10	–
	159	Jelly	5	Made up (in fluoridated tap water)
CONFECTIONERY	148	Chocolate confectionery	10	–
	149	Non-chocolate confectionery	10	–
	150	Plain chocolate	10	–
	151	Cocoa powder	4	–
	152	Drinking chocolate powder	10	–
	153	Chocolate/nut spreads	10	–

Food category	FSAI code	Food group	Number of subsamples per sample	Food preparation applied
BEVERAGES	160	Tap water	n/a	–
	161	Lager	10	–
	162	Stout	10	–
	163	Wine (white/red/rosé)	10	–
	164	Spirits	10	–
	165	Cider	10	–
	166	Carbonated soft drinks	10	–
	167	Soft drinks (wine substitutes, etc.)	10	–
	168	Sports drinks	10	–
	169	Squashes	10	–
	170	Apple juice (fresh)	10	–
	171	Orange juice (fresh)	10	–
	172	Apple juice (from concentrate)	10	–
	173	Orange juice (from concentrate)	10	–
	174	Fruit nectars	10	–
	175	Cranberry juice	10	–
	176	Multivitamin juices	10	–
	177	Other fruit juices	10	–
	178	Tea	1	Infused (in deionised water)
	179	Tea	1	Infused (in deionised water)
	180	Tea	1	Infused (in deionised water)
	181	Tea	1	Infused (in deionised water)
	182	Tea	1	Infused (in deionised water)
	183	Tea	1	Infused (in deionised water)
	184	Tea	1	Infused (in deionised water)
	185	Tea	1	Infused (in deionised water)
	186	Tea	1	Infused (in deionised water)
	187	Tea	1	Infused (in deionised water)

Food category	FSAI code	Food group	Number of subsamples per sample	Food preparation applied
	188	Tea (aggregate sample)	10	Infused (in deionised water)
	189	Tea (aggregate sample) (dry leaves)	10	–
	190	Tea with milk	10	Infused (in deionised water)
	191	Tea with milk	10	Infused (in deionised water)
	192	Tea with milk	10	Infused (in deionised water)
	193	Instant coffee	10	–
	194	Percolated coffee	10	Percolated (in deionised water)
	195	Herbal tea	10	Infused (in deionised water)
	196	Bottled water	10	–
OTHER	143	Vinegar	10	–
FOOD FOR INFANTS AND YOUNG CHILDREN	206	Infant formula (powder)	10	–
	207	Infant formula (dairy alternatives) (powder)	10	–
	208	Growing-up milk (powder)	5	–
	209	Infant-specific beverages	10	–
	210	Infant food (meat-based)	10	–
	211	Infant food (fish-based)	6	–
	212	Infant food (fruit-based)	10	–
	213	Infant food (vegetable-based)	10	–
	214	Infant food (cereal-based)	10	–
	215	Infant food (dairy-based)	10	–
	216	Baby snacks	10	–
COMPOSITE FOOD	202	Pizza	10	Baked
	204	Rice pudding	10	–

APPENDIX II ANALYTICAL METHODOLOGY

The 216 samples were analysed for fluoride concentration after overnight hexamethyldisiloxane (HMDSO) acid diffusion at room temperature using a Fluoride-Ion Selective Electrode (F-ISE) (Model 9609, Orion) and a meter (Model 900A, Orion) (Martínez-Mier *et al.*, 2011; Taves, 1968) at the Fluoride Laboratory, Centre for Oral Health Research, School of Dental Sciences, Newcastle University, UK.

Fluoride concentrations of samples were calculated from the constructed calibration (standard) curve. The concentration of the fluoride standards, for constructing the calibration curve, must cover the concentration range expected in the samples to be analysed. As a routine practice, a new set of fluoride standards was analysed every time a new set of analyses was undertaken to construct the calibration curve. Since the fluoride concentrations of the samples were not known, five fluoride standards (0.01, 0.1, 1, 10, 100 ppm fluoride) were analysed with every set of analyses. Then, after entering all data, the three fluoride standards which covered the fluoride concentration range of the samples in the set were selected to construct the calibration curve and calculate the final fluoride concentrations. The food and beverage and fluoride-standard sample analyses were carried out in triplicate and the results checked for acceptable repeatability between replicates. A minimum of two replicate fluoride concentration values, for each sample, were used to calculate the average fluoride concentration of the corresponding sample; if the replicate results did not have acceptable repeatability, the analysis was repeated.

