

1 **Development of a database of the fluoride content of selected drinks and foods in the UK**

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18 **Abstract**

19 It is important to monitor systemic fluoride (F) intake from foods, drinks and inadvertent toothpaste
20 ingestion in order to minimise risk of dental fluorosis while maximising caries prevention. In the UK,
21 a F database containing the F content of commercially available foods and drinks was compiled from
22 518 products analysed using an acid-diffusion method and F-Ion-Selective Electrode. Individual
23 products analysed ranged from $<0.01\mu\text{gF}/100\text{g}$ for butter/margarine (Miscellaneous foods group) to
24 $1054.20\mu\text{gF}/100\text{g}$ for canned sardine (Fish group). These findings, along with the wide range of F
25 contents found within food groups highlight the need for comprehensive F content labelling of food
26 and drink products.

27 **Introduction**

28 Extensive topical use of fluorides has been well documented as an important reason for the global
29 decline in dental caries prevalence [Featherstone, 1999]. Conversely, excessive chronic systemic
30 exposure to fluorides - from multiple sources - has been suggested as the main reason for the rise in
31 the prevalence of dental fluorosis in both water-fluoridated and non- water-fluoridated communities
32 [Pendrys and Stamm, 1990; Szpunar and Burt, 1990; Beltran-Aguilar et al., 2010].

33 Diet including fluoridated waters, foods and drinks prepared with waters or which contain fluoride (F)
34 naturally, and inadvertent ingestion of F-containing dental products (primarily toothpastes) are the
35 main sources of systemically ingested F in children. Studies in children have shown that up to 70% of
36 total F intake could be from diet, including water [Levy et al., 2001; Levy et al., 2003; Maguire et al.,
37 2007; Zohoori et al., 2013]. Data from F intake studies of infants in the UK have indicated that diet
38 was the sole source of F intake for 87% of infants aged 1-12 months [Zohoori et al., 2014].

39 Regarding western diets, a positive association between powdered infant milk formula (IMF)
40 reconstituted with water and mild dental fluorosis prevalence has been demonstrated [Levy et al 2010,
41 Do et al 2012], indicating the effect of the F concentration of the water used in the preparation of IMF
42 as well as the effect of the F content of the IMF itself [McKnight-Hanes et al., 1988; Hujoel et al.,
43 2009]. Additionally it has been reported that certain Latino diet foods such as a meal of rice and beans
44 prepared with fluoridated water and soy-based processed infant foods could contribute up to 29% and
45 45%, respectively, of the threshold F dose of 0.07 mgF/kg body weight/day in terms of clinically
46 acceptable dental fluorosis [Casarin et al., 2007].

47 Information on total F intake is essential when planning effective community-based F therapy for the
48 prevention of dental caries and/or dental fluorosis. A F database containing information on the F
49 content of commonly consumed food and drink items is a pre-requisite to enable assessment of dietary
50 F intake at individual and/or community levels. In 2005, a F database of selected US beverage and
51 food samples was published by the US Department of Agriculture [U.S. Department of Agriculture,
52 2005], in which the mean F contents of 427 beverages and food samples were reported; the values for
53 F content of many items being taken from different data sources published between 1977 and 2003.
54 Over time, with enhancement of the analytical methods used for F analysis with improved detection
55 limits, as well as dietary changes towards consumption of more ready-to-eat food/drink products, a F
56 database needs to remain current and it is important that it reflects items commonly consumed by the
57 population it serves. A range of research projects on F intake of infants and young children, conducted
58 in the UK since 2003 [Zohoori et al., 2003; Zohoori et al., 2006; Maguire et al., 2007; Maguire et al.,
59 2012; Zohoori et al., 2012; Zohoori et al., 2013; Zohoori et al., 2014], provided the information and
60 opportunity to derive a current and comprehensive F database. This paper briefly describes the

61 analysis of the F contents of 518 different food and drink products, available on the UK market, and
62 subsequent development of the F database (Zohoori and Maguire 2015).

63 **Material and Methods**

64 The selection of food and drink items for F analysis was determined by collecting dietary information
65 from parents of infants and children younger than 7 years of age, using a validated 3-day food diary
66 [Zohoori et al., 2006; Maguire et al., 2007]. The food diaries were collected during studies conducted
67 between 2003 and 2014 [Zohoori et al., 2006; Maguire et al., 2007] and consequently analysed to
68 identify the most consumed food/drink items and the core contributors to dietary F intake in this age
69 group [Zohoori et al., 2003; Zohoori et al., 2006; Maguire et al., 2007; Maguire et al., 2012; Zohoori
70 et al., 2012; Zohoori et al., 2013; Zohoori et al., 2014; Omid et al., 2015].

