

## FLUORIDE CONSUMPTION: THE EFFECT OF WATER FLUORIDATION

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**SUMMARY:** Complete fluoride intake data from the 2000-2003 UK National Diet and Nutrition Survey were used to examine questions outstanding from the Fluoridation of Water Supplies review process. In a quarter of the population, fluoride consumption from all sources exceeds the Safe Intake defined by the Committee on the Medical Aspects of Food Policy, regardless of water fluoride concentration. In areas supplied with fully fluoridated water, fluoride intake exceeds the recommended safe maximum in nearly two thirds of consumers. The implications are discussed and recommendations are made.

Keywords: Fluoride intake; Safe Intake of fluoride; UK National Diet and Nutrition Survey.

### INTRODUCTION

The York University *Systematic Review of Water Fluoridation*<sup>1</sup> identified a need for information about consumption of fluoride from all sources. During the consequent Medical Research Council (MRC) Working Party review, the results of the 2000 National Diet and Nutrition Survey (NDNS)<sup>2</sup> were awaited. This periodic survey of a random sample of people in England and Scotland included for the first time a 24-hour urine collection in which fluoride concentration was determined. The MRC working group commented: “Additional recommendations for future research will depend to some extent on whether results (from the NDNS urinary fluoride analyses) are in line with existing estimates of total fluoride intake.”<sup>3</sup> The MRC Working Party review panel suggested, whatever the outcome from the NDNS, that:

- periodic 24-hour urinary fluoride sampling should remain a feature of at least some national diet surveys
- fluoride ingestion (from all sources) . . . and fluoride retention should be measured in children
- the relative importance of water as a source of fluoride ingestion in children should be determined.

### REVIEW METHOD

As a member of both the Advisory Panel to the original systematic review, and of the MRC Working Group, the present author obtained and re-analysed the raw data on urinary analytes from the NDNS.<sup>3</sup>

An error, implying that all ingested fluoride is excreted in urine so that fluoride excretion can be taken to be equal to fluoride intake, came to light in the interpretation of urinary fluoride concentration,<sup>4</sup> which The Food Standards Agency acknowledged and corrected.<sup>5</sup> The erratum noted this was incorrect as in adults only ca. 50% of ingested fluoride is excreted in urine.<sup>6</sup> Therefore the estimate that 1% of men and 3% of women had intakes above 0.05 mg F/kg/day,

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the Safe Intake level set out by the Committee on the Medical Aspects of Food Policy for the UK, in its 1991 Dietary Reference Values report as amended in 1994, was likely to be too low. Two more recent reports giving higher upper limits for fluoride intake were also noted in the erratum: in 1997 the US Institute of Medicine recommended an upper limit of 10 mg F/day for adults and children aged nine and over,<sup>7</sup> and in 2006 the European Food Safety Authority recommended an upper limit of 0.12 mg F/kg/day for adults and children aged nine and over, equivalent to an upper limit of 7 mg F/day for a 60-kg adult.<sup>8</sup> On a body weight basis the European report proposed tolerable upper intake levels for fluoride for children aged 9–14 years of 5 mg F/day and for those aged 15 years and older of 7 mg F/day.

The value for the percentage of ingested fluoride that is excreted in urine used in the erratum of ca. 50% has been used in recalculating the proportion of subjects in the NDNS who have a fluoride intake exceeding the safe level.<sup>6</sup> This figure of 50% is given in a 1993 National Research Council publication, *Health Effects of Ingested Fluoride*, where it is noted that “Approximately 75–90% of the fluoride ingested each day is absorbed from the alimentary tract. Because of its chemical affinity for calcium compounds, about half of that fluoride becomes associated with teeth and bones within 24 hours of ingestion. In growing children, even more of the ingested fluoride is retained because of the large surface area provided by numerous and loosely organized bone crystallites. The remaining fluoride is eliminated almost exclusively by the kidneys, and the rate of renal clearance is directly related to urinary pH. As a result, diet, drugs, metabolism, and other factors can affect the extent to which fluoride is retained in the body.”<sup>9</sup>

Tap water fluoride concentration, which had been measured for most of the NDNS sample, had not been further analysed by Henderson et al.,<sup>6</sup> but the original data were obtained by courtesy of the Chief Dental Officer and merged with the urine data. The sample populations were compared with the provisional findings for the 2001 Census, stratified to match the sampling frame of the NDNS. The consolidated data were then tabulated according to total fluoride consumption and water fluoride concentration.