In addition to fluoride analysis of the 216 samples, recovery and reanalysis procedures were undertaken. In terms of fluoride recovery, 10% of the samples within each food/beverage category were randomly selected and spiked with a known amount of fluoride to determine the fluoride recovery on the same day. The initial decision on the appropriate amount of fluoride to be added to the samples for recovery measurements was based on the previously reported fluoride concentration of the relevant samples in the literature. In addition, 10% of the samples within each category were randomly selected to be reanalysed on another day, and 20% of these reanalysed samples within each food/beverage category were randomly selected and spiked with a known amount of fluoride to check the fluoride recovery from the reanalysed samples on the same day as the reanalysis. For fluoride recovery of reanalysed samples, the decision on the appropriate amount of fluoride to be added to the samples for recovery measurements was based on the previously measured fluoride concentrations in these samples.

APPENDIX III ANALYTICAL CONCENTRATION DATA

Table A2: Fluoride concentration data in mg/kg

Category	Description	Comment	Fluoride mg/kg
CEREALS	White flour	Concentration as determined in the sample	0.133
	Wholemeal flour	Concentration as determined in the sample	0.041
	White bread/rolls	Concentration as determined in the sample	0.305
	Granary/wholegrain breads	Concentration as determined in the sample	0.44
	Brown bread and rolls	Concentration as determined in the sample	0.284
	Plain frozen pastry	Concentration as determined in the sample	0.196
	Plain biscuits	Concentration as determined in the sample	0.109
	Chocolate biscuits	Concentration as determined in the sample	0.101
	Other biscuits	Concentration as determined in the sample	0.198
	Cakes	Concentration as determined in the sample	0.107
	Other cakes, buns and pastries	Concentration as determined in the sample	0.209
	Noodles (unflavoured) (boiled)	Concentration as determined in the sample	0.658
	Noodles (unflavoured) (dry)	Concentration as determined in the sample	0.193
	Pasta (cooked)	Concentration as determined in the sample	0.579
	Pasta (dry)	Concentration as determined in the sample	0.139
	Rice (cooked)	Concentration as determined in the sample	0.7
	Rice (dry)	Concentration as determined in the sample	0.065
	Couscous	Concentration as determined in the sample	0.691
	Cornflakes	Concentration as determined in the sample	0.063
	Bran flakes	Concentration as determined in the sample	0.061
	Wheat-type cereals	Concentration as determined in the sample	0.082
	Muesli	Concentration as determined in the sample	0.077
	Oat flakes	Concentration as determined in the sample	0.006
	Rice-type cereals	Concentration as determined in the sample	0.085
	Rice cakes	Concentration as determined in the sample	0.166
	Cornflour	Concentration as determined in the sample	0.167
DAIRY	Whole milk	Concentration as determined in the sample	0.009
	Low-fat, skimmed and fortified milk	Concentration as determined in the sample	0.008
	Cream	Concentration as determined in the sample	0.006
	Cheese (hard)	Concentration as determined in the sample	0.011
	Cheese (continental style)	Concentration as determined in the sample	0.054
	Cheese (soft and semi-soft)	Concentration as determined in the sample	0.007
	Yogurts	Concentration as determined in the sample	0.029
	Fromage frais	Concentration as determined in the sample	0.046

