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# Fluoride: Exposure and Relative Source Contribution Analysis

**Health and Ecological Criteria Division**
**Office of Water**

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## PREFACE

In March, 2006, the National Academy of Sciences National Research Council (NRC) released the Report entitled "Fluoride in Drinking Water: A Scientific Review of EPA's Standards". The NRC stated that "in light of the collected evidence on various health endpoints and total exposure to fluoride, the committee concludes the EPA's MCLG of 4 mg/L should be lowered". They further suggested that, in order to develop an MCLG (Maximum Contaminant Level Goal) that is protective against severe enamel fluorosis, clinical stage II skeletal fluorosis and bone fractures, EPA should:

- Develop better estimates of total exposure for individuals,
- Use current approaches for quantifying risk,
- Consider susceptible populations, and
- Characterize uncertainties and variability.

In response to the NRC (2006) recommendations, the Office of Water (OW) collected available data on the various media that contribute to fluoride exposure in the United States for the purpose of estimating total exposures for children during the period of sensitivity to severe dental fluorosis (six months to 14 years). Data were also collected to develop an exposure estimate for the adult population. This document presents the exposure analysis.

The objective of the OW's exposure and relative source contribution analysis was to quantify the fluoride exposures for children and adults in the United States to accomplish the following:

- Determine sources of fluoride exposure for the U.S. population.
- Quantify exposures where possible for the age groups of concern.
- Compare oral intake estimates to the reference dose established in the companion dose-response assessment.
- Estimate the relative source contribution for each exposure source.
- Provide information for use in characterizing opportunities for reducing population risk from fluoride in public drinking water systems and facilitating any necessary adjustment in the regulatory non-enforceable Maximum Contaminant Level Goal (MCLG).

This document addresses the relative source contribution for drinking water from public systems and contains information from peer-reviewed publications on multiple topics; these topics include concentrations of fluoride in foods and beverages, estimated dietary exposure estimates for fluoride, concentrations of fluoride in tap water delivered by public drinking water systems, estimated fluoride intakes from toothpaste, and estimated fluoride exposures from sulfuryl fluoride (a pesticide).

In addition, this report presents background on the analytical methods used to measure fluoride in various media, as well as approaches applied in developing dietary exposure assessments. The background information is included to provide perspective on how methods of analysis used in individual studies have impacted the exposure estimates.

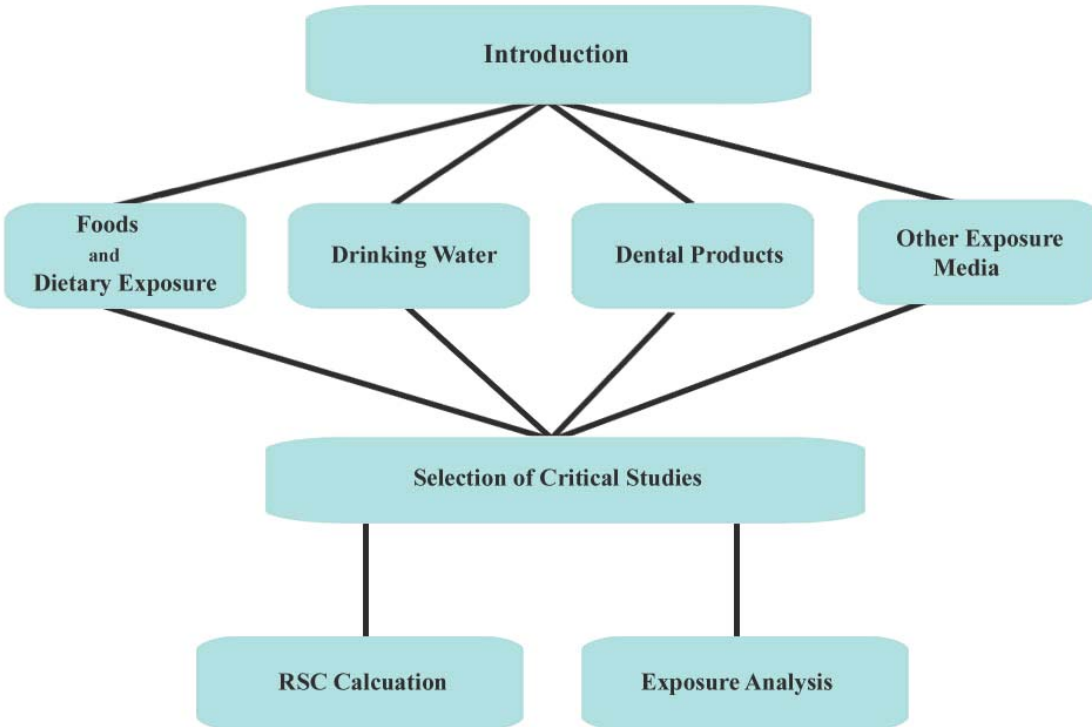
There are a number of factors that the reader should consider when reviewing this document characterizing the relative source contribution from tap water to total fluoride intake:

- Only peer-reviewed and published data from the United States and Canada were used in the assessment (excepting the Information Collection Request data collected by Office of Water for the second six-year review of its regulations).
- Water intakes are those for public water systems and consumers only (EPA, 2004).
- EPA conducted no independent study measuring dietary exposure; data employed are from the published papers.
- Exposure estimates for sulfuryl fluoride were prepared for the OW by the Office of Pesticide Programs.
- Office of Water policies applied in the relative source contribution analysis are those presented in EPA (2000b).
- The age groupings selected were those used by Ershow and Cantor (1987). The Ershow and Cantor (1987) publication provided the best available water intake data for the time period of the critical study (1930-1940).

The document is structured (see map below) to present the published information available on fluoride in foods (Chapter 2), fluoride in drinking water from public and nonpublic sources (Chapter 3), fluoride in toothpaste (Chapter 4), and fluoride from more minor exposure sources (Chapter 5). Each chapter also presents published exposure estimates applicable to each exposure medium when available. Chapters 2 and 3 include background data on the analytical methods used for the analyses in the cited studies and the experimental approaches used to assess dietary exposures.

Chapter 6 identifies the studies selected for the quantitative exposure analysis and the reasons supporting their selection. The RSC calculations and sensitivity analysis are found in Chapter 7 while Chapter 8 compares current exposure values to the reference dose from the dose-response document (EPA, 2010a) and nutritional guidelines from the Institute of Medicine (IOM, 1997). Appendices A and B located at the end of this document (beginning at page 127), were provided by the Office of Pesticide Programs and retain their original pagination.

The OW has also prepared and peer reviewed a second document that provides an estimate of the RfD for fluoride. This second document, *Fluoride: Dose-Response Analysis for Non-cancer Effects* (EPA Report No. 820-R-10-019), can be accessed through the following url: http://water.epa.gov/action/advisories/drinking/fluoride_index.cfm

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# ACKNOWLEDGMENTS

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## LIST OF ACRONYMS

| | |
|---|---|
| ADA | American Dental Association |
| ANOVA | Analysis of Variance |
| ATSDR | Agency for Toxic Substances and Disease Registry |
| AWQC | Ambient Water Quality Criteria |
| BW | Body weight |
| CAMP | Continuous Air Monitoring Project |
| CDC | Centers for Disease Control |
| CI | Confidence Interval |
| CSFII | Continuing Survey of Food Intake by Individuals |
| CTE | Central Tendency Exposure |
| DI | Drinking water intake |
| F | Fluoride |
| FDA | Food and Drug Administration |
| GI | Gastrointestinal |
| HMDS | Hexamethyldisiloxane |
| IOM | Institute of Medicine (of The National Academies) |
| ISE | Ion-selective electrode |
| MCL | Maximum contaminant level |
| MCLG | Maximum contaminant level goal |
| NDL | Nutrient Data Laboratory (U.S. Department of Agriculture) |
| NF | Non-fluoridated |
| NFCS | Nationwide Food Consumption Survey |
| NHANES | National Health and Nutrition Examination Survey |
| NIDR | National Institute of Dental Research |
| NIPDWR | National Interim Primary Drinking Water Regulations |
| nmole | Nanomole |
| NRC | National Research Council (of The National Academies) |
| OR | Odds ratio |
| OPF | Optimal fluoride level |
| OSHA | Occupational Safety and Health Administration |
| RDA | Recommended Daily Allowance |
| RfD | Reference dose (in mg/kg/day) |
| RMCL | Recommended Maximum Contaminant Level |
| RME | Reasonable Maximum Exposure |
| RSC | Relative Source Contribution |
| SD | Standard Deviation |
| SE | Standard Error |
| SEM | Standard Error of the Mean |
| SDWA | Safe Drinking Water Act |
| SMCL | Secondary Maximum Contaminant Level |
| USDA | U.S. Department of Agriculture |

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# EXECUTIVE SUMMARY

In response to the 2006 National Research Council (NRC) report: *Fluoride in Drinking Water: A Scientific Review of EPA's Standards*, the U.S. EPA Office of Water (OW) initiated an examination of dose-response data for critical noncancer effects of fluoride on teeth and bone in light of the NRC (2006) conclusion that "the EPA's MCLG of 4 mg/L should be lowered," so as to reduce the risk of severe enamel fluorosis and to minimize the risk for bone fractures and skeletal fluorosis in adults. Dose-response assessment for fluoride was updated using current approaches for quantifying risk with consideration given to susceptible populations as well as uncertainties and variability in the data (U.S. EPA, 2010a).

One goal of the exposure and relative source contribution (RSC) analyses was to obtain and evaluate available U.S. domestic exposure data that could be used by the Office of Water during its reconsideration of the current USEPA Maximum Contaminant Level Goal (MCLG; nonenforceable) for fluoride. This assessment examined data on the concentrations of fluoride in foods and beverages, available dietary exposure estimates for fluoride, concentrations of fluoride in the tap water delivered by public drinking water systems, incidental ingestion of fluoride from toothpaste use and potential exposures to fluoride from sulfuryl fluoride (pesticide) applications. The information utilized was largely drawn from peer-reviewed published literature that examined the exposure of US domestic or in some cases Canadian populations and communities.

Once information on the various media contributing to fluoride exposure were assembled and analyzed, total exposures for the period of sensitivity to severe dental fluorosis (six months to 14 years) were estimated. An exposure estimate was also developed for the adult population. The RSC determination followed the methodology established by the OW for chemicals found in drinking water which use average exposures for all media except residential drinking water from public drinking water systems. The drinking water component of the relative source analysis is based on the average fluoride concentration (~0.9 mg/L) from public drinking water systems that reported detectable levels of fluoride during the second six-year review of U.S. EPA drinking water regulations and the intake data (direct and indirect) for the 90$^{th}$ percentile consumer from the U.S. Department of Agriculture (USDA) Continuing Survey of Food Intake by Individual (CSFII). The analysis considers susceptible populations, and the impact of the uncertainties and variability in the data as part of the RSC analysis.

Among the age groups evaluated, the RSC values for drinking water range from 40 to 70 percent, with the higher values associated with infants fed with powdered formula or concentrate reconstituted with residential tap water (70%) and adults (60%). Comparison of the age-specific total estimated exposure for the 90$^{th}$ percentile drinking water consumer to the daily reference dose suggests that some children at ages less than seven years old may be at risk for severe dental fluorosis. The major contributors to total daily fluoride intakes for these age groups are their drinking water, commercial beverages, solid foods and swallowed toothpaste.

In addition to the exposure information, this report presents background information on the strengths and weaknesses of the analytical methods that were used to measure fluoride in various media for the key critical studies and the approaches applied in developing dietary exposure assessments.

# 1. Introduction

## 1.1. Background

In 2006, the National Research Council (NRC) released: *Fluoride in Drinking Water: A Scientific Review of EPA's Standards*, a three year effort to examine the health effects of ingested fluoride in drinking water. The development of the NRC (2006) report was funded by the U. S. EPA Office of Water (OW). The project was initiated as a result of the 2002/2003 review of the Maximum Contaminant Level Goal (MCLG) and the Maximum Contaminant Level (MCL) for fluoride.

NRC (2006) concluded that EPA's current MCLG of 4 mg/L for fluoride should be lowered to reduce the risk of severe enamel fluorosis and minimize the risk for bone fractures and skeletal fluorosis in adults. In response, the U.S. EPA OW initiated an examination of the dose-response data for the critical noncancer effects of fluoride on teeth and bone. The dose-response assessment for fluoride was updated using current approaches for quantifying risk with consideration given to susceptible populations and the uncertainties and variability in the data (U.S. EPA, 2010a).

The U.S. EPA (2010a) report identifies the fluoride concentration in drinking water that was not associated with an increased risk of severe dental fluorosis in 99.5% of children in selected towns distributed across the United States (Dean (1942) prior to the introduction of fluoridation and fluoridated dental products. The U.S. EPA (2010a) report includes an estimated Reference Dose (RfD) for severe dental fluorosis derived from the Dean (1942) data. It also determined that the RfD associated with severe dental fluorosis is similar to or lower than that associated with an increased risk of bone fracture or Stage III skeletal fluorosis.

At the time the dose-response data were collected the fluoride in drinking water was largely determined by local geological composition of the soils and bedrock; there was no intentional fluoridation of public drinking water supplies and no commercial fluoride-containing dental products. Currently, exposures to fluoride come from drinking water, foods, beverages, dental products (toothpaste, mouth rinses), supplements, industrial emissions, pharmaceuticals, and pesticides. In the case of young children, ingestion of fluoride-containing soil is another source of exposure. These exposure pathways are discussed in this report and quantified where possible. The data presented include some of the studies that were considered by the NRC (2006) report in their analysis of relative source exposures as well as additional published papers identified by the OW.

The ratio between exposure from drinking water and total exposure is called the relative source contribution (RSC). The OW traditionally uses the RSC in the derivation of noncancer MCLGs for a drinking water regulation. Section 1.2 below describes OW RSC policies.

## 1.2. U.S. EPA RSC Policies

The OW RSC policies have evolved gradually over the more than twenty years since fluoride was regulated. The derivation of the fluoride MCLG did not include an RSC, in part because the data supporting the critical effect of crippling skeletal fluorosis in adults were derived primarily from the ingestion of fluoride from drinking water. The diet was assumed to have a minimal contribution to total intake and was not reported in the critical studies. The MCLG was derived from an estimated 20 mg/day chronic fluoride intake divided by a drinking water intake of 2 L/day and a 2.5 safety factor yielding an MCLG of 4 mg/L. This same approach was used in determining the MCLG values for a few other contaminants (i.e. nitrate, copper, barium) because the exposure and toxicology data applied to drinking water and did not include background intakes from other sources.

Currently calculation of the MCLG for noncancer endpoints, in almost all cases, utilizes the Reference Dose (RfD) as the point of departure. The reference dose is defined as: "an estimate (with uncertainty spanning perhaps an order of magnitude) of a daily oral exposure to the human population (including sensitive subgroups) that is likely to be without an appreciable risk of deleterious effects during a lifetime" (U.S. EPA, http://www.epa.gov/ncea/iris/help_ques.htm#rfd).

The MCLG is usually derived from the RfD using the following equation.

$$\mathbf{MCLG = \frac{RfD \times BW \times RSC}{DI}}$$

where:

BW = Average body weight (70 kg for an adult)
DI = 90$^{th}$ percentile drinking water intake (2 L/day for an adult)
RSC = Relative Source Contribution

Prior to the 1998 Stage I Disinfectants and Disinfection Byproducts rule [Fed. Reg. 63(241):69389-69476], a 20% default RSC was applied for the majority of MCLGs for noncancer effects. The few exceptions to this practice were those cases where published data were used to support an alternate RSC. A shift away from automatically defaulting to 20% was an outgrowth of the 2000 publication of the Ambient Water Quality Criteria (AWQC) for Human Health which included a peer reviewed decision tree approach for determining the RSC (EPA, 2000b). The human health AWQC applies to the intake of a contaminant from both drinking water and fish/shellfish from ambient surface waters of interest. It is easily adapted for use with drinking water alone scenarios. It was used for determining the RSC values for chloroform, monochloroacetic acid, and trichloroacetic acid in the 2003 Stage II Disinfection By-product Rule [Fed. Reg. 71(2):387-493]. The MCLGs for all three compounds are based on the RfD rather than a cancer endpoint. The MCLG for all carcinogens is currently zero and does not require an RSC.

Key features of the Human Health AWQC Decision Tree as applied to drinking water can be summarized as follows:

- The RSC value used is determined by the type and amount of data available. The data should be representative of the population of concern (adults, child, pregnant woman, etc).

- The RSC is based on national exposure estimates that, at a minimum, provide average values and associated confidence bounds. Knowledge of the properties of the chemical and more limited exposure data can be used when nationally representative data are not fully available.
- All known exposure routes and media are considered.
- The lowest RSC is 20% based on the assumption that regulatory or guideline values for chemicals with exposures that are less than 20% of the total will not provide a meaningful opportunity to reduce risk for the population. In these cases the greatest health benefit can be achieved by establishing guidance or regulations for the medium that contributes the major portion of the total exposure.
- The highest allowable RSC is 80% based on the assumption that there may be many minor sources of exposure that will not be captured by the available data.
- Subtraction and percentage options are available but are bounded by the 20% floor and 80% ceiling. There are policy limitations on the use of the subtraction approach. It can be applied only under circumstances where the MCLG is the sole health-based U.S. EPA criterion for the contaminant. For example, the subtraction approach is not possible for fluoride because pesticides containing fluoride have established tolerances for food crops.
- Average exposure values are used to represent the contribution from the diet, ambient air, soil ingestion, and other exposure media.
- The drinking water intake contribution to total exposure is represented by the 90$^{th}$ percentile value and the average analyte drinking water concentration because drinking water is the exposure route of concern for the OW.
- The body weight is an average for the population of interest (e.g. adults, infants, and children).
- Exposures to drinking water contaminants that occur during showering, bathing, laundry, etc. are not included as part of drinking water ingestion intake. They are included in the other sources of exposure.
- In determining the RSC as a percentage, the estimated daily analyte intake from a 90$^{th}$ percentile tap water consumption estimate at the average analyte concentration from public water systems is divided by the total uptake into the body from all quantified exposure routes.

The OW is in the process of considering refinements to the 2000 decision tree methodology for ambient water and drinking water. However, those modifications were not available for the fluoride exposure assessment. Accordingly, the RSC for fluoride has been developed using human health AWQC methodology framework.

## 2. Exposure from Foods and Beverages

In the 30-year period covered by the data in this report, there have been changes in analytical methods and instrumentation that have led to improvements in the accuracy and precision of measurements of fluoride in food and beverages. The early studies usually relied on colorimetric techniques for the measurement of fluoride; such techniques were subject to interference from other elements in the food matrix. Most later studies employed a fluoride ion-specific electrode in the measurement of fluoride. Some changes in the measured levels of fluoride in foods and beverages over the years covered in this report and in EPA (2010a) appear to be a consequence of differences in the analytical methodologies used to measure fluoride as well as changes in food consumption patterns. Section 2.1 below provides background historic information on analytical methods used to measure fluoride in food. The methodological impacts on measurements of fluoride concentrations in foods are discussed in Sections 2.1, 2.2.3 and 2.3.3.

### 2.1. Analytical Methods

Procedures for the determination of fluoride in foods typically exhibit three distinct phases: digestion, isolation and quantification. In the digestion phase, samples are ashed in the presence of a caustic agent such as concentrated calcium or sodium hydroxide. The caustic agent not only serves to help digest the organic sample, but also acts as a trapping agent for fluoride ion. Several methods can be used in the isolation phase. In one method, the ashed residue is dissolved in concentrated acid. Fluoride ion is converted to volatile hydrofluoric or hydrofluosilicic acid, distilled from the residues, and collected in a clean aqueous distillate. Successful isolation of fluoride ion from the ashed residues has also been accomplished by merely allowing the analyte to diffuse through a membrane into a strongly basic trapping medium. Hexamethyldisiloxane accelerates this process significantly. Once the fluoride is isolated, several approaches can be employed in the quantification phase, including (a) titration with colorimetric reagents; (b) reaction with a colored reagent followed by spectrophotometric measurement; (c) measurement using a fluoride ion-selective electrode, and (d) gas chromatography. Further details about each of these basic steps are provided below.

#### 2.1.1. Sample Preparation

Food samples are primarily composed of bulk organic matter which is largely insoluble in water, with small concentrations of inorganic species of interest. The bulk organic matrix must be removed while inorganic analytes such as fluoride are retained. In many cases, some form of “trapping” medium is required to ensure that fluoride is not lost during the digestion process.

Mineralization by ashing of the food sample is used to remove the organic matrix. The process was initially described in AOAC (1945), and has been virtually unchanged in more than fifty years (AOACI, 2000). Modest quantities of dry material, liquid samples, and undried food products or plant material are selected for analysis, depending upon the expected fluoride content and interferences. The sample is mixed with a calcium hydroxide (lime) suspension or sodium hydroxide, dried and ashed in a muffle furnace at $600^{o}$ C. Variations of the official methods can employ smaller samples, different trapping agents, or both (Malde et. al., 2001; Venkateswarlu, 1975; Singer and Ophaug, 1979).

Venkateswarlu (1975) compared "closed ashing", employing a standard oxygen bomb technique, with the "open ashing" employing a muffle furnace, described above. Both procedures are applicable to solid samples, soft tissue, and liquid samples that are low in organic matter. In all cases, the "closed ashing" approach required that samples be pressed into pellets containing up to 1 g of solid. Blank values were typically < 0.05 µg fluoride. The recoveries of fluoride from bovine albumin and serum at concentrations of 0.28 and 0.05 µg fluoride per gram sample exceeded 95%. A comparison between results obtained from "closed" vs. "open" ashing for bovine and human sera samples suggested that the values obtained by the open ashing process are frequently lower than those obtained with closed ashing in an oxygen-enriched chamber.

Several authors have described procedures for quantifying fluoride in food matter that do not involve ashing; these changes are a reflection of improvements in analytical instrumentation. Pesselman et al. (1989) described an alternative preparation for soluble samples such as cocoa powder that did not involve ashing. Small samples were mixed with doubly-deionized water and blended using a simple Waring blender. The product was then centrifuged and vacuum filtered. Nedeljković et al. (1991) homogenized food samples and transferred the resulting slurry to a "microdiffusion" cell, described below, containing sodium hydroxide solution as the trapping medium. Both methods employed a fluoride ion selective electrode, described below, for final measurement of fluoride concentration in solution.

### 2.1.2. Fluoride Recovery

***Distillation.*** In the classical approach for isolating fluoride from interfering elements in the mineralized ash, fluoride is converted to hydrofluosilicic acid by adding perchloric or sulfuric acid to water (the former is preferable, since nearly all of the perchlorates are very soluble). This method was described by Willard and Winter (1933) almost seventy-five years ago. Several pieces of glass are added to the sample in a distillation flask, and distilled. When only a small quantity of fluorine (10 mg or less) is present in the sample and the temperature is not allowed to rise above approximately 125 ºC, the pieces of glass appear to supply the silica necessary to combine with the fluorine to form hydrofluosilicic acid, and there is no noticeable etching of the flask. The authors presented data showing that 7-10 mg of fluoride (as sodium fluoride) could be recovered with >95% efficiency from a variety of matrices, such as plant ash, gelatinous silica, boric acid, and aluminum chloride. A similar approach is presented in APHA/AWWA/WEF (2005) as "Method 4500 $F^-$. B. Preliminary Distillation Step."

***Microdiffusion and Trapping of Fluoride.*** Many investigators have reported concerns with the standard method, including losses of a volatile fluoride species through the ground glass joints, the possibility of a perchlorate explosion, the obvious skill needed to make the distillation work properly, and the time and effort required for a proper isolation. For these reasons, investigators have tried to develop simpler and faster isolation methods with analyte recovery comparable to that of the standard method. Most of these involve the diffusion of hydrogen fluoride through modified polypropylene "Conway cells" (Öbrink, 1955; Conway, 1950), a specific microdiffusion cell design.

The "Conway cell" operates in the following manner: According to Singer and Armstrong (1965), a strongly-basic "trapping solution" for HF, e.g., 2.5 N sodium hydroxide is placed into the center well ("inner chamber") of the diffusion cell. The sample containing fluoride is acidified strongly with perchloric acid and placed into the sample compartment ("outer chamber"). The cell is then sealed using a small Petri dish, heated to 55-60 °C, and left undisturbed for 22 hours. Hydrogen fluoride diffuses from the sample compartment into the headspace of the sealed cell, and is collected in the trapping solution contained in the inner chamber. The authors demonstrated that the recovery of 0.1-2 µg fluoride from samples of rat liver, beef liver, and beef muscle, by use of this diffusion procedure was virtually identical to that obtained using the Willard and Winter (1933) distillation procedure. This new method was considered a reasonable substitute for the traditional distillation procedure because both approaches produced the same results at or below the microgram level for fluoride. Additional experiments with 0.5 or 1 µg of $F^{18}$, a radioactive tracer, confirmed that the recoveries of fluoride typically exceeded 95% from human plasma, saliva, and urine when using the microdiffusion cell to isolate the analyte.

Taves (1968a) found that the diffusion of fluoride increased if silicone grease was used to seal the Conway cells. In follow-up work, the author used 6 M hydrochloric acid saturated with 0.5 mL hexamethyldisiloxane (HMDS) in the "outer chamber" of the Conway cell, and examined the rate of fluoride diffusion into a variety of trapping agents with and without HMDS present (Taves, 1968b). All recovery measurements were performed using the radioactive tracer $F^{18}$. Without the HMDS present, there was practically no diffusion of fluoride. When the HMDS-saturated hydrochloric acid was present, but not in contact with sample, one-third of the fluoride diffused to the trapping solution in 10 minutes, owing to the volatilization of the HMDS. Mixing the solutions increased the rate of diffusion appreciably and continuous rotary motion resulted in over 80% recovery of the radioactive tracer in only 10 minutes, a very rapid process. In one hour at room temperature, >97% tracer recovery was attained by this method (Taves, 1968b). HMDS is presumed to accelerate the diffusion of fluoride by formation of trimethylfluorosilane.

### 2.1.3. Measurement and Quantitation of Fluoride Ion

***Ion-selective electrode (ISE).*** The fluoride ion-selective electrode (ISE) was introduced in the mid-1960s (Buck and Lindner, 2001), and quickly became the industry-wide standard for the accurate determination of fluoride concentrations. When this electrode is compared to later spectrophotometric methods, such as that employing the SPADNS [sodium 2-(parasulfophenylazo)-1, 8-dihydroxy-3, 6-naphthalene disulfonate] reagent, the former exhibits superior selectivity when challenged with chloride, chlorine, color and turbidity, iron, phosphate, sulfate, and aluminum (APHA/AWWA/WEF, 2005).

The key element in the fluoride electrode is the laser-type doped lanthanuim fluoride crystal across which a potential is established by fluoride solutions of different concentrations. The crystal contacts the same solution at one face and an internal reference solution at the other. Strictly speaking, the fluoride electrode measures the ion activity of fluoride in solution rather than concentration. Fluoride ion activity depends on the solution total ionic strength and pH, and on fluoride complexing species. For that reason, adding an appropriate buffer provides a nearly uniform ionic strength background, adjusts pH, and breaks up complexes so that, in effect, the electrode measures concentration (APHA/AWWA/WEF, 2005; Omega, 1993). The literature documents successful use of the fluoride ISE in quantifying this analyte in many different foods and materials.

Singer and Ophaug (1979) presented a side-by-side comparison of fluoride concentration results obtained using both a colorimetric and fluoride ISE approach. The results were entirely comparable for strained meats (chicken and beef with respective broths), milk-based infant formula, and vegetables (green beans, peas, and spinach) at concentrations ranging between 0.1-6 mg F/kg sample. When fruits (pears, applesauce, peaches, etc.) were analyzed, substantially (~20 times) higher levels were observed with the colorimetric method. It thus appears that reagents employing eriochromecyanin R (see "Spectrophotometric determination", below) to determine fluoride in the diffusates of unashed foods may result in erroneously high values.

***Spectrophotometric Determination.*** The introduction of the Beckman Model DU spectrophotometer, an instrument which could measure absorbance in both the ultra-violet and visible ranges, in 1941 (Simoni et. al., 2003) rendered the classic titration-methods for quantifying fluoride ion obsolete. Spectrophotometric procedures frequently employed a zirconium-alizarin or eriochromecyanin R (*syn.* Eriochrome Cyanine R) lake dye, whose absorbance fades with increasing fluoride ion concentration (Singer and Armstrong, 1959; Megregarian and Maier, 1952) over the range of 0–4 ppm. This method was highly dependent upon the presence of phosphate and iron, but relatively insensitive to bicarbonate, chloride, and sulfate. Grutsch et al. (1953) demonstrated that the interferences from iron, manganese, and chlorine could be eliminated by adding thioglycolic acid to the aqueous samples.

The above approach is still one of those accepted for the determination of fluoride ion, albeit in a modified form (APHA/AWWA/WEF, 2005). The SPADNS colorimetric method (Bellack and Schouboe, 1968) is based on the reaction between fluoride and a zirconium-dye lake. Fluoride reacts with the dye lake, dissociating a portion of it into a colorless complex anion, $(ZrF_6)^{2-}$. As the amount of fluoride increases and reacts with the dye, the color produced becomes progressively lighter; absorbance is measured at 570 nm.

Taves (1968c) described a related approach, in which the concentration of fluoride ion was related to the fluorescence quenching of a Morin (pentahydroxyflavone aluminum complex)-thorium complex, rather than the change in absorbance described above. The standard quenching curve was linear between 0-10 nmoles of fluoride ion, after which significant deviations from linearity were observed. The Morin-thorium reagent was also more sensitive to the fluoride than to the sulfate and phosphate ions by factors of twenty and forty, respectively. Nitrate has no immediate effect, but has a marked effect within 18 hours. When the same amount of acid is used, the effect of chloride, perchlorate, and nitrate is only 1/80,000 that of fluoride.

The method described in Elvove (1933) uses "Nessler" color comparison tubes and the human eye as the detector. Known quantities of fluoride, typically ranging between 0 and 55 µg/mL, were mixed with a fixed quantity of zierconium-alizarin reagent, permitted to stand undisturbed, and then compared with the color of unknowns prepared in the same fashion.

***Titration.*** The classical titration of fluoride ion is based upon a two-step process. Initially, fluoride ion (colorless) reacts with a zirconium-alizarin lake dye (red) to form a zirconium-fluoride complex (colorless) and free alizarin (yellow) (Grutsch et. al., 1953). The resulting solution is then back-titrated with a standardized solution of thorium nitrate, which decomposes the zirconium-fluoride complex and permits the zirconium-lake complex (red) to reform. The endpoint of the titration is the faint permanent reappearance of the lake color. (Willard and Winter, 1933). Willard and Winter (1933) employed this procedure for quantifying fluoride accurately at the milligram level. This procedure was modified to employ "Nessler tubes" for color comparison in the Official Methods of Analysis (AOAC, 1945; AOACI, 2000), and has not been changed in more than fifty years.

The procedures discussed above all employed distillation of volatile hydrogen fluoride prior to titration. Singer and Armstrong (1965) described a titration of fluoride collected in the "trapping solution" of a "Conway cell", described above, using hydrochloric acid as the titrant to a simple phenolphthalein endpoint. The authors do mention a Beckman model B spectrophotometer in the method, but it is not clear how this instrument was used.

***Gas Chromatography.*** In this procedure, as described by Fresen et al. (1968), an alkyl or arylchlorsilane (e.g., trimethylchlorsilane) is converted by water into the corresponding silanol which then reacts selectively with fluoride to form fluorsilane. The fluorsilane is extracted from an acidified (1 M HCl) sample with an organic solvent such as benzene. The amount of fluoride still present in the aqueous layer after extraction is negligible. The fluorsilane is injected into a gas chromatograph using an internal standard such as isopentane. The relative peak height (corrected with a blank value) is linearly proportional to the fluoride content in the sample. A solution of 0.6 mg trimethylchlorsilane per mL benzene is sufficient to determine amounts of fluoride from 0.01 to 10 μg.

### 2.1.4. Confidence in Analytical Results

Analytical procedures for the determination of fluoride in foods and drinking water samples have been studied, evaluated, and improved since the 1930's. During all of that time, questions and concerns raised by analytical chemists have remained the same, viz., (a) accuracy, (b) precision, (c) detection limit, (d) calibration range, (e) low blank, and (f) interferences. It is certainly true that current methods employing the fluoride ion-selective electrode, for example, are easier to use, exhibit a lower blank, and are more selective than predecessor methods. However, from the onset, it is evident that investigators were keenly aware of technology limitations, and made strenuous attempts to correct or account for potential interferences. Investigators tried to simplify or eliminate the traditional "open ashing" procedure, which reduces the mass and volume of the sample matrix and the ensuing distillation procedure for further isolating fluoride.

The method used to detect fluoride can often be predicted based upon the date of the research. Prior to approximately 1950, the only method available was based upon titrations employing a zirconium-alizarin reagent first, followed by back-titration with thorium nitrate. Between approximately 1950 and 1965, the preferred method was spectrophotometry using the zirconium-alizarin reagent alone. Work reported after 1965 almost always employs the fluoride ion-selective electrode.

The results obtained using the earlier titration, spectrophotometric or colorimetric procedures exhibited sufficient precision and accuracy to support a reasonable estimate for the concentration of fluoride in environmental media. For example, McClure (1939) employed a titration-based method to evaluate the fluoride content in a very wide variety of foods. With the exception of items grown in a fluoride-containing area or sprayed with a fluoride-containing pesticide, McClure (1939) reported typical concentrations below 10 ppm, and frequently below 1 ppm, however levels of detection appear to have been better with some food matrices than others. Forty years later, Singer and Ophaug (1979) employed an ion-selective electrode-based method and reported very similar results for a smaller variety of foodstuffs. Taken together, the newer methods may be easier to perform, are faster, more selective, and more sensitive than their earlier counterparts. In some cases the results from the older methods are comparable to those from the newer ones but that is not always the situation. A number of interferences and methodological variables can result in reported concentrations in foods being lower or higher than the actual concentration.

The most current methods of analysis for fluoride adopted by the Association of Official Analytical Chemists International (AOACI) include ion chromatography for inorganic fluoride in water; the ion selective electrode for fluoride in wine and other beverages, and the distillation method for fluorine in food (see: http://www.eoma.aoac.org/methods).