## RESULTS

### *Excessive fluoride intake*

The NDNS sample of 1725 subjects who completed a food diary was confirmed not to be significantly different in respect of age and gender from the expected population reconstructed from the 2001 Census ( $\chi^2$  11.37 with 7 degrees of freedom,  $p > 0.1$ ).

Fewer males aged 35–49 had completed diaries than had provided urine collections, so the two samples were somewhat separate. The 1459 subjects for whom urine data were complete, and the 1395 for which both urine data and tap water fluoride were known, both differed very significantly from the interview sample ( $\chi^2$  80 with 7 degrees of freedom,  $p < 0.001$ ). Men up to age 34 were

under-represented, in favour of those aged 35–49. The difference from census expectation was less marked ( $\chi^2$  54) but still highly significant.

The 1395 subjects providing both urine and tap water samples were no different from the 1459 providing urine only ( $\chi^2$  0.197,  $p > 0.995$ ). Because of the way the samples were identified it was not possible for the author to check the age or gender of 170 individuals providing tap water but not a urine sample.

A corrected version of Table 4.4 in Volume 3 of the NDNS is given in Table 1, using the higher NDNS value of Safe Intake (SI) only for direct comparability. The author disputes this threshold, 3  $\mu\text{M}$  F/kg/day (0.057 mg F/kg/day), for which no authority is cited and which is 14% higher than the value of 0.05 mg F/kg/day established by the Committee on the Medical Aspects of Food Policy (CoMA) for the UK.<sup>10</sup>

**Table 1.** Proportion of National Diet and Nutrition Survey (NDNS) urine collection subjects consuming more than 3  $\mu\text{M}$  F/kg/day (0.057 mg F/kg/day), the Safe Intake (SI) specified in the NDNS, if the following conditions, proposed by the author (PM) based on the current literature,<sup>9,13</sup> apply: 90% of ingested fluoride is assimilated, 50% of that is sequestered in calcified tissues, and only the remainder is excreted in the urine

Age/gender group	No. $\leq$ SI (NDNS)	No. $>$ SI (NDNS)	Total	% $>$ SI (NDNS)
Male 19–24	39	4	43	9.3
Male 25–34	114	11	125	8.8
Male 35–49	224	48	272	17.6
Male 50–64	152	52	204	25.5
All males	529	115	644	17.9
Female 19–24	56	5	61	8.2
Female 25–34	136	31	167	18.6
Female 35–49	240	77	317	24.3
Female 50–64	180	60	240	25.0
All females	612	173	785	22.0
Total	1141	288	1429	20.2

This finding, that the Safe Intake is exceeded by a total of 20.2% (17.9% for males and 22.0% for females) is an order of magnitude higher than the figures (1% for males, 3% for females) published in the original NDNS report. But the proportion of subjects consuming more than the lower, better-established CoMA definition of Safe Intake was higher still, at 25.1% (Table 2).

### *Relevance of water fluoride*

Data from 1373 subjects were available for cross-tabulation of tap water fluoride concentration and daily fluoride consumption. Of these, only 75 lived in fully fluoridated areas of the West Midlands, with water supplies containing 0.8 parts per million fluoride (ppm F) or more. Another 38 received water at around 0.4 ppm F, mainly in North Tyneside. The remaining 1260 subjects received tap water of lower fluoride concentration.

Table 2 summarises total fluoride intake in the sample ranked according to tap water F concentration.

**Table 2.** Proportion of National Diet and Nutrition Survey (NDNS) urine collection subjects consuming more than 0.05 mg F/kg/day, the Safe Intake (SI) specified by the Committee on the Medical Aspects of Food Policy for the UK (CoMA), if the following conditions, proposed by the author (PM) based on the current literature,<sup>9,13</sup> apply: 90% of ingested fluoride is assimilated, 50% of that is sequestered in calcified tissues, and only the remainder is excreted in the urine, with the total fluoride intake in the sample ranked according to tap water F concentration

Tap water F concentration (ppm)	No.	No. > SI (CoMA)	% > SI (CoMA)
≤0.3	1260	275	21.8
0.3<0.8	38	20	52.6
≥0.8	75	49	65.3
Total	1373	344	25.1

At any level of water fluoride above 0.3 ppm the proportion of consumers receiving more than the CoMA Safe Intake of fluoride rises steeply. The differences are highly significant ( $\chi^2$  65 with 2 degrees of freedom,  $p < < 0.001$ ). The 1260 recipients of lower tap water fluoride concentrations were subdivided into 0.02 ppm F intervals but showed no trend in relation to their total fluoride intake.