Category	Description	Comment	Fluoride mg/kg
	Custard	Concentration as determined in the sample	0.017
	Vanilla ice cream	Concentration as determined in the sample	0.028
	Butter	Concentration as determined in the sample	0.006
	Dairy and non-dairy spreads	Concentration as determined in the sample	0.005
	Other ice creams	Concentration as determined in the sample	0.119
	Other non-dairy milk (e.g. rice milk)	Concentration as determined in the sample	0.213
	Soya milk	Concentration as determined in the sample	0.324
	Coconut milk/Cream	Concentration as determined in the sample	0.223
EGGS	Eggs (fried)	Concentration as determined in the sample	0.007
VEGETABLES	Potatoes without skin (boiled)	Concentration as determined in the sample	0.382
	Potatoes without skin (raw)	Concentration as determined in the sample	0.018
	Potatoes (baby) with skin (microwaved)	Concentration as determined in the sample	0.045
	Chips	Concentration as determined in the sample	0.139
	Garlic	Concentration as determined in the sample	0.005
	Onion	Concentration as determined in the sample	0.005
	Tomatoes	Concentration as determined in the sample	0.019
	Canned tomatoes	Concentration as determined in the sample	0.058
	Tomatoes (canned/concentrate)	Concentration as determined in the sample	0.263
	Peppers	Concentration as determined in the sample	0.038
	Cucumber	Concentration as determined in the sample	0.006
	Mushrooms	Concentration as determined in the sample	0.008
	Canned sweetcorn	Concentration as determined in the sample	0.034
	Carrots (boiled)	Concentration as determined in the sample	0.65
	Carrots (raw)	Concentration as determined in the sample	0.007
	Celery	Concentration as determined in the sample	0.038
	Peas (boiled)	Concentration as determined in the sample	0.244
	Peas (raw)	Concentration as determined in the sample	0.045
	Canned peas	Concentration as determined in the sample	0.39
	Green beans (boiled)	Concentration as determined in the sample	0.379
	Green beans (raw)	Concentration as determined in the sample	0.029
	Baked beans	Concentration as determined in the sample	0.469
	Legumes (excluding peas) (boiled)	Concentration as determined in the sample	0.948
	Legumes (excluding peas) (raw)	Concentration as determined in the sample	0.01
	Canned legumes (excluding peas)	Concentration as determined in the sample	0.288
	Cabbage (raw)	Concentration as determined in the sample	0.05

Category	Description	Comment	Fluoride mg/kg
	Cabbage (boiled)	Concentration as determined in the sample	0.958
	Broccoli (boiled)	Concentration as determined in the sample	0.401
	Broccoli (raw)	Concentration as determined in the sample	0.02
	Cauliflower (boiled)	Concentration as determined in the sample	0.529
	Cauliflower (raw)	Concentration as determined in the sample	0.005
	Root vegetables (excluding carrots) (boiled)	Concentration as determined in the sample	0.374
	Root vegetables (excluding carrots) (raw)	Concentration as determined in the sample	0.005
	Stir-fry vegetables	Concentration as determined in the sample	0.235
	Lettuce	Concentration as determined in the sample	0.037
	Spinach and other leaves	Concentration as determined in the sample	0.078
	Avocado	Concentration as determined in the sample	0.021
	Pumpkin/Squash	Concentration as determined in the sample	0.005
	Seaweed (Irish moss, Kombu, Nori) (dry)	Concentration as determined in the sample	0.806
	Aubergine/Courgette	Concentration as determined in the sample	0.022
	Oriental vegetables (water chestnuts/bamboo shoots)	Concentration as determined in the sample	0.172
FRUIT	Apples	Concentration as determined in the sample	0.005
	Citrus fruit	Concentration as determined in the sample	0.005
	Bananas	Concentration as determined in the sample	0.005
	Grapes	Concentration as determined in the sample	0.028
	Pears	Concentration as determined in the sample	0.006
	Peaches and nectarines	Concentration as determined in the sample	0.006
	Kiwis	Concentration as determined in the sample	0.005
	Plums	Concentration as determined in the sample	0.005
	Berries	Concentration as determined in the sample	0.008
	Other fruit	Concentration as determined in the sample	0.007
	Canned fruit	Concentration as determined in the sample	0.055
	Melons	Concentration as determined in the sample	0.005
	Olives	Concentration as determined in the sample	0.456
	Smoothies	Concentration as determined in the sample	0.141
	Dried raisins/sultanas/currants	Concentration as determined in the sample	0.222
	Apricots, prunes, and other dried fruit	Concentration as determined in the sample	0.064
	Figs and dates	Concentration as determined in the sample	0.089
	Glacé cherries and mixed peel	Concentration as determined in the sample	0.413

