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

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14   **Declaration of interests**

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 <0.01µgF/100g for butter/margarine (Miscellaneous foods group) to

24   1054.20µgF/100g 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.

Introduction

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

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

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

Information on total F intake is essential when planning effective community-based F therapy for the prevention of dental caries and/or dental fluorosis. A F database containing information on the F content of commonly consumed food and drink items is a pre-requisite to enable assessment of dietary F intake at individual and/or community levels. In 2005, a F database of selected US beverage and food samples was published by the US Department of Agriculture [U.S. Department of Agriculture, 2005], in which the mean F contents of 427 beverages and food samples were reported; the values for F content of many items being taken from different data sources published between 1977 and 2003. Over time, with enhancement of the analytical methods used for F analysis with improved detection limits, as well as dietary changes towards consumption of more ready-to-eat food/drink products, a F database needs to remain current and it is important that it reflects items commonly consumed by the population it serves. A range of research projects on F intake of infants and young children, conducted in the UK since 2003 [Zohouri et al., 2003; Zohoori et al., 2006; Maguire et al., 2007; Maguire et al., 2012; Zohoori et al., 2012; Zohoori et al., 2013; Zohoori et al., 2014], provided the information and 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 [Zohouri 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 Widowson'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 (μg/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   (μg/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   [Martínez-Mier et al., 2011; Zohoori et al., 2014]. F content per 100g of each sample (μg/100g) was

88   then calculated by multiplying F concentration (μg/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 Widowson'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 µgF/100g of food. Descriptive analyses were used to report the mean (SD),

95   minimum, maximum and quartile F content values (µg/100g) 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 (7.90-1054.20 µg/100g) (~ 0.007 – 1.054 ppmF) was found for the

109   "fish and fish products" category for older children (Table 2) with a median of 10.54 µg/100g (~

110   0.010 ppm) and a mean of 148.70 µg/100g (~ 0.149 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 µg/100g ~ 0.019 ppm), and the

114   lowest F content was found for the "bottled drinking waters" category with a median F content of 2.80

115   µg/100g (~ 0.003 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 µg/100g ~ 0.149

117   ppm), whereas the highest median F content was found for the "cereal and cereal products" category

118   (13.10 µg/100g ~ 0.013 ppm).The lowest mean F content was found for the "vegetables, herbs and

119   spices" category (6.61 µg/100g ~ 0.007 ppm), whereas "fruit and nuts" had the lowest median F

120   content (2.55 µg/100g ~ 0.002 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.

194 **Author contributions:**

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

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275   Table 1. Mean (SD), minimum, maximum and quartile distributions of F contents (µg/100g) of 251 <u>infant</u> food and drink products, as purchased, across 9

276   categories[1].

Food group	Number of products	F content (µg/100g)					
		Mean (SD)	Minimum	25th Percentile	50th Percentile	75th Percentile	Maximum
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.

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281 Table 2. Mean (SD), minimum, maximum and quartile distributions of F contents (µg/100g) of 267 food and drink products as purchased, <u>consumed by</u>

282 <u>children</u>, across 9 categories[1]

Food group	Number of products	F content (µg/100g)					
		Mean (SD)	Minimum	25th Percentile	50th Percentile	75th 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

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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