The data were then ranked in order of total fluoride intake to set water fluoride in perspective. The results are shown in Table 3.

The steeply rising proportion of higher-consuming subjects receiving fluoridated water is highly significant ( $\chi^2$  120 with 8 degrees of freedom,  $p < < 0.001$ ). Almost all subjects consuming 5 mg F/day or more are ingesting more than either definition of Safe Intake.

**Table 3.** Proportion of National Diet and Nutrition Survey (NDNS) urine collection subjects consuming more than 3 µm F/kg./day (0.057 mg F/kg/day), the Safe Intake (SI) specified in the NDNS, if the following conditions, proposed by the author (PM) based on the current literature,<sup>9,13</sup> apply: 90% of ingested fluoride is assimilated, 50% of that is sequestered in calcified tissues, and only the remainder is excreted in the urine, with the data ranked in the order of the total fluoride intake to set the contribution from water fluoride in perspective

Daily F intake (mg)	No.	No. > SI (NDNS)	%>SI (NDNS)	F in the water supply (ppm)			
				0–0.29	≥0.30	Total	% ≥0.30
≥ 10.0	19	19	100.0	11	7	18	38.9
8.0–9.9	28	28	100.0	19	7	26	26.9
6.0–7.9	80	78	97.5	56	19	75	25.3
5.0–5.9	80	70	87.5	63	15	78	19.2
4.0–4.9	117	41	35.0	97	16	113	14.2
3.0–3.9	174	25	14.4	147	22	169	13.0
2.0–2.9	325	1	0.3	300	15	315	4.8
1.0–1.9	376	0	0.0	346	10	356	2.8
<1.0	232	0	0.0	217	2	219	0.9
Total	1431	262	18.3	1256	113	1369	8.3

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### DISCUSSION

Although the figure of 45% has been used for the proportion of the ingested fluoride which is sequestered in the calcified tissues and which is excreted in the urine<sup>4</sup> (50% of the 90% that is assimilated and not sequestered in the calcified tissues), in line with the figure used in the erratum of the Food Standards Agency<sup>6</sup> and the 1993 review by the National Research Council,<sup>9</sup> the percentage is actually variable among individuals. Largent and Heyroth found, in 1949, that while the amount of ingested fluoride that was retained is usually between 37% and 48%, a wide individual variation was present.<sup>11</sup> Waldbott found, in 1961, that the proportion of a 6.8 mg dose of fluoride that was eliminated in the urine within 24 hours varied from 3.6% to 99.5%.<sup>12</sup> A recent 2010 review of data for 212 children, aged less than 7 years, and 283 adults, aged 18–75 years, found the limiting fractional fluoride retention (FFR) values, assuming an average fluoride absorption of 90%, to be 0.55 for children and 0.36 for adults, when the total daily fluoride intake (TDFI) was above 0.5 mg and 2 mg respectively.<sup>13</sup> These figures

correspond to 10% of the ingested fluoride not being absorbed, 55% being retained by children and 36% being retained by adults, and the percentage of the ingested fluoride excreted in the urine being 35% for children and 54% for adults.

Because fluoride was only one of several analytes of interest in the urine study, it seems unlikely that any error from biased selection, due, for example, to interest in the issue of water fluoridation, would account for the difference between these subjects and the population at large. It seems most likely that collection of a 24-hr urine sample raised insuperable difficulties for some subjects who were in full-time employment, in both the private and public sectors.

These results are in line with a recent Irish study that replicated the NDNS method in three fully-fluoridated neighbourhoods of County Donegal. Fluoride consumption for 22 (73%) of the 30 subjects was at or above the NDNS Safe Intake.<sup>14</sup>

Various authors have related fluoride exposure to different degrees of impairment, but recently most refer to Smith and Hodge,<sup>15</sup> who were chiefly concerned with rapid occupational exposures. They noted an asymptomatic stage, with some radiological signs, at concentrations up to 5500 ppm fluoride in bone ash. Joint pain and stiffness, with radiological osteosclerosis, were noted at concentrations around 6000–7000 ppm. More chronic symptoms with ligamentous calcification supervened between 7500 and 9000 ppm fluoride. Severe disability began at and above this level.