71 The identified items of food and drink were purchased from all main supermarket chains, convenience
72 stores, grocers shops and other retail outlets in the north east of England. For each item, 30 samples in
73 total (ten different brands and for each brand three different batches) were purchased to cover any
74 possible within-product variability. The selection of the brands, for each item, was based on the
75 records in the food diaries (i.e. the ten most frequently recorded brands) and the selection of the
76 batches was based on the availability when the store was visited. Following the method used in
77 *McCance and Widowsen's 'The Composition of Foods'*, a commonly used UK food composition table
78 [Food Standard Agency, 2002], the samples of each individual brand and batch were not analysed
79 separately but were pooled before analysis. Each resultant composite sample therefore consisted of up
80 to 30 individual samples (i.e. 10 different brands x 3 batches/brand). Each composite was prepared by
81 mixing equal weights/volumes of each specific purchased product/brand together.

82 F concentration ($\mu\text{g/g}$) of each non-milk-based drink composite was measured, in triplicate, directly
83 using a fluoride-ion-selective electrode (Orion Research, model 96-09) after adding a total ionic
84 strength adjustment buffer [Martinez-Mier et al., 2004; Zohoori et al., 2006]. The F concentration
85 ($\mu\text{g/g}$) of each composite of milk and milk-based drinks as well as food samples and infant milk
86 formula was measured using the hexamethyldisiloxane (HMDS)-facilitated diffusion method
87 [Martinez-Mier et al., 2011; Zohoori et al., 2014]. F content per 100g of each sample ($\mu\text{g}/100\text{g}$) was
88 then calculated by multiplying F concentration ($\mu\text{g/g}$) by 100. Reliability and validity of the analytical
89 methods were confirmed by i) re-analysis of 10% of the samples and ii) measurement of recovery of a
90 known amount of F added to 10% of samples.

91 The food and drink samples were categorised in groups according to type; similar to the food and
92 drink groups used in *McCance and Widowsen's 'The Composition of Foods'*. As with most food
93 composition tables, the F values in the tables were quoted per 100g edible portion and the F content

94 was expressed as $\mu\text{gF}/100\text{g}$ of food. Descriptive analyses were used to report the mean (SD),
95 minimum, maximum and quartile F content values ($\mu\text{g}/100\text{g}$) for each group.

96 **Results:**

97 The overall mean (SD) recovery of F added to the 10% of randomly selected samples was 98.5%
98 (2.4%), representing good validity for the method of F analysis used. The correlation between analysis
99 and re-analysis (conducted on a separate day) of the 10% of randomly selected samples was strong (R
100 = 97%) indicating excellent reproducibility.

101 In total, 518 food/drink items were analysed, of which 251 items were infant food and drinks. The full
102 list of the 518 individual items with their F contents, is deliberately reported elsewhere [Zohoori and
103 Maguire, 2015; <http://tees.openrepository.com/tees/handle/10149/581272> and
104 http://eprint.ncl.ac.uk/pub_details2.aspx?pub_id=216711] in view of the restrictions associated with a
105 short communication. The compiled database shows a very wide range in F content both between and
106 within the different food and drink groups. The F contents of a range of categories of food and drinks,
107 “as purchased”, are summarised in Tables 1 and 2. In general, the data were not normally distributed
108 and the highest range in F content ($7.90\text{--}1054.20 \mu\text{g}/100\text{g}$) ($\sim 0.007 - 1.054 \text{ ppmF}$) was found for the
109 “fish and fish products” category for older children (Table 2) with a median of $10.54 \mu\text{g}/100\text{g}$ (\sim
110 0.010 ppm) and a mean of $148.70 \mu\text{g}/100\text{g}$ ($\sim 0.149 \text{ ppm}$). Across Tables 1 and 2, the minimum F
111 content was observed for butter/margarine within the “miscellaneous foods” category for older
112 children (Table 2). Among the categories for infant food and drink products (Table 1), the “cereal and
113 cereal products” category had the highest median F content ($19.55 \mu\text{g}/100\text{g} \sim 0.019 \text{ ppm}$), and the
114 lowest F content was found for the “bottled drinking waters” category with a median F content of 2.80
115 $\mu\text{g}/100\text{g}$ ($\sim 0.003 \text{ ppm}$). Among the categories of food and drink products for older children (Table 2),
116 the “fish and fish products” category contained the highest mean F content ($148.70 \mu\text{g}/100\text{g} \sim 0.149$
117 ppm), whereas the highest median F content was found for the “cereal and cereal products” category
118 ($13.10 \mu\text{g}/100\text{g} \sim 0.013 \text{ ppm}$). The lowest mean F content was found for the “vegetables, herbs and
119 spices” category ($6.61 \mu\text{g}/100\text{g} \sim 0.007 \text{ ppm}$), whereas “fruit and nuts” had the lowest median F
120 content ($2.55 \mu\text{g}/100\text{g} \sim 0.002 \text{ ppm}$).