### 2.2. Natural Fluoride Levels in Solid Foods

Several studies suggest that natural fluoride in foods may not be as bioavailable as that from inorganic fluoride compounds. IOM (1997) notes that when a soluble inorganic fluoride compound such as sodium fluoride is ingested with milk, baby formula, or foods with high concentrations of calcium, or certain other divalent or trivalent ions that form insoluble compounds, absorption may be reduced by 10 to 25%. Trautner and Siebert (1986) investigated the bioavailability in food products rich in natural fluoride (bone meal, fish bone meal, seaweed flour, canned sardines, chicken bone meal, tea, krill). Fluoride concentrations in plasma and saliva over 8 hr, as well as 24 hr urinary fluoride excretions, were determined in healthy adult volunteers receiving single oral doses containing between 2 and 10 mg F. Comparisons were made with sodium fluoride (administered as 2, 5, 7.5 and 10 mg doses in NaF solution, or 2, 5 and 8 mg doses as NaF tablets) which was assumed to be 100% bioavailable as reported by Ekstrand et al. (1978). Plasma, saliva, and urinary fluoride levels were determined with a fluoride ion-specific electrode. Fluoride in food items was determined by gas chromatography after extraction with HCl. Bioavailability (B) of fluoride from different substances (sub) was calculated from the plasma data as:

$$\mathbf{B\%} = \frac{\mathbf{\Delta AUC_{sub} \times D_{NaF} \times 100}}{\mathbf{\Delta AUC_{NaF} \times D_{sub}}}$$

where: D = the quantity of the substance administered or present in the NaF reference sample, and ΔAUC is the net area under the fluoride plasma concentration curve minus the background fluoride levels in the control samples. The same procedure was used to calculate bioavailability from values of urinary fluoride:

$$\mathbf{B\%} = \frac{\mathbf{\Delta U_{sub} \times D_{NaF} \times 100}}{\mathbf{\Delta U_{NaF} \times D_{sub}}}$$

where: ΔU is the net amount of fluoride excreted in the urine during 24 hr.

The tested foods and beverages varied widely in their bioavailabilities. Relative to sodium fluoride, bones of mammals, chicken, and fish, whole fish, were poor sources of fluoride (bioavailability less than one-fourth that of sodium fluoride). In contrast, tea had a bioavailability close to that of sodium fluoride.

Spak et al. (1982) evaluated the bioavailability of fluoride added to baby formula and milk. Three different 500 mL solutions (water, milk or formula) containing 10 ppm F (from sodium fluoride) were administered to volunteers aged 23-25 yr. Fluoride levels in plasma and urine were determined with a modified microdiffusion technique (Taves, 1968b). The results indicated that 72% of the fluoride in milk and 65% in the baby formula were absorbed.

### 2.2.1. Fluoride in Infant Foods

***Breast Milk***. Concentrations of fluoride in human breast milk are very low. Dabeka et al. (1986) analyzed 210 samples of breast milk from Canadian women and found detectable concentrations in 92 (44%). Fluoride concentrations ranged from <0.002 to 0.097 mg/L. The mean concentration in milk from mothers in fluoridated communities (1 mg F/L water) was 0.0098 mg/L; in nonfluoridated communities the mean was 0.0044 mg/L. Fluoride concentrations in breast milk were directly related to the fluoride concentration in the mother's drinking water ($p = 0.007$). The IOM (1997) reported that concentrations in human milk ranged from 0.007 to 0.011 mg/L based on data from Ekstrand et al. (1984), Esala et al. (1982) and Spak et al. (1982).

***Infant Formula***. Infant formula varies in fluoride content, depending on the type of formula and the water with which it is prepared.

Dabeka and McKenzie (1987) analyzed fluoride levels in about 115 samples of infant formulas. Fluoride content was determined by micro-diffusion and a fluoride ion-specific electrode. Results are shown in Table 2-1.

**Table 2-1. Fluoride Concentrations in Infant Formula (Dabeka and Mckenzie, 1987)**

| Category | Number of No. Samples | Fluoride Concentration (mg/kg food) | |
|---|---|---|---|
| | | Mean | Range |
| Ready to use formula, all: | 41 | 0.79 | 0.15–2.31 |
| Ready to use formula, Canadian | 34 | 0.90 | 0.35–2.31 |
| Ready to use formula, US | 7 | 0.23 | 0.15–0.28 |
| Ready to use formula, glass; all: | 23 | 0.75 | 0.28–1.13 |
| Canadian | 20 | 0.82 | 0.46–1.13 |
| US | 3 | 0.28 | 0.28–0.28 |
| Ready to use formula, canned; all: | 18 | 0.84 | 0.15–2.31 |
| Canadian | 14 | 1.02 | 0.35–2.31 |
| US | 4 | 0.19 | 0.15–0.26 |
| Conc. liquid formula | 33 | 0.60 | 0.15–1.47 |
| Powdered infant formula | 18 | 1.13 | 0.14–5.53 |
| Milk, evaporated | 9 | 0.23 | 0.06–0.55 |

Mean fluoride levels ranged from 0.23 mg/kg for evaporated milk to 1.13 mg/kg for powdered formula concentrate. Dabeka and McKenzie (1987) note that a major source of fluoride in infant formula appeared to be the processing water used by the manufacturer. The concentrations of fluoride in the U.S. products appear to be lower than those in the Canadian products.

Johnson and Bawden (1987) analyzed fluoride levels in infant formulas obtained from local supermarkets in 7 cities across the U.S. (Minneapolis, Los Angeles, New York, Dallas, Seattle, Largo, Florida, and Chapel Hill, North Carolina). Between 7 and 24 products were collected in each location. Concentrated and powdered formulas were reconstituted with de-ionized water according to manufacturers' directions. Those from Chapel Hill were also reconstituted with fluoridated tapwater (1.1 mg F/L). Fluoride was analyzed using the Taves microdiffusion method and a fluoride ion-specific electrode. The mean fluoride concentration in ready-to-feed formulas ranged from 0.06 to 0.38 mg/L. Mean fluoride levels in liquid concentrates reconstituted with de-ionized water ranged from 0.04 to 0.32 mg/L, whereas the Chapel Hill formulas reconstituted with tapwater containing 1.1 mg F/L ranged from 0.60 to 0.72 mg F/L. For the powder concentrates reconstituted with de-ionized water, mean fluoride levels were 0.03 to 0.24 mg/L, whereas those from Chapel Hill reconstituted with tapwater containing 1.1 mg F/L ranged from 1.00 to 1.25 mg/L. The overall mean fluoride concentration was 0.21 mg/L for ready-to-feed formulas, 0.10 mg/L for liquid concentrates and 0.12 mg/L for powder concentrates.

McKnight-Hanes et al. (1988) analyzed fluoride levels in infant formulas purchased in the Rochester, NY, area. The formulas were prepared with de-ionized water or with water containing 0.15 mg F/L or 1.0 mg F/L. Fluoride was separated from 3 mL of the prepared formula as hydrofluoric acid, appropriately buffered, and analyzed directly using a fluoride ion-specific electrode. The Taves method was used to separate the acid-diffusible fluoride from the sample. Results are shown in Table 2-2. Results indicate that there is a significantly greater amount of fluoride in the ready-to-eat soy-based formula and the liquid concentrate soy-based formula than the corresponding milk-based formulas.

**Table 2-2. Mean Fluoride Concentrations (mg/L) in Infant Formulas (McKnight-Hanes et al., 1988)**

| Type | Dilutent | | | Student's t-Test |
|---|---|---|---|---|
| | Deionized Water | 0.15 mg F/L | 1.0 mg F/L | |
| **Milk-based formulas:** | | | | |
| Ready-to-use[a] | 0.127 | – | – | t = 3.3 |
| Liquid concentrates[a] | 0.121 | 0.196 | 0.621 | t = 2.9 |
| Powdered concentrates[a] | 0.055 | 0.170 | 0.825 | t = 1.4 |
| **Soy-based formulas:** | | | | |
| Ready-to-use[a] | 0.305 | – | – | $p < 0.01$[b] |
| Liquid concentrates[a] | 0.242 | 0.317 | 0.742 | $p < 0.02$[b] |
| Powdered concentrates[a] | 0.084 | 0.200 | 0.854 | N.S. |

**SOURCE:** McKnight-Hanes et al., 1988.
[a]Undiluted.
[b]Significantly greater than milk-based, ready-to-use formula.
[c]Significantly greater than milk-based liquid concentrate formula.

Van Winkle et al. (1995) analyzed fluoride levels in water and formula fed to 1,308 children younger than 2 years of age who were participants in the Iowa Fluoride Study. Mothers of newborns completed questionnaires and 3-day food and beverage and dental care diaries which were used to document fluoride intake from diet, supplements and dentifrice. Information was obtained at 6 weeks, when the children were 3 months old, and every 3–4 months thereafter. Water sources other than unfiltered public water supplies were assayed for fluoride using a fluoride ion-specific electrode. All formulas that appeared in the diaries were purchased and analyzed for fluoride using direct readout (DR) from a fluoride ion-specific electrode (milk-based formulas) or the modified Taves microdiffusion method (soy-based formulas) followed by fluoride ion-specific electrode analysis. All powder and liquid concentrates were reconstituted with distilled water (0 mg F/L). Fluoride levels in the various types of formula are given in Table 2-3. Fluoride levels in soy-based formulas were higher than those in milk-based formulas.

**Table 2-3. Fluoride Concentrations in Infant Formulas (Van Winkle et al., 1995)**

| Category | Number of No. Samples | Fluoride Concentration (mg/L) | |
|---|---|---|---|
| | | Mean (mg/L) | Range (mg/L) |
| **Milk-based formulas:** | | | |
| Ready-to-use | 16 | 0.17 | 0.04–0.55 |
| Liquid concentrates[a] | 14 | 0.12 | 0.04–0.19 |
| Powdered concentrates[a] | 17 | 0.14 | 0.05–0.28 |
| **Soy-based formulas:** | | | |
| Ready-to-use | 5 | 0.30 | 0.17–0.38 |
| Liquid concentrates[a] | 6 | 0.24 | 0.04–0.47 |
| Powdered concentrates[a] | 6 | 0.24 | 0.19–0.28 |

**SOURCE:** Van Winkle et al., 1995.

[a]Reconstituted with distilled water.

Siew et al. (2009) analyzed fluoride concentrations of 27 powdered and 13 liquid infant formula concentrates and nine ready-to-feed formulas purchased in the Chicago area. The formulas included both milk-based and soy-based varieties. The powdered and liquid concentrate formulas were reconstituted with deionized water according to the manufacturers' instructions. Powdered formulas were reconstituted by adding 2 ounces of deionized water to one scoop of formula, and liquid concentrates were reconstituted 1:1 with deionized water. The total fluoride content of the formulas was analyzed using a modified Taves diffusion method and a fluoride ion-specific electrode. Results are shown in Table 2-4.

**Table 2-4. Fluoride Levels in Infant Formulas Reconstituted with Deionized Water (Siew et al., 2009)**

| Formula type | Base | Range of values (ppm) | N | Mean ±SD (ppm)[a] | *P* Value[b] |
|---|---|---|---|---|---|
| Powdered concentrate | Milk | 0.03–0.27 | 21 | 0.12 ±0.08 | 0.44 |
| | Soy | 0.06–0.29 | 6 | 0.16±0.09 | |
| Liquid concentrate | Milk | 0.07–0.48 | 8 | 0.27±0.18 | 0.01 |
| | Soy | 0.41–0.57 | 5 | 0.50±0.08 | |
| Ready-to-feed | Milk | 0.08–0.23 | 6 | 0.15±0.06 | 0.46 |
| | Soy | 0.13–0.32 | 3 | 0.21±0.10 | |
| **Overall mean** | | | **49** | **0.1976 ± 0.15** | |

**SOURCE**: Siew et al., 2009.

[a]Mean fluoride concentrations for milk-based formulas are compared with those for soy-based formulas.
[b]The P value is based on a *t* test (unpaired data) comparing the mean values for milk and soy-based formulas.

In general, soy-based formulas were higher in fluoride content than milk-based formulas. This difference was not statistically significant for powdered concentrate and ready-to-feed formulations; however, the fluoride concentration of the liquid concentrate soy formulas tested was significantly higher than that of milk-based liquid concentrate formulas ($P < 0.05$, $t$ test analysis for unpaired data). The fluoride content in different batches of the same product was fairly consistent.

***Infant Foods.*** Singer and Ophaug (1979) measured fluoride levels in a variety of infant foods including meats, vegetables, and fruits using a fluoride ion-specific electrode. Results are shown in Table 2-5. The highest fluoride concentration was found in strained chicken with broth (mean 5.29 mg/kg; range 1.94–10.64 mg/kg). Mean F concentration in vegetables ranged from 0.15-0.43 mg/kg, and those in fruits 0.017-0.078 mg/kg. Fluoride levels in dry cereal varied depending on whether fluoridated water was used in the processing facility. Mixed cereals, oatmeal, rice and barley cereals from facilities using non-fluoridated water contained 0.93, 0.98, 2.11, and 1.99 mg F/kg, respectively, whereas levels in the same types of cereals from plants using fluoridated water were 3.85, 4.87, 6.35, and 4.30 mg/kg, respectively. Mean fluoride levels in fruit juices made with non-fluoridated water ranged from 0.014 to 0.14 mg/L; juices prepared with fluoridated water contained 0.15 to 1.48 mg F/L. Similarly, mean fluoride levels in milk formulations were 0.08–0.31 mg/L when prepared with non-fluoridated water and 0.57–0.66 mg/L when prepared with fluoridated water.

**Table 2-5. Fluoride Concentrations in Infant Foods as Reported by Singer & Ophaug, 1979**

| Food Type | Number of Plants | Fluoride Concentration (mg/kg food) | |
|---|---|---|---|
| | | Mean | Range |
| Strained meats | | | |
| Chicken and broth | 4 | 5.29 | 1.94–10.64 |
| Turkey and broth | 2 | 0.39 | 0.34–0.43 |
| Beef and broth | 3 | 0.19 | 0.17–0.21 |
| Lamb and broth | 2 | 0.29 | 0.16–0.42 |
| Liver and broth | 1 | 0.14 | 0.14 |
| Veal and broth | 1 | 0.40 | 0.40 |
| Pork and broth | 1 | 0.23 | 0.23 |
| **Overall mean** | | **0.99** | |
| Vegetables | | | |
| Carrots | 6 | 0.23 | 0.022–0.53 |
| Peas | 4 | 0.18 | 0.038–0.34 |
| Squash | 4 | 0.15 | 0.046–0.34 |
| Spinach | 2 | 0.43 | 0.18–0.67 |
| Green beans | 3 | 0.16 | 0.036–0.33 |
| Beets | 3 | 0.23 | 0.13–063 |
| **Overall mean** | | **0.24** | |
| Fruits | | | |
| Pears | 7 | 0.057 | 0.012–0.13 |
| Peaches | 5 | 0.017 | 0.003–0.034 |
| Applesauce | 7 | 0.078 | 0.016–0.23 |
| **Overall mean** | | **0.051** | |

**SOURCE:** Singer and Ophaug, 1979.

A group of 206 commercially available, ready-to-eat infant foods purchased in Iowa City, Iowa, were studied by Heilman et al. (1997) using a modified version of the Taves microdiffusion method coupled with a fluoride ion-specific electrode. Fluoride levels ranged from 0.01 to 8.38 mg/kg. A summary of the results by food type is provided in Table 2-6.

**Table 2-6. Fluoride Concentrations in Infant Foods as Reported by Heilman et al., 1997**

| Food Type | No. Samples | Fluoride Concentration (mg/kg food) | | |
|---|---|---|---|---|
| | | Range | Median | Mean |
| Fruits and desserts | 88 | 0.01-0.49 | 0.03 | 0.10 |
| Vegetables | 48 | 0.01-0.42 | 0.08 | 0.12 |
| Mixed foods | 42 | 0.01-0.63 | 0.13 | 0.21 |
| Meats[a] | 19 | 0.01-8.38 | 0.05 | 1.46 |
| Chicken | 6 | 1.05-8.38 | 4.04 | 4.40 |
| Cereals | 9 | 0.01-0.31 | 0.02 | 0.08 |

**SOURCE:** Heilman et al., 1997.

[a]Includes poultry.

The highest fluoride concentrations were found in chicken (1.05-8.38 mg/kg); concentrations in other meats ranged from 0.01 mg/kg in veal to 0.66 mg/kg in turkey. Heilman et al. (1997) reported that the substantial variation in fluoride levels within a given type of food was due primarily to different fluoride concentrations in the water used to process the foods. High fluoride levels in chicken were attributed to the processing methods (mechanical deboning) that leave some skin and residual bone particles in the meat. It was estimated that an infant consuming 2 oz (about 60 g) of chicken containing 8 mg F/kg would have a fluoride intake of about 0.48 mg (Heilman et al., 1997).

Heilman et al. (1997) also analyzed the fluoride content of 32 dry infant cereals and found that the fluoride content ranged from 0.10-0.40 mg/kg. The study authors note that a considerable amount of fluoride may be added to the cereal during manufacturing when the cereal is processed as a slurry which is then dried, leaving any contained fluoride from the process-water behind. Additional fluoride may be later added when the dry cereal is reconstituted with water containing fluoride.

In 2005, the U.S. Department of Agriculture published a National Fluoride Database (USDA, 2005). This database summarizes and critically evaluates the quality of published and unpublished information on the fluoride content of selected foods and beverages from a variety of sources including data from some of the studies cited in this report. The database also includes the results of USDA sampling of food and beverage products at 144 locations across the U.S. (Pehrsson et al., 2000) as well as upublished data from several research projects. The USDA samples were analyzed using a fluoride ion-specific electrode with direct readout for clear liquids, and a microdiffusion method for other foods. The ranges of mean values for various infant foods and beverages are shown in Table 2-7. The USDA data for foods consumed by adults are given in Table 2-15.

| Table 2-7. Fluoride Concentrations in Infant Foods as Summarized by USDA (2005) | |
|---|---|
| Category/food group | Range of Mean Fluoride Concentrations (mg/kg) |
| Cereals | 0.01–0.16 |
| Desserts | 0.02–0.18 |
| Dinners | 0.02–0.29 |
| Fruit | 0.01–0.36 |
| Juice | 0.10–0.70 |
| Meat | 0.02–0.44 |
| Vegetables | 0.01–0.32 |

## 2.2.2. Fluoride in Foods of Children and Adults

San Filippo and Battistone (1971) calculated the fluoride content of representative food items obtained in Baltimore, MD from an FDA "market basket program" on four separate occasions (four diets) in 1967 and 1968. Each diet represented the 2-week food and beverage intake of 16-19 year old males. The food items were placed into 12 commodity groups and analyzed on a composite basis using microdiffusion and spectrophotometry (using erichrome cyanine R and zirconyl chloride). Items were prepared in a manner representative of preparation in the home. Fluoride concentrations are shown in Table 2-8. The highest fluoride levels were found in beverages (beverages included tea, coffee, soft drinks and drinking water). Analysis of the drinking water in the study area indicated that the fluoride level ranged from 0.99 to 1.0 mg/L.

| Table 2-8. Fluoride Content of Food Commodity Groups | | | | |
|---|---|---|---|---|
| Commodity Group | Sample #1 (ppm) | Sample #2 (ppm) | Sample #3 (ppm) | Sample #4 (ppm) |
| Dairy products | 0.19 | 0.22 | 0.15 | 0.11 |
| Meat, fish and poultry | 0.55 | 1.00 | 1.04 | 0.42 |
| Grain and cereal products | 0.49 | 0.44 | 0.26 | 0.59 |
| Potatoes | 0.13 | 0.17 | 0.17 | 0.45 |
| Leafy vegetables | 0.46 | 0.15 | 0.13 | 0.85 |
| Legume vegetables | 0.24 | 0.19 | 0.24 | 0.25 |
| Root vegetables | 0.08 | 0.06 | 0.07 | 0.22 |
| Garden fruits | 0.41 | 0.09 | 0.07 | 0.18 |
| Fruits | 0.11 | 0.10 | 0.11 | 0.10 |
| Oils, fats, shortenings | 0.45 | 0.24 | 0.25 | 0.12 |
| Sugar and adjunct | 0.44 | 0.30 | 0.33 | 0.56 |
| Beverages[a] | 1.22 | 1.07 | 1.10 | 1.10 |

**SOURCE:** San Filippo and Battistone, 1971.

[a]Tea, coffee, soft drinks and drinking water.

Singer et al. (1980) evaluated fluoride concentrations in 117 food items placed in 12 composite food groups for four geographic regions of the United States. Fluoride in the food items was determined by four methods: ashed and unashed samples quantified using a fluoride ion-specific electrode and colorimetric analysis (eriochromecyanine R procedure). The results from the ion-specific electrode were found to be more accurate than the colorimetric method, especially for unashed samples. Mean fluoride levels in the composite food groups are shown in Table 2-9. Beverages represented the single highest source of fluoride (0.82–1.35 ppm).

**Table 2-9. Fluoride Content of Composite Food Groups for Four Geographic Regions of the U.S.**

| Commodity Group | San Francisco | Buffalo | Atlanta | Kansas City |
|---|---|---|---|---|
| | ppm F | ppm F | ppm F | ppm F |
| Dairy | 0.05 | 0.05 | 0.07 | 0.05 |
| Meats, fish, poultry | 0.22 | 0.22 | 0.92 | 0.32 |
| Grain and cereal products | 0.34 | 0.39 | 0.41 | 0.29 |
| Potatoes | 0.14 | 0.08 | 0.13 | 0.14 |
| Leafy vegetables | 0.13 | 0.13 | 0.15 | 0.10 |
| Legume vegetables | 0.15 | 0.24 | 0.39 | 0.31 |
| Root vegetables | 0.09 | 0.10 | 0.10 | 0.09 |
| Misc. vegetables | 0.15 | 0.14 | 0.06 | 0.17 |
| Fruits | 0.06 | 0.13 | 0.07 | 0.06 |
| Oils, fats | 0.24 | 0.13 | 0.15 | 0.15 |
| Sugars, adjuncts | 0.21 | 0.24 | 0.32 | 0.35 |
| Beverages[a] | 1.35 | 0.82 | 1.54 | 0.83 |

**SOURCE:** Singer et al., 1980.

[a]Includes tea, coffee, soft drinks, and water.

Food items in FDA toddler "Market Basket" collections made in 1977 and 1978 were analyzed for fluoride by Ophaug et al. (1980b). The food items were placed in 11 composite groups for four cities of the United States. Results are shown in Table 2-10. In all four locations, beverages contained the highest concentrations of fluoride (0.54–1.19 ppm).

Taves (1983) measured fluoride levels in 93 foods and beverages included in a standard hospital diet. The study authors note that the hospital was in a fluoridated area and consequently any foods prepared with water reflect this factor. The concentration of fluoride in the tapwater was not reported. Inorganic and total fluoride levels were determined on ashed and unashed samples using the hexamethyldisiloxane (HMDS) microdiffusion (Taves) method coupled with a fluoride ion-specific electrode. Range of mean levels in various food groups were reported as nanomole per gram food and were converted to measures of mg/kg or mg/L by the IOM (1997). The transformed results are given in Table 2-11. The highest fluoride concentration (144 nm/g; about 2.7 mg/L) was found in tea.

| Table 2-10. Fluoride Content of Four Representative Diets for 2-Year-Olds | | | | |
|---|---|---|---|---|
| Commodity Group | Orlando | Grand Rapids | Philadelphia | Los Angeles |
| | ppm F | ppm F | ppm F | ppm F |
| Drinking water | 0.67 | 1.04 | 0.66 | 0.37 |
| Whole milk | 0.02 | 0.02 | 0.02 | 0.02 |
| Other dairy | 0.10 | 0.19 | 0.14 | 0.08 |
| Meats, fish, poultry | 0.48 | 0.44 | 0.37 | 0.22 |
| Grain and cereal products | 0.23 | 0.30 | 0.27 | 0.47 |
| Potatoes | 0.04 | 0.12 | 0.11 | 0.19 |
| Vegetables | 0.24 | 0.17 | 0.22 | 0.18 |
| Fruits and juices | 0.22 | 0.25 | 0.11 | 0.15 |
| Oils, fats | 0.29 | 0.45 | 0.24 | 0.15 |
| Sugars, adjuncts | 0.24 | 0.44 | 0.25 | 0.36 |
| Beverages[a] | 0.94 | 1.19 | 0.55 | 0.54 |

**SOURCE:** Ophaug et al., 1980b.

[a]Includes carbonated and noncarbonated soft drinks, Kool-Aid, and tea.

| Table 2-11. Fluoride Concentrations in Food Products | | |
|---|---|---|
| Category | Fluoride Concentration (mg/kg or mg/L) | |
| | Mean | Range |
| Dairy products | 0.25 | 0.02–0.82 |
| Meat, fish and poultry | 0.22 | 0.04–0.51 |
| Grains and cereal products | 0.42 | 0.08–2.01 |
| Potatoes | 0.49 | 0.21–0.84 |
| Leafy vegetables | 0.27 | 0.08–0.70 |
| Legume vegetables | 0.53 | 0.49–0.57 |
| Root vegetables | 0.38 | 0.27–0.48 |
| Fruits | 0.06 | 0.02–0.08 |
| Sugars, etc. | 0.28 | 0.02–0.78 |
| Beverages[a] | 0.76 | 0.02–2.74 |
| Fats and oils | 0.25 | 0.02–0.44 |
| Miscellaneous | 0.59 | 0.29–0.87 |

**SOURCE:** Taves, 1983, as modified in IOM, 1997.

[a]Does not include drinking water, but does include beverages made with tapwater.

Fluoride was determined with an ion-specific electrode after microdiffusion separation in various foods obtained in 1987 in Winnipeg, Canada and reported in Dabeka and McKenzie (1995). Mean fluoride levels for various food groups ranged from 0.095 mg/kg for fruits and fruit juices to 2.1 mg/kg for fish (Table 2-12). The highest single items were cooked veal (1.2 mg/kg), canned fish (4.6 mg/kg), shellfish (3.4 mg/kg), cooked wheat cereal (1.0 mg/kg), and tea (5.0 mg/kg). The mean for all samples was 0.325 mg/kg and the range was <0.011 to 4.97 mg/kg.

**Table 2-12. Fluoride Concentrations in Foods Obtained in Winnipeg, Canada**

| Category | Number of Samples | Fluoride Concentration (mg/kg food) | |
|---|---|---|---|
| | | Mean | Range |
| Milk and milk products | 12 | 0.189 | <0.012–0.797 |
| Meat and poultry | 17 | 0.251 | 0.037–1.230 |
| Fish | 4 | 2.118 | 0.213–4.567 |
| Soups | 4 | 0.606 | 0.412–0.836 |
| Vegetables | 38 | 0.146 | <0.011–0.678 |
| Fruits and fruit juices | 25 | 0.095 | <0.011–0.582 |
| Bakery goods and cereal | 24 | 0.402 | <0.011–0.678 |
| Fats and oils | 3 | 0.096 | 0.046–0.132 |
| Sugar and candies | 7 | 0.111 | <0.016–0.275 |
| Beverages | 7 | 1.148 | 0.213–4.970 |
| Miscellaneous[a] | 7 | 0.564 | 0.075–1.000 |

**SOURCE:** Dabeka and McKenzie, 1995.

[a]Includes tapwater (Dabeka and McKenzie, 1995, Table 2).

A recent study by Jackson et al. (2002) surveyed adolescents 12–14 years old to determine the foods and beverages most commonly consumed by this age group. As a result, a total of 441 brand-name food items were purchased in both a non-fluoridated community (Connersville, IN; fluoride 0.16 ±0.01 mg/L drinking water) and in a fluoridated community (Richmond, IN; 0.90 ±0.05 mg/L). The foods and beverage items were placed into dietary groups according to USDA guidelines, and the most up-to-date methods for analyzing for fluoride were used. Fluoride in water and carbonated beverages was analyzed directly by using a combined fluoride ion-specific electrode and pH/ion meter. Measurements were compared to a series of standards. Fluoride in foods, juices and milk was analyzed with the HMDS silicon-facilitated microdiffusion method of Taves (1968b) as modified by Rojas-Sanchez (1999). This method, which does not require pre-ashing of the sample, was recommended in 1981 by the Association of Official Analytical Chemists as the separation method of choice for analyzing fluoride in infant foods. The method was validated in a series of spike and recovery tests. The Intraclass Correlation Coefficient of the measurements was calculated to assess the reliability of the analyses (Bartko and Carpenter, 1976). The fluoride content of food and beverage items that were not cooked or reconstituted with tap water did not vary significantly between the two towns; therefore, measurements of the fluoride content of these items were assessed together (Table 2-13). However, fluoride content of some food items reconstituted with or prepared in tap water (beverages and grain products) was significantly different for several food categories (Table 2-14). The beverage items prepared with the local Richmond tap water (0.9 mg/L) had significantly higher fluoride concentrations then those prepared with the Connersville tap water (0.16 mg/L).

| Table 2-13. Fluoride Concentrations of Noncooked and Nonreconstituted Foods and Beverages Consumed by Adolescents 12-14 Years Old[a] | | | | | |
|---|---|---|---|---|---|
| Category[e] | N | Fluoride Concentration (mg/kg) | | | |
| | | Mean | Minimum | Maximum | 95% CI |
| Grains and cereal products[b] | 129 | 0.49±0.25 | 0.007 | 1.36 | 0.44, 0.53 |
| Vegetables[c] | 78 | 0.25±0.28 | 0.003 | 1.93 | 0.18, 0.31 |
| Fruits | 26 | 0.12±0.21 | 0.01 | 0.84 | 0.04, 0.20 |
| Dairy products | 30 | 0.31±0.29 | 0.23 | 1.36 | 0.20, 0.42 |
| Meat, poultry | 55 | 0.36±0.30 | 0.03 | 1.41 | 0.28, 0.44 |
| Nuts and seeds | 4 | 0.16±0.03 | 0.13 | 0.19 | 0.12, 0.20 |
| Fats and oils | 14 | 0.24±0.17 | 0.05 | 0.62 | 0.15, 0.34 |
| Sugars and sweets | 15 | 0.29±0.19 | 0.07 | 0.60 | 0.19, 0.40 |
| Beverages[d] | 32 | 0.55±0.26 | 0.04 | 0.93 | 0.46, 0.65 |

**SOURCE:** Jackson et al., 2002.

[a]Combined data for foods and beverages purchased in Connersville and Richmond, IN.
[b]Excludes foods prepared with water.
[c]Excludes foods cooked with water.
[d]Excludes reconstituted and fountain beverages.
[e]USDA food categories.

| Table 2-14. Fluoride Concentrations (mg/kg) of Drinking Water and Foods and Beverages Reconstituted in or Cooked in Tapwater | | | | | | | |
|---|---|---|---|---|---|---|---|
| Food Group[d] | Connersville, IN | | | Richmond, IN | | | P value |
| | N | Mean | SD | N | Mean | SD | |
| Water[e] | 3 | 0.16 | 0.01 | 3 | 0.90 | 0.05 | <0.01 |
| Beverages | | | | | | | |
| Bottled fruit drinks | 4 | 0.44 | 0.40 | 4 | 0.65 | 0.39 | 0.49 |
| Bottled carbonated beverages | 12 | 0.58 | 0.21 | 12 | 0.53 | 0.24 | 0.59 |
| Reconstituted/fountain carbonated beverages[a] | 10 | 0.16 | 0.04 | 12 | 0.78 | 0.29 | <0.01[e] |
| Grain products[b] | 13 | 0.26 | 0.11 | 11 | 0.86 | 0.47 | 0.01[e] |
| Vegetables[c] | | | | | | | |
| Raw | 3 | 0.10 | 0.06 | 3 | 0.10 | 0.10 | 0.99 |
| Cooked | 3 | 0.08 | 0.06 | 3 | 0.73 | 0.70 | 0.18 |

**SOURCE:** Jackson et al., 2002.

[a]Includes juices, powdered drinks and fast food fountain drinks.
[b]Includes cooked cereals, pastas, soups.
[c]Includes carrots, cauliflower, broccoli.
[d]USDA food categories.
[e]Significantly different between Connersville and Richmond.

The USDA (2005) database on foods provided information on foods consumed by the general population as well data on infant foods (see Table 2-7). The ranges of mean values for various food categories are shown in Table 2-15. The food categories in Table 2-15 are the headings used in the database.

This database is the most comprehensive source of information on the concentrations of fluoride in foods, but is incomplete because many foods found in an average U.S. diet are not included. The database was developed from the data reported in many of the publications cited in this report after critical review of the data and supplemented by data for foods collected and analyzed by USDA (mostly beverages) during development of the database. USDA used a "key foods" approach when selecting the materials they sampled for creation of the database; giving consideration to previously published fluoride data for foods, beverages, and drinking water as well as the respective patterns of consumption of these dietary items. Mean estimates of fluoride concentration and variability in drinking water, beverages and foods that are the chief contributors to dietary fluoride in the United States were developed from analyses of representative samples.

**Table 2-15. Fluoride Concentrations in Foods as Summarized by the USDA, 2005**

| Category/food group | Range of Mean Fluoride Concentrations (mg/kg) |
|---|---|
| Baked goods | 0.13–0.69 |
| Beef products | 0.05–0.22 |
| Breakfast cereals | 0.17–0.72 |
| Cereal grains and pastas | 0.06–0.41 |
| Dairy and egg products | 0.01–0.35 |
| Cream substitute-powdered | 1.12 |
| Fast foods | 0.13–1.15 |
| Fats and oils | 0.01–0.27 |
| Finfish and shellfish | 0.18–2.10 |
| Fruits and fruit products | 0.01–2.34 |
| Lamb, veal and game | 0.05–0.32 |
| Legume and legume products | 0.02–0.54 |
| Meals, entree, side-dishes | 0.13–0.84 |
| Nuts | 0.10 |
| Pork products | 0.04–0.38 |
| Poultry | 0.15–0.21 |
| Sausages and luncheon meats | 0.16–0.48 |
| Snacks | 0.06–1.06 |
| Soups, sauces and gravies | 0.04–1.32 |
| Spices and herbs | 0.02–0.34 |
| Sweets | 0.01–0.89 |
| Vegetables and vegetable products | 0.01–0.55 |

**SOURCE:** USDA, 2005.

### 2.2.3. Summary of the Data on Fluoride in Solid Foods

There is some consistency in the data on the concentrations of fluoride in foods despite the differences in analytical methods, preparation, and sampling practices. The solid foods highest in fluoride are fish and shellfish, reflective of the fluoride found in ocean water (~13 ppm). Most samples of fish and shellfish that have been analyzed contain greater than 1 ppm (Jackson et al., 2002; Singer et al., 1980; USDA, 2005). More recent analyses of fish samples are lower than those from the early studies (range 0.18–2.10; USDA, 2005). Choice of analytical methods can account for some of these differences as can the presence or absence of bone fragments in the sample analyzed.