Category	Description	Comment	Fluoride mg/kg
NUTS AND SEEDS	Tree nuts	Concentration as determined in the sample	0.053
	Peanuts (roasted salted)	Concentration as determined in the sample	1.257
	Seeds	Concentration as determined in the sample	0.045
HERBS AND SPICES	Herbs	Concentration as determined in the sample	0.437
	Spices	Concentration as determined in the sample	0.325
MEAT	Pork	Concentration as determined in the sample	0.005
	Ham	Concentration as determined in the sample	0.126
	Pork sausage	Concentration as determined in the sample	0.157
	Bacon rashers	Concentration as determined in the sample	0.049
	Beef	Concentration as determined in the sample	0.005
	Beef mince	Concentration as determined in the sample	0.027
	Beefburger	Concentration as determined in the sample	0.11
	Chicken	Concentration as determined in the sample	0.005
	Turkey	Concentration as determined in the sample	0.018
	Lamb	Concentration as determined in the sample	0.057
	Offal (kidney)	Concentration as determined in the sample	0.198
	Offal (liver)	Concentration as determined in the sample	0.005
	Pudding, black	Concentration as determined in the sample	0.102
	Pudding, white	Concentration as determined in the sample	0.202
	Liver pâté (pork/chicken/duck)	Concentration as determined in the sample	0.024
FISH	Cod	Concentration as determined in the sample	2.175
	White fish (other than cod)	Concentration as determined in the sample	0.533
	Oily fish other than salmon	Concentration as determined in the sample	1.168
	Salmon	Concentration as determined in the sample	0.036
	Tinned tuna	Concentration as determined in the sample	0.341
	Tinned fish (excluding salmon and tuna)	Concentration as determined in the sample	5.8
	Tinned salmon	Concentration as determined in the sample	4.511
	Mussels	Concentration as determined in the sample	0.714
	Prawns	Concentration as determined in the sample	1.659
	Crab	Concentration as determined in the sample	0.474
	Fish fingers	Concentration as determined in the sample	0.159
SOUPS, SAUCES AND CONDIMENTS	Stock cubes, concentrated savoury spreads and essences	Concentration as determined in the sample	0.365
	Soup (fresh) (Tetra Paks and tubs)	Concentration as determined in the sample	0.109
	Soups (canned)	Concentration as determined in the sample	0.115
	Soups (dried packet)	Concentration as determined in the sample	0.56

Category	Description	Comment	Fluoride mg/kg
	Tomato sauce	Sample result unreliable and excluded from analysis	0
	Mayonnaise	Concentration as determined in the sample	0.014
	Gravy	Concentration as determined in the sample	0.129
	Cook-in sauces (other)	Concentration as determined in the sample	0.299
	Cook-in sauces (tomato-based)	Concentration as determined in the sample	0.097
	Dry sauces	Concentration as determined in the sample	0.455
	Other sauces and condiments	Sample result unreliable and excluded from analysis	0
	Soy sauce	Sample result unreliable and excluded from analysis	0.359
	Mustard	Sample result unreliable and excluded from analysis	0
	Salad dressings	Sample result unreliable and excluded from analysis	0
	Pickled vegetables	Sample result unreliable and excluded from analysis	0
	Pickles, chutneys and relish	Sample result unreliable and excluded from analysis	0
CONFECTIONERY	Chocolate confectionery	Concentration as determined in the sample	0.034
	Non-chocolate confectionery	Concentration as determined in the sample	0.137
	Plain chocolate	Concentration as determined in the sample	0.022
	Cocoa powder	Concentration as determined in the sample	0.005
	Drinking chocolate powder	Concentration as determined in the sample	0.163
	Chocolate/nut spreads	Concentration as determined in the sample	0.018
SUGAR/PRESERVES	Sugar	Concentration as determined in the sample	0.005
	Marmalade	Concentration as determined in the sample	0.05
	Jam	Concentration as determined in the sample	0.033
	Honey	Concentration as determined in the sample	0.605
	Sugar syrups	Concentration as determined in the sample	0.052
	Jelly	Concentration as determined in the sample	0.641
BEVERAGES	Tap water	Average fluoride concentration in fluoridated tap water in Ireland	0.65
	Lager	Concentration as determined in the sample	0.125
	Stout	Concentration as determined in the sample	0.176
	Wine (white/red/rosé	Concentration as determined in the sample	0.181
	Spirits	Concentration as determined in the sample	0.033
	Cider	Concentration as determined in the sample	0.135
	Carbonated soft drinks	Concentration as determined in the sample	0.079
	Soft drinks (wine substitutes, etc.)	Concentration as determined in the sample	0.155