121 **Discussion:**

122 Monitoring F intake in infancy and early childhood is necessary to ensure that total F intake from all
123 sources does not exceed certain thresholds and limits [Institute of Medicine, 1997] to minimise risk of
124 development of dental fluorosis due to chronic excessive ingestion of F. Depending on the age of the
125 child and their toothbrushing habits, diet may be the predominant source of F intake for some
126 children. This F database, which covers a substantial number of food and drink products and is based

127 on an analysis of F content of food/drink items actually consumed by children, provides a useful
128 information source for estimation of dietary F intake in children. Although the samples were
129 purchased in the UK, most were brands manufactured by leading companies in the European food
130 market [Maguire et al., 2012; Zohoori et al., 2012], therefore, the F content values are likely to be of
131 relevance to other European countries. To ensure the F database remains up-to-date, new products
132 will be analysed and added and the current items will be reanalysed periodically.

133 However, as with any food database, there are some limitations with its content. The F content of any
134 particular similar food or drink product might not be exactly as that recorded in the database because
135 of the natural variability in the nutrient composition of the product. Attempts were made to minimise
136 the impact of this “natural” variation by deriving the database from careful selection and analyses of
137 several representative samples of each product to cover variability in composition. In general, the
138 natural (intrinsic) variation in the nutrient composition of products may arise from, for example; age,
139 feeding routine and season which could influence the composition of animal products. Alternatively,
140 the country of origin and local growing conditions could affect the composition of plant products.
141 Additionally, the F content of a particular product might change due to the alteration in recipes,
142 amounts and types of ingredients, storage time and even composition of the cooking vessel. For
143 example, using aluminium pots for cooking can reduce the F content of food, whereas using Teflon-
144 coated vessels can increase the F content [Full and Parkins, 1975]. Methods of food preparation by
145 manufacturer and/or consumer can also change the F content of the products [Oelschlager, 1970;
146 Nanda, 1972]. The F contents of food/drink requiring water for preparation/cooking are also affected
147 by the F concentration of the water used for preparation. Although the focus of this report was on the
148 F contents of products “as purchased”, the full database [Zohoori and Maguire 2015] provides the F
149 contents of the 518 individual items “as purchased” as well as the 71 items that required water for
150 preparation/cooking before consumption (e.g. vegetables/rice/pasta boiled in water; reconstituted
151 infant milk formula and gravy instant granules, made up with water) which confirms the effect of F
152 concentration of water on the F contents of the prepared food/drink.

153 An increase in F content of a dish through the inclusion of animal or fish bone fragments or bone dust
154 during preparation [Walters et al., 1983] has been confirmed in the compiled database [Zohoori and
155 Maguire, 2015] in which the highest F content (1054.20 µgF/100g) was found for sardines, canned in
156 brine. The very wide range in the concentrations of F between and within the different food groups,
157 reported in the UK database [Zohoori and Maguire 2015], is in agreement with the wide range of F
158 values reported in the US F database [U.S. Department of Agriculture, 2005].

159 Another limitation of use of any nutrient database, when using it to correlate intake with development
160 of a condition (in this case, dental fluorosis) is that the total content of F in a particular food ingested

161 may not be a true indication of the systemically absorbed amount. Several physiologically-related
162 factors, such as age and, for children and young adults, their stage of development (e.g. whether they
163 are undergoing a period of active growth) can influence absorption and retention of F in the body
164 [Whitford, 1996]. In addition, the bioavailability of F can be influenced by dietary-related factors
165 which include the physical and chemical form of F in a food and its solubility as well as the presence
166 of enhancers or inhibitors of F absorption. For example, when F, as NaF, is ingested with water,
167 almost 100% is absorbed but when it is taken with milk or infant formula or with foods containing
168 divalent or trivalent cations (such as calcium), the degree of F absorption is decreased [Whitford,
169 1996]. A mixed diet may reduce F absorption by 47% [Ekstrand and Ehrnebo, 1979; Shulman and
170 Vallejo, 1990], whereas a diet high in fat content may increase F absorption due to a reduction in the
171 rate of gastric emptying [Whitford, 1996].

172 A wide variation in the bioavailability of F from different individual foods has been reported in
173 human adults; 12% for fish-bone meal, 23% for canned sardines, 24% for chicken-bone meal, 79%
174 for Krill and 89% for tea [Trautner and Siebert, 1986]. Absorption of F given with milk can be
175 reduced by 13-30% compared with F absorption when given with water [Spak et al, 1982; Shulman
176 and Vallejo, 1990]. In children, F bioavailability from fluoridated milk given to children and from
177 infant milk formula reconstituted with water has been reported to be 72% and 65%, respectively
178 [Spak et al, 1982].