When foods were grouped for analysis, the inclusion of fish in a grouping with meat and poultry tended to lead to a higher mean value than found for meat or poultry alone or combined (Dabeka and McKenzie, 1995; Jackson et al., 2002; USDA, 2005). Chicken had a relatively high fluoride content (>1 ppm) in baby foods (1.20–8.38 ppm; Heilman et al., 1997). However, in the USDA (2005) database chicken has a far lower average value (0.15 ppm) than reported in some of the earlier studies. The USDA value for chicken is attributed to Featherstone (1988), Jackson (2002) and Ophaug (1983–1987).

The majority of vegetables, be they leafy, root, legumes, green or yellow, have a relatively low fluoride concentration (<0.5 ppm; IOM 1997; Singer et al., 1980; USDA, 2005). The concentrations for fruits were generally lower than those in vegetables (< 0.2 ppm) in most assays (IOM, 1997; Singer et al., 1980; Singer and Ophaug, 1979; USDA, 2005). Table 2-15 derived from the USDA database indicated a concentration range of 0.01 to 2.34 ppm for fruits. However, in this case, the high end measure (raisins) was an outlier reflecting the use of cryolite as a pesticide on grapes and concentration through drying. Fresh grapes had 0.08 ppm fluoride; the concentration in all other fresh fruit was < 0.04 ppm.

Cereals, baked goods, breads, and other grain products tended to have fluoride concentrations between about 0.5 and 1 ppm (IOM, 1997; Jackson et al., 2002; Singer et al., 1980). Dairy product fluoride concentrations, as reported by IOM (1997), Singer et al. (1980), and USDA (2005) were low (<0.5 ppm).

Infant foods have a tendency to have a higher liquid content than foods for toddlers through adults in order to minimize chewing and increase the ease of swallowing. When fluoridated water is used in their preparation, this can add to the total fluoride concentration. Most infant foods studied had concentrations less than 0.5 ppm if they were cereal, fruit or vegetable based and less than 1 ppm if they contained meat. Products containing chicken had a higher fluoride concentration than those with meat or turkey in several studies (Heilman et al., 1997; Singer and Ophaug, 1979). However, this was not the case with the meat and poultry containing infant foods in the USDA (2005) database where concentrations were less than 0.5 ppm. The USDA data for the meat and poultry-containing baby foods was attributed to unpublished data from Steven Levy (University of Iowa).

In a study of maternal milk by Dabeka et al. (1986) fluoride levels were below detection in 56% of the 210 samples tested and the mean concentration in areas without fluoridated water was 0.0044 mg/L while those receiving fluoridated water (1 mg/L) was 0.0098 mg/L. These differences were significant (p = 0.007). The IOM (1997) reported that the fluoride concentration in human breast milk ranged from 0.007 to 0.011 ppm.

The fluoride concentration in infant formula is difficult to assess and depends on the brand and form of the formula product (i.e., liquid, concentrate, powder; Dabeka and McKenzie, 1987) and the protein source (milk protein or soy protein; Van Winkle et al, 1995). In US products analyzed by Dabeka and McKenzie (1987) the mean fluoride levels ranged from 0.23 mg/kg for ready-to-serve products to 1.13 mg/kg for powdered formula concentrate. Fluoride from the dilution water further increases the total fluoride from formula (as served) in the case of concentrated and powdered products. For milk-based formulas Van Winkle et al. (1995) reported mean values of 0.17 mg/L for ready-to-use products, 0.12 mg/L for liquid concentrates, and 0.14 mg/L for powdered concentrates. In the case of soy formulas, the comparable values were 0.30 mg/L (ready-to-use) and 0.24 mg/L (liquid and powdered concentrate. Distilled water was used to prepare the samples for analysis.

In the most recent analysis of U.S. infant formula, Siew et al. (2009) reported mean values of 0.15, 0.27 and 0.12 ppm for milk-based, ready-to-use, liquid concentrates, and powdered concentrates, respectively, and 0.21, 0.50, and 0.16 ppm for soy-based, ready-to-use, liquid concentrates and powdered concentrates, respectively. The overall mean for all products combined was 0.198 ppm.

It is difficult to tell if changes in analytical methods over time have influenced the results from studies of fluoride in foods. Singer et al. (1980 found that the results with an ion-specific electrode were more accurate than a colorimetric method and that ashed samples gave different results from unashed samples for some food groups but not for others. Table 2-16 compares the results from several studies conducted over the past 30 plus years that grouped the foods in the same manner. No pattern is apparent in the results reported. The analytical results are likely influenced by the products represented in a food group, food growth and preparation practices, as well a variety of other variables that are difficult to quantify.

**Table 2-16. Comparison of Food Group Measures over a 30-Year Period**

| Food group | San Filippo and Battistone, 1971[a,b] (mg/kg) | Singer et al., 1980[b] (mg/kg) | Taves, 1983/ IOM, 1997 (mg/kg) | Jackson et al., 2002 (mg/kg) | USDA 2005 |
|---|---|---|---|---|---|
| Dairy products | 0.17 | 0.06 | 0.25 | 0.31 | 0.01-0.33 |
| Meat fish poultry | 0.75 | 0.24 | 0.22 | 0.36 | 0.04-2.10 |
| Grains and cereals | 0.45 | 0.36 | 0.42 | 0.49 | 0.06-0.72 |
| Leafy vegetables | 0.4 | 0.13 | 0.27 | 0.25 | 0.01-0.55 |
| Legume vegetables | 0.23 | 0.27 | 0.53 | | |
| Root vegetables | 0.10 | 0.1 | 0.38 | | |
| Fruits | 0.11 | 0.08 | 0.06 | 0.12 | 0.01-.0.13[c] |

[a]Average of 4 measurements.
[b]Colorimetric method used by San Fillipo and Battistone (1971) has a tendency to give higher results than the ion-specific electrode used by the other researchers (Singer et al., 1980).
[c]Raisins not included.

Cooking and preparing foods with water that contains fluoride increases the fluoride content of the food as served (Marier and Rose, 1966). This is true for home-prepared and commercial foods. However, the uptake of fluoride from the process water varies with the food product. This may relate to the presence of cations in the water that form poorly soluble fluoride salts such as calcium fluoride reducing fluoride uptake into the finished product to a greater extent than those like sodium that form soluble salts or from fluoride in the water reacting with these same ions in the food and increasing the fluoride from water retained in the cooked product.

Maier and Rose (1966) analyzed the fluoride content of canned vegetables processed at plants using low-fluoride water and plants using municipal water with 1 mg F/L using a micro-distillation method coupled with colorimetric/spectrophotometric detection. Use of fluoridated process water increased the fluoride content of the vegetables by 0.34 to 0.75 mg/kg (average 0.5 mg/kg) (Table 2-17). Although the values measured are likely to be high because of the colorimetric quantification, they do illustrate the impact of processing foods with fluoridated water. However, Ophaug (1985) reported that there was not a strong correlation between the local fluoride drinking water concentration and total fluoride intake from solid foods in market basket studies, most likely reflecting the combination of purchased and home prepared foods in a normal diet.

**Table 2-17. Fluoride Content of Canned Vegetables**

| Food | Average Fluoride Content (mg/kg)[a] | | | |
|---|---|---|---|---|
| | Non-fluoridated Process Water | | Fluoridated Process Water (1 mg F/L) | |
| | Liquid | Solid | Liquid | Solid |
| Mixed vegetables | 0.30 | 0.37 | 1.03 | 1.05 |
| Green beans | 0.14 | 0.20 | 0.71 | 0.89 |
| Whole potatoes | 0.13 | 0.38 | 0.87 | 0.76 |
| Diced carrots | 0.30 | 0.19 | 0.55 | 0.61 |
| Kernel corn | 0.10 | 0.20 | 0.48 | 0.56 |
| Green peas | 0.15 | 0.10 | – | – |
| Wax beans | – | – | 0.49 | 0.60 |

**SOURCE:** Marier and Rose, 1966.

[a]Results are averages of single determinations for duplicate samples.

## 2.3. Fluoride in Beverages

Beverages are a major source of human dietary exposure to fluoride, especially after fluoridation of public drinking water became widespread and before the growth in bottled water intake. Exposure from plain (e.g., non-beverage) drinking water is summarized in Section 3 of this report. Data on other beverages are presented below.

### 2.3.1. Non-Alcoholic Beverages

Clovis and Hargreaves (1988) published data (Table 2-18) from a study by Hargreaves which measured fluoride levels in beverages in two towns in Canada. One town had a fluoridated water supply (average adjusted fluoride concentration of 1.08 mg/L) and the other had a natural fluoride level of 0.23 mg/L. Fluoride levels in commercially prepared beverages were similar in the two towns; however, fluoride levels in home-prepared beverages were substantially higher in the community with the fluoridated water supply.

Stannard et al. (1991) measured fluoride levels in 43 ready-to-drink fruit juices purchased in the Boston area; however, the products were bottled in various locations around the U.S. Fluoride was measured using a fluoride ion-specific electrode. Fluoride concentrations ranged from 0.15 to 6.80 ppm. Forty-two percent of the samples had a fluoride content of greater than 1 ppm. Grape juice had the highest levels of fluoride (1.94–6.80 ppm), most likely reflecting the use of cryolite as a pesticide on grapes.

Fluoride concentrations were measured in 532 different juices and juice-flavored drinks (including five teas) purchased in Iowa City by Kiritsy et al. (1996). Many of the products were distributed nationally or internationally. Frozen-concentrated beverages were reconstituted with distilled water before analysis. The fluoride concentration ranged from 0.02 to 2.8 mg/L (mean, 0.56 mg/L). Upper limits on most kinds of juices exceeded 1.50 mg/L. The highest mean fluoride concentration (1.45 mg/L) was found in white grape juice.

**Table 2-18. Fluoride Concentrations in Beverages in Two Canadian Towns**

| Category | Fluoride Concentration (ppm) | |
|---|---|---|
| | Town #1 (1.08 mg/L) | Town #2 (0.23 mg/L) |
| Milk | 0.03 | 0.03 |
| Carbonated beverages | 0.80 | 0.80 |
| Commercially prepared juice | 0.80 | 0.80 |
| Home prepared juice | 1.06 | 0.21 |
| Soups | 1.06 | 0.21 |
| Tea | 2.18 | 1.33 |
| Coffee | 1.08 | 0.23 |
| Other beverages prepared with tapwater | 1.08 | 0.23 |
| Misc., prepared with 0.1 ppm water | 0.10 | 0.10 |

**SOURCE:** Hargreaves, unpublished, as cited in Clovis and Hargreaves, 1988.

The fluoride content of 332 carbonated beverages was measured by Heilman et al. (1999). The beverages were purchased in Iowa, but produced at 17 different locations. Mean concentrations of fluoride ranged from 0.04 mg/L to 1.06 mg/L (overall mean 0.72±0.34 mg/L).

Turner et al. (1998) reported fluoride levels of 0.68–0.91 ppm (mean 0.78±0.07 ppm) in carbonated drinks bought in Houston, TX, and 0.0–0.73 ppm (mean 0.33±0.28 ppm) in carbonated drinks bought in San Antonio, TX. Levels of fluoride in ready-to-drink juice drinks were 0.28–1.08 ppm (mean 0.77 ±0.21 ppm) in Houston and 0.16–1.02 ppm (mean 0.58 ±0.38 ppm) in San Antonio. Fluoride determinations were made with a fluoride ion-specific electrode.

Various brands and kinds of coffee sold in the Houston area were analyzed for fluoride by Warren et al. (1996). All samples were prepared with deionized distilled water. Fluoride levels ranged from 0.10 to 0.58 mg/L. The mean concentration for decaffeinated coffee was 0.14 mg/L and that for caffeinated 0.17 mg/L. Instant coffee had a mean fluoride content of 0.30 mg/L.

The USDA (2005) database contains mean values for a variety of beverage categories as shown in Table 2-19. The results are consistent with those reported in other publications. In order to examine more closely the possible relationship between the concentrations of fluoride in carbonated beverages and possible use of tap water containing fluoride in the production of such beverages, the OW evaluated the mean and maximum concentration for the large sample sets (28–72 samples/set) in the USDA (2005) fluoride database. The mean of the means for six different carbonated beverage sets (4 colas) was 0.53 mg/L while the mean of the maximum values was 0.97 mg/L. Since the ingredients other than water in such beverages are not notably rich in fluoride, much of the fluoride present appears to come from the water component of the beverage.

**Table 2-19. Fluoride Levels in Beverages as Summarized by the USDA, 2005**

| Category/food group | Range of Mean Fluoride Concentrations (ppm) |
|---|---|
| Carbonated, non-alcoholic drinks | 0.14–0.84 |
| Carbonated flavored water | 0.84–1.05 |
| Chocolate, ready-to-drink | 0.87 |
| Coffee | 0.52–0.91 |
| Grain-based coffee substitute | 1.25 |
| Fruit juices and drinks | 0.08–1.09 |

Tea is a rich source of fluoride; concentrations vary depending on the type of tea, its source, and the age of the leaves. The fluoride content of buds and young leaves ranges from 100 to 430 mg/kg, whereas that of older leaves ranges from 530 to 2350 mg/kg (Lu et al., 2004). Data on the fluoride concentration of teas are summarized in Table 2-20.

**Table 2-20. Fluoride Concentration in Tea as Served**

| Study | Type | Concentration (ppm) | Notes |
|---|---|---|---|
| Cao et al., 2006 | Black tea sticks | 0.95–1.41 | |
| | Black tea granules | 0.7–2.44 | |
| | Black tea bags | 1.15–6.01 | Aged tea leaves |
| Chan and Koh, 1996 | Caffeinated | 0.34–3.71 | 44 brands; brewed 5 to 120 minutes |
| | Decaffeinated | 1.10–5.2 | |
| | Herbal | 0.02–0.14 | |
| USDA, 2005 | Caffeinated | 3.10–3.93 | |
| | Decaffeinated | 2.47–2.93 | |
| | Iced Tea | 0.72–1.23 | |
| | Green Tea | 1.15–2.72 | Caffeinated and decaffeinated |
| | Herbal | 0.13–0.90 | Chamomile, peppermint |
| Whyte et al., 2005 | Caffeinated and decaffeinated | 1–6.5 | Prepared with distilled water |

### 2.3.2. Alcoholic Beverages

Fluoride is present in a number of alcoholic beverages, especially wines, due to the use of cryolite as a pesticide on grapes. Burgstahler and Robinson (1997) reported fluoride levels of 0.23–2.80 ppm (mean 1.02 ppm) in California wines. Seven of 19 samples tested above 1 mg/L. Fluoride was determined using a fluoride ion-specific electrode. Martínez et al. (1998) reported mean fluoride concentrations ranging from 0.08 to 0.68 mg/L in 70 wines from the Canary Islands. The overall mean concentration was 0.16 mg/L. USDA (2005) found a mean concentration of 1.05 ppm from 14 red wine samples and 2.02 ppm for 17 white wine samples.

Warnakulasuriya et al. (2002) reported mean fluoride concentrations of 0.08–0.71 mg/L in eight kinds of beers available in Great Britain. The concentrations were the equivalent of 0.03–0.31 mg fluoride in one 440 mL can. USDA (2005) reported a mean of 0.45 ± 0.023 ppm for 142 light beer samples and 0.44±0.025 ppm for 102 regular beer samples. The average fluoride in distilled alcoholic beverages was 0.08 ppm in the USDA (2005) database.

### 2.3.3. Summary for Fluoride in Beverages

The fluoride in commercial products tends to reflect the water source at the plant where juices and carbonated beverages are processed. In most instances concentrations in carbonated beverages ranged between 0.7 and 1 ppm, reflecting the concentrations in fluoridated water (Clovis and Hargreaves, 1988; Heilman et al., 1999; Schulz et al., 1976; Turner et al., 1998, USDA, 2005). Commercial fruit juices have the same or slightly lower means (Clovis and Hargreaves, 1988; Kiritsy et al., 1996; Stannard et al., 1991, USDA, 2005), although the means for grape-based products can be higher. USDA (2005) reported the mean concentration of grape juice as 0.77 mg/kg for 20 samples of regular grape juice and 2.13 mg/kg for 12 samples of white grape juice. Home-prepared products appear to reflect the concentration of the local water supply (Clovis and Hargreaves, 1988; Jackson et al., 2002).
Tea is a rich source of fluoride, especially when made from aged leaves (Cao et al., 2006). Herbal teas do not have the high fluoride content of real teas (Chan and Koh, 1996). All of the samples of brewed black tea analyzed by USDA (2005) had a mean fluoride concentration of > 3 ppm. Brewed herbal teas and green teas had lower concentrations. Three popular brands of bottled commercial ice teas had means between 0.72 and 1.23 mg/L (USDA, 2005).

Among alcoholic beverages, wines have the highest fluoride levels (usually 1–2 ppm) likely reflecting the cryolite use in the growing of grapes (Burgstahler and Robinson, 1997; Martinez et al., 1998; USDA, 2005). Levels of fluoride in distilled alcoholic beverages are low (<0.1 ppm; USDA, 2005) and those in beer are intermediate, about 0.4 to 0.5 ppm for U.S. products (USDA, 2005).

## 2.4. Indirect Exposure from Pesticide Residues on Food

**Cryolite.** Cryolite (sodium aluminofluoride; $Na_3AlF_6$) was first registered for use as a pesticide in the U.S. in 1957 (U.S. EPA, 1996). It is used on fruits, vegetables and ornamental plants to protect against leaf eating insects. The major products treated with cryolite are grapes, citrus fruits, and potatoes. Applications rates are frequently high, and application can occur several times during a growing season (U.S. EPA, 1996).

According to NRC (2006), the high fluoride content of grape juices (and grapes, raisins, and wines), even when little or no manufacturing water is involved, is thought to be due to a cryolite used in grape growing (Stannard et al., 1991; Kiritsy et al., 1996; Burgstahler and Robinson, 1997). The water-extractable fluoride in five brands of California raisins ranged from 0.83 mg/kg to 5.20 mg/kg (mean 2.71 mg/kg). Soaking the raisins in distilled water for 1–2 hr resulted in the release of 70–90% of the fluoride, suggesting that the fluoride was concentrated on the skin of the fruit (Burgstahler and Robinson, 1997).

One study reported by Waldbott (1963) showed that celery leaves sprayed with cryolite had fluoride residues of 77.0–135.0 ppm F whereas the normal levels of fluoride in celery were reported to be 0.7–5.7 ppm. Similarly, 2.0–4.5 ppm F was found on sprayed apples compared with 0.04–1.3 ppm F on unsprayed apples.

The market basket dietary data reported in this document include fluoride exposure from cryolite because of its long history of use on a variety crops. To avoid counting the exposure to fluoride from cryolite twice, the additional estimates of cryolite residue values provided by OPP (U.S. EPA, 2009, see Appendix A) were not directly incorporated into the EPA exposure assessment.

There is uncertainty surrounding the OPP estimation of fluoride exposure through cryolite, because the current analytical methods are unable to differentiate the various aluminum fluoride species in each product and instead report total fluoride. Thus, it is possible that the residue estimates could represent an overestimate. In the OPP assessment (U.S. EPA, 2009), the highest level of fluoride residues was contributed by the OPP "other" food group which includes grape and grape juice among other miscellaneous commodities such as coco beans, and coconut. About 60% of the total fluoride residue in the "other" group comes from cryolite rather than sulfuryl fluoride (See Appendix A).

**Sulfuryl Fluoride.** Sulfuryl fluoride, initially also known as Vikane, is a pesticide that was not registered for food use when the studies reported in Section 2.2.2 were conducted. Therefore, fluoride residues from its use are not included in the data presented. Sulfuryl fluoride was developed by Dow Chemical in the late 1950s as a structural fumigant. It was first registered by the OPP in December 1959 and first marketed in the United States in 1961. Sulfuryl fluoride is now produced and sold by several manufacturers, under various brand names.

Sulfuryl fluoride is highly reactive and breaks down to form sulfate and fluoride anions. Parent sulfuryl fluoride and the fluoride anion are the OPP's residues of concern for both tolerance expression and risk assessment. It is considered to be an effective replacement for ozone depleting methyl bromide, the conventional pesticide that had been used for structure fumigation.

On February 7, 2002 the Federal Register established temporary tolerances for residues of sulfuryl fluoride and inorganic fluoride in or on walnuts and raisins. The temporary tolerances were established to support an Experimental Use Permit (EUP) that involved testing a possible alternative to methyl bromide in the post-harvest fumigation of stored walnuts and raisins (Fed Reg. 67(59):14713–14714). The temporary tolerances supported a 3-year EUP effective between March 1, 2002 and March 1, 2005. There was no apparent exercise of the EUP. An 18-month period was given to allow the treated commodities to clear commerce, meaning the temporary tolerances expired on September 1, 2006.

The OPP was later petitioned by Dow AgroSciences (DAS) to register sulfuryl fluoride to control pests in storage and processing facilities as well as to establish permanent tolerances for residues of sulfuryl fluoride and the fluoride anion on cereal grains, dried fruits, and tree nuts. In 2004 the Health Effects Division (HED) of OPP conducted a human health risk assessment for sulfuryl fluoride. Time-limited tolerances were granted for the requested commodities and facilities, with the understanding that when the National Academies of Sciences (NAS) review of fluoride for the OW was completed, the proposed tolerances were to be revisited.

In January of 2006, HED released a risk assessment that postdated a 2005 FR notice establishing tolerances for residues of sulfuryl fluoride and fluoride resulting from fumigation of additional foods (i.e. milk powder, eggs. cocoa, cheese, meat, coffee) and for food processing facilities. Health-effects related limitations for fluoride exposure were based on the OW's MCLG for fluoride in drinking water (4 mg/L and 2L drinking water per day). The MCLG was under evaluation by NAS at that time. The OPP risk assessment stated that the tolerances would be reevaluated once the NAS report was published.

HED performed a dietary exposure assessment for fluoride from treated food products for this effort at the request of the OW (U.S. EPA, 2009). The analysis incorporates the most recent residue data submitted to the agency by the registrant (Dow AgroSciences). The current analysis is intended to replace the exposure projections from the 2006 OPP risk assessment. The HED report to OW is found as Appendix A in this report.

The OPP report to the OW does not include any experimental data on residues in foods. The OPP exposure analysis is based on residue data from select foods commodities extrapolated to similar foods in deriving exposure estimates for humans. Intake data for food groups were derived from the USDA's Continuing Survey of Food Intakes by Individuals (CSFII). All the OPP exposure estimates utilize percent crop treated information.

The OPP analysis (U.S. EPA, 2009) used percent crops treated values for exposures during fumigation that ranged from 0.1% to 100% for food fumigation based on reports of methyl bromide usage by USDA. Use rate information was incorporated in the analysis to derive anticipated residue values. One hundred percent of the dried beans and legumes (except chick pea and cow pea) were assumed to be fumigated using sulfuryl fluoride as were cocoa beans and a high percentage of walnuts, dates, prunes, raisins, and figs. The percent of crop treated for coarse grains and wheat by-products such as flour was 0.1% and that for rice was 3%. For most nuts, 10% of the crop was estimated to be treated based on the data for methyl bromide.

The OPP (U.S. EPA, 2009) food group exposure estimates are summarized in Table 2-21. Food groups have been consolidated from the OPP tables to be consistent with groups reported in other publications. Twenty-one food groups were reduced to twelve in this process. The values reported are exposures of the general US population to fluoride from sulfuryl fluoride in each food group. The "Other" group includes but is not limited to, cocoa beans, coconut, cranberry, grape, and grape juice products. Based on the data from OPP, grains, legumes, and fruits including fruit juices appear to be the major contributors of fluoride in the U. S. diet through the tolerances granted to sulfuryl fluoride. The "other" food group is another large contributor but is varied in its composition. The data from OPP were reported in units of mg/kg/day. They were converted to mg/day values for this report using a 70 kg body weight consistent with OW policies for the general U.S. population.

**Table 2-21. Estimated Food Group Exposures of the General U.S. Population to Fluoride from Sulfuryl Fluoride Tolerances**

| Food Groups | mg/day[a] |
|---|---|
| Dairy products | 0.0002 |
| Meat & Poultry | 0.0007 |
| Cereal Grains | 0.0297 |
| Leafy vegetables | 0.0016 |
| Legume vegetables | 0.0370 |
| Root, tuber, bulb vegetables | 0.0015 |
| Cucurbit and Fruiting Vegetables | 0.0017 |
| Fruits & Fruit Juices | 0.0044 |
| Tree nuts | 0.0011 |
| Herbs and spices | 0.0002 |
| Oil seeds | <0.0001 |
| Other[b] | 0.0646 |

**SOURCE:** U.S. EPA, 2009.

[a]Based on a 70 kg body weight.

[b]The "other" category applies to foods not captured in one of the other groups including but not limited to cocoa beans, coconut, cranberry, grapes and grape juice.

## 2.5. Estimates of Dietary Fluoride Intake

### 2.5.1. Exposure Assessment Methodologies

Estimates of dietary fluoride exposure are based on studies using several analytical approaches. In reviewing the data it is important to understand the technical framework for each approach as well as its strengths and limitations. The studies included in this Section have relied on combinations of several methods for collecting dietary data for use in an exposure analysis:

- Dietary records
- Dietary recalls
- Food frequency recall
- Market Basket or Total Diet Study (TDS) surveys
- Duplicate plate-type analyses.

The following paragraphs provide background information on each of the methods that were used in generating the fluoride exposure estimates. To facilitate evaluation of the resultant exposure estimate, the studies are grouped by method for three age groupings: infants, children ≤ 14 years, adolescents and adults. Studies examining intakes for children less than 6 month of age are not included because this age group was not identified as a sensitive population in the fluoride dose-response assessment (U.S. EPA, 2010a).

**Dietary Records.** Dietary record studies require participants to keep a diary of the amounts and kinds of foods they consume daily. This approach is useful for assessment of individual or group intakes. Generally a minimum of three days is recommended (Guthrie, 1989), often two week days and one week-end day. Compliance with recording intake tends to decline as the number of days and complexity of record keeping increase.

The accuracy of dietary records is dependant on the literacy and commitment of the participants. Failure to record condiments and other foods taken in small amounts is common. With busy individuals, record keeping can regress to end of the day recall as the study progresses. Some people may fail to record foods they think they should not be eating and favor recording intakes of foods they feel are nutritious.

The dietary record is applicable to other groups who share the characteristics (i.e. age, sex, and ethnicity) of the population that participated in the study, but not to groups with different demographics. They provide information on nutrient intake when they are coupled with food composition databases or analytical data on the amounts of a nutrient in specific foods. Three-day records are best for studies of macronutrient intakes and less-well suited for studies of micronutrients (Nutrition Quest, 2008).

**Dietary Recall.** Dietary recalls are the preferred method for population studies but can also be used for evaluation of individual intakes. The difference between the recall and record approach is the use of a trained interviewer for collection of the recall data. The interview is structured to stimulate the responder's memory. The interviewer has a set of props to assist the respondent in quantifying portion sizes. The use of the interview reduces the requirement for participant literacy and widens the pool of potential participants.

Single 24-hour recalls can be used to describe the average intake of a group or to determine if two groups have similar mean intakes. A single day 24-hour recall is not appropriate for epidemiology studies or for assessing the quality of an individual's diet (Nutrition Quest, 2008). Two- and three-day recalls are popular durations for the recall approach. As was the case for the dietary record, a three-day recall will often target two week days and one weekend day.

Recall intake data are coupled with food composition information from nutrient databases or food analysis information to generate exposure estimates. Studies show that large portion sizes are generally underestimated and small portion sizes overestimated in recall studies (Guthrie, 1989). The recall approach lessens the record-keeping fatigue problems encountered with the dietary record approach.

Two large-scale, recall-based studies in the United States are the National Health and Nutrition Examinations Survey (NHANES) and the Continuing Survey of Food Intake by Individuals (CSFII). NHANES is periodically updated by the National Center for Health Statistics of the Centers for Disease Control and Prevention (CDC). In the NHANES, dietary data are gathered through a 24-hour recall interviews conducted by a trained professional.

The CSFII was conducted by the U.S. Department of Agriculture on a periodic basis. One purpose of this survey was to provide information on the kinds and amounts of food eaten by the U.S. population. Each survey covered 3 years. In each of the survey years, a nationally representative sample of the population was interviewed to provide information on 2 non-consecutive days of food intake using the 24-hour recall approach. The direct tap water intake data reported in USEPA (2000a, 2004) were derived from the CSFII.

**Food Frequency Recall**. In a food frequency recall the subject is asked how frequently foods from a defined list are consumed over a specific time period (i.e. per day, week or month). The list of foods is selected based on the objective of the study, generally targeting foods that are a source of a particular nutrient or group of nutrients. The food frequency questionnaire can be administered by an interviewer or self administered (Nutrition Quest, 2008). Frequency recall data can be used in the development of analytical market baskets that reflect food preferences for age groups of interest, but need to be combined with national intake data for foods or food groups as collected by CSFII or NHANES in order to quantify food group intakes applicable to the population studied.

Food frequency recalls are well suited to examining food preferences focused on intakes of specific nutrients. For example, if there is concern about vitamin A intake of elderly adults, a food frequency recall tool can be developed that focuses on foods know to be high in vitamin A. The population status can be estimated by the frequency at which such foods are consumed (i.e. daily vs. once per week). The food frequency tool is not as well suited to an evaluation of the nutritional status of an average daily diet.

**Market Basket Survey.** A Market Basket Study relies on chemical analysis of a typical diet using foods purchased (market baskets) at different locations and during different seasons of the year. The U.S. Food and Drug Administration uses a Market-Basket approach to track the intake of nutrients and contaminants in the U.S. diet in their Total Diet Study (TDS, Egan et al., 2007). Several of the studies in Section 2.2 used a Market Basket approach for collecting and grouping of foods. The results from those studies provided the data for some of the exposure estimates reported in this section.

The Market Basket approach combines food recall data with chemical analysis of foods that are representative of dietary intakes for different age/gender groups plus the geographic diversity and seasonal influences that influence the foods purchased. The composition of representative diets is derived from food intake studies such as the CSFII Survey. The foods are purchased in different locations and prepared as they would be served. Individual food samples are pooled, homogenized, and analyzed to obtain representative aliquots for the analytes of interest (Egan, 2002). Intake from water that does not become incorporated in the foods as served is not captured by this analysis although analytes transferred to food from water during preparation are captured. Foods can be analysed individually or in narrow food groups such as "white breads" or "cooked apple products" (Egan et al., 2007). The most recent FDA TDS included 280 foods from 12 broad food groups and covered 15 age/gender groupings (Egan et al., 2007).

The TDS represents the typical US diet. It does not provide estimates for individual or population exposure distributions (Egan et al., 2007) unless coupled with the intake distributions of a national survey such as NHANES or CSFII. The TDS data can identify the food groups that are the major source of exposure to a nutrient or contaminant.

**Duplicate Plate or Duplicate Diet Analysis**. In a duplicate plate or duplicate diet study the participants set aside an equivalent weighed portion of each of the foods they consume for analysis. The plate terminology is appropriate in cases where two identical servings of each meal are prepared in a food service setting such as a hospital kitchen. One plate is served and consumed and the other is used for analysis. In the case of a duplicate diet study sometimes duplicate portions of each food consumed are placed directly in one or more dedicated collection vessels and preserved for later analysis (Thomas et al., 1997). Often there are separate collection vessels for solid and liquid foods. At the end of the collection period the foods are homogenized and several aliquots are harvested for chemical analysis of the analytes of interest.

The analyte concentration per mass of the aliquot when scaled to the mass of food collected/consumed produces the estimate of the analyte intake for the day. The estimate of intake is rather accurate for each individual and can be averaged for the participating group providing a mean, median, standard deviation, and range for intake of the analyte. Intake from direct tap water ingestion is not usually captured by this analysis.

The data from a duplicate diet study are limited if they do not identify and quantify the foods contributing the analyte to the diet, and do not easily extrapolate to groups with other dietary habits and/or demographic characteristics (age, gender, etc.). Intake estimates are also impacted if the consumer eats more or less than was placed on the duplicate plate or in the collection vessel.

Carrying out a duplicate diet study is resource intensive (Thomas et al., 1997; Martinez-Mier et al., 2008). It requires dedicated participants if the collection period lasts for more than a day or two. When participants are in a free-living setting they must prepare their foods, record and weigh what they consume, collect the duplicate portion in the dedicated collections vessels and keep the collected foods under conditions that will preserve the analytes and prevent spoilage. Special plans must be made for measuring, collecting and preserving any foods consumed away from the home setting.

Several exposure estimates reported in Section 2.2 involve plate analyses from hospital kitchens. This type of analysis represents foods served but not necessarily food consumed unless there is a correction for plate waste. The majority of duplicate plate or diet studies included in this report did not require that the participants prepare, record, measure, and preserve the foods they ate for later analysis in a free-living setting. Most of the studies cited were conducted in a hospital or school-like setting.

There are strengths and weakness to each of the dietary methodologies that impact the study outcomes. Martinez-Mier et al. (2008) conducted a pilot study that compared the results of 3-day duplicate diets with 3-day diary records for 12 children (ages 18 to 25 months). Adults (parents and/or caregivers) kept the diaries and collected the food and beverage samples. The 3-day averages for each child differed for the two approaches with the differences ranging from 0.01 to 0.4 mg F/day. Both approaches suffered from protocol compliance problems, and large variations in daily fluoride intake from both beverages and food were observed between and within children.

The majority of the published studies that provided estimated oral fluoride intakes from the diet for this report utilized a market basket approach coupled with recall records collected and analyzed by the U.S. Departure of Agriculture. The date and title of the USDA study varies and is provided in the study descriptions that follow. In one case the market basket was developed from a food frequency recall but it too used food group intake values from USDA. Fewer studies were identified that used a diary approach or a duplicate diet approach. In one instance the diary record was used to construct a market basket for analysis.

