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

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Running title: Fluoride database

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Declaration of interests

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Abstract

It is important to monitor systemic fluoride (F) intake from foods, drinks and inadvertent toothpaste ingestion in order to minimise risk of dental fluorosis while maximising caries prevention. In the UK, a F database containing the F content of commercially available foods and drinks was compiled from 518 products analysed using an acid-diffusion method and F-Ion-Selective Electrode. Individual products analysed ranged from <0.01µgF/100g for butter/margarine (Miscellaneous foods group) to 1054.20µgF/100g for canned sardine (Fish group). These findings, along with the wide range of F contents found within food groups highlight the need for comprehensive F content labelling of food 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
analysis of the F contents of 518 different food and drink products, available on the UK market, and subsequent development of the F database (Zohoori and Maguire 2015).

**Material and Methods**

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

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

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

The food and drink samples were categorised in groups according to type; similar to the food and drink groups used in McCance and Widowsen’s ‘The Composition of Foods’. As with most food composition tables, the F values in the tables were quoted per 100g edible portion and the F content...
was expressed as µgF/100g of food. Descriptive analyses were used to report the mean (SD), minimum, maximum and quartile F content values (µg/100g) for each group.

**Results:**

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

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

**Discussion:**

Monitoring F intake in infancy and early childhood is necessary to ensure that total F intake from all sources does not exceed certain thresholds and limits [Institute of Medicine, 1997] to minimise risk of development of dental fluorosis due to chronic excessive ingestion of F. Depending on the age of the child and their toothbrushing habits, diet may be the predominant source of F intake for some children. This F database, which covers a substantial number of food and drink products and is based
on an analysis of F content of food/drink items actually consumed by children, provides a useful information source for estimation of dietary F intake in children. Although the samples were purchased in the UK, most were brands manufactured by leading companies in the European food market [Maguire et al., 2012; Zohoori et al., 2012], therefore, the F content values are likely to be of relevance to other European countries. To ensure the F database remains up-to-date, new products will be analysed and added and the current items will be reanalysed periodically.

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

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

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

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

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

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Author contributions:

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


Zohoori, FV; Maguire, A (2015): Database of the Fluoride (F) content of Selected Drinks and Foods in the UK. Teesside University and Newcastle University (URLs:
http://tees.openrepository.com/tees/handle/10149/581272 and
Table 1. Mean (SD), minimum, maximum and quartile distributions of F contents (µg/100g) of 251 infant food and drink products, as purchased across 9 categories¹.

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


* Products which were specifically labelled for infants; including plain and flavoured drinking water.
Table 2. Mean (SD), minimum, maximum and quartile distributions of F contents (µg/100g) of 267 food and drink products as purchased, consumed by children, across 9 categories

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


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

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

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