Category	Description	Comment	Fluoride mg/kg
	Sports drinks	Concentration as determined in the sample	0.18
	Squashes	Concentration as determined in the sample	0.169
	Apple juice (fresh)	Concentration as determined in the sample	0.027
	Orange juice (fresh)	Concentration as determined in the sample	0.01
	Apple juice (from concentrate)	Concentration as determined in the sample	0.46
	Orange juice (from concentrate)	Concentration as determined in the sample	0.473
	Fruit nectars	Concentration as determined in the sample	0.295
	Cranberry juice	Concentration as determined in the sample	0.475
	Multivitamin juices	Concentration as determined in the sample	0.344
	Other fruit juices	Concentration as determined in the sample	0.23
	Tea (average)	Combination of average concentration as determined in 10 samples plus tap water average	5.405
	Instant coffee	Sample result unreliable and excluded from analysis	0
	Percolated coffee	Sample result unreliable and excluded from analysis; only the tap water proportion included	0.65
	Herbal tea	Combination of concentration as determined in sample plus tap water average	2.299
	Bottled water	Concentration as determined in the sample	0.196
FATS/OILS	Olive oil	Concentration as determined in the sample	0.005
	Vegetable oil	Concentration as determined in the sample	0.005
	Animal fat	Concentration as determined in the sample	0.005
SNACKS	Crisps	Concentration as determined in the sample	0.191
	Other savoury snacks	Concentration as determined in the sample	0.237
OTHER	Vinegar	Sample result unreliable and excluded from analysis	0.005
COMPOSITE FOOD	Pizza	Concentration as determined in the sample	0.208
	Rice pudding	Concentration as determined in the sample	0.052
INFANT FOODS	Infant formula (powder)	Proportional combination of concentration as determined in sample plus tap water average	0.592
	Infant formula (dairy alternatives) (powder)	Proportional combination of concentration as determined in sample plus tap water average	0.622
	Growing-up milk	Proportional combination of concentration as determined in sample plus tap water average	0.576
	Infant-specific beverages	Concentration as determined in the sample	0.14
	Infant food (meat-based)	Concentration as determined in the sample	0.208
	Infant food (fish-based)	Concentration as determined in the sample	0.172
	Infant food (fruit-based)	Concentration as determined in the sample	0.116

Category	Description	Comment	Fluoride mg/kg
	Infant food (vegetable-based)	Concentration as determined in the sample	0.23
	Infant food (cereal-based)	Concentration as determined in the sample	0.143
	Infant food (dairy-based)	Concentration as determined in the sample	0.141
	Baby snacks	Concentration as determined in the sample	0.203

APPENDIX IV MANAGEMENT OF FOOD CONSUMPTION DATA

All food consumption data collected are stored in relational SPSS databases. The primary food consumption files (food files) provide food intake on an individual level, with each individual line in the files representing one single eating occasion recorded by the participant. Food items are coded following the food classification system as described in *McCance and Widdowson's The Composition of Foods* and supplementary editions (Holland *et al.,* 1988, 1989, 1991, 1992a, 1992b, 1993; Chan *et al.,* 1994, 1995, 1996; FSA 2002; MAFF 1998) and supplemented with additional national codes developed by the Irish Universities Nutrition Alliance (IUNA) for foods for which no matching code was available. In tandem with these surveys, information on recipes and food ingredients was also collected when available and entered into two databases as follows:

- **Recipe database**

The recipe database contains information on constituent ingredients of composite foods recorded in the consumption surveys. Contribution to the total weight of the composite food is recorded for each ingredient based either on recipes as recorded by the survey participants or standard recipe information available from *McCance and Widdowson's The Composition of Food*s and supplementary editions (Holland *et al.,* 1988, 1989, 1991, 1992a, 1992b, 1993; Chan *et al.,* 1994, 1995, 1996; FSA 2002; MAFF 1998).

- **Irish National Food Ingredients Database**

The Irish National Food Ingredients Database (INFID) was established in 1999 by researchers at University College Dublin (UCD) and University College Cork (UCC) and includes data on food additives, packaging type and ingredient data for brand foods consumed by food consumption survey participants. Since then, updated versions accompanying each new food consumption survey have become available and are to be used in tandem with the survey for which they were collected (IUNA, 1999; IUNA, 2006b; IUNA, 2010; IUNA, 2012a).

Modification of the food consumption database

For the purposes of estimating exposure to fluoride, the food files were manipulated to convert the food consumption data as described below. The final food files used in the exposure assessment models comprised intake data from food consumed as such and composite food disaggregated into ingredients.

Disaggregation of recipes/composite foods into ingredients

Recipes or composite dishes were disaggregated into single ingredients using the recipe database. Where no recipe was available for a particular composite food, recipe fractions were either derived based on comparable composite foods for which information was available, extrapolated from the list of ingredients using the INFID or derived from ingredient information available on the labels of pre-packaged food sold in Ireland.

Table A3 provides an example of the composite food 'Apple Slices/Lattice', which contains seven ingredients, of which ingredient number 1 contains a further six sub-ingredients.

Table A3: Example of disaggregated composite food

Food code	Ingredient number	Sub-ingredient number	Ingredient (Food code)	Sub-ingredient (Food code)
APPLE SLICES/ LATTICE/ TURNOVER	1	1	Flaky pastry, raw	Wheat flour, white, plain
	1	2	Flaky pastry, raw	Margarine
	1	3	Flaky pastry, raw	Butter
	1	4	Flaky pastry, raw	Salt
	1	5	Flaky pastry, raw	Water
	1	6	Flaky pastry, raw	Lemon juice, fresh
	2	–	Wheat flour, white, plain	n/a
	3	–	Cooking apples, raw, peeled	n/a
	4	–	Sugar, white	n/a
	5	–	Cinnamon, ground	n/a
	6	–	Lemon juice, fresh	n/a
	7	–	Eggs, chicken, whole, raw	n/a

On finalisation of this procedure, rigorous quality checks were performed on all data to test against coding and weight conversion errors. This included reaggregation of the entire database to compare aggregated weight for each food code (addition of recipe ingredient weights) against the weight recorded in the original database, and the checking of aggregated recipe information (food codes and ingredient fractions) against information held in the recipe databases.

Application of weight loss factors

Heat treatment of food, such as frying, baking, boiling, etc., may result in a change of composition, in particular gain or loss of water and gain or loss of fat. The change in water status is usually covered by weight loss or weight gain factors to be applied in tandem with recipe fractions, and recipe ingredients are typically recorded in their raw state (see Table A4).