The study summaries that follow, with the exception of some of the duplicate plate analyses, are suitable for estimating dietary population intakes of fluoride within the limitations that apply to the methods described in preceding paragraphs. Where possible, EPA chose to rely most heavily on studies that obtained the fluoride concentration information from a market basket analysis because such studies were considered to be more nationally representative than a study based duplicate diet analyses.

### 2.5.2. Infants

Each of the studies assessing fluoride intake by infants used a market basket-type approach where analysis of the fluoride content of foods was combined with estimates of food intake from a recall, record, or intake recommendation, and measured or assumed drinking water concentration, in order to arrive at an estimate for fluoride exposure.

Singer and Ophaug (1979) estimated maximum and minimum total fluoride intake of 6-month old infants on diets prepared with fluoridated water or non-fluoridated water (Table 2-22). Commercial manufacturers of infant foods provided samples of foods and milk formulations produced at each of their domestic plants. Each sample was "closely examined for the fluoride content of the water used in processing it" (actual fluoride concentrations in the processing water were not reported). The food samples were fixed with magnesium oxide and then ashed. Fluoride was isolated by diffusion and analyzed with a fluoride ion-specific electrode. Separate samples were unashed and analyzed for fluoride by a colorimetric technique and an ion-specific electrode. While the results with the electrode were in good agreement with both ashed and unashed samples, the colorimetric method gave substantially higher fluoride readings-presumably due to interfering substances.

Food consumption estimates (milk, formula, and "beikost") were based on the total caloric intake for six month old infants according to the estimates of Fomon (1975). Beikost is a term that refers to solid or semi-solid baby foods other than milk or formula. The quantity of each food consumed was calculated by dividing the caloric intake supplied by each food item (kcal/day) by average values of caloric density (kcal/gm) as given by Wiatrowski et al. (1975). The total fluoride intake was calculated using the mean fluoride values for various food groups. In estimating fluoride intakes, maximum values were based on foods obtained from the plant using fluoridated water using the assumption that the infant's drinking water would contain 1.0 mg F/L; minimum intake values were based on data from the non-fluoridated plant using the assumption that the infant's drinking water would contain only 0.1 mg F/L. For 6-month-old infants (bw 8.1 kg) the minimum fluoride intake was 0.153 mg/day, and the maximum intake was 0.763 mg/day.

**Table 2-22. Fluoride Intake of Infants 6 Months Old (Singer and Ophaug, 1979)**

| Food Item | Caloric Intake[a] (kcal)/day | Food Consumption (g)[b] | Maximum Fluoride Intake[c] | Minimum Fluoride Intake[d] |
|---|---|---|---|---|
| | | | mg/day (mg/kg/day) | mg/day (mg/kg/day) |
| Milk formula | 444 | 663 | 0.451 | 0.020 |
| Cereals | 57 | 15 | 0.073 | 0.023 |
| Fruits | 93 | 109 | 0.006 | 0.006 |
| Vegetables | 62 | 138 | 0.033 | 0.033 |
| Juices | 22 | 34 | 0.023 | 0.002 |
| Meats | 62 | 58 | 0.057 | 0.057 |
| Water | | 120 | 0.120 | 0.012 |
| **Total** | **740** | | **0.763 (0.094)** | **0.153 (0.019)** |

**SOURCE:** Singer and Ophaug, 1979.

[a]From Fomon, 1975.

[b]Consumption based on daily caloric intake and the following caloric densities (kcal/g): milk formula, 0.67; cereals 3.74; fruits, 0.85; vegetables, 0.45; meats, 1.06; and juices, 0.65.

[c]Mean fluoride content were: milk formulations – 0.68 mg/L; cereal – 4.84 mg/kg, and juices – 0.67 mg/L processed in plants using fluoridated water, and fruits – 0.051 mg/kg, vegetables – 0.24 mg/kg, meats – 0.99 mg/kg and water – 1.0 mg/L.

[d]Mean fluoride content of 1.5 mg/kg for cereal and 0.061 mg/L for juices processed in plants using nonfluoridated water, and 0.03 mg/L for human or bovine milk , 0.051 mg/L for fruit, 0.24 mg/L, for vegetables, 0.99 mg/kg for meats, and 0.1 mg/L for water.

Ophaug et al. (1980a) estimated the daily fluoride intake of 6-month-old infants for four geographic regions of the United States. The study was based on the FDA market basket food collections for 1977 and 1978. The foods were placed in 11 composite food groups. The composites were prepared according to Shopping and Compositing Guides representing an average 14-day consumption of a 6-month-old infant in Orlando, Philadelphia, Grand Rapids, and Los Angeles. The first three cities reportedly had fluoridated water supplies (1.07 mg/L was the maximum value reported, which was for Grand Rapids). The fluoride concentration in the Los Angeles water system at the time of the study was reported to be 0.37 mg/L. The Shopping and Composite Guides are based on data obtained by the U.S. Department of Agriculture survey of food consumption made in 1965 to 1966 for each of the geographic regions (USDA, 1968). The fluoride levels in all composites except one were analyzed by ashing, followed by diffusion and detection by a fluoride ion-specific electrode. The oils and fats composite was analyzed by an oxygen bomb reverse extraction procedure (Venkateswarlu, 1975). The total daily fluoride intake ranged from a high of 0.541 mg/day in Orlando to a low of 0.207 mg/day in Grand Rapids (Table 2-23). Using an estimated body weight of 8.1 kg for a 6-month-old infant, Ophaug et al. (1980a) calculated a fluoride intake of 0.026 to 0.067 mg/kg/day.

| Table 2-23. Fluoride Intake (mg F/day) by Infants 6 Months Old in Four Regions of the U.S. | | | | |
|---|---|---|---|---|
| **Food Item** | **South (Orlando)** | **North central (Grand Rapids)** | **Northeast (Philadelphia)** | **West (Los Angeles)** |
| Water | 0.295 | 0.092 | 0.077 | 0.108 |
| Milk | 0.013 | 0.015 | 0.017 | 0.007 |
| Other dairy and formula | 0.060 | 0.024 | 0.016 | 0.073 |
| Meats, fish, poultry | 0.024 | 0.006 | 0.009 | 0.022 |
| Grains/cereals | 0.077 | 0.011 | 0.026 | 0.102 |
| Potatoes | 0.000 | 0.001 | 0.001 | - |
| Vegetables | 0.026 | 0.044 | 0.057 | 0.021 |
| Fruits/juices | 0.028 | 0.014 | 0.011 | 0.012 |
| Oils/fats | - | - | 0.005 | - |
| Sugars, etc. | 0.002 | 0.000 | 0.008 | 0.001 |
| Beverages | 0.016 | - | 0.045 | 0.008 |
| **Total** | **0.541** | **0.207** | **0.272** | **0.354** |

**SOURCE:** Ophaug, 1980a.

Ophaug et al. (1985) estimated dietary fluoride intake of 6-month-old infants in 20 cities across the U.S. The cities were grouped in one of four geographic regions. The survey used the same market basket and same composite food groupings as those used in the authors' 1980 publication. Fluoride levels were determined with a fluoride ion-specific electrode in all but one case; fluoride level in oils and fats was determined using the oxygen bomb reverse extraction procedure. Dietary fluoride intake from each composite was calculated by multiplying its fluoride content by an estimate of the amount consumed daily. The fluoride content of the drinking water in the cities where the market baskets were collected ranged from 0.05 to 1.04 mg/L. Specific information on food and drinking water intakes was not reported.

A summary of the estimated fluoride intakes for 6-month-old infants for each of the study sites in the Ophaug et al. (1985) study is shown in Table 2-24. Fluoride intake for infants was estimated from an analysis of commercial infant foods processed in fluoridated and non-fluoridated plants. Within each region total fluoride intake was correlated with water fluoride concentration. The highest dietary intake of fluoride occurred in the southern region. The daily fluoride intake from foods (total intake minus that from water and beverages) averaged 0.171 ±0.012 (SE) mg/day and was not correlated with water fluoride level.

Ophaug et al. (1985) assessed their results by concentration of fluoride in drinking water. The mean total dietary intake of fluoride (including beverages) for 6-month old infants ranged from 0.226 mg/day where the fluoride level in drinking water was less than 0.3 mg/L to 0.418 mg/day in areas where the fluoride level was greater than 0.7 mg/L (Table 2-25).

| Table 2-24. Estimated Fluoride Intake of 6-Month Old Infants in Different Regions of the U.S. | | | | |
|---|---|---|---|---|
| Region/city (year of sample) | Water F Level | Total F intake | | F Intake in Foods |
| | mg/L | mg/day | mg/kg | mg/day |
| **Northeast:** | | | | |
| Boston, MA (1980) | 1.00 | 0.307 | 0.038 | 0.130 |
| Hartford, CT (1978) | 0.93 | 0.369 | 0.033 | 0.091 |
| Philadelphia, PA (1977) | 0.66 | 0.272 | 0.034 | 0.150 |
| Boston, MA (1977) | 0.10 | 0.305 | 0.038 | 0.227 |
| Manchester, NH (1980) | 0.10 | 0.220 | 0.027 | 0.140 |
| **North Central:** | | | | |
| Grand Rapids, WI (1978) | 1.04 | 0.207 | 0.026 | 0.115 |
| Akron, OH (1981) | 1.01 | 0.251 | 0.031 | 0.162 |
| Fargo, ND (1981) | 0.91 | 0.178 | 0.022 | 0.098 |
| Kansas City, KS (1982) | 0.54 | 0.097 | 0.012 | 0.049 |
| **South:** | | | | |
| Louisville, KY (1980) | 1.00 | 0.642 | 0.079 | 0.164 |
| Chattanooga, TN (1982) | 1.00 | 0.650 | 0.080 | 0.188 |
| Columbia, SC (1979) | 0.80 | 0.582 | 0.072 | 0.208 |
| Orlando, FL (1976) | 0.67 | 0.541 | 0.068 | 0.230 |
| Baton Rouge, LA (1980) | 0.30 | 0.265 | 0.033 | 0.123 |
| **West:** | | | | |
| Boise, ID (1979) | 1.00 | 0.549 | 0.068 | 0.257 |
| Boise ID (1980) | 1.00 | 0.504 | 0.062 | 0.210 |
| Denver, CO (1977) | 0.71 | 0.456 | 0.056 | 0.242 |
| Phoenix, AZ (1982) | 0.50 | 0.354 | 0.044 | 0.205 |
| Los Angeles, CA (1977) | 0.37 | 0.354 | 0.044 | 0.238 |
| Fresno, CA (1981) | 0.10 | 0.239 | 0.030 | 0.201 |
| Tacoma, WA (1981 | 0.05 | 0.204 | 0.025 | 0.179 |
| Sacramento, CA (1980) | 0.05 | 0.163 | 0.020 | 0.147 |
| **OVERALL MEAN** | | | | **0.171** |

**SOURCE:** Ophaug et al., 1985.

| Table 2-25. Mean Dietary Fluoride Intake of 6-Month-Old Infants (Ophaug et al., 1985) | | | | | | |
|---|---|---|---|---|---|---|
| Fluoride Conc. | n | mg/day | SEM | n | mg/kg/day | SEM |
| <0.3 ppm | 5 | 0.226[a] | ±0.023 | 5 | 0.028[b] | ±0.003 |
| 0.3-0.7 ppm | 6 | 0.314 | ±0.059 | 6 | 0.039 | ±0.007 |
| >0.7 ppm | 11 | 0.418[a] | ±0.054 | 11 | 0.052[b] | ±0.007 |

**SOURCE:** Ophaug et al., 1985.
[a]Statistically different at p <0.025.
[b]Statistically different at p <0.025.

Fomon and Ekstrand (1999) estimated fluoride intakes of infants from birth to age 10 months. Fluoride concentrations in infant foods were derived from an earlier study (Fomon and Ekstrand, 1993b), as were estimates of mean energy intakes for specific age groups (Fomon and Ekstrand, 1993a). Fomon and Ekstrand (1999) give an estimate of 120 mL/kg/day for milk or formula intake by "older infants" (although a specific age range is not given, the implication in the text is that these are infants 4–10 months old). The study authors note that the older infants would also be consuming a small amount of beikost (weaning food) which they estimated would increase in fluoride intake by an average 20 µg/kg/day in most cases. Estimates of fluoride intake from milk and formulas only are shown in Table 2-26.

**Table 2-26. Estimated Fluoride Intake of 4–10 Month Old Infants with Varying Intakes of Milk or Formula**

| Milk/ Formula | F Concentration (µg/L) | | | F Intake (µg/kg/day) for a Formula Intake of 120 mL/kg/day |
|---|---|---|---|---|
| | Formula | Water | As Fed | |
| Human milk | | | 6 | 1 |
| Cow's milk | | | 40 | 5 |
| **Formula**: | | | | |
| Ready to feed-milk-based | 200 | – | 200 | 24 |
| Conc. liquid-milk-based | 200 | 200 | 200 | 24 |
| | 200 | 1000 | 600 | 72 |
| Isolated soy protein-based | 240 | 200 | 270 | 22 |
| | 240 | 1000 | 620 | 74 |
| Powdered milk-based | 690[a] | 200 | 276[b] | 33 |
| | 690 | 600 | 700 | 84 |
| | 690 | 1000 | 980 | 118 |

**SOURCE:** Fomon and Ekstrand, 1993b; as modified by Fomon and Ekstrand, 1999.

[a]µg/kg of formula powder.

[b]Assumes that 145 g of formula diluted with 880 mL of water to make 1 liter.

Fomon and Ekstrand (1999) note that infant feeding patterns have changed from the 1960s and 70s to the 1980s and 90s with a trend toward more extended feeding of formula. As a result, prolonged intake of fluoride from formula became more common. A comparison of infant fluoride intakes during these two periods for infants from 4 to 10 months old is shown in Table 2-27. The study authors note that the estimates for the 1960s and early 1970's were based on measurements of fluoride levels in milk and formula made by Fomon and Ekstrand (1993b); and not on measurements from the 1960s and 1970s, and that values are therefore somewhat less than would be the case if calculations had been based on concentrations of fluoride in formulas actually marketed in the 1960s and 1970s.

**Table 2-27. Dietary Fluoride Intake of Infants 4-10 Months Old from the 1960s to the 1990s**

| Diet | 1960s-and Early 1970s[a] | | 1980s and Early 1990s | |
|---|---|---|---|---|
| | F Intake[b] (µg/kg/day) | Estim. % of Infants | F Intake[b] (µg/kg/day) | Estim. % of Infants |
| Human milk | – | – | 1–37 | 15 |
| Infant formula | 24–118[a] | <20 | 24–118 | 55 |
| Cow's milk | 5 | >80 | 5 | 30 |

**SOURCE:** Fomon and Ekstrand, 1999.

[a]Based on measurements of fluoride levels in milk and formula made by Fomon and Ekstrand (1993); and not on measurements from the 1960s and 1970s. The study authors note that the values listed are therefore somewhat less than would be the case if calculations had been based on concentrations of fluoride in formulas actually marketed in the 1960s and 1970s.

[b]Fluoride intakes by exclusively breast-fed infants do not exceed 1 µg/kg/day; however, many breast-fed infants also receive formula and the range of intakes in the table includes those of partially breast-fed infants.

Using the same assumptions concerning energy intakes of infants and energy equivalents of infant foods, Fomon et al. (2000) updated the estimates of fluoride intake by infants that were reported by Fomon and Ekstrand (1999). The study authors also included estimates of fluoride intake from formulas prepared at home with evaporated milk. These estimates are shown in Table 2-28.

**Table 2-28. Updated Estimated Fluoride Intake of 4-10 Month Old Infants with Varying Intakes of Milk and Formula (Fomon et al., 2000)**

| Milk/ Formula | F Concentration (µg/L) | | | F Intake (µg/kg/day) |
|---|---|---|---|---|
| | Formula | Water | As Fed | 120 mL/kg/day |
| Human milk | | | 6 | 1 |
| Cow's milk | | | 40 | 5 |
| **Formulas:** | | | | |
| Home prepared evaporated milk formula[a] | 90 | 200 | 155 | 19 |
| | 90 | 1000 | 632 | 76 |
| Ready to feed-milk-based | – | – | 200 | 24 |
| Conc. liquid-milk-based | 200 | 200 | 200 | 24 |
| | 200 | 1000 | 600 | 72 |
| Isolated soy protein-based | 250 | 200 | 225 | 27 |
| | 250 | 1000 | 625 | 75 |
| Powdered milk-based | 690[b] | 200 | 262[c] | 31 |
| | 690[b] | 1000 | 966[c] | 116 |

[a]Assumes 0.39L of evaporated milk to 0.57 L of water (also includes formulas made with fresh milk)

[b]µg/kg of formula powder.

[c]Assumes that 125 g of formula diluted with 880 mL of water makes 1 liter of formula as fed.

Siew et al. (2009) measured fluoride levels in different types of infant formula (see Table 2-4), and estimated the daily fluoride intake for age-groups from birth to 12 months. Based on body weight and formula intake data for male and female infants (Table 2-29), Siew et al. (2009) reported that female infants would have a slightly greater intake of fluoride than male infants.

**Table 2-29. Volume of Formula Consumed and Body Weights from Birth to 12 Months (Siew et al., 2009)**

| Age (months) | Formula intake (ounces)[a] | Body weights (kg) | | | | | |
|---|---|---|---|---|---|---|---|
| | | Girls | | | Boys | | |
| | | 10th Percent. | 50th Percent. | 90th Percent. | 10th Percent. | 50th Percent. | 90th Percent. |
| 0–4 | 21–29 | 2.7–5.2 | 3.4-6.2 | 4.0-7.1 | 2.8-5.7 | 3.6-6.7 | 4.2-7.8 |
| 4–6 | 29–32 | 5.2–6.2 | 6.2-7.2 | 7.1-8.4 | 5.7-6.8 | 6.7-7.9 | 7.8-9.2 |
| 6–9 | 30–32 | 6.2–7.4 | 7.2-8.5 | 8.4-9.8 | 6.8-8.0 | 7.9-9.3 | 9.2-10.8 |
| 9–12 | 24–30 | 7.4–8.3 | 8.5-9.5 | 9.8-11.0 | 8.0-9.0 | 9.3-10.3 | 10.8-11.9 |

**SOURCE:** Siew et al. 2009.

[a] Derived from Hendricks and Duggan's Manual of Pediatric Nutrition.

Total fluoride intake for female infants was then calculated from both the amount of fluoride ingested from the water used to reconstitute the formula (0.0, 0.4, 0.5, 0.7 or 1.0 ppm fluoride) and from the formula itself. Results showed that the formulas themselves did not contain fluoride at levels high enough to exceed an intake of 0.10 mg/kg/day with normal consumption.

It was estimated that a minimal risk of exceeding 0.1 mg/kg/day would exist with a fluoride drinking water level of 0.5 ppm. If the drinking water contained 1 ppm fluoride, infants consuming powdered formula reconstituted with this water would exceed a fluoride intake of 0.1 mg/kg/day. However, it should be recognized that fluoride is a nutrient and reconstitution of infant formulas with water containing lower levels of fluoride may result in infants not consuming the Adequate Intake for fluoride (0.5 mg/day) established by the Institute of Medicine (1997). The American Dental Association (2007) recommends "Parents and caregivers should consult with their dentist, pediatrician or family physician regarding the most appropriate water to use in their area to reconstitute infant formula". The ADA (2007) publication informs users of liquid concentrate or powdered infant formula as the primary source of nutrition that can "be mixed with water that is fluoride free or contains low levels of fluoride to reduce the risk of fluorosis".

### 2.5.3. Children to 14 Years of Age

The fluoride exposure estimates for children up to 14 years of age come from three types of studies. Most have used the Market Basket approach but there are also two that use dietary records of beverage intake to estimate the fluoride from beverages only and two that employed a duplicate plate methodology. The Market Basket-type studies are presented first followed by the two using the dietary records and then the duplicate plate studies.

**Market Basket Studies**. Ophaug et al. (1980b) estimated the daily fluoride intake of 2-year-old children residing in four regions of the United States (Table 2-30).

**Table 2-30. Fluoride Intake (mg F/day) by Children 2 Years Old in Four Regions of the U.S.**

| Food Item | South (Orlando) | North central (Grand Rapids) | Northeast (Philadelphia) | West (Los Angeles) |
|---|---|---|---|---|
| Water | 0.274 | 0.302 | 0.206 | 0.136 |
| Milk | 0.009 | 0.011 | 0.011 | 0.010 |
| Other dairy and formula | 0.005 | 0.013 | 0.011 | 0.006 |
| Meats, fish, poultry | 0.060 | 0.051 | 0.057 | 0.023 |
| Grains/cereals | 0.023 | 0.042 | 0.029 | 0.055 |
| Potatoes | 0.001 | 0.005 | 0.004 | 0.006 |
| Vegetables | 0.016 | 0.011 | 0.016 | 0.016 |
| Fruits/juices | 0.021 | 0.042 | 0.020 | 0.020 |
| Oils/fats | 0.004 | 0.008 | 0.002 | 0.002 |
| Sugars, etc. | 0.008 | 0.014 | 0.008 | 0.010 |
| Beverages | 0.133 | 0.111 | 0.046 | 0.031 |
| **Totals (mg/day)** | **0.554** | **0.610** | **0.410** | **0.315** |
| **(mg/kg/day)** | **0.044** | **0.049** | **0.033** | **0.025** |

**SOURCE:** Ophaug, 1980b.

This study was identical in methodology to that conducted by Ophaug et al. (1980a) for 6-month-old infants, but was based on the FDA toddler market basket food collections for 1977 and 1978.

The foods were placed in the same 11 composite food groups according to Shopping and Compositing Guides representing an average 14-day consumption of 2-year-old children. The Shopping and Composite Guides were based on data obtained by the U.S. Department of Agriculture survey of food consumption made in 1965 to 1966 for each of the geographic regions. The fluoride levels in all composites except one were analyzed by ashing, followed by diffusion and detection by a fluoride ion-specific electrode. The oils and fats composite was analyzed by an oxygen bomb reverse extraction procedure (Venkateswarlu, 1975). The total daily fluoride intake ranged from a low of 0.315 mg/day for Los Angeles to a high of 0.610 mg/day for Grand Rapids. The intake per unit body weight ranged from 0.025 mg/kg/day to 0.049 mg/kg/day.

Ophaug et al. (1985) continued the studies of Ophaug et al. (1980b) by evaluating dietary fluoride intake of 2-year-old children in 20 cities across the U.S. (Table 2-31).

**Table 2-31. Dietary Fluoride Intake of an Average 2-Year-Old Child**

| City (year of sample) | Water F Level | Total F intake | | Foods |
|---|---|---|---|---|
| | (mg/L) | (mg/day) | (mg/kg) | (mg/day) |
| **Northeast:** | | | | |
| Boston, MA (1980) | 1.00 | 0.475 | 0.038 | 0.125 |
| Hartford, CT (1978) | 0.93 | 0.507 | 0.041 | 0.141 |
| Philadelphia, PA (1977) | 0.66 | 0.410 | 0.033 | 0.158 |
| Boston, MA (1977) | 0.10 | 0.348 | 0.028 | 0.314 |
| Manchester, NH (1979) | 0.10 | 0.182 | 0.014 | 0.132 |
| **North Central:** | | | | |
| Grand Rapids, WI (1978) | 1.04 | 0.607 | 0.049 | 0.194 |
| Akron, OH (1981) | 1.01 | 0.682 | 0.055 | 0.190 |
| Fargo, ND (1981) | 0.91 | 0.504 | 0.040 | 0.155 |
| Kansas City, KS (1982) | 0.54 | 0.376 | 0.040 | 0.150 |
| **South:** | | | | |
| Louisville, KY (1980) | 1.00 | 0.880 | 0.070 | 0.150 |
| Chattanooga, TN (1982) | 1.00 | 0.784 | 0.063 | 0.191 |
| Columbia, SC (1979) | 0.80 | 0.718 | 0.057 | 0.211 |
| Orlando, FL (1976) | 0.67 | 0.554 | 0.044 | 0.147 |
| Baton Rouge, LA (1980) | 0.30 | 0.310 | 0.025 | 0.107 |
| **West:** | | | | |
| Boise, ID (1979) | 1.00 | 0.537 | 0.043 | 0.127 |
| Boise ID (1980) | 1.00 | 0.568 | 0.045 | 0.173 |
| Denver, CO (1977) | 0.71 | 0.566 | 0.045 | 0.244 |
| Phoenix, AZ (1982) | 0.50 | 0.350 | 0.028 | 0.138 |
| Los Angeles, CA (1977) | 0.37 | 0.315 | 0.025 | 0.148 |
| Fresno, CA (1981) | 0.10 | 0.197 | 0.016 | 0.144 |
| Tacoma, WA (1981 | 0.05 | 0.162 | 0.013 | 0.116 |
| Sacramento, CA (1980) | 0.05 | 0.146 | 0.012 | 0.124 |
| **OVERALL MEAN** | | | | **0.163** |

**SOURCE:** Ophaug et al., 1985.

This study was based on FDA market food basket collections obtained during 1977–1982. The methodology was the same as that described above. The fluoride content of the drinking water in the cities where the market baskets were collected ranged from 0.05 to 1.04 mg/L. A summary of the fluoride intakes is given in Table 2-31. Fluoride dietary intake was highly correlated with water fluoride level with correlation coefficients ≥ 0.72. The highest dietary intake occurred in the southern region, and was reported to be a reflection of greater consumption of water and beverages (551 g/day vs. 383-426 g/day in the other regions). The daily fluoride intake from foods (total intake minus that from water and beverages) averaged 0.161±0.010 (SE) mg/day, and was not correlated with water fluoride level.

Mean total dietary intakes (including beverages) based on fluoride levels in drinking water are shown in Table 2-32. Mean fluoride intake increased with increase in fluoride concentration in drinking water.

**Table 2-32. Mean Dietary Fluoride Intake of 2-Year-Olds**

| Fluoride Concentration in Drinking Water | n | mg/day | SEM | n | mg/kg/day | SEM |
|---|---|---|---|---|---|---|
| <0.3 ppm | 5 | 0.207[b,c] | ±0.036 | 5 | 0.017[e,f] | ±0.003 |
| 0.3-0.7 ppm | 6 | 0.386[a,c] | ±0.037 | 6 | 0.031[d,f] | ±0.003 |
| >0.7 ppm | 11 | 0.621[a,b] | ±0.039 | 11 | 0.050[d,e] | ±0.003 |

**SOURCE:** Ophaug et al., 1985.

[a]Statistically different at the p <0.0025.
[b]Statistically different at the p <0.0005.
[c]Statistically different at the p <0.005.
[d]Statistically different at the p <0.0025.
[e]Statistically different at the p <0.0005.
[f]Statistically different at the p <0.005.

Dabeka and McKenzie (1995) surveyed fluoride levels in various foods obtained in 1987 in Winnipeg, Canada. The foods were prepared for consumption and combined into 113 composites and 39 composite subsets using a Total Diet Study approach. The water used to prepare the foods contained 1 mg F/L. Fluoride was determined with a fluoride ion-specific electrode after microdiffusion. As reported in Dabeka et al. (1993), food intake data (g/person/day) for each of food composites was obtained from the Nutrition Canada Survey (Bureau of Nutritional Sciences, 1977) for the age groups of 1–4, 5–11, and 12–19 yr. Total dietary intake of fluoride (excluding plain drinking water) was estimated to be 0.353 mg/day for boys and girls 1–4 years old; 0.530 mg/day for boys and girls 5–11 years old; 1.025 mg/day for boys 12–19 years old; and 0.905 mg/day for girls 12–19 years old.

The fluoride content of 441 brand-name food items purchased in both a non-fluoridated community (Connersville, IN, fluoride 0.16 ±0.01 mg F/L) and in a fluoridated community (Richmond, IN, 0.90 ±0.05 mg F/L) (see Section 3.1.2) were evaluated by Jackson et al. (2002). A modified validated Food Frequency Questionnaire was administered to determine the 75 foods and beverages most commonly eaten by adolescents (ages 12–14) in these communities. Frequency of ingestion was weighted from 1 for less than monthly to 9 for two or more times per day. Parents of the children were interviewed to determine the outcome of the Frequency Recall and asked to identify the brand names of the foods and beverages most often purchased. Food samples were purchased in each community (grocery stores and restaurants) and prepared using community water in cases where preparation was necessary. Foods were grouped for analysis based on the USDA classification (1998). [Note: According to USDA (1998), the beverages group excludes plain water and noncarbonated bottled water]. Homogenates of each food group were analyzed for their fluoride content and used to estimate exposure for 3–5 year old children.

Mean fluoride intakes were derived from the fluoride content of each food group homogenate using age and gender-specific mean food intakes from the Midwest regional data from the USDA (1998) CSFII survey (Table 2-33). Mean fluoride intake was 0.454 mg/day in Connersville, IN and 0.536 mg in Richmond, IN (Note: fluoride intake from consumption of drinking water was not included in the calculation).

**Table 2-33. Estimated Fluoride Intake of 3- to 5-Year-Old Children Living in a Nonfluoridated and Fluoridated Community**

| Food Category | Food intake (g/day)[a] | Connersville, IN (F = 0.16 mg/L) | | Richmond, IN (F = 0.9 mg/L) | |
|---|---|---|---|---|---|
| | | F Content (µg/g)[b] | F Intake (µg/day)[b] | F Content (µg/g)[b] | F Intake (µg/day)[b] |
| Grains and cereal products | 264 | 0.44 | 116.16 | 0.55 | 145.20 |
| Vegetables | 90 | 0.26 | 23.40 | 0.28 | 25.20 |
| Fruits | 213 | 0.13 | 27.69 | 0.11 | 23.43 |
| Dairy Products | 387 | 0.35 | 135.45 | 0.28 | 108.36 |
| Meat, poultry | 64[d] | 0.35 | 22.40 | 0.37 | 23.68 |
| Nuts and seeds | 5 | 0.14 | 0.70 | 0.18 | 0.90 |
| Fats and oils | 5 | 0.24 | 1.20 | 0.25 | 1.25 |
| Sugars and sweets | 45 | 0.24 | 10.80 | 0.35 | 15.75 |
| Beverages[c] | 291 | 0.40 | 116.40 | 0.66 | 192.06 |
| **Total** | **1010** | | **454.20** | | **535.83** |

**SOURCE:** Jackson et al., 2002.
[a]USDA, 1998.
[b]Mean values.
[c]Plain drinking water is not included in the category according to USDA (1998).
[d]USDA (1998) lists 99 grams/day for this age group for the mid-west region of the US; however, in Jackson et al. (2002) it is given as 64 grams/day.

Since the USDA (1998) percentile intake distributions for food groups were not available to the researchers, an upper bound estimate of fluoride intake was calculated using the mean intake and the 90$^{th}$ percentile data for fluoride concentration in each food group. Jackson et al. (2002) calculated that the upper bound fluoride intake would be 0.925 mg/day (0.058 mg/kg/day) in Connersville and 0.999 mg/day (0.062 mg/kg/day) in Richmond.

Jackson et al. (2002) determined exposure data only for the 3–5 year old age group because of their vulnerability to dental fluorosis. However, because the food frequency recall data that supported the market baskets were collected from adolescents, the analytical data on fluoride levels in the food groups can be combined with food group intake information from USDA (1998) to provide estimates for the 6–11 and 12–19 year age groups. Tables 2-34 and 2-35 below provide the results of these calculations for Connersville and Richmond. The extrapolated estimates for both age groups are supported by the results from the Dabeka and McKenzie (1995) Canadian Study.

**Table 2-34. Estimated Fluoride Intake of 6 to 11 and 12 to 19 Year Olds Living in a Nonfluoridated Community**

| Food Category[a] | Mean F Content (µg/g)[b] | 6-11 yr olds (average for males and females | | 12-19 yr olds (average for males and females) | |
|---|---|---|---|---|---|
| | | Food intake[c] (g/day) | Fluoride Intake[d] (µg/day) | Food Intake[c] (g/day) | Fluoride Intake[d] (µg/day) |
| Grains/cereal products | 0.44 | 309 | 136 | 363 | 159.7 |
| Vegetables | 0.26 | 119 | 31.0 | 170.5 | 44.3 |
| Fruits | 0.13 | 163.5 | 21.3 | 144.5 | 18.8 |
| Dairy Products | 0.35 | 435 | 152.3 | 403.5 | 141.2 |
| Meat, poultry | 0.35 | 139.5 | 48.8 | 227 | 79.5 |
| Nuts and seeds | 0.14 | 5 | 0.7 | 3 | 0.4 |
| Fats and oils | 0.24 | 8.5 | 2.0 | 11.5 | 2.8 |
| Sugars and sweets | 0.24 | 53 | 12.7 | 42.5 | 10.2 |
| **BEVERAGES**[e] | 0.40 | 407.5 | **163** | 959.5 | **383.7** |
| **TOTAL F INTAKE** | | | **567.9** | | **840.7** |
| **F INTAKE FROM FOOD [(Total) – (beverages)]** | | | **404.8** | | **457** |

[a]Food categories of eggs and legumes are listed in USDA (1998), but are not included in Jackson et al. (2002)
[b]Mean values (Jackson et al., 2002).
[c]USDA (1998; survey data from 1994-1996; mean food intake values).
[d]Fluoride intake calculated as fluoride concentration in food category (µg/g) multiplied by food intake (g/day).
[e]Plain drinking water is not included in the beverage category according to USDA (1998).