179 In conclusion, a F database, such as the one introduced here could be a useful tool to facilitate
180 monitoring of dietary F intake in children and therefore to help maximise the oral health benefits of
181 fluorides while minimising the risk of development of dental fluorosis. The wide range of F contents
182 found within groups and the potential for natural (e.g. product origin) and extrinsic (e.g. alteration in
183 recipe) variations in the composition of each individual sample highlights the need for comprehensive
184 F labelling of food and drink products, particularly those targeted for use by infants and very young
185 children. Improved labelling will help consumers make more informed choices to help ensure an
186 optimal F intake.

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194 **Author contributions:**

195 FVZ and AM designed the studies and supervised the laboratory analysis of samples; FVZ and AM
196 drafted the paper.

197

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273 http://eprint.ncl.ac.uk/pub_details2.aspx?pub_id=216711) accessed 4th April 2016.

274

275 Table 1. Mean (SD), minimum, maximum and quartile distributions of F contents ($\mu\text{g}/100\text{g}$) of 251 infant food and drink products, as purchased across 9
 276 categories¹.

Food group	Number of products	F content ($\mu\text{g}/100\text{g}$)					Maximum
		Mean (SD)	Minimum	25 th Percentile	50 th Percentile	75 th Percentile	
Cereal and cereal products	50	36.53 (40.72)	3.00	11.00	19.55	51.47	171.00
Bottled drinking waters*	7	4.11 (2.17)	2.20	2.20	2.80	6.80	6.90
Fish and fish products	7	14.89 (4.78)	10.50	10.67	13.60	18.30	22.93
Fruits and nuts	22	15.21 (25.52)	2.00	3.55	5.95	13.15	117.60
Infant milk formula	29	7.76 (7.64)	0.90	1.60	3.00	15.15	25.20
Meat products and dishes	64	15.64 (15.49)	4.27	9.40	12.13	15.57	120.00
Milk products	16	10.08 (9.52)	3.30	3.82	5.60	14.02	37.38
Miscellaneous foods	29	22.28 (27.87)	3.00	5.90	10.00	24.95	103.40
Vegetable dishes	27	12.62 (6.07)	4.00	7.60	11.05	17.32	30.90

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278 ¹ Zohoori and Maguire, 2015. URLs: <http://tees.openrepository.com/tees/handle/10149/581272> and http://eprint.ncl.ac.uk/pub_details2.aspx?pub_id=216711

279 * Products which were specifically labelled for infants; including plain and flavoured drinking water.

280

281 Table 2. Mean (SD), minimum, maximum and quartile distributions of F contents ($\mu\text{g}/100\text{g}$) of 267 food and drink products as purchased, consumed by
 282 children, across 9 categories¹

Food group	Number of products	F content ($\mu\text{g}/100\text{g}$)					
		Mean (SD)	Minimum	25 th Percentile	50 th Percentile	75 th Percentile	Maximum
Cereal and cereal products	62	21.58 (20.00)	3.30	8.74	13.10	26.47	75.30
Fish and fish products	13	148.70 (297.80)	7.90	8.07	10.54	203.35	1054.20 ⁱ
Fruits and nuts	26	6.90 (9.63)	0.60	1.87	2.55	7.55	45.00
Meat products and dishes ^a	26	11.40 (11.62)	1.60	2.80	6.91	15.50	49.40
Meat, poultry and game ^b	15	7.91 (8.09)	2.50	3.10	3.70	6.00	23.50
Milk products and eggs	43	10.46 (13.65)	0.05	0.88	2.80	23.00	58.50
Miscellaneous foods ^c	56	13.17 (20.56)	<0.01 ⁱⁱ	2.85	6.65	13.61	90.00
Vegetables dishes	9	10.71 (11.53)	0.67	0.73	9.10	19.90	31.40
Vegetables, herbs and spices	17	6.61 (6.43)	0.65	1.02	5.10	11.10	23.80

283

284 ¹ Zohoori and Maguire, 2015. URLs: <http://tees.openrepository.com/tees/handle/10149/581272> and http://eprint.ncl.ac.uk/pub_details2.aspx?pub_id=216711

285 ^a This group represents the dishes which contain meat and other ingredients such as sausages, Bolognese sauce, beef in sauce with vegetables etc

286 ^b This group represents the dishes which mainly contain meat such as steak, chicken thigh, oxtail etc

287 ^c This group includes “fat and oils”, “sugar, preserves and confectionery”, “beverages”, “soups, sauce and pickles” etc

288 ⁱ Canned sardine;

289 ⁱⁱ Butter/margarine