The rate of exposure required to achieve these accumulated concentrations has been controversial. Hodge maintained that it would take consumption of 20–80 mg F/day for 10–20 years to produce crippling skeletal fluorosis. He repeatedly quoted this figure up to 1979<sup>16,17</sup> and attributed it to Møller.<sup>18</sup> However, in the same paragraph he acknowledged Roholm's<sup>19</sup> contrasting estimate of 0.2–0.35 mg F/kg body weight/day. He may have mistakenly equated this to Møller's figure, by using weights in pounds (100–229) rather than kilograms (45–100). In 1979 he eventually adjusted his figure without explanation to 10–25 mg F/day for 10–20 years.<sup>20</sup> Later authorities<sup>21</sup> have followed Roholm more closely, at 10–20 mg F/day for 10 years or more. This figure is five times lower than Møller's 1932 figure.

In view of this body of opinion, the data presented here strongly suggest that the extent and danger of public exposure to fluoride have been seriously underestimated. In 14% of this sample, regardless of the level of water fluoride, 5 mg or more of fluoride is consumed daily, and in 1.3% the daily intake of fluoride is 10 mg or more, which is indisputably sufficient to cause concern. Lesser chronic exposures are likely to have consequences in proportion. There is no definite level of exposure below which fluoride accumulation in bone might not eventually impair bone health during a productive life spanning six decades or more.

Besides all these considerations, if the chemical mechanism of fluoride toxicity is related in part to disruption of hydrogen bonding, vague global debility may well be a further insidious result. This kind of ill health is usually unexplained, and

seems to be increasing in prevalence. A possible connection with fluoride accumulation has so far not received serious consideration.

The reaction of the Food Standards Agency, on discovering that their data implied such high levels of exposure, was to widen the goalposts by quoting higher levels of Safe Intake maintained in other jurisdictions.<sup>7,8</sup> These run as high as 10 mg per day,<sup>7</sup> which would give a therapeutic ratio no higher than 2. Much higher ratios would be mandatory in regulated pharmaceuticals for medical or dental prescriptions. These fluoride exposures are therefore out of proportion to the measures designed to safeguard public health and are out of control and unmonitored. So low a margin of safety in such circumstances is completely unacceptable.

The Safe Intake (SI) specified by the NDNS of 3  $\mu\text{M}$  F/kg/day (0.057 mg F/kg/day) is 57% of the minimum level of 0.1 mg F/kg of body weight found by Akiniwa to be associated with acute fluoride toxicity.<sup>22</sup>

The data presented in Table 3 suggest that a daily fluoride consumption of 5 mg or more is likely to exceed the Safe Intake as defined in the UK by the NDNS. Table 2 suggests that fluoridating water pushes the majority of consumers into excessive fluoride intake.

Moreover, data from the 2000 NDNS cannot answer questions about children. Between age 6 months and six years the Safe Intake of fluoride is considered by CoMA to be 0.12 mg F/kg/day, and in younger infants 0.22 mg F/kg/day. The practice of relating fluoride intake to body weight is questionable. In childhood it complicates more than it clarifies. Small children retain ingested fluoride at a higher rate than adults, making them more vulnerable to the long-term toxic effects of over-consumption.<sup>13</sup>

It is, however, practical to measure urinary fluoride concentration at any age, a short series of which in any individual will seldom give totally misleading results.

### CONCLUSIONS

1. Fluoride exposure from all sources in the UK is an order of magnitude higher than previously estimated.
2. Fluoridation of a water supply makes most of the population excessive consumers of fluoride.
3. There is no modern information about what adverse health effects this excessive fluoride intake may have. A surveillance program should begin urgently in the West Midlands to relate the total fluoride consumption of individuals to their health experience
4. It would be highly desirable to add a test square to detect fluoride concentration to the strips produced for routine multi-testing of urine samples. The feasibility of this should be explored with manufacturers.
5. No further water fluoridation schemes should be started until results from No. 3 above are available.

## ACKNOWLEDGEMENTS

Thanks are due to the staff of Economic and Social Data Service, University of Essex, and the Chief Dental Officer, Department of Health, for provision of raw data from Reference 3.

The Food Standards Agency (FSA) have critically reviewed a draft of this paper and examined the author's working spreadsheets. No fault was found. The small size of some of the cells in the tabulations was mentioned. Nonetheless, high levels of statistical significance are achieved and do not diminish when cells are combined. The FSA declined to publish the data themselves.

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