**Dietary Record Studies.** Three-day beverage records of Grade 6 children (average age 11.94 yr) in two towns in Canada were used to document daily means of the highest and lowest fluoride intake from beverages (Clovis and Hargreaves, 1988). The study was conducted in a town with a fluoridated water supply (average adjusted fluoride concentration of 1.08 mg/L) and in one without fluoride added to the water (0.23 mg F/L). The three highest and three lowest beverage intakes (including drinking water) were used to estimate the range of fluoride intakes in the two communities. In the nonfluoridated community, the probable fluoride intake ranged from 0.00–0.03 mg from milk, 0.02–0.43 mg from water, 0.08–0.69 mg from carbonated beverages; 0.01–0.14 mg from reconstituted juices; and 0.08–0.09 mg from other types of drinks. The total fluoride intake from beverages ranged from 0.02 to 0.82 mg. For the fluoridated community, the probable fluoride intake ranged from 0.00–0.05 mg from milk, 0.07–0.25 mg from water, 0.1–0.93 mg from carbonated beverages; 0.21–0.35 mg from reconstituted juices; and 0.07–1.76 mg from other types of drinks. The total fluoride intake from beverages ranged from 0.40 to 2.45 mg.

| Table 2-35. Estimated Fluoride Intake of 6 to 11 and 12 to 19 Year Old Children Living in a Fluoridated Community | | | | | |
|---|---|---|---|---|---|
| Food Category[a] | Mean F Content (µg/g)[b] | 6-11 yr olds (average for males and females) | | 12-19 yr olds (average for males/females) | |
| | | Food intake[c] (g/day) | Fluoride Intake[d] (µg/day) | Food Intake[c] (g/day) | Fluoride Intake[d] (µg/day) |
| Grains/cereal products | 0.55 | 309 | 170.0 | 363 | 199.7 |
| Vegetables | 0.28 | 119 | 33.3 | 170.5 | 47.7 |
| Fruits | 0.11 | 163.5 | 18.0 | 144.5 | 15.9 |
| Dairy products | 0.28 | 435 | 121.8 | 403.5 | 113.0 |
| Meat, poultry | 0.37 | 139.5 | 51.6 | 227 | 84.0 |
| Nuts and seeds | 0.18 | 5 | 0.9 | 3 | 0.5 |
| Fats and oils | 0.25 | 8.5 | 2.1 | 11.5 | 2.9 |
| Sugars and sweets | 0.35 | 53 | 18.6 | 42.5 | 14.9 |
| **BEVERAGES**[e] | 0.66 | 407.5 | **269.0** | 959.5 | **633.3** |
| **TOTAL F INTAKE** | | | 685.3 | | 1111.9 |
| **F INTAKE FROM FOOD [(Total) – (beverages)]** | | | **416.3** | | **478.6** |

[a]Food categories of eggs and legumes are listed in USDA (1998), but are not included in Jackson et al. (2002)
[b]Mean values (Jackson et al., 2002).
[c]USDA (1998; survey data from 1994-1996; mean food intake values).
[d]Fluoride intake calculated as fluoride concentration in food category (µg/g) multiplied by food intake (g/day).
[e]Plain drinking water is not included in the beverage category according to USDA (1998).

Pang et al. (1992) studied fluoride intake of 225 children, ages 2–10 years, living in North Carolina. Data on beverage intake was collected by means of three-day diary records kept during April, May or June, 1990. Concentrated fruit juices, fruit drinks, and teas were prepared with deionized water. Total fluid intake was 970–1,240 mL/day, and consumption of soft drinks, juices, tea, and other beverages 585–756 mL/day. Of the total fluid consumption, milk and water constituted 36–40%. Fluoride was determined by the microdiffusion method and a fluoride ion-specific electrode. Fluoride concentrations in the beverages ranged from nondetectible to 6.7 mg/L; mean concentrations were 0.74 mg/L for soda, 0.36 mg/L for juices, 0.33 mg/L for punches, 2.56 mg/L for teas, and 0.85 mg/L for Gatorade. The estimated average fluoride intake (±SD) from beverages (excluding milk, plain water and beverages listed less than five times in the diaries) for children ages 2–3, 4–6, and 7–10 years were 0.36±0.31, 0.54±0.52, and 0.60±0.48 mg/day, respectively. The maximum fluoride intakes for individual children within these groups were 1.40, 2.39, and 2.00 mg/day, respectively. The study authors note that fluoride levels were high in grape juice (maximum 1.6 ppm) and also in teas (mostly 2–3 ppm, and with a maximum of 6.5 ppm).

Levy et al. (2003a) estimated an average fluoride intake of 0.2 mg/day from beverages, not including plain drinking water, for 785 three to six year olds. Parents were asked to periodically

complete modified food frequency questionnaires which assessed numbers and sizes of daily servings of different categories of beverages and foods made with water.
There was no direct verification of the data reported by the parents in the questionnaires. The 90th percentile estimate was about 0.5 mg/day.

**Duplicate Diet/Plate Analyses.** Rojas-Sanchez et al. (1999) estimated fluoride intakes from foods and beverages (and dentifrice) consumed by children (16–40 months old; about 1.3 to 3.3 years old) living in three different communities using a duplicate plate methodology. The three communities differed in the fluoride concentration of their water supply: 1) a low-fluoride community (San Juan, Puerto Rico; ≤0.3 mg F/L); 2) a fluoridated community (Indianapolis, IN, 0.8–1.2 mg F/L); and 3) a "halo" community (Connersville, IN, ≤0.3 mg F/L) in the distribution region for Indianapolis). All participating children were required to be healthy, attend a certified, commercial-, community- or church-based day-care center on a full-time basis, and have parental consent and cooperation. The day-care water source was required to have a fluoride concentration similar to the community water supply. Duplicate plate samples of all foods consumed (after visual adjustment for plate waste) on one or two day-care days and one weekend home day were collected and conglomerated for analysis. Beverages were kept separate from solid foods.

Water samples were analyzed for fluoride using a combined fluoride ion-specific electrode and pH/ion meter calibrated with a series of standards. Food samples were analyzed for fluoride using the HMDS-microdiffusion method of Taves (1968b) as modified by Dunipace et al. (1995). All samples were analyzed in duplicate, and the reliability of measurements was determined using the Intraclass Correlation Coefficient, the values of which were estimated from the variance components of an ANOVA model. Mean fluoride intake from food was 0.116-0.146 mg/day (Table 2-36) with no significant difference between communities. Intake from beverages (including drinking water) was estimated to be 0.103, 0.257, and 0.396 mg/day for the low-fluoride, halo, and fluoridated communities; differences between the towns were statistically significant ($p < 0.05$) as determined by one-way ANOVA. Based on mean values, total dietary fluoride intake (including drinking water) was 0.219 mg/day in San Juan, 0.389 mg/day in Connersville, and 0.544 mg/day in Indianapolis.

**Table 2-36. Dietary Fluoride Intake of 16-40 Month Old Children**

| City | F in DW | N | F Intake[a] from Foods (µg/day) | F Intake[a] from Beverages (µg/day) | Total Dietary F Intake (µg/day)[d] |
|---|---|---|---|---|---|
| San Juan, PR | ≤0.3 mg/L | 11 | 116±24[b] | 103±22[c] | 219 |
| Connersville, IN | ≤0.3 mg/L | 14 | 132±16[b] | 257±59[c] | 389 |
| Indianapolis, IN | 0.8-1.2 mg/L | 29 | 146±17[b] | 396±52[c] | 542 |

**SOURCE:** Rojas-Sanchez et al., 1999.
[a]Mean ± SEM.
[b]Not significantly different from each other ($p > 0.05$; one-way ANOVA).
[c]Significantly different, $p < 0.05$.
[d]Total of mean values.

Using a duplicate plate method, Brunetti and Newbrun (1983), evaluated the fluoride dietary intake and output of a group of 10 children (4 boys and 6 girls) ages 3 and 4 years, living in an optimally fluoridated community (study location and fluoride concentration in drinking water not reported). The diet of the children was unrestricted except that they were not allowed to chew gum. Duplicates of all food and fluid served and any leftovers by each child were collected and pooled every 24 hr. Intake was measured by subtracting leftovers from food served. Samples were assayed for fluoride using a diffusion method. The reported average dietary intake of fluoride was 0.33 (±0.14) mg F/day (food and beverage). Fluoride output (assumed to be based on urinary excretion) was reported to be 0.28 (±0.08) mg F/day.

### 2.5.4. Older Children and Adults

Exposure estimates for older children and adults are based on market basket and duplicate-plate types of studies. As was the case in Sections 2.5.2 and 2.5.3, the data from Market Basket-type analyses are presented first. Summaries that utilized the duplicate plate-type of approach follow.

**Market Basket Studies.** San Filippo and Battistone (1971) calculated the fluoride content of representative diets of 16–19 year-old males. Food items were obtained in Baltimore, MD from a market basket program conducted by the FDA on four separate occasions in 1967 and 1968. The food items were purchased in local supermarkets and prepared "in a manner representative of preparation at home" using the local fluoridated water. The food items for a two-week period were weighed to the nearest gram (wet weight) and then separated into 12 commodity groups. The commodity groups were homogenized and analyzed on a composite basis using microdiffusion and a colorimetric analysis. For most groups, the final values were averages of triplicate analyses. Results are shown in Table 2-37. Analysis of the Baltimore water supply indicated that the fluoride level ranged from 0.99 to 1.1 mg/L.

The daily contribution of each commodity group was an average of the two-week content. The data indicated an average total daily intake of 2.09–2.34 mg fluoride. Beverages contributed 61% to the total (1.28–1.46 mg/day), and all other food stuffs including those prepared with milk or water contributed 39% (0.78–0.9 mg/day). San Filippo and Battistone (1971) note that in their study the fluoride intake from the food ranged from 0.8 to 0.9 mg/day, an increase of about 0.5 mg over the intake from areas containing low fluoride in the drinking water reported in other studies (McClure, 1949 and Cholak, 1959).

Singer et al. (1980) evaluated the total daily dietary fluoride intake of 16–19 year-old males living in four geographic regions of the United States. Fluoride content of FDA composite "market basket collections" made in 1975 and 1977 were used in the analysis (USFDA, 1977). Food collections consisted of 117 items placed in 12 composite groups. The diets were based on Department of Agriculture (USDA, 1968, 1972) regional food consumption surveys. Fluoride in the food items was determined for ashed and unashed samples using ion-specific electrode and colorimetric (eriochromecyanine R) procedures. Total daily dietary fluoride intake, excluding drinking water (see Singer et. al., 1985), ranged from 0.912 mg in Kansas City to 1.720 mg in Atlanta (Table 2-38). Average and total mean fluoride intake for all four cities combined is 1.211 mg/kg/day (Table 2-39). Beverages contributed 65% of the total.

| Table 2-37. Fluoride Content of Four Two-week Representative Diets for Teens 16-19 Years Old | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| **Commodity Group** | **Diet #1** | | **Diet #2** | | **Diet #3** | | **Diet #4** | |
| | **mg F /2 wk** | **mg F /day** | **mg F /2 wk** | **mg F /day** | **mg F /2 wk** | **mg F /day** | **mg F /2 wk** | **mg F /day** |
| Dairy | 2.47 | 0.18 | 2.66 | 0.19 | 1.86 | 0.13 | 1.34 | 0.10 |
| Meats, fish, poultry | 2.03 | 0.15 | 4.20 | 0.30 | 4.38 | 0.31 | 1.52 | 0.11 |
| Grain and cereal products | 3.06 | 0.22 | 2.77 | 0.20 | 1.64 | 0.12 | 4.09 | 0.29 |
| Potatoes | 0.34 | 0.02 | 0.49 | 0.04 | 0.49 | 0.04 | 1.32 | 0.09 |
| Leafy vegetables | 0.52 | 0.04 | 0.37 | 0.03 | 0.34 | 0.02 | 0.94 | 0.07 |
| Legume vegetables | 0.28 | 0.02 | 0.14 | 0.01 | 0.18 | 0.01 | 0.28 | 0.02 |
| Root vegetables | 0.09 | 0.01 | 0.05 | 0.01 | 0.07 | 0.01 | 0.27 | 0.02 |
| Garden fruits | 1.11 | 0.08 | 0.19 | 0.01 | 0.14 | 0.01 | 0.45 | 0.03 |
| Fruits | 0.43 | 0.03 | 0.50 | 0.04 | 0.56 | 0.04 | 0.49 | 0.04 |
| Oils, fats and shortening | 1.27 | 0.09 | 0.34 | 0.02 | 0.36 | 0.03 | 0.18 | 0.01 |
| Sugar, salt, candy | 0.53 | 0.04 | 0.83 | 0.06 | 0.92 | 0.07 | 0.68 | 0.05 |
| Beverages: tea, coffee, soft drinks, water | 20.38 | 1.46 | 17.89 | 1.28 | 18.31 | 1.31 | 18.51 | 1.32 |
| **Totals** | **32.51** | **2.34** | **30.43** | **2.18** | **29.25** | **2.09** | **30.07** | **2.15** |

**SOURCE:** San Filippo and Battistone, 1971.

| Table 2-38. Average Daily Fluoride Intake of 16-19 Year Olds Residing in Four Cities | | | | |
|---|---|---|---|---|
| **Commodity Group** | **San Francisco, CA** | **Buffalo, NY** | **Atlanta, GA** | **Kansas City, KS** |
| | **mg F/day** | **mg F/day** | **mg F/day** | **mg F/day** |
| Dairy | 0.035 | 0.039 | 0.052 | 0.040 |
| Meats, fish, poultry | 0.058 | 0.058 | 0.239 | 0.084 |
| Grain and cereal products | 0.138 | 0.167 | 0.168 | 0.126 |
| Potatoes | 0.022 | 0.013 | 0.021 | 0.022 |
| Leafy vegetables | 0.007 | 0.007 | 0.009 | 0.005 |
| Legume vegetables | 0.011 | 0.017 | 0.032 | 0.023 |
| Root vegetables | 0.003 | 0.004 | 0.003 | 0.003 |
| Misc. vegetables | 0.011 | 0.011 | 0.011 | 0.013 |
| Fruits | 0.013 | 0.031 | 0.015 | 0.013 |
| Oils, fats | 0.017 | 0.011 | 0.011 | 0.011 |
| Sugars, adjuncts | 0.018 | 0.020 | 0.026 | 0.028 |
| Beverages | 0.882 | 0.610 | 1.133 | 0.544 |
| **Totals[a]** | **1.215** | **0.988** | **1.720** | **0.912** |

**SOURCE:** Singer et al., 1980.

[a]Singer et al., 1985, state that the total daily fluoride intake reported in Singer et al., 1980, did not include fluoride ingested in drinking water.

| Table 2-39. Daily Fluoride Intake Based on Composite Diets | | |
|---|---|---|
| **Category** | **Fluoride Intake (mg/day)** | |
| | **Mean** | **SD** |
| Dairy products | 0.042 | 0.007 |
| Meat, fish and poultry | 0.110 | 0.087 |
| Grains and cereal products | 0.150 | 0.021 |
| Potatoes | 0.020 | 0.004 |
| Leafy vegetables | 0.007 | 0.002 |
| Legume vegetables | 0.021 | 0.009 |
| Root vegetables | 0.003 | 0.001 |
| Garden Fruits | 0.012 | 0.001 |
| Fruits | 0.018 | 0.009 |
| Oils and fats | 0.013 | 0.003 |
| Sugars, etc. | 0.023 | 0.005 |
| Beverages | 0.792 | 0.270 |
| **Total Intake** | **1.211** | |

**SOURCE:** Singer et al., 1980.

In a continuation of the studies of Singer et al. (1980), Singer et al. (1985) utilized 24 FDA market basket collections made between 1975 and 1982 to again evaluate total daily fluoride intake of 15–19 year-olds living in the same four geographic regions of the United States. Food collections (24 "market baskets") consisted of 117 items placed in the same 12 composite groups. The diets used by Singer et al. (1985) were based on the USDA's Food Consumption Surveys of 1968 and 1972, and the USFDA (1977) Compliance Program Guidance Manual, extrapolated to reflect the Recommended Daily Allowance (RDA) for the average young adult male (2800 kcal). Fluoride in the composites was determined on diffusates of ashed samples with a fluoride ion-specific electrode.

Total fluoride intake ranged from 0.46 to 2.04 mg/day in eight cities in the West; 0.93 to 2.45 mg/day for four cities in the South; 0.80 to 1.92 mg/day for four cities in the North Central part of the country; and 1.47 to 1.94 mg/day for 3 cities in the North East. Singer et al. (1985) separated their data on the basis of the fluoride level in the municipal drinking water of each city to determine the impact of fluoride concentration in tap water on total fluoride intake (Table 2-40). Foods, exclusive of beverages and drinking water, contributed a mean fluoride intake of 0.27–0.37 mg/day (overall mean 0.33 mg/day). Singer et al. (1985) noted that a basal diet in a nonfluoridated region of the United States contained 0.43 mg of fluoride as reported by Maheshwari et al., 1981.

| Table 2-40. Average Daily Fluoride Intake (mg/day) of 16-19 Year Olds | | | | |
|---|---|---|---|---|
| Commodity Group | Fluoride Concentration of Municipal Water | | | |
| | < 0.3 mg/L | 0.3-0.7 mg/L | >0.7 mg/L | <0.1 to 1.3 mg/L |
| | (0.14 ±0.03)[a] (n = 5)[b] | (0.56 ±0.05)[a] (n = 11)[b] | (1.04 ±0.05)[a] (n = 8)[b] | (n =24)[b] |
| Total dietary | 0.86 ±0.14 | 1.39 ±0.13 | 1.85 ±0.11 | NR |
| Beverages and Water | 0.59 ±0.12 | 1.06 ±0.11 | 1.48 ±0.08 | NR |
| Food only | 0.27 ±0.03 | 0.33 ±0.03 | 0.37 ±0.05 | 0.33 ±0.02 |

**SOURCE:** Singer et al., 1985.

NR. Not reported.

[a]Mean F concentration ±SEM.

[b]Number of market baskets.

Based on a 6-day survey of a regular hospital diets (location not specifically mentioned, but presumed to be in the Rochester, NY area, as the affiliation of the researchers was the University of Rochester), and using information on fluoride levels in 93 foods and beverages (see Section 2.2.2), Taves (1983) calculated a mean total daily fluoride intake of 1.783 mg, of which 1.383 mg (78%) was provided by beverages (Table 2-41). Tea was the major contributor to the intake from beverages. The author notes that drinking water was not taken into account in the study, but that the tap water used in the preparation of the hospital foods was fluoridated. The fluoride level in the tap water was not reported.

| Table 2-41. Daily Fluoride Intake based on 6-day Hospital Diets | | |
|---|---|---|
| Category | Fluoride Intake (mg/day) | |
| | Mean | SD |
| Dairy Products | 0.013 | 0.000 |
| Meat, fish and poultry | 0.044 | 0.035 |
| Grains and cereal products | 0.241 | 0.153 |
| Potatoes | 0.018 | NR |
| Leafy vegetables | 0.027 | 0.019 |
| Legume vegetables | 0.037 | NR |
| Root vegetables | 0.010 | NR |
| Garden Fruits | 0.000 | NR |
| Fruits | 0.006 | NR |
| Oils and fats | 0.003 | 0.001 |
| Sugars, etc. | 0.001 | 0.000 |
| Beverages | 1.383 | 0.041 |
| **Total Intake** | **1.783** | |

**SOURCE:** Taves, 1983.

NR, Not reported

Dabeka and McKenzie (1995) surveyed fluoride levels in various foods obtained in 1987 in Winnipeg, Canada. The foods were prepared for consumption and combined into 113 composites and 39 composite subsets using a Total Diet Study approach. The concentration of fluoride in tapwater was reported to be 1 mg/L. Fluoride was determined with a fluoride ion-specific electrode after microdiffusion. As reported in Dabeka et al. (1993), food intake data (g/person/day) for each of composites was obtained from the Nutrition Canada Survey (Bureau of Nutritional Sciences, 1977) for the age groups of 1–4, 5–11, and 12–19, and 20+ years. Total dietary fluoride intake was 1.025 mg/day for 12–19 yr old males and 0.905 mg/day for 12–19 year old females. For the age groups of 20+ years, the fluoride intake ranged from 2.17 to 3.03 mg/day. Over all ages (including the 20+ yr groups) and both sexes, the estimated average dietary intake of fluoride was 1.76 mg/day; the food category contributing most to the estimated intake was beverages (80%).

**Duplicate Diet/Plate Methods.** The fluoride content of the strictly controlled metabolic diets that were used over a six-year period at a VA hospital in the Chicago area during 1967–72 were analyzed by Osis et al. (1974b). The house diets served to patients in the same hospital were also analyzed using the same approach. Fluoride concentrations were determined by the diffusion method of Singer and Armstrong (1965) with spectrophotometric analysis. Osis et al. (1974a) reported a coefficient of variation of 4.3% for this method. The daily intake of fluoride of individuals on the metabolic diet, as shown in Table 2-42, averaged 1.56–1.91 mg/day. During the course of the study, fluoridation of the tap water was temporarily discontinued. As a result, it was possible for the study authors to compare the fluoride content of the general hospital diet when “non-fluoridated” water (0.27 mg F/L) was used in the preparation of meals with that prepared with fluoridated water (about 0.9 mg F/L). The results, shown in Table 2-43, indicate that the average fluoride intake was reduced more than 50% when the “nonfluoridated” water was used in the preparation of the meals.

**Table 2-42. Fluoride Intake of Individuals on a Metabolic Diet over a Six-Year Period**

| Year | Average ±SD (mg/day)[a] | Range |
|---|---|---|
| 1967 | 1.91 ±0.42 | 1.47–3.08 |
| 1968 | 1.60 ±0.15 | 1.26–1.83 |
| 1969 | 1.56 ±0.18 | 1.21–2.30 |
| 1970 | 1.76 ±0.15 | 1.46–2.06 |
| 1971 | 1.74 ±0.16 | 1.28–2.07 |
| 1972 | 1.60 ±0.15 | 1.33–1.88 |

**SOURCE:** Osis et al., 1974b.

[a]Water used in the preparation of the meals contained about 0.9 mg/L fluoride.

| Table 2-43. Fluoride Intake from a General Hospital Diet Prepared with and without Fluoridated Water | | | |
|---|---|---|---|
| **Meal** | **No.** | **Average ±SD (mg/day)** | **Range** |
| **Diet prepared with fluoridated water[a]** | | | |
| Breakfast | 5 | 0.65 ±0.17 | 0.47–0.86 |
| Lunch | 5 | 0.75 ±0.28 | 0.42–1.16 |
| Dinner | 5 | 0.57 ±0.15 | 0.34–0.71 |
| **Total F (mg/day)[b]** | | **1.96 ±0.48** | **1.23–2.41** |
| **Diet prepared with non-fluoridated water[c]** | | | |
| Breakfast | 5 | 0.29 ±0.06 | 0.21–0.37 |
| Lunch | 5 | 0.32 ±0.06 | 0.25–0.37 |
| Dinner | 5 | 0.25 ±0.02 | 0.22–0.27 |
| **Total F (mg/day)[b]** | | **0.86 ±0.08** | **0.73–0.94** |

**SOURCE**: Osis et al., 1974b.

[a]Water used in the preparation of the meals contained 0.9 mg F/L.

[b]The total daily dietary fluoride represents the range from the lowest to the highest intakes per day, and does not represent the sum of the individual meals listed.

[c]Water used in the preparation of the meals contained about 0.27 mg F/L.

Kramer et al. (1974) analyzed the fluoride content of diets obtained from hospitals in 16 cities in the United States, 12 cities where the drinking water was fluoridated and 4 cities where the drinking water was not fluoridated (Table 2-44). The diets were normal in composition and provided 2,400 to 2,600 kcal/day. Most of the diets were collected as separate, individual meals, breakfast, lunch and dinner; although in some cases the food items making up the diet for the entire day were obtained. The compositions of each individual meal and of the total diet were determined. Beverages, including coffee and tea, were included, but not plain drinking water. Fluoride was analyzed by the method of Singer and Armstrong (1965). Dietary fluoride was lowest in those communities having the lowest fluoride levels in drinking water. The mean fluoride content of the diet was generally greater in fluoridated areas than in nonfluoridated areas, however, there was not a linear relationship between the fluoride concentration of the drinking water supply and that of the diet. The highest level of dietary fluoride was that for a community with a 0.6 mg/L fluoride concentration, not the system with the highest drinking water fluoride concentration (1.27 mg/L).

| Table 2-44. Dietary Fluoride Intake in Sixteen U.S. Cities | | |
|---|---|---|
| **City** | **F in Drinking water (mg/L)** | **Daily Dietary F Intake (mg)** |
| Birmingham, AL | 0.08 | 0.78 |
| Iron Mountain, MI | 0.08 | 1.03 |
| Chicago, IL | 0.33 | 0.86[a] |
| Houston, TX | 0.44 | 0.95 |
| Durham, NC | 0.53 | 2.62 |
| Corvallis, OR | 0.60 | 3.44 |
| Tuscaloosa, AL | 0.76 | 2.94 |
| Martinez, CA | 0.81 | 1.73 |
| Milwaukee, WI | 0.85 | 3.41 |
| New York, NY | 0.88 | 2.55 |
| St. Louis, MO | 0.91 | 2.10 |
| Chicago, IL | 0.95 | 1.97 |
| Madison, WI | 1.11 | 2.88 |
| Louisville, KY | 1.14 | 1.98 |
| Lexington, KY | 1.15 | 2.84 |
| Cleveland, OH | 1.27 | 3.05 |

**SOURCE:** Kramer et al., 1974.

[a]Average of five diets analyzed at a time when the water was not fluoridated.

### 2.5.5. Combined Exposure Estimates for Age Groups of Concern

The OW has used the dietary exposure data to estimate fluoride intakes for the age groups identified in the OW dose-response assessment (U.S. EPA, 2010a). The data summarized in Table 2-45 come from the U.S. assessments discussed in Section 2.5.2 through 2.5.4 that were based on analytical data from foods and TDS or duplicate diet estimates. Table 2-45 does not include intake from drinking water or the beverage grouping where possible. The beverage data are summarized in Table 2-46. Study conditions are described in the notes field of the table.

Evaluation of the food and exposure data support several conclusions related to fluoride intake via the diet.

- The use of fluoridated water in processing and preparing food increases the fluoride content of the diet for both home prepared and commercial foods but not in predictable linear fashion (Maier and Rose, 1966; Ophaug et al., 1985).
- The relationship between the fluoride in local tap water and intake from beverages displays a linear relationship (≥ 0.72 correlation coefficient; Ophaug et al., 1985).
- Analytical methods influence the results. The older colorimetric methods appear to be less reliable than more recent methods (Singer at al., 1980).

- Concentration of fluoride appears to be related to food group as follows: protein foods > grains and vegetables, > fruits, > beverages.

**Table 2-45. Summary of Daily Dietary Fluoride Intakes for Age Groups of Concern**

| Age years | Fluoride Exposure Estimate (mg/day) | Notes |
|---|---|---|
| 0.5 - <1 | 0.171 ±0.012 | Ophaug et al., 1985 – Overall mean of 44 market baskets, and national food intake data; does not include F from water and beverages; 6 months old age group. |
| 1-<4 | 0.161 ±0.010 | Ophaug et al., 1985 – Overall mean of 22 market baskets, and national food intake data; does not include water and beverages; 2 years old age group. |
| | 0.116 ±0.024<br>0.132 ±0.016<br>0.146 ±0.017 | Rojas-Sanchez et al., 1999 – Duplicate plate analysis (n=54; mean ±SEM ) for three cities; excludes beverages and drinking water (≤0.3 mg F/L DW in 2 cities and 0.8-1.2 mg F/L DW in the third); 1.3-3.3 year old age group. |
| | 0.33 ±0.14 | Brunetti and Newbrunn, 1983 – Duplicate plate analysis (n=10, for 1-4 days); estimate for all foods and fluids consumed 3-4 year old age group |
| 4-<7 | 0.33 ±0.14 | Brunetti and Newbrunn, 1983 – Duplicate plate analysis (n=10, for 1-4 days); estimate for all foods and fluids consumed; 3-4 year old age group. |
| | 0.338 | Jackson et al., 2002 – Analysis of 75 most commonly consumed foods and beverages of 12-14 yr olds placed in 9 composites and USDA food consumption data for 3-5 year olds. Does not include water and beverages; 0.16 mg F/L DW; fluoridated water concentration 0.16 mg/L. |
| | 0.344 | Jackson et al., 2002 – Analysis of 75 most commonly consumed foods and beverages of 12-14 yr olds placed in 9 composites and USDA food consumption data for 3-5 year olds. Does not include water and beverages; fluoridated water concentration 0.9 mg/L |
| 7-<11 | 0.35 | No U.S. data for age group. The estimate is based on the analytical food group fluoride data from Jackson et al., (2002) and USDA data on food group intakes for 6-11 year olds. Does not include water and beverages; fluoridated water concentration 0.9 mg/L. |
| 11-<14 | 0.405 | No U.S. data for age group. The estimate is based on the analytical food group fluoride data from Jackson et al., (2002) and USDA data on food group intakes for 12-19 year olds. Does not include water and beverages; fluoridated water concentration 0.9 mg/L |
| > 14 | 0.83 | San Filippo and Battistone, 1971 – Four market baskets, and FDA food intake data; does not include water and beverages; 16-19 years old. |
| | 0.424 | Singer et al., 1980. Market baskets from 4 regions of the country; beverages and plain drinking water not included; 16-19 years old. |
| | 0.33 | Singer et al., 1985. 24 market baskets from different areas of the country; beverages and plain drinking water not included; 16-19 years old. |
| | 0.403 | Taves, 1983 – Six-day hospital diet; does not include beverages and plain drinking water; Adults. |

| Table 2-46. Estimates of Daily Dietary Fluoride from Beverages for Age Groups of Concern | | |
|---|---|---|
| **Age (yr)** | **Fluoride Exposure Estimate (mg/day)** | **Notes** |
| 0.5- <1 | 0.14 (mg/L) | Van Winkle et al., 1995. Concentration for powdered formula prepared with distilled water. |
| 0.5–1 | 0.09–0.12 | Siew et al. 2009. Estimates (from graphical presentation of data) of range of fluoride intake from powdered formula prepared with distilled water, based on estimates of formula intake for female infants 6 to 12 months old. |
| 1 - <4 | 0.36 ±0.31 | Pang et al., 1992 – Three-day drink diaries (n=57); beverages only, excluding milk, water and those listed fewer than five times; home-prepared beverages made with de-ionized water; 2–3 years old. |
| | 0.257 ±0.059 | Rojas-Sanchez et al., 1999 – Duplicate plate study (n=14; mean ±SEM); beverages and drinking water; ≤0.3 mg F/L DW; 1.3–3.3 years old. |
| | 0.396 ±0.052 | Rojas-Sanchez et al., 1999 – Duplicate plate study (n=29; mean ±SEM); beverages and drinking water; 0.8 mg F/L DW; 1.3–3.3 years old. |
| 4-<7 | 0.54 ±0.52 | Pang et al., 1992 – Three-day drink diaries (n=79); beverages only, excluding milk, water and those listed fewer than five times; home-prepared beverages made with de-ionized water; 4–6 years old. |
| | 0.116 | Jackson et al., 2002 – Analysis of 75 most commonly consumed foods and beverages of 12–14 yr olds placed in 9 composites and USDA food consumption data for 3–5 yr olds; beverages only, plain DW not included; low F location (0.16 mg/L); 3–5 years old. |
| | 0.192 | Jackson et al., 2002 – Analysis of 75 most commonly consumed foods and beverages of 12–14 yr olds placed in 9 composites and USDA food consumption data for 3–5 yr olds; beverages only, plain drinking water not included; fluoridated location (0.9 mg/L); 3–5 years old. |
| | 0.2 | Levy et al., 2003a. Estimate of average intake from beverages, not including plain drinking water for 3-6 year olds derived from questionnaires completed by the parents and historical data on fluoride concentrations in the beverages. The 90$^{th}$ percentile estimate was 0.5 mg/day. |
| 7-<11 | 0.60±0.48 | Pang et al., 1992 – Three-day drink diaries (n=89); beverages only, excluding milk, water and those listed fewer than five times; home-prepared beverages made with de-ionized water; 7-10 years old. |
| | 0.216 | This estimate is based on the means from two market baskets in the study by Jackson et al. (2002) and USDA data on beverage intakes. It does not include drinking water. Ages 6–11. |
| 11- <14 | 0.509 | This estimate is based on the means from two market baskets in the study by Jackson et al. (2002) and USDA data on beverage intakes. It does not include drinking water. Ages 12–19. It is supported by the average (0.51 mg/L) from a Canadian dietary record survey by Clovis and Hargreaves (1988); (range 0.02-0.82 mg/day). |
| ≥14 | 1.34 | San Filippo and Battistone, 1971 – Four market baskets, and FDA intake data; includes beverages and plain drinking water; 16–19 years old. |
| | 0.792 | Singer et al., 1980. Market baskets from 4 regions of the country16–19 years old. |
| | 0.59 | Singer et al., 1985. 5 Market baskets from different areas of the country; plain drinking water not included. Drinking water used to prepare beverages low in fluoride (0.14 mg/L ± 0.03); 15–19 years old. |
| | 1.383 ±0.041 | Taves, 1983 – beverages; does not including plain drinking water; derived from a duplicate plate hospital study; adults. |

The U. S. EPA assessment of dose-response for severe dental fluorosis (U.S. EPA, 2010a) divided the population into age groups that correlate with those used in the Ershow and Cantor (1989) analysis of drinking water intakes because they represented the water intake data that were closest (1977-1978) to those likely to have occurred at the time of the Dean (1942) publication. The age groupings reported in the published papers summarized above are not always congruent with those used by EPA (2010a). For this reason Tables 2-45 and 2-46 array the published data according to the age groups used for the dose-response assessment. As a result, each study was placed according to its best fit with the drinking water age groups.

Except for Brunetti and Newbrun (1983), Table 2-45 on intakes from solid foods does not include intakes from beverages. Milk and fruit juices are included in the solid foods grouping because of their placement in a market basket survey in the dairy and fruit groups, respectively. Table 2-46 is a summary of the data reported for other beverages as a separate market basket item. In Table 2-46, no attempt was made to separate fluoride that may have originated from local tap water used in making tea, coffee or powdered juice drinks from the commercial beverages. Two of the studies (Pang et al., 1992; Van Winkle et al., 1995) used deionized water in the home preparation of beverages.

There is variability in the results reported for the fluoride in beverages with Pang et al. (1992) generally reporting higher levels for the 4 to <7 year old group and the 7 to < 11 year old group than other studies. The Pang et al. (1992) study used a record keeping approach (3-days) to determining the kinds and amounts of beverages consumed by children in North Carolina in April, May and June. The ages of the participants, diary approach, location (southern U.S.), and time of year (Spring and early Summer) could have influenced these results.