Table A4: Example of recipe with recorded weight loss factor

Composite food	Raw ingredients	Ingredient portion (g)	Raw ingredient weight (g)	Ingredient fraction	Weight loss after cooking (%)
HOMEMADE LENTIL, ONION AND CARROT SOUP	Lentils, red, split, dried, raw	250	1720	0.145	0.28
	Onions, raw	250	1720	0.145	0.28
	Carrots, old, raw	420	1720	0.244	0.28
	Stock cubes, chicken	5	1720	0.003	0.28
	Water	795	1720	0.462	0.28

APPENDIX V EXPOSURE FUNCTION

Exposure to fluoride was calculated by multiplying the concentration (o_f) for each food group (f belonging to the F set of food groups) by the respective consumption amount ($c_{f,d,i}$) per kg body weight (bw_i) separately for each individual (i belonging to the I set of individuals) in the database, calculating the sum of exposure for each survey day (d belonging to the D_i set of days surveyed for an individual i) and then deriving the daily average for the survey period. (The operation $|D_i|$ represents the number of days in the survey for each individual.) Mean, P95 and P97.5 exposures were calculated for the total survey population separately for each survey.

The method used can be described according to the following equation calculating the individual exposure [($\overline{o}_f$) for the food group 'tea', (o_f) for all remaining food groups]:

$$\overline{e}_i = \frac{\displaystyle\sum_{d \in D_i} \sum_{f \in F} \overline{o}_f \cdot c_{f,d,i}}{|D_i| \cdot bw_i}$$

APPENDIX VI SUBSTITUTION MODEL RESULTS

As described in Section 2.5, a substitution model was used to estimate the potential impact of excluding samples for which results did not meet the required analytical quality criteria. For these samples, substitute values were derived from the literature and included in the substitution model as discussed in the following paragraphs.

Coffee

Substitute values for coffee were derived from Warren *et al.* (1996), who analysed 40 different brands and types of coffee obtained from supermarkets and specialty coffee stores. As in the present study, coffee was prepared using deionised distilled water, using comparable amounts (data provided in cubic centimetres, which was converted using Food and Agriculture Organization (FAO) density database factors (FAO, 2012).

- Fluoride 'Instant coffee' substitute value: 14.7 mg/kg in powder [based on 0.3 mg/kg fluoride in a made up sample containing 15 cc coffee (=3.75 g at a density of 0.25g instant coffee/cc) per 180 mL distilled water]
- Fluoride 'Percolated coffee' substitute value: 0.82 mg/kg in coffee as consumed [0.17 mg/kg in percolated coffee (based on sample containing 15 cc coffee (=5.25 at a density of 0.35g roasted beans/cc) per 180 ml distilled water) + 0.65 mg/kg fluoride from tap water]

Sauces

Substitute values for sauces were derived from the USDA National Fluoride Database (USDA, 2005):

- Tomato sauce: 0.15 mg/kg
- Other sauces and condiments: 0.3 mg/kg
- Mustard: 0.01 mg/kg
- Salad dressings: 0.27 mg/kg
- Pickled vegetables: 0.3 mg/kg
- Pickles, chutneys and relish: 0.3 mg/kg

Results

As can be seen from Table A5, the results indicate that the exclusion of certain samples from the exposure assessment had no impact on the total dietary exposure to fluoride, which is in the main influenced by consumption of tea and water (see Section 3.3).

Table A5: Comparison of total dietary fluoride exposure results (µg/kg bw/day) derived using the standard versus the substitution model