In order to refine the fluoride estimate from beverage ingestion, EPA examined the list of market basket foods and their categories in the 1990 and 2003 FDA market basket lists (Egan, 2009). Most fruit juices were included in the fruit rather than the beverage group. The beverage group included carbonated beverages, coffee, tea products reconstituted or prepared using tap water, and alcoholic beverages. Based on information obtained from FDA, beverages containing commercial water contributed 53 to 74 % of the total mass intake from the beverage category in the TDS based on the 1987–1988 CSFII and 65 to 77 % for the TDS based on the 1994–1998 CSFII (bottled water and alcoholic beverages excluded) for the age groups of interest. The remainder would be contributed by the indirect use of tap water explaining the strong correlation between local levels in drinking water and the market basket results for beverages. In general, the commercial water contribution to a market basket beverage intake increases with age (Egan, Personal Communication, 2009). This is consistent with the higher intakes of carbonated and other commercial beverages by the older age groups.

The San Filippo and Battistone (1971) results for those >14 include plain drinking water but are similar to the Taves (1983) results which do not. However, Taves (1983) explains that the hospital diets studied included orange juice, coffee, and two servings of tea on a daily basis as well as other juices. The analytical data from the Taves (1983) study show that the tea was the major contributor to the fluoride from beverages. For that reason the Singer et al. (1980, 1985) results are considered to be more representative of the general population when plain drinking water is excluded.

### 2.5.6. Fluoride Exposures from Sulfuryl Fluoride Use

At the request of the Office of Water, OPP (U.S. EPA, 2009, 2010b) provided estimates of exposures to fluoride from the tolerances granted to sulfuryl fluoride (SuF in Table 2-47). The OPP data were generated using the DEEM exposure program that integrates residue data from representative commodities with age-specific food group intakes from CSFII (1998). Exposure estimates were provided by age group and whether the residues were the result of fumigation of food storage facilities or fumigation of food processing structures (U.S. EPA, 2010b).

**Table 2-47. Summary of Pesticidal Fluoride Contributions to Dietary Fluoride Exposure**

| Population Group | Exposure Estimates, mg/day | | |
|---|---|---|---|
| | SuF Structural Fumigations | SuF Food Fumigations | Total |
| 0.5-1 year | 0.0087 | 0.0213 | 0.0300 |
| Children 1-<4 yrs | 0.0121 | 0.0329 | 0.0450 |
| Children 4-<7 yrs | 0.0153 | 0.0466 | 0.0619 |
| Children 7-<11 yrs | 0.0170 | 0.0544 | 0.0714 |
| Youth 11-<14 yrs | 0.0182 | 0.0675 | 0.0857 |
| Adults >14 | 0.0187 | 0.0576 | 0.0763 |

**SOURCE:** U.S. EPA, 2010b

The age groups generated by the OPP exposure assessment (U.S. EPA, 2010b) are congruent with those used by OW for this report. OPP (U.S. EPA, 2010b) also reported the exposure estimates in terms of mg/day. The 11 to <14 year age group appears to have the highest estimated total exposure from sulfuryl fluoride residues.

## 3. Exposure from Drinking Water

Fluoride occurs naturally in water. Levels in drinking water can range from insignificant to unacceptably high depending on the water source and the extent of treatment. In many locations where the fluoride levels are naturally low, fluoride is intentionally added to water supply systems to reduce the occurrence and severity of dental caries in children. Community water fluoridation at a concentration of about 1 ppm was initiated in 1945 (Ripa, 1993). Based on data collected in 1999 from 24 locations nation-wide, Miller-Ihli et al. (2003) concluded that 40% of the U.S. water supplies were fluoridated (mean concentration 1.01±0.15 mg/L). Currently CDC (2008) records indicate that about 69% of the population obtains its water from systems that fluoridate.

### 3.1. Analytical Methods

Methods used to analyze for fluoride in drinking water have changed over time. In the 1930s and early 1940s, colorimetric methods required visual comparison of the color of samples with a set of standard solutions to identify the fluoride concentration in the sample. In the Elvove (1933) method, water samples were acidified with hydrochloric acid and mixed with a dye complex such as zirconium oxychloride and alizarin sodium monosulphonate mixed to produce a colored solution from binding of the fluoride with the reagent. A series of solutions containing varying known amounts of sodium fluoride are mixed with the reagent to produce a series of colored standards. The test samples were then visually compared to the standards (in "Nessler" tubes) to estimate the concentration by a match of the sample color with that of the color of the closest standard. Elvove (1933) reported that as little as 0.01 mg of fluorine in 50 cc, or 0.2 mg/L could be differentiated from a corresponding control with this method. This method was used by Dean (1942) in evaluating the fluoride content in water supplies of 22 U.S. cities. The Dean (1942) report states that the sensitivity of the analytical method was about 0.1 mg/L. The Dean (1942) study is the basis of the dose-response assessment for severe dental fluorosis in U.S. EPA (2010a).

The Elvove (1933) colorimetric method is subject to error caused by interfering substances such as sulfate, chloride, bicarbonate, iron, manganese, and aluminum when these substances exceed specific concentrations. Nevertheless, colorimetric methods for fluoride determination are still considered Standard methods today, albeit using spectrophotometric instrumentation and standard curves for determining concentrations.

The most recent standard colorimetric methods employ two reagents related to those used by Elvove (1933). One employs an acidic reagent containing zirconyl chloride and the complexing agent SPADNS [sodium 2-(parasulfophenylazo)-1, 8-dihydroxy-3, 6-naphthalene disulfonate] (APHA/AWWA/WEF, 2005). The other employs both alizarin and lanthanum nitrate to form a blue complex in an automated system.

In the mid-1960s a fluoride ion-specific electrode was developed which allowed direct detection and measurement of fluoride concentrations in water by means of a potentiometer (see Section 2.1.3 for further discussion). The concentration of the fluoride ion was in direct proportion to the current generated. Compared to colorimetric methods, the fluoride ion-specific electrode exhibits superior selectivity when challenged with chloride, chlorine, color and turbidity, iron, phosphate, sulfate, and aluminum (*Standard Methods,* APHA/AWWA/WEF, 2005). *Standard Methods* clearly indicates the electrode and colorimetric methods are most satisfactory

Ion-specific chromatography can also be used to analyze fluoride in aqueous solution, and although this method has a high level of sensitivity and specificity for fluoride, it has only rarely been used in the studies discussed in this report.

### 3.2. Natural Sources

Drinking water can be obtained from non-fluoridated municipal systems, private wells, cisterns, springs, or from bottled water. The fluoride levels in these sources may vary considerably depending on the source, time of year, and the level of treatment. Certain geological formations are rich in fluoride-containing minerals from which fluoride can leach into surrounding groundwater or surface water. According to Fleischer et al. (1974), some groundwaters average as much as 8 ppm of fluoride or more. Groundwater from the Wilcox Basin in Southeastern Arizona can contain up to 282 ppm fluoride (Kister et al., 1966). Most water from this basin is used primarily for irrigation. However, it is also the water source for several public drinking water systems (Towne and Freark, 2001).

Fluoride levels in groundwater in the coterminous United States were mapped by the U.S. Geological Survey (see Figure 3-1). Some of the areas indicated in Figure 3-1 correspond to areas of aridity as shown in Figure 3-2 (McGinnies et al., 1968). In these areas drinking water consumption rates may be greater than average, and combined with the high levels of fluoride in groundwater, may contribute to higher than normal exposures to fluoride from private drinking water systems and more frequent exceedences of the SMCL. States that have reported MCL violations most frequently to the Safe Drinking Water Information System – Federal (SDWIS/FED) during the period from 1998 to 2006 are Arizona, Florida, Montana, New Mexico, Texas and Virginia. All states have some areas with high levels of geological fluoride.

In 1993, the CDC reported on naturally occurring fluoride levels in U.S. water sources. Although there is a range in fluoride concentrations within each state, in most cases the maximum reported concentrations correspond fairly well with the areas predicted to have high levels of fluoride in groundwater (Fig. 3-1). According to CDC (1993), maximum concentrations of 7 mg/L or greater were reported for Arizona, Colorado, Idaho, Iowa, Montana, New Mexico, North Dakota, Oklahoma, and Texas. Seventeen states had maximum concentrations exceeding 4.0 mg/L, and 32 states had maximum concentrations ≥2.0 mg/L in some localities. In most cases only a small proportion of the total sampled population was located in areas where the fluoride levels were high. The CDC (1993) estimated that of the approximately 10 million people in the U.S. with naturally fluoridated public drinking water, approximately 67% had fluoride concentrations of ≤1.2 mg/L; about 14% had concentrations of 1.3–1.9 mg/L; 14% had concentrations of 2.0–3.9 mg/L and 2% had levels of ≥4.0 mg/L.

Due to the differences in groundwater fluoride, private water sources (particularly well-water) are likely to have highly variable fluoride concentrations. Felsenfeld and Roberts (1991) reported one case of fluoride-associated osteosclerosis in an individual whose drinking water well had an average concentration of about 8 mg F/L.

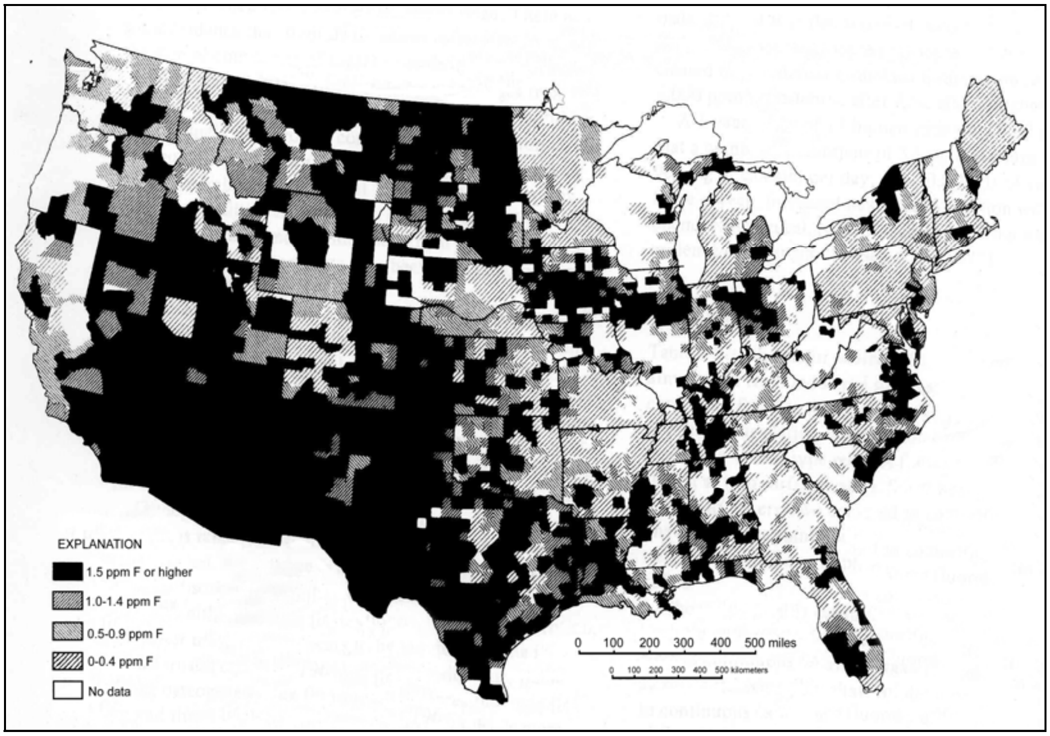


**Figure 3-1. Fluoride Levels in Groundwater in the U.S. (Fleischer et al., 1974).**

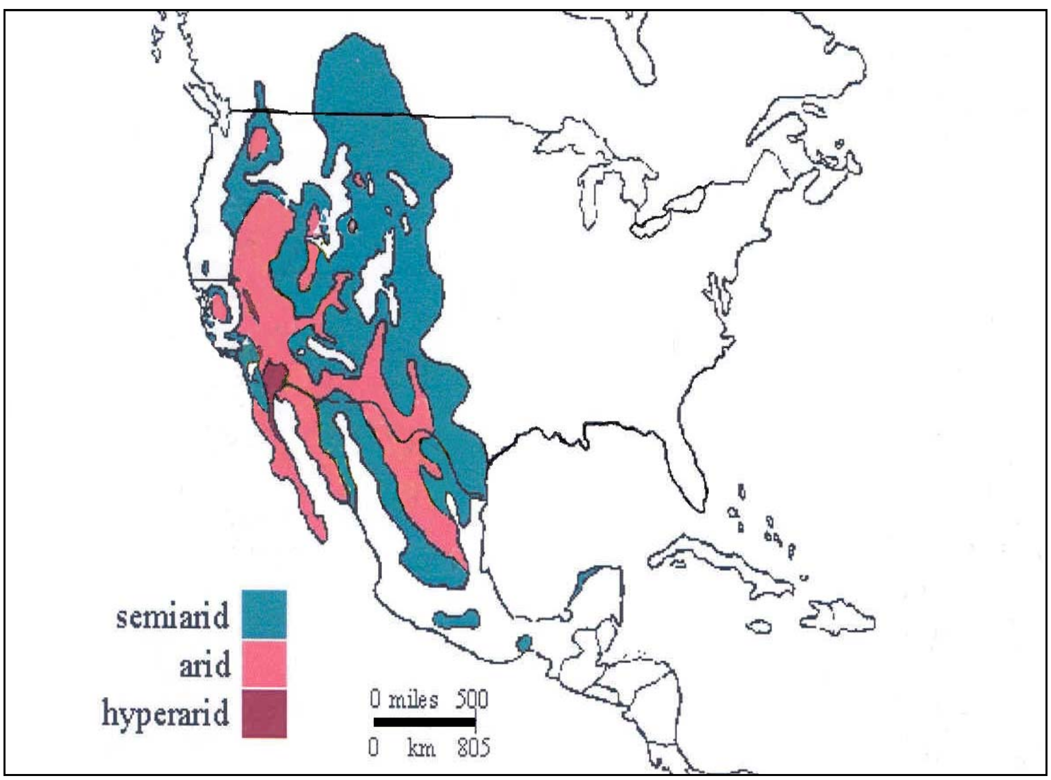


**Figure 3-2. Arid Regions in the U.S. (McGinnies et al., 1968).**

### 3.3. Public Drinking Water Systems

Public drinking water systems are required to monitor finished water for fluoride on defined schedules determined by whether or not the level detected exceeds the MCL, and to report the results to the state. If there is no exceedence of the MCL, surface water systems monitor once a year while groundwater systems monitor only once every three years unless granted a waiver by States to further reduce monitoring. The monitoring identifies whether or not there has been an exceedence of the MCL and SMCL. Exceedences are reported to consumers in their required yearly drinking water quality Consumer Confidence Report and trigger a return to quarterly monitoring. When the yearly average fluoride concentration exceeds the MCL (4 mg/L) the Consumer Confidence Report is required to include the following language regarding health effects:

> *Some people who drink water containing fluoride in excess of the MCL over many years could get bone disease, including pain and tenderness of the bones. Fluoride in drinking water at half the MCL or more may cause mottling of teeth, usually in children less than nine years old. Mottling, also known as dental fluorosis, may include brown staining and/or pitting of the teeth and occurs only in developing teeth before they erupt from the gums.* (40CFR141, subpart O, App. A).

In cases where the yearly average fluoride concentration exceeds the SMCL (2 mg/L), the following message must be sent to consumers within 12 months of the exceedence. This can be accomplished by including the warning in the annual Consumer Confidence Report. Exceedence of the SMCL is more frequent than exceedence of the MCL; ground water systems are affected to a greater extent than surface water systems. Exceeding the SMCL does not require a return to quarterly monitoring.

> *This is a notification about your drinking water and a cosmetic dental problem that might affect children under nine years of age. At low levels fluoride can help prevent cavities, but children drinking water containing more than 2 mg/L of fluoride may develop cosmetic discoloration of their permanent teeth (dental fluorosis). The drinking water provided by your community water system [name] has a fluoride concentration of [insert number] mg/L.*

> *Dental fluorosis, in its moderate or severe forms, may result in brown staining or pitting of the permanent teeth. This problem occurs only in developing teeth before they erupt from the gums. Children under nine should be provided with alternative sources of drinking water or water that has been treated to remove the fluoride to avoid the possibility of staining and pitting of the permanent teeth. You may also want to contact your dentist about proper use by young children of fluoride-containing products. Older children and adults may safely drink the water.*

> *Drinking water containing more than 4 mg/L fluoride (the U.S. Environmental Protection Agency's drinking water standard) can increase your risk of developing bone disease. Your drinking water does not contain more than 4 mg/L*

*fluoride, but we are required to notify you when we discover fluoride levels in your drinking water that exceed 2 mg/L because of this cosmetic dental problem.* (40CFR141.208).

In conjunction with the second six-year review of the National Primary Drinking Water Regulations, EPA conducted an Information Collection Request (ICR). Through this process EPA asked that all States and primacy entities voluntarily submit their SDWA compliance monitoring data. This request was for the submission of compliance monitoring data collected between January 1998 and December 2005 for 79 regulated contaminants. A total of 52 States and entities provided compliance monitoring data that included all analytical detection and non-detection records. These data represent the national occurrence of regulated contaminants in public drinking water systems. Through extensive data management efforts, quality assurance evaluations, and communications with State data management staff, EPA established a high quality dependable contaminant occurrence database consisting of data from 46 States. Details of the data management and data quality assurance evaluations are available in the supporting document (U.S. EPA, 2008b).

The contaminant occurrence data from the States and entities comprise more than 17 million analytical records from approximately 136,000 public water systems. Approximately 265 million people are served by these public water systems nationally. The number of States and public water systems represented in the data set varies across contaminants because of variability in voluntary State data submissions and contaminant monitoring schedules. This is the largest, most comprehensive set of drinking water compliance monitoring data ever compiled and analyzed by EPA.

EPA used a two-stage analytical approach to analyze these data and characterize the national occurrence of contaminants. The first stage of analysis provides a straightforward evaluation of contaminant occurrence. This stage is a simple, non-parametric count of occurrence for regulated contaminants in public water systems. A typical stage 1 occurrence analysis generates a count of the number (or percentage) of systems with at least one analytical detection of a specific contaminant at a concentration above the concentration of interest (i.e., the SMCL). This approach generates a conservative (i.e., upwardly biased) estimate of the number of potential systems having contaminant occurrence at levels of interest. It is the appropriate metric for a contaminant such as fluoride where intakes above the threshold of concern over even a limited period of time can have an impact on the development of enamel on the secondary teeth forming only during the time of the exposure.

ICR data for fluoride were examined on the basis of all samples and all systems as well as for only those systems that reported at least one sample with a concentration ≥2 mg/L during the 8-year reporting period. The results are summarized in Tables 3-1 and 3-2, and include conservative estimates of the total populations exposed during the monitoring period. According to information extracted from the U.S. Census Bureau's web site, there were 60.3 million children under the age 14 in the U.S. in 2010, approximately 21.4% of the total U.S. population. The period from 6 months to 14 years is the age period for enamel formation for secondary teeth, including the third molars (Massler and Schour, 1958).

The data set for fluoride included some entries with apparent unit discrepancies. Fluoride concentrations were designated as mg/L values but appear to have actually been µg/L values based on the other reported measures from the same utility. If the actual levels were truly mg/L measures, the high fluoride concentrations would have caused adverse effects among the exposed population (gastrointestinal irritation; see NRC, 2006 for review).

Values for detections reported as < 0.002 mg/L and greater than 40 mg/L were considered as outliers and eliminated from the analysis. Values reported as greater than 20 mg/L are also suspect based on historic records for the United States, but have been included in the analysis presented in Tables 3-1 and 3-2. A total of 426 entries were considered as anomalously high and eliminated from the analysis; six values between 40 and 100 mg/L and 420 values equal to or greater than 100 mg/L.

The ICR data set also included results from some transient noncommunity systems. The Agency excluded these samples from the analysis presented in Tables 3-1 and 3-2 because federal fluoride regulations do not apply. The Agency also excluded all samples that could be identified as source water quality samples that do not represent water quality at the entry point to the distribution system (e.g., water quality prior to treatment or fluoridation). The data in Tables 3-1 and 3-2 also do not include samples reporting fluoride as not detected in the determination of mean, median and 90$^{th}$ percentile values. There are variations in the number of samples and systems across the monitoring period. These variations reflect differences in the monitoring schedule and the number of States providing data. Systems that fluoridate are required to report fluoride levels monthly to the appropriate organization within their state (often the state dental officer) but have no obligation to report those monthly measurements to EPA.

The number of quarterly samples analyzed over the 8 years of monitoring ranged from about 7,000 to 12,000, with 2.3 to 5.6 % of these samples ≥ 2 mg/L. Monitoring data were analyzed for four quarters per year, but the data have been compressed in Table 3-1 and 3-2 to show only the range across the four quarters. The systems reporting each quarter are not consistent because surface water systems with mean average annual concentrations below 4 mg/L have to report only once per year or once every three years for a groundwater system.

Table 3-1 suggests the possibility of a trend towards an increase in the percent of samples with detections of 2 mg/L or higher across the 8-year monitoring period. In the first 4 years the percent of detections for the subset ≥ 2 mg/L exceeded 4% for two of the 16 quarters. In the second 4 years, the frequency increased to all 16 quarters. The percent of systems reporting a concentration of ≥2 mg/L ranged from 4.1 % to 5.6 % in the first four years of monitoring and 4.6% to 8.3% in the second four years. Close inspection of the ICR results indicates that the apparent trend was the result of an increase in the number of states included in the data set. The later years include states with high geological levels of fluoride (Florida, Texas, and Virginia) that did not submit data for the early years of the monitoring period.

The mean, median, and 90$^{th}$ percentile concentrations were determined for each of the monitoring quarters. Over the first four years of monitoring the high end of the range for the mean was 0.85 or 0.86 mg/L while in the second 4 years it increased to a maximum of 0.95 mg/L. In the last four years, the range for the means is consistently higher than that for the medians reflecting positively skewed distribution (i.e., having a longer right tail with higher F concentrations). A similar trend is reflected in the 90$^{th}$ percentile values, which have also increased over the 8 years of monitoring. The means and medians remain at a concentration within the recommended range for fluoridation and the 90$^{th}$ percentile value, although consistently above the upper end of the fluoridation range, never exceeded the 2 mg/L SMCL. The average quarterly mean for the 8 years reported is 0.85 mg/L and that for the 2002–2005 period is 0.87 mg/L. The corresponding average quarterly 90$^{th}$ percentile values are 1.39 mg/L and 1.43 mg/L, respectively.

Table 3-2 represents only the systems that were at 2 mg/L or higher for at least one quarter during the eight year monitoring period. In parallel with the pattern observed in Table 3-1, Table 3-2 shows that the number of systems that measure a concentration of 2 mg/L or above in a given year is increasing from around 500 in the early years of the ICR time span to above 800 in the last two years. This too reflects an increase in the number of states reporting. The samples from systems that have reported levels ≥ 2 mg/L come from 26 to 46% of the systems in each quarter. This difference between the percent of systems affected and percent of samples can reflect sampling at multiple entry points for the system or the taking of a second sample for confirmation of the original result. It is important to remember when looking at the percent data, that the reporting of a value of ≥2 mg/L does not require a system to begin monitoring on a quarterly basis. The system can maintain their yearly or triennial monitoring schedule, but are required to report the exceedence of the SMCL in their consumer confidence report. Some systems may increase their monitoring for fluoride when the concentration reaches 2 mg/L.

In examining the mean and median of the concentrations reported by the systems that had at least 1 sample with a concentration of 2 mg/L or higher, all of the median values are still within the fluoridation range, while all of the means lie above the fluoridation range but are lower than the SMCL. The ranges for the 90th percentile values are consistently above the SMCL but below the MCL. For the last four years of the ICR monitoring (2001–2005) the average quarterly fluoride concentration was 1.76 mg F/L and the 90th percentile value was 3.84 mg F/L. Over the ICR reporting period from 1.8 million to 6.4 million individuals could have been exposed in a given year to a concentration of 2 mg/L or higher for at least a short period of time. It is not possible to estimate how many of these individuals may have been exposed during a period of vulnerability for severe dental fluorosis.

**Table 3-1. Public Water System Monitoring Data 1998–2005**
**Ranges Across Quarterly Data in Each Year; Nondetect Values Not Included in Samples, Mean, Median and 90th Percentile**

| Year | 1998 | 1999 | 2000 | 2001 | 2002 | 2003 | 2004 | 2005 |
|---|---|---|---|---|---|---|---|---|
| **Samples** | 6,566 - 7,288 | 6,783 - 6,991 | 6,990 - 8,049 | 6,559 - 8,961 | 6,126 - 8,295 | 6,910 - 8,562 | 8,231 - 9,580 | 7,051 - 9,635 |
| **% samples ≥2 mg/L** | 3.2% - 3.6% | 2.8% - 3.0% | 2.7% - 3.3% | 3.1% - 4.5% | 4.0% - 5.1% | 5.2% - 6.2% | 4.9% - 6.4% | 5.4% - 6.8% |
| **Systems** | 3,263 - 3,973 | 3,134 - 3,322 | 3,489 - 3,873 | 3,972 - 4,480 | 3,541 - 4,563 | 4,054 - 4,981 | 5,007 - 5,700 | 3,869 - 5,472 |
| **% systems ≥2 mg/L** | 4.8% - 5.6% | 4.5% - 4.9% | 4.1% - 4.7% | 4.5% - 5.5% | 4.6% - 5.8% | 6.1% - 7.2% | 5.6% - 7.7% | 6.9% - 8.3% |
| **Mean (mg/L)[a]** | 0.81 - 0.85 | 0.83 - 0.85 | 0.82 - 0.86 | 0.81 - 0.86 | 0.78 - 0.89 | 0.86 - 0.93 | 0.80 - 0.90 | 0.84 - 0.95 |
| **Median (mg/L)[a]** | 0.83 - 0.86 | 0.88 - 0.92 | 0.87 - 0.90 | 0.77 - 0.87 | 0.70 - 0.85 | 0.80 - 0.85 | 0.69 - 0.80 | 0.75 - 0.86 |
| **90th percentile (mg/L)[a]** | 1.32 - 1.36 | 1.34 - 1.37 | 1.30 - 1.38 | 1.33 - 1.40 | 1.40 - 1.44 | 1.40 - 1.47 | 1.40 - 1.50 | 1.40 - 1.50 |
| **Population** | 40,455,048 - 52,890,715 | 41,810,370 - 70,262,253 | 43,543,007 - 70,200,938 | 45,062,700 - 82,331,386 | 50,333,719 - 82,609,244 | 44,398,104 - 87,126,153 | 47,726,060 - 86,715,548 | 58,824,170 - 102,533,400 |

**SOURCE:** The monitoring data used in this analysis were collected through information collection request for EPA's second Six-Year Review under the provisions of the Paperwork Reduction Act, 44 U.S.C. 3501 et seq.; Office of Management and Budget (OMB) control number 2040-0275.

[a]Mean, median and 90th percentile based on all detections (modal minimum reporting level (MRL) = 0.1 mg/L).

**Table 3-2. A Summary of Public Water System Fluoride Monitoring Data from Systems for Systems with at Least One Detection of 2 mg/L or Higher during the Year of Monitoring**

**Ranges Across Quarterly Data in Each Year; Nondetect Values Not Included in the Sample, Mean, Median and 90th Percentile**

| Year | 1998 | 1999 | 2000 | 2001 | 2002 | 2003 | 2004 | 2005 |
|---|---|---|---|---|---|---|---|---|
| **Samples from systems that ever had a detection ≥ 2 mg/L** | 1,380 - 1,513 | 1,372 - 1,494 | 1,432 - 1,527 | 1,225 - 1,762 | 1,138 - 1,473 | 1,409 - 1,603 | 1,557 - 1,951 | 1,521 - 1,713 |
| **% samples with at least one detection ≥2 mg/L** | 15.3% - 17.4% | 13.3% - 14.7% | 14.5% - 16.5% | 16.4% - 24.0% | 24.9% - 27.5% | 29.6% - 31.8% | 27.7% - 33.9% | 30.5% - 31.8% |
| **Systems that ever had a detection ≥ 2 mg/L** | 499 - 563 | 528 - 549 | 541 - 586 | 563 - 656 | 579 - 668 | 687 - 763 | 756 - 843 | 754 - 822 |
| **% systems with at least one detection≥2 mg/L** | 32.3% - 36.9% | 26.5% - 29.5% | 27.3% - 32.0% | 31.4% - 36.3% | 32.3% - 35.6% | 40.5% - 44.3% | 42.3% - 48.3% | 42.6% - 45.9% |
| **Mean (mg/L)** | 1.27 - 1.43 | 1.32 - 1.37 | 1.32 - 1.43 | 1.33 - 1.60 | 1.60 - 1.69 | 1.75 - 1.84 | 1.65 - 1.86 | 1.73 - 1.86 |
| **Median (mg/L)** | 1.05 - 1.10 | 1.10 - 1.10 | 1.10 - 1.11 | 1.10 - 1.20 | 1.20 - 1.29 | 1.20 - 1.30 | 1.15 - 1.30 | 1.20 - 1.23 |
| **90th percentile (mg/L)** | 2.40 - 2.65 | 2.20 - 2.40 | 2.21 - 2.46 | 2.60 - 3.10 | 3.10 - 3.40 | 3.80 - 4.39 | 3.70 - 4.18 | 3.90 - 4.24 |
| **Population-served by systems that ever had a detection ≥ 2 mg/L** | 2,513,263 - 3,887,873 | 1,864,149 - 4,703,418 | 2,429,353 - 3,215,929 | 3,088,021 - 4,450,151 | 3,563,761 - 5,402,152 | 3,820,278 - 4,793,365 | 3,849,780 - 5,242,650 | 4,326,194 - 6,405,661 |

**SOURCE:** The monitoring data used in this analysis were collected through information collection request for EPA's second Six-Year Review under the provisions of the Paperwork Reduction Act, 44 U.S.C. 3501 et seq.; Office of Management and Budget (OMB) control number 2040-0275.

[a]Mean, median and 90th percentile based on only detections from systems that ever had a sample detection of 2 mg/L or higher (modal minimum reporting level (MRL) = 0.1 mg/L).

## 3.4. Fluoridation Contributions

The U.S. Public Health Service (CDC, 1995) recommends that fluoride levels in municipal drinking water be maintained in the range of 0.7 to 1.2 mg/L. The exact level is determined by the annual average of maximum daily ambient air temperatures (Table 3-3). The linkage between fluoridation levels and ambient air temperatures was based on the hypothesis that drinking water intake is increased in areas with warmer climates requiring less fluoride in the water to achieve the same average population dose.

**Table 3-3. CDC Recommendations for Optimal Fluoride Concentrations in Public Water Supply Systems**

| Annual Average of Maximum Daily Air Temperatures[a] | Community Water Systems Fluoride Concentration (mg/L) |
|---|---|
| 50.0–53.7°F | 1.2 |
| 53.8–58.3°F | 1.1 |
| 58.4–63.8°F | 1.0 |
| 63.9–70.6°F | 0.9 |
| 70.7–79.2°F | 0.8 |
| 79.3–90.5°F | 0.7 |

**SOURCE:** Adapted from CDC, 1995.
[a]Based on 5 years of temperature data.

In the past, school drinking water fluoridation programs targeted areas where the municipal water was not fluoridated (naturally or intentionally). CDC (2001) updated the school fluoridation recommendation because of the widespread use of fluoride toothpaste and, availability of other fluoride-treatment modalities that can be delivered in the school setting. CDC (2001) recommends that decisions to initiate or continue school fluoridation programs be based on an assessment of present caries risk in the target school(s) and alternative preventive modalities that might be available accompanied by periodic evaluation of program effectiveness.

Several studies have indicated that current drinking water consumption rates may not be as affected by climatic conditions as they once were thought to be, suggesting that the temperature-related guidelines for fluoride concentrations in drinking water may need to be reevaluated (NRC, 2006). Heller et al. (1999) examined drinking water intake estimates documented in the 1994–1996 CSFII and compared these data to information from the 1977–78 Nationwide Food Consumption Survey and found no "obvious strong or consistent association between water intake and month or season."

Using 24-hr recall data from the third National Health and Nutrition Examination Survey (NHANES III, 1988–1994), Sohn et al. (2001), reported that for children aged 1–10 years there was no significant relationship (based on multiple regression analysis) between total fluid intake or plain water intake and mean daily maximum temperature, either before or after controlling for sex, age, socioeconomic status, and race or ethnicity. Fluid intake was significantly associated with age, sex, socioeconomic status, and race and ethnicity. Estimates of total fluoride intake and plain water intake by geographic region are shown in Table 3-4. However, the NHANES survey was designed to avoid interviewing people in extremely hot or cold weather conditions.

This could be a limitation on the applicability of results from this analysis to the entire U. S. population. The mean maximum temperatures used in the analysis (based on the average of daily maximum temperatures during 1960–1990 for the survey month) ranged from 53.4°F to 89.3°F. The majority of temperatures were distributed within the range of 65.0° to 85°F.

**Table 3-4. Estimated Daily Fluid and Plain Water Regional Intake in Children 1–10 Years Old**

| Region | No. | Total Fluid Intake[b] | | Plain Water Intake | |
|---|---|---|---|---|---|
| | | mL/day±SE | mL/kg/day±SE | mL/day±SE | mL/kg/day±SE |
| Northeast | 679 | 1,734.8 ±30.7 | 86.9 ±2.3 | 568.2 ±52.1 | 26.4±2.1 |
| Midwest | 699 | 1,734.4 ±45.3 | 83.7 ±1.5 | 639.7 ± 53.8 | 28.9 ±1.8 |
| South | 869 | 1,739.4 ±31.2 | 83.2 ±2.2 | 612.9 ±24.1 | 27.6 ±1.3 |
| West | 1,622 | 1,737.4 ±24.5[a] | 81.1 ±1.7 | 624.4 ±44.2 | 27.0 ±1.9 |

**SOURCE**: Sohn et al., 2001; NHANES III, 1988–1994.

[a]A value of 734.4 is given in Sohn et al., 2001; however, based on the consumption per unit body weight, it appears that this data point should actually be 1,737.4 mL/day, as shown here.

It should be noted that the CDC recommendations for temperature-dependent optimal fluoride concentrations in municipal drinking water are still in effect (CDC, 1995) but are an issue of current interest as indicated by Heller et al. (1999) and Sohn et al. (2001). NRC (2006) and CDC (2001) have also recommended a reevaluation of the ambient air temperature-based guidelines.

## 3.5. Bottled Water

Fluoride content of bottled water varies considerably with brand, source, and time of packaging. Nowak and Nowak (1989) analyzed the fluoride content of 19 types of bottled water obtained in the Iowa City area using a fluoride ion-specific electrode and found that the F concentration ranged from 0.004 to 0.33 mg/L. Chan et al. (1990) analyzed the fluoride content of twenty-two types of bottled water originating from nine different regions of the US and three regions of France. Eighteen of the samples had fluoride levels below 0.3 mg/L; and the highest fluoride level was 0.79 mg/L. Stannard et al. (1990) tested 24 brands of domestic and imported bottled waters for fluoride using an ion-specific electrode. The fluoride levels ranged from a trace amount (two samples less than 0.1 mg/L) to 1.25 mg/L. The average was calculated to be 0.33 mg/L, assuming the two samples to have 0 mg/L fluoride.

Among 78 commercially available bottled waters sampled in Iowa, Van Winkle et al. (1995) found that fluoride levels ranged from 0.2 mg/L to 1.36 mg/L with a mean of 0.18 mg/L; 83% ranged from 0.02 to 0.16 mg/L, 7% from 0.34 to 0.56 mg/L, 1% had a fluoride level of 0.88 mg/L and 9% had levels >1 mg/L. Van Winkle et al. (1995) reported that 340 of 1308 homes (26%) used bottled water.

Allen et al. (1989) analyzed the chemical composition of 37 brands of imported and domestic bottled mineral water. Fluoride was analyzed with an ion-selective electrode. Fluoride concentrations ranged from <0.01 mg/L to 7.9 mg/L. In an earlier study MacFadyen et al. (1982) reported fluoride levels of <0.1 mg/L to 5.8 mg/L in 26 bottled spring waters.

The National Fluoride Database (USDA, 2005) includes data on the concentrations of fluoride in several brands of bottled water. Samples were collected in up to 144 locations across the country, depending on the level of contribution to fluoride intake as previously determined by the USDA. Differences in geographical location were incorporated into the sampling strategy. Fifteen brands and one to 20 samples per brand were assayed using a fluoride ion-specific electrode. The range of mean values for various types of bottled water was 0.02-0.78 mg/L. The one brand containing fluoride at a level within the fluoridation range was a product intended to supply fluoride. The mean concentration for most of the remaining samples tended to be below 0.2 mg/L F. According to U.S. EPA (2004), bottled water accounts for 3 mL/kg/day of total ingested water from all sources (equal to 210 mL/day for a 70 kg adult), or about 18 % of mean adult total water intake.

The fluoride concentrations in bottled water products vary substantially. Some products can contain fluoride at levels that exceeded the levels recommended for fluoridation; a few mineral or spring waters exceeded the MCL for fluoride.

## 3.6. Exposure from Drinking Water

Estimated exposures from public drinking water sources have been calculated using the average and 90$^{th}$ percentile age-related water consumption estimates derived from U.S. EPA (2004), and the average national concentration of fluoride reported in the ICR monitoring data (Section 3.3). The average water concentration used for this calculation, 0.87 mg/L, is the average of the averages from the data submitted to EPA for the 16 monitoring quarters from 2002 through 2005. The data used to determine the average concentration are reported in Table 3-1.

Mean water consumption (direct and indirect) and mean fluoride intake for all individuals (consumers and nonconsumers) for specific age groups and the entire population, using the average fluoride concentration of 0.87 mg/L, are shown in Table 3-5.

**Table 3-5. Fluoride Intake from Consumption of Municipal Water (Direct and Indirect[a]) at the Average Concentration ( 0.87 mg/L) Determined from Monitoring Records for 2002 through 2005**

| Group | Water Consumption (mL/day)[b] | | Fluoride Intake (mg/day)[b] | |
|---|---|---|---|---|
| | Mean | 90 % C.I. Upper bound | Mean | 90% C.I. Upper bound |
| Infants <0.5 yr | 296 | 329 | 0.26 | 0.29 |
| 0.5–0.9 | 360 | 392 | 0.31 | 0.34 |
| 1–3 yrs | 311 | 324 | 0.27 | 0.28 |
| 4–6 yrs | 406 | 426 | 0.35 | 0.37 |
| 7–10 yrs | 453 | 485 | 0.39 | 0.42 |
| 11-14 yrs | 594 | 642 | 0.52 | 0.56 |
| 15-19 | 761 | 823 | 0.66 | 0.72 |
| 20+ | 1,098 | 1127 | 0.96 | 0.98 |
| **Total Pop.** | **926** | **949** | **0.81** | **0.83** |

**SOURCE:** Adapted from U.S. EPA, 2004. Table 5.1.A1.
[a]Indirect consumption refers to intake through beverages and foods that include fluoridated drinking water as an ingredient.
[b]Based on an average fluoride concentration of 0.87 mg/L.

U.S. EPA (2004) reported that during a 2-day survey period for the CSFII survey, it was determined that 5% of the individuals older than 1 year and 25% of infants younger than 1 yr did not drink community water. If these individuals are excluded from the average intake calculations, then the average amounts of municipal water consumed increase as do the fluoride exposures. U.S. EPA (2004) calculated water consumption levels for the group "consumers only" in order to adjust for those that did not report drinking water intake during the two days of dietary data reported. These data are most important for infants who consume formula reconstituted using tap water on a daily basis but whose formula intake is not recognized as a source of tap water in the survey records. Estimated fluoride exposure at the mean fluoride concentration (0.87 mg/day) and the consumer-only mean and 90$^{th}$ percentile intakes for the six month to < 1 year age group are 0.41 mg/day and 0.84 mg/day (water intake = 971 mL) , respectively (see Table 3-6). For the 1 to < 3 year old group they are 0.30 mg/day and 0.63 mg/day (water intake = 723 mL), respectively.

For comparison with the estimates in Table 3-5, Table 3-7 presents the estimated average fluoride exposures for all individuals (consumers and nonconsumers) with average drinking water consumptions of direct and indirect water who consume water that is at the 90$^{th}$ percentile fluoride concentration (1.43 mg/L) for a sustained period of time. The 90$^{th}$ percentile concentration used for this analysis is the average of the 90$^{th}$ percentile values for the 16 quarters reported to EPA between 2002 and 2005. Average consumers of drinking water from public systems representative of the 90$^{th}$ percentile fluoride concentration have higher daily intakes of fluoride from drinking water than those with 90$^{th}$ percentile intakes of drinking water at an average fluoride concentration. However, only ten percent of the population will have water at or greater than the 90$^{th}$ percentile concentration.

**Table 3-6. Consumers Only Fluoride Intake from Consumption of Municipal Water (Direct and Indirect[a]) at the Average Concentration (0.87 mg/L) Determined from Monitoring Records for 2002 through 2005**

| Group | Water Consumption[b] (mL/day) | | Fluoride Intake[b] (mg/day) | |
|---|---|---|---|---|
| | Mean | 90% Percentile | Mean | 90% Percentile |
| Infants <0.5 yr | 548 | 985 | 0.48 | 0.86 |
| 0.5–0.9 | 467 | 971 | 0.41 | 0.84 |
| 1–3 yrs | 349 | 723 | 0.30 | 0.63 |
| 4–6 yrs | 442 | 943 | 0.38 | 0.82 |
| 7–10 yrs | 487 | 993 | 0.42 | 0.86 |
| 11-14 yrs | 641 | 1415 | 0.56 | 1.23 |
| 15-19 | 817 | 1671 | 0.71 | 1.45 |
| 20+ | 1176 | 2284 | 1.02 | 1.99 |
| **Total Pop.** | **1000** | **2069** | **0.87** | **1.80** |

**SOURCE:** Adapted from U.S. EPA, 2004. Table 5.2.A1.

[a]Indirect consumption refers to intake through beverages and foods that include fluoridated drinking water as an ingredient.
[b]Based on an average fluoride concentration of 0.87 mg/L .

| Table 3-7. Fluoride Intake From Average Drinking Water Consumption and 90th Percentile Fluoride Concentration (1.43 mg/L) Determined from Monitoring Records for 2002 through 2005 | | |
|---|---|---|
| **Group** | **Water Consumption** | **Fluoride Intake** |
| | **Average Total mL** | **mg/day total** |
| Infants <0.5 yr | 296 | 0.42 |
| 0.5–0.9 | 360 | 0.51 |
| 1–3yrs | 311 | 0.44 |
| 4–6yrs | 406 | 0.58 |
| 7–10 yrs | 453 | 0.65 |
| 11-14 yrs | 594 | 0.85 |
| 15-19 | 761 | 1.09 |
| 20+ | 1098 | 1.57 |
| **Total Pop.** | **926** | **1.32** |

**SOURCE:** Adapted from U.S. EPA, 2004, Table 5.1.A1.

As noted by NRC (2006), fluoride exposures from drinking water depend on individual water intakes, fluoride concentration in the water, and whether water purification or filtration systems are used to remove fluoride. Some individuals may have substantially higher intakes of fluoride from their drinking water as a result of specific types of activities that increase water intake (e.g., athletes or outdoor laborers in warm climates), life stage (e.g., pregnant or lactating women), or as a result of medical conditions such as diabetes mellitus, diabetes insipidus, or renal problems.

## 4. Fluoride in Dental Products

### 4.1. Toothpaste

According to Newbrun (1992), more than 95% of all toothpaste sold in the United States contains fluoride. Results of the 1983 National Health Interview Survey showed that 67.8% of children younger than 5 years old used fluoridated tooth paste and 95.5% of those 5–9 years old (Ismail et al., 1987). As many as 15% to 20% of children in some age groups studies by Wagener et al., (1992) used fluoride supplements or mouth rinses.

The total daily amount of fluoride ingested and systemically absorbed following tooth brushing with a fluoride toothpaste will vary with: 1) the concentration of fluoride in the toothpaste, 2) the amount of toothpaste used; 3) the frequency of brushing; 4) the amount of rinsing; 5) the swallowing control of the individual; and 6) the time of brushing relative to the time the last meal was eaten. Most toothpaste sold in North America contains fluoride ion at a concentration of 1000–1100 ppm (Levy, 1993). Toothpastes with lower concentrations of fluoride (250–500 ppm) are sold specifically for use by children (Newbrun, 1992) in other countries but are not generally available in the United States. Some products without added fluoride are available in the United States,

Fluoridated toothpastes (gel or paste products) in the United States are required to include guidance to users on their product label (USFDA, 2009). Children under the age of 6 are to be instructed in "good brushing and rinsing habits to minimize swallowing" and supervised "as necessary until capable of using without supervision". It is recommended that a dentist or pediatrician be consulted about toothpaste use for children under 2. The label should identify toothpaste as a product intended for adults and children 2-years of age and older. Brushing is recommended after every meal or twice per day. In a study discussed later, Levy et al. (1997) found that 31.7% of parents surveyed reported use of fluoridated toothpaste by their children by the time they were one year old, suggesting that many individuals do not follow the label guidance.

The amount of toothpaste used per brushing, the frequency of brushing and the amount of rinsing are expected to be highly variable factors which can substantially impact the amount of toothpaste ingested. In studies conducted in Europe, Cochran et al. (2004) and O'Mullane et al. (2004) found that 60% of 1.5–2.5 year-olds swallowed between 70% and 100% of the toothpaste placed on the brush. Borysewicz-Lewicka et al., (2007) reported that children swallowed on average 17% of the fluoride used in brushing with a gel containing 1.25% fluoride. Baxter (1980) reported that children 5-6 years of age ingested an average of about 0.27 g per brushing; older children ingested less.

Levy (1993) noted that a full strip of toothpaste covering a child's size toothbrush is 0.75 to 1.0 g which could result in a fluoride intake as high as 1 mg per brushing. Based on the literature available at the time, Levy (1993, 1994) estimated that children 2–3 years old would ingest about 0.3 g per brushing, equivalent to 59-65% of the amount used. At one time a complete ribbon of toothpaste across the surface of the toothbrush was recommended. However, more recent guidelines stress the application of a pea-sized portion. Levy et al. (1992) found that children using flavored toothpastes marketed specifically for children used higher amounts of toothpaste than those using regular toothpaste.

Levy (1993) also reported that 49% of 59 children aged 1–4 years did not rinse or expectorate when brushing and an additional 27% rinsed but ingested almost all of the rinse water. Only 5% of the children under the age of 2.5 years spit after brushing. In reviewing the available literature, Levy (1993, 1994) noted that children who did not rinse after tooth brushing ingested 75% more toothpaste than those who rinsed. Swallowing control is especially weak in younger children, and Levy et al. (2001) note that several studies have shown that younger children may ingest more than half of the toothpaste used per brushing. In studies on young adults, Sjögren and Melin (2001) found that oral retention of fluoride following brushing can be substantially reduced by more than 50% by increased rinsing.

Following ingestion, fluoride absorption in the GI tract has been found to be close to 100% (Ekstrand and Ehrnebo, 1980); however, the total amount absorbed can be affected by the presence of certain foods in the stomach. Ekstrand and Ehrnebo (1979) reported that the absorption of fluoride from sodium fluoride tablets was reduced to 50–79% when co-administered with milk products. Cury et al. (2005) conducted a double-blind crossover study on eleven volunteers (six women and five men aged 17–20 yrs) who ingested toothpastes with fluoride concentrations of 0, 550 or 1100 μg F/g. The toothpastes were administered as a slurry (45 mg/kg body weight) while fasting or 15 min after a meal (breakfast or lunch). Fluoride levels were measured in unstimulated whole saliva for up to 3 hours post-exposure and in urine 24 hour pre-exposure and 24 hr post-exposure using an ion-selective electrode. Bioavailability was 61% and 71% after lunch and breakfast, respectively, compared to an assumed 100% after fasting for a toothpaste containing 1100 μg F/g, and 78% and 65%, respectively, for a toothpaste with 550 μg F/g.

Osuji et al. (1988) conducted a case-control study of children 8–10 years old (34 children with fluorosis and 34 controls) living in East York, Ontario, to determine the risk factors for dental fluorosis. Factors evaluated included: prematurity, low birth weight, breastfeeding, use of fluoride mouth rinses or supplements, residence history, medical and dental history (including history of tooth brushing), and consumption of formula, tea, fish, soft drinks, milk, water, and reconstituted juices. The only factors showing a significant association with fluorosis were ingestion of infant formula and early use of fluoride tooth paste. Children who brushed their teeth before age 25 months had 11 times the odds of developing fluorosis as those who began tooth brushing at a later age. Prolonged use of infant formulas (≥13 months) was associated with 3.5 times the risk of fluorosis compared with no or shorter duration of formula use. The odds ratio for developing fluorosis was 7.1 (95% C.L. = 1.14–44.45) for children with prolonged formula use, 13.8 (95% C.L. = 5.12–37.38) for children who had started brushing early, and 37.9 (95% C.L. = 10.60–134.52) for children who were in both groups.

Simard et al. (1989) evaluated tooth brushing habits, toothpaste use and its ingestion in a group of Canadian children 2 to 5 years old. All but one of the children used a fluoridated toothpaste. The majority (71.4%) brushed twice daily, 23.8% brushed three times daily, and 4.8% brushed only once daily. The study was conducted at a day care center where the children brushed with a toothpaste containing 0.24% NaF (1100 ppm F). Brushing habits at home were determined by a questionnaire filled out by the parents. The quantity of toothpaste used and ingested and the estimated amount of fluoride ingested are shown in Table 4-1. For all age groups combined the amount of fluoride ingested was 0.329 mg per brushing.

| Table 4-1. Toothpaste Use and Estimated Fluoride Ingestion by Children 2-5 Years Old | | | | | | | |
|---|---|---|---|---|---|---|---|
| Age (yr) | No of Subjects | Toothpaste Used Per Brushing (g) | | Toothpaste Ingested Per Brushing (g) | | Estimated Fluoride Ingested Per brushing (mg) | |
| | | Mean | SD | Mean | SD | Mean | SD |
| 2–3 | 5 | 0.464 | ±0.19 | 0.278 | ±0.13 | 0.304 | ±0.15 |
| 4 | 9 | 0.783 | ±0.28 | 0.390 | ±0.25 | 0.429 | ±0.27 |
| 5 | 9 | 0.651 | ±0.34 | 0.221 | ±0.12 | 0.243 | ±0.13 |
| All | 23 | 0.662 | ±0.30 | 0.299 | ±0.19 | 0.329 | ±0.20 |

**SOURCE:** Simard et al., 1989.

Fluoride retention following tooth brushing in nineteen 3–10-year-old children was evaluated by Salama et al. (1989). Each child brushed with 1.8 g of toothpaste (1043 ppm F as MFP). Fluoride recovered on the toothbrush and in expectorant was analyzed with a fluoride ion-specific electrode after HMDS diffusion. The average quantity of fluoride not recovered was 0.36 ± 0.05 mg (range 0.08 to 0.82 mg). The study authors concluded that fluoride intake from a single tooth brushing exceeds dietary intake in non-fluoridated areas and is equivalent to about 75% of dietary intake in fluoridated areas.

A pilot study was conducted to determine the tooth brushing habits of children 12–24 months old and used to estimate the quantity of fluoride that children in this age group would ingest during brushing (Simard et al., 1991). The study was conducted in the Quebec City region and involved 15 children. The authors used information from their earlier study (Simard et al., 1989) which indicated that children 2–3 years of age ingested about 60% of the toothpaste used to estimate fluoride exposures. A survey of the parents indicated that 60% of the children had their teeth cleaned once a day, 32% twice a day and 8% three or four times per day. The average amount of toothpaste used was 0.160 g. The assumption was made that the mean NaF concentration in the toothpaste was 0.243%. The amount of fluoride ingested was calculated by taking 60% of the quantity of toothpaste used per brushing per day multiplied by a conversion factor of 1.09 (to convert from NaF to mg F/g of toothpaste) multiplied by the number of times the child brushed each day. The estimated amount of fluoride ingested per day ranged from 0.02 to 0.33 mg (N=8) for those whose teeth were cleaned once, and from 0.05 to 0.55 mg (N=6) for those whose teeth were cleaned twice per day. The amount ingested by the one child who brushed three times per day was 0.07 mg. Simard et al. (1991) reported that 20% of the children ingested more than 0.25 mg of fluoride per day. The average amount of fluoride ingested by all 15 children was 0.15 mg/day.

Levy et al. (1995) summarized the results of studies conducted up to 1993 which evaluated the amounts of toothpaste ingested during tooth brushing for various age groups. Toothpaste ingestion per brushing for children 1–9 years old ranged from 0.11 to 0.39 g, with 90$^{th}$ percentile levels ranging from 0.08 to 0.73 g. Assuming 1.1 mg F/g toothpaste, this amount of toothpaste ingestion would result in a consumption of 0.12–0.43 mg F (90$^{th}$ percentile range of 0.09 to 0.8

mg F). Levy et al. (1995) estimated a mean fluoride intake from toothpaste of 0.01 mg (range 0–0.04 mg) for infants 6 months old, 0.07 mg (range 0.03–0.66 mg) for children 12 months old, and 0.25 mg (range 0.01–1.50 mg) for children 2 and 3 years old.

In a later study Levy et al. (1997) surveyed by questionnaire the parents of children born in eastern Iowa on the tooth brushing practices of their children up to 1 year of age (Table 4-2). If it is assumed that about 62.45% of the toothpaste reported as used in the 1997 paper is ingested, then the estimated amount of fluoride ingested is 0.13 mg for 6-mo-olds, 0.12 mg for 9-mo-olds; and 0.12 mg for 12-mo-olds. The estimate for ingestion comes from a Levy et al. (2000) study of 3-4 year old subjects. The percent of children who were reported as having their teeth brushed increased from 12.9% at six months to 64.5% at one year. The percent of parents that reported using fluoride-containing toothpaste increased from 1.9% at six months to 31.7 % at one year.

**Table 4-2. Toothpaste Use by Children 6 to 12 Months Old**

| Parameter | Age Groups | | |
|---|---|---|---|
| | 6 Months | 9 Months | `12 Months |
| Number of children | 899 | 665 | 508 |
| Percentage with erupted teeth | 34.6% | 83.6% | 98.0% |
| Percentage whose teeth were brushed | 12.9% | 36.7% | 64.5% |
| Percentage using fluoridated toothpaste | 1.9% | 11.7% | 31.7% |
| Mean amount of fluoride used per brushing | 0.11 mg (0.02–0.05)[a] | 0.14 mg (0.02–0.88)[a] | 0.17 (0.02–0.88)[a] |
| Mean amount of fluoride used per day | 0.21 mg (0.02–1.50)[a] | 0.20 (0.01–1.75)[a] | 0.19 (0.01–1.75)[a] |
| **Frequency of cleaning/brushing** | | | |
| Less than once per day | 31.4% | 33.2% | 37.0% |
| Once per day | 41.2% | 45.5% | 44.8% |
| Twice per day | 16.9% | 17.0% | 14.7% |
| Three times per day | 6.3% | 3.1% | 3.5% |
| More than three times per day | 6.3% | 1.1% | – |

**SOURCE:** Levy et al., 1997.

[a]Range.

Levy et al. (2000) further evaluated the tooth brushing habits of 28 U.S. preschoolers (3–4 years old; mean age 44 months). The average amount of toothpaste applied to the toothbrush was 0.256 g (range 0.035–0.620 g, SD = 0.177 g). The estimated mean amount of ingested fluoride was determined by subtracting the estimated amount expectorated from the amount of toothpaste applied to the brush. Fluoride was determined with a fluoride ion-specific electrode after diffusion using a modified Taves microdiffusion method. The mean amount of fluoride ingested was 0.17 mg per brushing (SD 0.15 mg; range 0.00–0.52 mg), equivalent to 62.45% of the initial amount in the toothpaste.

Only a few studies have given 90$^{th}$ and 95$^{th}$ percentile estimates for toothpaste and/or fluoride ingestion. Barnhart et al. (1974) measured toothpaste use and ingestion in four age groups; 2–4 yr olds (N=68), 5–7 yr olds (N=4); 11–13 yr olds (N=98); and 20–35 yr olds (N=70) under simulated home-use conditions. Chronic usage conditions were simulated with a statistical model to obtain realistic estimates of the 90th and 95th percentile ingestion. The mean amount of toothpaste used per brushing was 0.86 g for the 2–4 yr olds, 0.94 g for the 5–7 yr olds and 1.10 g for the 11–13 yr olds. Ingestion rates among the four groups are summarized in Table 4-3. Assuming 1000 ppm F in the toothpaste, these toothpaste ingestion rates would correspond to mean fluoride ingestion rates of 0.3 mg for the 2–4 yr olds, 0.13 mg for the 5–7 yr olds and 0.07 mg for the 11–13 yr olds.

**Table 4-3. Age-Related Estimates of Fluoride Ingestion from Toothpaste Use**

| Age (yr) | No. of Subjects | Toothpaste Used Per Brushing (grams)[a] | Toothpaste Ingestion (g) | | | Estimated Fluoride Ingestion (mg)[b] | | |
|---|---|---|---|---|---|---|---|---|
| | | | Mean | 90th Percentile | 95th Percentile | Mean | 90th Percentile | 95th Percentile |
| 2–4 | 62 | 0.86 | 0.30 | 0.73 | 0.82 | 0.3 | 0.73 | 0.82 |
| 5–7 | 56 | 0.94 | 0.13 | 0.27 | 0.44 | 0.13 | 0.27 | 0.44 |
| 11–13 | 73 | 1.10 | 0.07 | 012 | 0.21 | 0.07 | 0.12 | 0.21 |
| 20–35 | 60 | 1.39 | 0.04 | 0.12 | 0.13 | 0.04 | 0.12 | 0.13 |

**SOURCE:** Barnhart et al., 1974.

[a]Mean value.

[b]Assumes 1000 ppm F in toothpaste.

Environment Canada/Health Canada (1993) estimated the daily intake of fluoride from toothpaste (products for home use) for different age groups. Based on a mean inorganic fluoride concentration of 1000 ppm in most toothpaste products (Beltran and Szpunar, 1988; Whitford, 1987), and an estimated toothpaste intake of 0.26-0.78 g/day for children 7 months to 4 years of age, 0.22–0.54 g/day for children 5 to 11 years of age, 0.14 g/day for adolescents 12-19 years of age, and 0.08 g/day for adults 20+ years of age (Levy, 1993), and assuming an average of two brushing per day, the fluoride intakes for these age groups was estimated to be 0.02–0.06 mg/kg bw/day, 0.008–0.02, 0.00246, and 0.00114 mg/kg bw/day, respectively. Using an average body weight of 57 kg for the 12-19 yr-olds and 70 kg for the adults, the daily fluoride intakes for these two age groups can be calculated as 0.14 mg/day and 0.0798 mg/day, respectively.

The most important factor determining the quantity of fluoride ingested by children during tooth-brushing was the amount of toothpaste used according to a study of 405 children, ages 2–7 yr, enrolled in Quebec City schools (Naccache et al., 1992). The estimated amount of toothpaste used per brushing was determined by the difference between the amount used and the amount recovered from the toothbrush and rinse water. Fluoride was analyzed with an ion-specific electrode. The toothpaste contained 0.24% NaF. The amount of toothpaste used, the age of the children and the amount of rinsing were analyzed by multiple regression analysis. The amount of toothpaste used and the amount of fluoride ingested are shown in Table 4-4. On average, the amount of toothpaste used was 0.5 g per brushing. The mean amount of fluoride ingested was 0.229 mg per brushing. The amount ingested decreased with increasing age.

| Table 4-4. Toothpaste Use and Fluoride Ingestion in Children Two to Seven Years Old | | | | | |
|---|---|---|---|---|---|
| Age (yr) | No of Subjects | Toothpaste Used (grams)[a] | | Estimated Fluoride Ingested (mg per brushing) | |
| | | Mean | SD | Mean | SD |
| 2 | 36 | 0.618 | 0.976 | 0.358 | 0.363 |
| 3 | 56 | 0.529 | 0.424 | 0.280 | 0.218 |
| 4 | 81 | 0.446 | 0.269 | 0.241 | 0.184 |
| 5 | 77 | 0.516 | 0.366 | 0.227 | 0.174 |
| 6 | 78 | 0.484 | 0.254 | 0.180 | 0.127 |
| 7 | 77 | 0.497 | 0.401 | 0.175 | 0.194 |
| **Total** | **405** | **0.503** | **0.401** | **0.229** | **0.195** |

**SOURCE:** Naccache et al., 1992.

Rojas-Sanchez et al. (1999) estimated fluoride intake from toothpaste in groups of children, aged 16–40 months, from three communities; San Juan, Puerto Rico (n=11), Connersville, IN (n=14) and Indianapolis, IN (n=29). Intake was determined by subtracting the amount of toothpaste expelled and the amount left on the toothbrush from the amount initially placed on the toothbrush. The concentration of fluoride in the toothpaste was 0.10–0.11% (theoretical). Samples were analyzed for fluoride using the hexamethyldisiloxane microdiffusion method of Taves (1968b) as modified by Dunipace et al. (1995). Frequency of brushing equal to or greater than two times per day was 91% (n=11) in San Juan; 67% (n = 14) in Connersville; and 46% (n = 29) in Indianapolis. The mean amount (± SEM) of fluoride ingested in toothpaste each day was estimated to be 548 ±62 µg in San Juan, 576 ±86 µg in Connersville, and 424 ±73 µg in Indianapolis.

The patterns of fluoride ingestion from toothpaste use in children from shortly after birth (1.5 months) to an age of 36 months were reported by Levy et al. (2001). Information was obtained from questionnaires as part of the longitudinal Iowa Fluoride Survey. Estimates of the amount of toothpaste used were based on the parents selecting from pictures depicting children's toothbrushes with different quantities of toothpaste on them, and the amount ingested were based on estimates made by the parents. Estimates of the fluoride ingested were based on the manufacturers indication of the fluoride content of the toothpaste used (in most cases 1000-1100 ppm). Results are shown in Table 4-5.

Using the same methodology as that for children 0–36 months old (Levy et al., 2001, see above), Levy et al. (2003a) calculated fluoride ingestion from toothpaste use in children aged 36 to 72 months old. Results of the survey by fluoride source were presented by Levy et al. (2003a) in graphical form. As estimated from the graphical data, mean fluoride intake from toothpaste was about 0.28 mg/day at 36 months, 0.27 mg/day at 48 months, 0.20 mg/day at 60 months, and 0.17 mg/day at 72 months. Estimates of 90$^{th}$ percentile intakes from toothpaste ingestion for these same age groups were 0.76, 0.76, 0.50, and 0.50 mg/day, respectively.

| Table 4-5. Estimated Fluoride Intake from Toothpaste[a] in Children 1.5 to 36 Months Old | | |
|---|---|---|
| Age (months) | Intake (mg) | |
| | Mean (SD) | 90th Percentile |
| 1.5 | 0.000 | 0.000 |
| 3.0 | 0.000 | 0.000 |
| 6.0 | 0.002 (0.041) | 0.000 |
| 9.0 | 0.013 (0.081) | 0.000 |
| 12.0 | 0.038 (0.136) | 0.109 |
| 16.0 | 0.102 (0.207) | 0.250 |
| 20.0 | 0.191 (0.270) | 0.500 |
| 24.0 | 0.257 (0.312) | 0.656 |
| 28.0 | 0.267 (0.305) | 0.750 |
| 32.0 | 0.290 (0.315) | 0.750 |
| 36.0 | 0.278 (0.292) | 0.750 |

**SOURCE:** Levy et al., 2001.

[a]Portion of toothpaste ingested estimated from parent's report.

Participants in the Iowa Fluoride Study were evaluated to determine the effect of fluoride toothpaste ingestion on the occurrence of dental fluorosis (Franzman et al., 2006). The study utilized information derived from questionnaires filled out by the participants' parents concerning fluoride exposures and toothbrushing at ages 16, 24, and 36 months. The results of the survey on toothpaste use are shown in Table 4-6. The estimated percent of individuals ingesting 75% or more of the toothpaste was 82% at age 16 months, 85% at age 24 months, and 66% at age 36 months.

| Table 4-6. Toothpaste Use and Ingestion by Children Ages 16 to 36 Months | | | |
|---|---|---|---|
| Parameter | Percentage of Children (n = 343) | | |
| | 16 Months Old | 24 Months Old | 36 Months Old |
| Individuals who brush teeth | 90 | 100 | 100 |
| Use of fluoridated toothpaste | 65 | 90 | 96 |
| Brush teeth less than once/day | 35 | 25 | 18 |
| Brush teeth once/day | 48 | 51 | 57 |
| Brush teeth twice/day | 14 | 23 | 24 |
| Brush teeth more than twice /day | 4 | 2 | 1 |
| ≤25% toothpaste swallowed | 13 | 7 | 21 |
| 50% toothpaste swallowed | 5 | 9 | 13 |
| ≥75 toothpaste swallowed | 82 | 85 | 66 |

**SOURCE:** Franzman et al., 2006.

In an earlier study (Franzman et al., 2004) estimated that 51–59% of children 9–32 months old ingested 0.125–0.25 g of toothpaste per brushing, declining to 28% at 60 months. 12% ingested 0.5–0.75 g at 9 months, increasing to 64% at 60 months. The percentage using 0.875–1.0 g per brushing was <3% up to 28 months, 3–5% at 32–54 months and 7% at 60 months. Using the information from Franzman et al. (2004), Franzman et al. (2006) estimated the amount of fluoride ingested (per kg body weight) by children who were showing definitive signs of fluorosis on the incisors and those not showing signs of fluorosis. Results are presented in Table 4-7. Average body weights for each age group were not reported. For all but the 16-month children the fluoride ingestion per unit of body weight was higher for the children with dental fluorosis than those without.

**Table 4-7. Fluoride Ingestion from Toothpaste Use and Fluorosis**

| Age | Fluorosis Absent | | Fluorosis Present[a] | | P Value[b] |
|---|---|---|---|---|---|
| | Number | Median Daily Fluoride Ingestion (mg/kg bw) | Number | Median Daily Fluoride Ingestion (mg/kg bw) | |
| 16 mo | 220 | 0.002 | 89 | 0.002 | 0.61 |
| 24 mo | 220 | 0.010 | 89 | 0.017 | 0.02 |
| 36 mo | 220 | 0.012 | 89 | 0.016 | 0.02 |
| 16–36 AUC[c] | 220 | 0.011 | 89 | 0.013 | 0.02 |

**SOURCE:** Franzman et al., 2006.

[a]Two or more incisors with definitive fluorosis (fluorosis risk index of 2).
[b]Based on Wilcoxon rank sum test.
[c]AUC = Area under the curve, a measure of cumulative exposure.

Bohaty et al. (1989) evaluated topical and systemic fluoride supplement use and the prevalence of dental fluorosis in 300 children, aged 6–13 from 6 elementary schools, living in areas with optimal water fluoridation (location of the study sites and the fluoride level in the drinking water were not reported). Fluorosis was scored using Dean's system (subjects with fluorosis were considered those with a Dean score of 0.5 or higher). The data were categorized according to fluoride use, residential history, age, sex and geographic location. Differences in frequency of the categorized data were evaluated statistically with Chi-square analysis where the differences were considered significant at $p < 0.05$. There were no differences between tooth brushing frequency and fluorosis scores in any group.

## 4.2. Topical Applications and Mouth Rinses

Several studies have evaluated use of topical fluoride products and mouth-rinses. According to Levy and Zarei-M (1991), the Dental Care Supplement of the 1983 National Health Information survey found that 5% of children under age 5 yr and 17% of children 5–17 years old reportedly were using fluoride mouth-rinses.

Data from the 1986–87 National Institute of Dental Research U.S. Children's Survey revealed that 54% of children 5–17 years old without access to fluoridated drinking water received topical fluoride treatments at a dentist's office and 22% had received topical fluoride treatments through school-based programs. From data complied in the Iowa Fluoride Study, Levy et al. (2003b) found that only 6% of children surveyed had a fluoride treatment by age 3, 27% by age 4, 44% by age 5 and 66% by age 6 (Table 4-8). Children with dental caries were more likely to have had such a treatment.