Exposure calculation	Food category	Food group	Total population			% of total diet
			µg/kg bw/day			
			Mean	P95	P97.5	
ADULTS (18+ years)						
Excluding substitute values	Soups, sauces and condiments	Tomato sauce				
		Other sauces/condiments				
		Mustard				
		Salad dressings				
		Pickled vegetables				
		Pickles, chutneys and relish				
	Beverages	Instant coffee				
		Percolated coffee*	0.26	1.68	2.34	0.66
	Total exposure	**Total diet**	**40.27**	**94.08**	**111.35**	**100**
Including substitute values	Soups, sauces and condiments	Tomato sauce	0.003	0.02	0.03	0.01
		Other sauces/condiments	0.001	0.001	0.01	0.002
		Mustard	0.00002	0.0001	0.0002	0.00005
		Salad dressings	0.0001	0	0	0.0004
		Pickled vegetables	0.01	0.02	0.06	0.01
		Pickles, chutneys and relish	0.002	0.01	0.03	0.00
	Beverages	Instant coffee	0.27	1.13	1.51	0.65
		Percolated coffee	0.33	2.11	2.95	0.82
	Total exposure	**Total diet**	**40.62**	**94.27**	**111.49**	**100**
CHILDREN (5–12 years)						
Excluding substitute values	Soups, sauces and condiments	Tomato sauce				
		Other sauces/condiments				
		Mustard				
		Pickled vegetables				
		Pickles, chutneys and relish				
	Beverages	Instant coffee				
		Percolated coffee*	0.004	0.001	0.01	0.02
	Total exposure	**Total diet**	**16.87**	**39.38**	**52.12**	**100**
Including substitute values	Soups, sauces and condiments	Tomato sauce	0.01	0.05	0.06	0.06
		Other sauces/condiments	0.002	0.003	0.01	0.01
		Mustard	0.00004	0.0002	0.0003	0.00
		Pickled vegetables	0.002	0.01	0.01	0.01
		Pickles, chutneys and relish	0.001	0	0	0.003
	Beverages	Instant coffee	0.01	0	0.07	0.06
		Percolated coffee	0.005	0.001	0.01	0.03
	Total exposure	**Total diet**	**16.89**	**39.40**	**52.12**	**100**

Exposure calculation	Food category	Food group	Total population			
			µg/kg bw/day			% of total diet
			Mean	**P95**	**P97.5**	
		PRESCHOOL CHILDREN (1–4 years)				
Excluding substitute values	Soups, sauces and condiments	Tomato sauce				
		Other sauces/condiments				
		Mustard				
		Pickled vegetables				
		Pickles, chutneys and relish				
	Beverages	Instant coffee				
		Percolated coffee*	0.00006	0	0	0.0003
	Total exposure	**Total diet**	**23.23**	**46.46**	**56.54**	**100**
Including substitute values	Soups, sauces and condiments	Tomato sauce	0.01	0.07	0.10	0.05
		Other sauces/condiments	0.002	0	0.01	0.01
		Mustard	0.00002	0.0001	0.0002	0.00
		Pickled vegetables	0.003	0	0.003	0.01
		Pickles, chutneys and relish	0.001	0	0	0.004
	Beverages	Instant coffee	0.0002	0	0	0.001
		Percolated coffee	0.0001	0	0	0.0003
	Total exposure	**Total diet**	**23.25**	**46.46**	**56.54**	**100**

* The Percolated coffee sample was prepared with distilled water. The figures shown represent the tap water part of the final food.

APPENDIX VII UNCERTAINTY ANALYSIS

In accordance with the guidance provided in the EFSA opinion related to uncertainties in dietary exposure assessment (EFSA, 2006), the following sources of uncertainties have been considered and are summarised in Table A6.

Table A6: Qualitative evaluation of the influence of uncertainties on the dietary exposure estimate

Sources of uncertainties	Direction of impact on fluoride exposure estimate
Consumption data: different underreporting/misreporting/portion size estimation	+/−
Use of data from food consumption survey of a few days to estimate long-term (chronic) exposure for high percentiles (95th and 97.5th percentiles)	+
Use of recipe fractions and weight loss factors in disaggregation of food	+/−
Extrapolation from foods for which analyte concentration data were available to comparable foods where no analyte concentration were available	+/−
Variability in fluoride occurrence in food	+/−
Assumption that all tap water recorded in the food consumption surveys (as consumed or as an ingredient of a disaggregated composite food) was fluoridated water	+
Use of upperbound (<LOD=LOD) concentration data	+
For eight foods, the analytical results did not conform with quality criteria and were not included in the exposure assessment	−
In the substitution model, concentration data for eight foods were used from external sources	+/−
Water used for boiling certain foods had a slightly elevated concentration of fluoride compared to average water fluoridation levels	+

(+): uncertainty with potential to cause overestimation of exposure.
(−): uncertainty with potential to cause underestimation of exposure.

The conservative approach applied to the exposure estimate for fluoride is likely to have led to an overestimation of the exposure.