**Table 4-8. Percentage of Children Receiving Fluoride Treatments by Age Groups**

| Age (yr) | Number and/or time of surveys | Number of respondents | Reported fluoride treatments (%) | Mean (±SD) number of survey periods with fluoride treatments |
|---|---|---|---|---|
| ≤1 yr | 6, 9, 12 mo | 719 | 0 | 0 |
| 1–2 | 16, 20, 24 mo | 504 | <1% | 0.01 ±0.10 |
| 2–3 | 28, 32, 36 mo | 434 | 6% | 0.07±0.30 |
| 3–4 | 40, 44 48 or 42 and 48 mo | 404 | 28% | 0.41±0.74 |
| 4–5 | 52, 56, 60 mo or 54 and 60 mo | 432 | 46% | 0.74±0.90 |
| 5–6 | 66 and 72 mo | 490 | 58% | 0.93±0.87 |
| | | | | |
| 1–3 | 9 | 347 | 6% | 0.07 ±0.31 |
| 1–4 | 11–12 | 265 | 27% | 0.43 ±0.82 |
| 1–5 | 13–15 | 207 | 44% | 1.09 ±1.52 |
| 1–6 | 15–17 | 187 | 66% | 1.96 ±2.10 |

**SOURCE:** Levy et al. 2003b.

Levy and Zarei-M (1991) reviewed several earlier studies (Ekstrand and Koch, 1980; Ekstrand et al., 1981; Le Compte and Doyle, 1982; Le Compte and Rubenstein, 1984; Larsen et al., 1985; and Wei and Hattab, 1989) which indicated that topical applications of fluoride gel in a professional setting can lead to ingestion of 1.3–31.2 mg fluoride. They also noted that substantial ingestion of fluoride could occur in the home from the use of fluoride mouth-rinses and self-applied topical fluoride gels based on data reported by Ericsson and Forsman, (1969), Wei and Kanellis (1983) and Bell et al. (1985).

Heath et al. (2001) evaluated fluoride salivary retention and ingestion in young adults after application of topical gels using commercial or custom trays, toothbrushes or spatulas. The gels contained 0.62 mg fluoride (toothpaste) to 62.5 mg fluoride (1.23% gel applied with a commercial tray) and the amount ingested ranged from 0.3 to 6.1 mg of fluoride (5-29% of total applied). An additional 0.1–3.5 mg fluoride was retained in the saliva and presumed to have been swallowed.

Eklund et al. (2000) evaluated insurance claims for 15,190 children for treatment provided by 1,556 dentists and determined that the mean number of annual topical fluoride treatments per child was 1.18 (range 0.0–3.22). The age of the patients ranged from 4 to 14 years. The NRC

(2006) concluded that intakes from topical fluorides during professional treatment were unlikely to be significant contributors to chronic fluoride exposures because they are used only a few times per year.

### 4.3. Summary of Fluoride Exposure from Dental Products

Table 4-9 is a summary of studies that examined exposure to fluoride from toothpaste. With few exceptions all of these studies were published in the early to mid-1990s and are likely to not reflect changes in guidance on the amounts of toothpaste recommended for brushing (a pea-sized portion rather than a ribbon). Accordingly, they may overestimate current fluoride intakes from toothpaste. The data provided in Table 4-9 come only from studies that measured ingested fluoride by comparing the amount placed on the toothbrush to that left on the toothbrush and expectorated. Many of other studies reported estimates of ingestion based on questionnaires from parent reporting on toothpaste use. Data on ingestion estimates are not included in Table 4-9.

Use of fluoridated mouth washes on a daily basis in the home setting is likely to increase the daily dose of fluoride from dental products. Unfortunately no primary data on exposures from mouthwashes were identified. In 1983 less than 20% of children in the 6 months to 14 year age range of concern used mouthwashes. However, these data may very well not reflect current use patterns.

Fluoride is released from a number of dental devices, including composite resins, resin-based cements, resin-bonding agents, orthodontic bracket adhesives, pit and fissure sealants, glass ionomer cements, and cavity varnishes. However, the exposure dose is probably small (HHS, 2010).

**Table 4-9. Age-Related Exposure Estimates for Fluoride From Toothpaste**

| Age (yr) | Fluoride Intake[a] (mg/day) | Notes |
|---|---|---|
| 0.5 <1 | 0.01 | Levy et al., 1995 – mean; 6 month olds |
| | 0.07 | Levy et al., 1995 – mean; 12 month olds |
| 1<4 | 0.358 ±0.363 | Nacchache et al., 1992 – 2 year olds |
| | 0.280 ±0.218 | Nacchache et al., 1992 – 3 year olds |
| | 0.25 | Levy et al., 1995; 2-3 year olds |
| | 0.424, 0.576 | Rojas-Sanchez et al., 1999; 1.3–3.3 year olds. Average of values for two different locations |
| | 0.17 | Levy et al., 2000; 3–4 year olds. |
| 4<7 | 0.241 ±0.184 | Nacchache et al., 1992; 4 year olds |
| | 0.227 ±0.174 | Nacchache et al., 1992; 5 year olds |
| | 0.180 ±0.127 | Nacchache et al., 1992; 6 year olds |
| 7<11 | 0.175 ±0.194 | Nacchache et al., 1992; 7 year olds |
| 11 – 14 | 0.2 | Levy et al., 1995 – as adjusted by NRC; 13-19 year olds |
| >14 | No data | No data |

[a]Fluoride values represent one brushing per day.

Surveys of fluoride ingestion from tooth brushing are indicative of wide individual variability with standard deviations that are frequently greater than the mean values (Naccache et al., 1992). The studies are generally consistent in showing that mean fluoride intake from toothpaste decreases with age. This is likely due in some part to maturation of the swallowing reflex as well as improved rinsing and expectoration practices.

The number of times a child or adult brushes their teeth per day is an important variable in determining the fluoride ingested because of toothpaste use. Table 4-10 summarizes the data available from studies in children that recorded this parameter. Three of the studies were conducted in the United States (Levy et al., 1997; Rojas-Sanchez et al., 1999; Franzman et al., 2006) and two in Canada (Simard et al., 1989, 1991). In all the studies but 2 (Rojas-Sanchez et al., 1999, at one location; Simard et al., 1989), the percentage brushing their teeth one time per day was greater than that for more frequent brushings. The Simard et al., 1989 study covered the largest age range (2 to 5 years), suggesting that those results may easily have been influenced by a high representation of older children who brushed two or three times per day. Based on these data, the OW chose to use the data for one brushing per day to represent fluoride exposure from ingestion of toothpaste. There are no ingestion data for elementary-school age children, adolescents or adults. Although some of the cited data are from Canada, the values reported suggest that the FDA (2009) guidance that children younger than 2 years in age should not use toothpaste when brushing their teeth is not practiced by many.

**Table 4-10. Number of Tooth Brushings Per Day Reported for Children (Six Months to Five Years Old)**

| Study | N = | Age (years) | Percentages[a] | | |
|---|---|---|---|---|---|
| | | | 1 time/day | 2 times/day | 3 times/day |
| Simard et al, 1989 | 23 | 2 to 5 | 4.8 | 71.4 | 23.8 |
| Simard et al. 1991 | 15 | 1 to 2 | 60 | 32 | 8 |
| Levy et al., 1997 | 899 | 0.5 | 41.2 | 16.9 | 6.3 |
| | 665 | 0.75 | 33.2 | 17 | 3.1 |
| | 508 | 1 | 37 | 14.7 | 3.5 |
| Rojas-Sanchez et al., 1999 | 14 | 2.25 | 33[b] | 67[c] | |
| | 29 | 2.3 | 54[b] | 46[c] | |
| Franzman et al., 2006 | 90 | 1.3 | 48 | 14 | 4 |
| | 100 | 2 | 51 | 23 | 2 |
| | 100 | 3 | 51 | 24 | 1 |

[a]Some studies also reported those brushing their teeth less than once per day and more than three times per day. In these cases the percentages do not add up to 100%.
[b]Less than or equal to 1 time per day
[c]Equal to or greater than 2 times per day

The presence of food in the gastrointestinal tract decreases the bioavailability of fluoride from 30 to 40 % based on studies in which adults ingested a toothpaste slurry after eating a meal or after fasting (Cury et al., 2005). In the fasted state, bioavailability was assumed to be close to 100%, deceasing to 61 to 71% after meals if the toothpaste has the current conventional 1100 ppm fluoride concentration. These data are supported by a study of fluoride absorption after ingestion of tablets (2 mg) of sodium fluoride and sodium monofluorophosphate (Trautner and Einwag, 1989); both chemicals are used in toothpaste. Ingestion of the tablet with milk reduced peak plasma fluoride levels to 70% of the level when the tablet was taken with water (Trautner and Einwag, 1989).

## 5. Other Sources of Exposure

### 5.1. Exposure from Air

As noted by NRC (2006), fluoride is released to the atmosphere by natural sources such as volcanoes and also by various anthropogenic sources. Atmospheric releases of inorganic fluoride to the atmosphere can come from power plants burning coal, aluminum production plants, phosphate fertilizer plants, chemical production facilities, steel mills, magnesium plants, and manufacturers of brick and structural clay (ATSDR, 2003).

#### 5.1.1. Monitoring Data

Cholak (1960) reviewed pre-1951 data on atmospheric levels of fluoride ion in several non-industrial areas of the United States. Average concentrations ranged from 0.02 ppb in Logan, Utah, to 2 ppb in New York.

Thompson et al. (1971) reported on water-soluble fluoride concentrations in ambient air collected by the National Air Surveillance Network in 1966, 1967, and 1968. Fluoride levels were measured in water-extracted samples using a fluoride ion-specific electrode. Of a total of 9175 urban air samples, only 18 (2%) exceeded 1.0 $\mu g/m^3$, and the maximum concentration recorded was 1.89 $\mu g/m^3$ (mean concentrations were not reported). Of 2164 non-urban samples only 3 (1%) exceeded 0.1 $\mu g/m^3$, and the maximum concentration recorded was 0.16 $\mu g/m^3$.

Thompson et al (1971) also summarized the results of the Continuous Air Monitoring Project conducted in 1967 and 1968 in six major US cities (Chicago, Cincinnati, Denver, Philadelphia, St. Louis, and Washington, DC). Over 110 samples were analyzed from each city. The percentage of 1967 samples in which no fluoride could be detected (minimum detection limit 0.05 $\mu g/m^3$) ranged from 58% in Chicago to 98% in Washington, DC. The percentage of 1968 samples in which no fluoride could be detected ranged from 42% in St Louis to 84% in Cincinnati. The maximum recorded values were 1.90 $\mu g/m^3$ in St. Louis in 1967 and 0.55 $\mu g/m^3$ in Chicago in 1968.

Kelly et al. (1993) reported that ambient concentrations of hydrogen fluoride in the United States, as measured around 1983, ranged from 1.0 to 7.5 $\mu g/m^3$ (ATSDR, 2003).

Atmospheric concentrations of fluoride in most parts of Canada are generally low or undetectable ($<0.05$ $\mu g/m^3$) (Environment Canada/Health Canada, 1993). Atmospheric levels in a residential area near Toronto averaged (monthly) 0.03 $\mu g/m^3$.

Fluoride levels in the atmosphere can be unusually high in certain locations due to industrial activity and/or the burning of fluoride-rich coal. Ernst et al. (1986) reported that in 1981 the Surveillance Division of the Air Pollution Control Directorate-Canada measured an average atmospheric fluoride concentration (particulate and gaseous) of about 0.6 $mg/m^3$ downwind from an aluminum smelter located in a rural inhabited area on the U.S.-Canadian border.

### 5.1.2. Exposure to Airborne Fluoride

According to NRC (2006), exposure to airborne fluoride for most individuals in the United States is expected to be low compared with ingested fluoride as reported by U.S. EPA, (1988), with exceptions being populations living in heavily industrialized areas or having occupational exposure. Using inhalation rates of 10 $m^3$/day for children and 20 $m^3$/day for adults, NRC (2006) calculated that fluoride inhalation exposures in rural areas (<0.2 μg/$m^3$ fluoride) would be less than 2 μg/day for a child and 4 μg/day for an adult. In urban areas (<2 μg/$m^3$), fluoride exposures would be less than 20 μg/day for a child and 40 μg/day for an adult. Most of the data that support these estimates are 30 to 40 years old and were collected before restrictions were placed on many industrial releases of gases and particulate matter to ambient air. The NRC estimates are consistent with the older monitoring data reported in Section 5.1.1 but the 1993 Canadian data cited above suggest that ambient air concentration in the U.S. may now be lower than the values used by NRC (2006) in their assessment.

Airborne fluoride can indirectly contribute to human exposure as a result of secondary contamination of edible fruits and vegetables. In reviewing the data available at the time, Waldbott (1963) reported that peaches grown near an aluminum plant in Oregon contained 3.2–21.9 ppm fluoride, whereas those grown in an uncontaminated area contained only 0.21 ppm F. Similarly, carrots grown near an aluminum plant in Switzerland contained 5.0 ppm F, whereas uncontaminated carrots contained 0.22–2.0 ppm F. High levels of fluoride were also reported for orange juice (0.05–3.12 ppm F), milk (3.2 ppm F) and spinach (16.0 ppm F) obtained in Tampa, FL, near a phosphate fertilizer plant. The normal levels of fluoride in orange juice were reported to be 0.07–0.17 ppm, and that in milk 0.1–0.3 ppm.

## 5.2. Oral Supplements

Oral fluoride supplements are prescribed by physicians and dentists for children living in areas where the drinking water contains low levels of fluoride. The daily doses of supplemental fluoride recommended by the American Dental Association (as revised in 1994) call for no supplement use for children less than 6 months old and none for any child whose water contains more than 0.6 mg F/L (Table 5-1). Guidelines for other age groups and drinking water fluoride concentrations are summarized in Table 5-1.

**Table 5-1. Daily Fluoride Supplementation Recommended by the ADA and the American Academy of Pediatric Dentistry**

| Age | Fluoride Concentration in Local Water Supply | | |
|---|---|---|---|
| | **<0.3 ppm** | **0.3-0.6 ppm** | **>0.6 ppm** |
| 0–6 months | None | None | None |
| 6–36 months | 0.25 mg | None | None |
| 3–6 years | 0.50 mg | 0.25 mg | None |
| 6–16 yr | 1.00 mg | 0.50 mg | None |

**SOURCE:** ADA (http://www.ada.org/3088.aspx).

The Dental Care Supplement of the 1989 National Health Interview Survey reported that approximately 10.5% of 31,446 children under 18 yr of age had used fluoride supplements (CDC, 1989).

Levy and Muchow (1992) evaluated patterns of fluoride supplement use among 446 children and their siblings living in either Iowa or North Carolina. Fluoride intake through the use of supplements was compared to the fluoride levels of the municipal drinking water in the areas where the children lived. Results suggested that approximately one-third of the primary children and 42% of the siblings did not receive an adequate amount of fluoride.

A survey conducted by Pendrys and Morse (1990) of seventh and eighth grade children living in Massachusetts and Rhode Island found that 35.1% of 74 children who had lived in a fluoridated community for at least 3 years during their first 6 years of life were given fluoride supplements.

As reported in Section 4, the patterns of fluoride ingestion from toothpaste use in children from shortly after birth (1.5 months) to an age of 36 months were reported by Levy et al. (2001). Using information from the questionnaires provided by the parents, Levy et al., (2003a) calculated fluoride ingestion from dietary supplements in children ages 36 to 72 months old. Results were presented in graphic form. The estimates of mean fluoride intakes from supplements were less than 0.05 mg/day for all age groups (Table 5-2).

**Table 5-2. Fluoride Intake from Supplements in Children 1.5 to 36 Months Old**

| Age (months) | Intake (mg/day) | |
|---|---|---|
| | Mean (SD) | Maximum |
| 1.5 | 0.014 (0.045)[a] | 0.375 |
| 3.0 | 0.018 (0.060) | 0.833 |
| 6.0 | 0.019 (0.063) | 1.000 |
| 9.0 | 0.014 (0.052) | 0.500 |
| 12.0 | 0.015 (0.054) | 0.500 |
| 16.0 | 0.011 (0.054) | 1.000 |
| 20.0 | 0.008 (0.038) | 0.250 |
| 24.0 | 0.008 (0.052) | 1.000 |
| 28.0 | 0.012 (0.068) | 1.000 |
| 32.0 | 0.013 (0.079) | 1.000 |
| 36.0 | 0.013 (0.079) | 1.000 |

**SOURCE**: Levy et al., 2001.

Trautner and Einwag (1986) measured the bioavailability of fluoride in three health food products recommended for children. The net urinary excretion of fluoride in six children ages 15-16 years was measured after ingestion of bone meal tablets, calcium earth tablets or siliceous earth tablets with fluoride contents of 520, 100, and 115 mg F/kg. Urinary fluoride was measured with an ion-specific electrode. Mean relative bioavailability was found to be 53.9

±21.6% from bone meal tablets, 64.8 ±23.6% from calcium tablets, and 38.9 ±20.5% from siliceous earth tablets.

In a later study Trautner and Einwag (1989) measured the bioavailability of fluoride when administered as NaF or sodium monofluorophosphate tablets (2 mg F). The test subjects were 7-19 years old and were given the supplements while fasting, or with milk or with milk and food. Fluoride levels in blood samples were measured using an ion-specific electrode. In fasting subjects equal levels of bioavailability were seen for both fluoride compounds and assumed to be 100%. Ingestion of milk reduced peak plasma fluoride levels by 30% compared to that for fasting individuals, but this effect was not seen when the milk was consumed with food.

Bohaty et al. (1989) evaluated both topical and systemic fluoride supplement use and the prevalence of dental fluorosis in 300 children, aged 6-13 from 6 elementary schools, living in areas with optimal water fluoridation (location of the study sites and the fluoride level in the drinking water were not reported). Fluorosis was scored using Dean's system (subjects with fluorosis were considered those with a Dean score of 0.5 or higher). The data were categorized according to fluoride use, residential history, age, sex and geographic location. Differences in frequency of the categorized data were evaluated statistically with Chi-square analysis where the differences were considered significant at $p < 0.05$. Although there were no significant associations between the frequency of tooth brushing and dental fluorosis, for subjects from four schools ($n = 206$), the frequency of using fluoride supplements was significantly associated with fluorosis. Similarly, for subjects of three of these four schools, the use of fluoride gels and rinses was significantly associated with dental fluorosis.

### 5.3. Soil Ingestion by Children

Fluoride ranks 13$^{th}$ or 14$^{th}$ in terms of its elemental abundance in the earth's crust. Thus, fluoride in soil could be a source of inadvertent exposure, primarily for children. Typical fluoride concentrations in soil in the United States range from very low (<10 ppm) to as high as 7% (70,000 ppm) in some areas with high concentrations of fluorine-containing minerals (ATSDR, 2003). Mean or typical concentrations in the United States are on the order of 300–430 ppm. Soil fluoride content may be higher in some areas due to use of fluoride containing phosphate fertilizers or to deposition of airborne fluoride released from industry.

The EPA (2008) Child-Specific Exposure Factor's Handbook recommends use of a combined soil and outdoor dust ingestion rate of 60 mg/day for children < 1 year old and 100 mg/day for children 1 to < 21 years of age. Using an average fluoride concentration of 400 ppm, the oral intake from soils for an infant (<1 year) would be 0.02 mg/day and that for older children and adolescents would be 0.04 mg/day. The estimated intake for adults in the EPA (1997) Exposure Factors Handbook is 50 mg/day and equivalent to 0.02 mg F/day from soils with an average concentration of 400 ppm. Erdal and Buchanan (2005) estimated intakes of 0.0025 and 0.01 mg/kg/day for children (3–5 years), for mean and reasonable maximum exposures, respectively, based on a fluoride concentration in soil of 430 ppm. In their estimates, fluoride intake from soil was 5–9 times lower than that from fluoridated drinking water.

For children with pica (a condition characterized by consumption of nonfood items such as dirt or clay), an estimated value for soil ingestion is 10 g/day (U.S. EPA, 1997). For a 20-kg child with pica, the fluoride intake from soil containing fluoride at 400 ppm would be 4 mg/day or 0.2 mg/kg/day. Although pica in general is not uncommon among children, the prevalence is not known (U.S. EPA, 1997). Pica behavior specifically with respect to soil or dirt appears to be relatively rare but is known to occur (U.S. EPA, 1997). Fluoride intake from soil for a child with pica could be a significant contributor to total fluoride intake. For most children and for adults, fluoride intake from soil probably would be important only in situations in which the soil fluoride content is high, whether naturally or due to industrial pollution.

### 5.4. Pharmaceuticals

As noted by Müller et al. (2007), since 1957, over 150 fluorine-containing drugs have come to the marketplace and now make up about 20% of all pharmaceuticals. The presence of fluorine in a drug can enhance binding efficacy and selectivity (Müller et al., 2007). Typical fluorine-containing drugs include fluoxetine (antidepressant Prozac), atorvastatin (cholesterol-lowering drug Lipitor), and ciprofloxacin (antibacterial drug Ciprobay). Waldbott (1963) reported that certain fluoride-containing tranquilizers and steroids, when taken three times per day, can result in a daily intake of 0.8–1.0 mg F. Fluoride in such drugs is organically bound to carbon atoms. The extent that the fluoride becomes bioavailable as a result of the metabolism of these drugs is likely to vary from drug to drug. To assess the contribution of fluorine-containing drugs to the total body pool of fluoride ion, information is needed on the changes in concentration of fluoride ion in blood serum following ingestion of such drugs. NRC (2006) reported that there are slight, but not significant increases of inorganic fluoride in serum after ingestion of several organofluorine pharmaceuticals but only a limited number of such products have been evaluated.

Oral electrolyte solutions were sampled for fluoride and found to contain 0.01–0.15 mg F/kg by Dabeka and McKenzie (1987). Electrolyte solutions are used to replenish the fluids lost during episodes of severe diarrhea in children.

### 5.5. Occupational Exposures

Inhalation exposures to fluoride in the workplace are limited by regulations established by the Occupational Safety and Health Administration (OSHA). The OSHA 8-hr TWA exposure limit of for fluoride is 2.5 $mg/m^3$ (ATSDR, 2003). A person breathing at an average rate of 20 $m^3$ per day would inhale 16.8 mg during one 8-hr working shift (equivalent to 0.24 mg/kg/day for a 70 kg man).

### 5.6. Smoking

As noted by NRC (2006), heavy cigarette smoking could contribute as much as 0.8 mg of fluoride per day to an individual (0.01 mg/kg/day for a 70-kg person) (U.S. EPA, 1988).

## 6. Exposure Assessment Summary

As mentioned in the preceding sections of this report, fluoride concentrations in different media and resultant fluoride exposures vary for a number of reasons including the following:

- The methodologies used in conducting the studies differ in the ways the data were gathered, grouped and analyzed
- The size and composition of the study populations differ between studies.
- The analytical methods used to determine the concentration of fluoride in media of interest have evolved over time with the evolution of new methods that improved fluoride recovery and detection levels as well as reduced interference from other ions
- The amounts of fluoride present in the drinking water supply and soils differ with local geology and fluoridation practices.
- Available commercial food and beverage products and population dietary preferences are not constant over time
- Use of fluoridated water as process water by commercial food and beverage facilities can increase fluoride content to levels above that in the unprocessed product.
- Home cooking of foods in fluoride-containing water increases the fluoride content of the finished product but the increase varies with the food material prepared.

Each of these factors contributes to the differences observed when comparing data from the studies included in this report and to the uncertainty inherent in establishing an RSC for fluoride.

In developing the RSC for the fluoride from drinking water, EPA chose to focus on the following media as the major contributors to total intake:

- Drinking water from public drinking water systems.
- Solid foods from the diet including milk and juices not made from concentrate.
- Residues of the recently registered pesticide, sulfuryl fluoride.
- Beverages, both commercial and home-prepared using tap water (i.e. coffee, tea. reconstituted juices and powdered beverage mixes).
- Infant formula made from powdered concentrate for the six-month to less than one-year age group.
- Toothpaste swallowed during tooth brushing.
- Incidental ingestion of soil and outdoor dust.

There are other sources of fluoride exposure such as ambient air, dietary supplements, professional dental treatment products, and some pharmaceuticals. These sources make minimal contributions to daily intakes during the period of dental fluorosis vulnerability. NRC (2006) estimated that average exposures from ambient air would be 2 micrograms per day for children and 4 micrograms per day for adults. Supplements are not recommended for use in cases where water is fluoridated, and thus, would not be appropriate at the 0.87 mg/L concentration that represents the national average fluoride concentration for public water systems (Section 3.3) because it falls within the recommended fluoridation range. Professional dental fluoride treatments are episodic and do not contribute greatly to the average daily intake when normalized across time. The major chronic-use, fluoride-containing pharmaceuticals (i.e. Zocor and Prozac) do not include young children among their target population. Intakes of the antibiotic Ciptoflaxozin (Cipro) by children would be episodic rather than chronic. In addition, the covalently-bound fluoride in pharmaceuticals does not appear to be bioavailable (NRC, 2006).

After consideration of the strengths and weaknesses of the studies presented in the preceding sections of this report, EPA selected the data from one or two studies to represent the fluoride intake for each of the age groups used in assessing the dose-response for severe dental fluorosis (U.S. EPA, 2010a). In making the selection of the representative study EPA applied the following guidelines:

- Where possible a study from the United States was selected over a study from Canada.
- The publication had to report that plain water was not included in the market basket or duplicate diet.
- Where there was no study that clearly eliminated plain water from the market basket in the study description, the study location with the lowest drinking water fluoride concentration was selected and the uncertainty introduced noted.
- Market basket approaches were preferred over duplicate diet or recall studies because they were considered to be more geographically representative.
- Studies considered for use as representative for toothpaste were those where the ingested toothpaste was measured.
- The study methodology and the ages of the children studied were both considered: methodology was given a higher weight in the selection process than age in situations where there were several study options for an age range.

The value selected and the rationale for its selection are provided in Tables 6-1, 6-2, 6-3, and 6-4 for solid foods, beverages, plain drinking water, and toothpaste, respectively. Soil ingestion by young children was determined using an average soil concentration of about 400 ppm (see Section 5.3) and the EPA estimates of 60 or 100 mg/day for soil ingestion by young children (U.S. EPA, 2008). Each value is reported to a hundredth of a mg/day due to the analytical limitations inherent in the representative values.

### 6.1. Dietary Intake

**Foods.** The food category includes milk and fruit and vegetable juices that are not made from concentrate. Milk and such juices are not categorized as beverages by the FDA Total Diet Study (Egan et al., 2007).

Data from Ophaug et al., (1985) and Jackson et al. (2002) were selected as representative for all but adults in Table 6-1 below. These studies used a market basket approach in the analysis of food for their fluoride content. Intakes for the non-beverage food groups come from the USDA (1998) Continuing Survey of Food Intakes by Individuals (Jackson et al., 2002) or its precursor USDA (1968) survey of food consumption. The Ophaug et al. (1985) data are used for the 0.5- to <1-year and 1- to <4-year age groups. The Jackson et al. (2002) data are used for the 4- to < 7-year age group. Duplicate plate data were available from Brunetti and Newbrunn (1983) and Rojas-Sanchez et al., (1999). OW determined that these were less representative of the age group than the data selected because of the study design and the small number of participants. In addition, Brunetti and Newbrunn (1983) did not separate the fluoride from beverages from that for solid foods.

| Table 6-1. Estimated Daily Dietary Fluoride Intakes from Solid Foods for Age Groups of Concern | | |
|---|---|---|
| **Age (years)** | **Exposure Estimate (mg/day)** | **Rationale** |
| 0.5 – <1 | 0.25 | Ophaug et al., 1985 – Overall mean (0.17 mg/day) from 22 market baskets, and national food intake data (Table 2-24); does not include F from plain water and beverages. These data were adjusted by subtracting the average for the milk and the average for formula and other dairy products of 0.06 mg/day from (Ophaug, 1980a Table 2-23) and replacing it with 0.14 mg/day from the powdered formula (Van Winkle et al., 1995) [ 0.17 - 0.06 + 0.14 = 0.25]. The Ophaug et al. (1985) data apply to 6-month-old infants. |
| 1 – <4 | 0.16 | Ophaug et al., 1985 – Overall mean of 22 market baskets, and national food intake data; does not include plain water and beverages (see Table 2-31). Based on 2-year-old children. This value is slightly greater than the average of the means (0.13 mg/day) from the less representative Rojas-Sanchez et al. (1999) duplicate plate analysis using data for 54 children from three cities covering a larger segment of the age range 1.3–3.3 years. The Ophaug et al. (1985) estimate, although an older study, had a broader geographic representation. |
| 4 – <7 | 0.35 | Jackson et al. (2002). Average of 2 market basket values (0.350 and 0.357 mg/day) excluding plain drinking water and beverages. Based on 3- to 5-year-old children. |
| 7 – <11 | 0.41 | The estimate for this age group is based on the mean F concentration for food groups from two market baskets in the study by Jackson et al. (2002) and USDA (1998) data on food group intakes for the 6–11 year age group. It does not include plain drinking water or beverages. |
| 11 – 14 | 0.47 | The estimate for this group is based on the mean F concentration for food groups from two market baskets in the study by Jackson et al. (2002), and USDA (1998) data on food group intakes for the 12–19 year age group. It does not include plain drinking water or beverages. |
| >14 | 0.38 | Average of the exposure estimates of Singer et al., (1980, 1985) and Taves (1983). All estimates but Taves (1983) are based on 15- or 16- to 19-year-old males. The Taves study was a six-day duplicate diet type representing an adult regular hospital diet. |

In the case of the 0.5 to one year old group, the exposure value for foods applies to formula-fed children in cases where the formula is a powdered product reconstituted with tap water. Only the fluoride in the powdered formula, not that in the tap water used to reconstitute the formula, is included in the food value in Table 6-1. Powdered formula is the most prevalent product chosen (~90%) by parents who use formula according to the HHS Infant Feeding Practices Study (Table 3-15, CDC, 2009).

As described in the table, the food value for this age group was calculated by adding the average of the four city average intakes from milk to the average of the four city average intakes from other dairy products and formula (0.06 mg/day; Table 2-23) and subtracting the sum from the food total (0.17 mg/day; Table 2-24), and then adding the amount from the powdered formula (0.14 mg). The water added to reconstitute the formula is included in the drinking water exposure (Table 6-3). The relative contribution of fluoride in the powdered formula versus the added water depends on the concentration present in the water as well as the concentration in the powder. When the water is fluoridated it accounts for more of the exposure than the powdered formula.

The reported food value in Table 6-1 represents a child with no intake of fluoride from milk or other dairy products. In many cases children begin to consume milk rather than formula as they approach their first birthday. Total fluid feeding of infants begins to decline at about 5-months as the intake of solid foods increases; at 9 months only about 10% of infants are being given a fluid-only diet. (Grummer-Strawn et al., 2008) This is not reflected in Table 6-1 making the estimate a conservative one.

Using the data from Van Winkle et al. (1995 Table 2-3) to represent a soy-based powdered formula will increase the fluoride intake for soy-based, formula-fed children 0.5 to 1 years old by 0.1 mg/day to 0.36 mg/day. The mean value for powdered soy-formula preparations reported by Siew et al. (2009, Table 2-4) had lower fluoride concentration and would lower the total fluoride from the powdered concentrate by 0.03 mg/day.

There is a lack of appropriate data from published studies of the 7- to <11-year and 11- to 14-year age groups. Accordingly, local fluoride food-group concentrations from the Jackson et al. (2002) study were combined with national USDA (1998) food intake data for the closest age range and used to represent these age groups (Tables 2-34). Food product information in the USDA (2005) database is too limited to support OW development of a market basket to apply with age groups that lack primary data. The value for the 11 to <14 year old age group is a conservative estimate since the USDA (1998) food intake data apply to the 12 to 19 year age group. High food intakes associated with the teenage growth spurt will tend to cause averages for 12–19 year old children to be higher than those for 11 to <14 year old children.

The adult data available for fluoride intake from foods were limited to an analysis based on hospital diets of limited scope (Taves, 1983) and three market basket surveys (San Filippo and Battistone, 1971, Singer et al., 1980, 1985). Each of the market basket surveys was based on teen-aged male adolescents as the population of interest. This age group tends to have a higher caloric and food intake than adults > 20 years old. Singer at al. (1980) found that the colorimetric method used for fluoride analysis by San Filippo and Battistone (1971) produced higher fluoride concentrations than those obtained for the same homogenates using an ion-specific electrode. This is likely the reason that the San Filippo and Battistone results are about 0.3 to 0.4 mg/day higher than those from the other three studies. Because of the weaknesses in the San Filippo and Battistone (1971) data, EPA chose to average the results of the other three studies together as representative of average adult intakes from foods (0.40 + 0.42 + 0.33 mg/day ÷ 3 = 0.38 mg/day).

The uncertainties in the exposure estimates in Table 6-1 are acknowledged. However, despite the limitations found in the available data set, the pattern of fluoride intake is consistent with the expected pattern for food and calorie intakes that apply to the individual age groups. The mg/day intake for infants whose primary food source is formula made from a powdered concentrate was higher than that for the 1-3 years age group with a more mixed diet. The estimates for the other age groups were higher than that for infants, increasing with age as caloric requirements and food intake levels increase.

**Beverages.** As was the case for the food estimates, EPA selected a single value from the beverage data (Table 2-46) to represent the intakes for each age group of interest. The values selected and selection rationales are presented in Table 6-2. Estimates represent a combination of fluoride from commercial beverages and beverages prepared at home using tap water. As was the case for the food, the beverage estimates are given to the hundredth of a mg/day in recognition of the analytical limitations for the studies that provided the representative values.

| **Table 6-2. Estimated Daily Fluoride Intake from Beverages Only for Age Groups of Concern** | | |
|---|---|---|
| **Age (years)** | **Exposure Estimate (mg/day)** | **Rationale** |
| 0.5 – <1 | – | No value. All fluoride intake was considered to be from powdered formula (Table 6-1) prepared with tap water (The tap water fluoride concentration is in Table 6-3). |
| 1 – <4 | 0.36 | Mean value from Pang et al., (1992); 3-day drink diaries (n=57); beverages store-bought or made with de-ionized water. Milk and plain drinking water were excluded. Based on 2–3 year olds. |
| 4 – <7 | 0.54[c] | Mean value from Pang et al., (1992); 3-day drink diaries (n=79); beverages store-bought or made with de-ionized water. Milk and plain drinking water were excluded. Based on 4–6 year olds. |
| 7 – <11 | 0.60 | Mean value from Pang et al., (1992); 3-day drink diaries (n=89); beverages store-bought or made with de-ionized water. Milk and plain drinking water were excluded. Based on 7–10 year olds. |
| 11 – 14 | 0.38 | Derived from Jackson et al. (2002). Data for fluoride in the beverage food group were combined with USDA (1998) data on beverage intakes to estimate fluoride exposure. Does not include plain drinking water. Applies to ages 12-19. The Jackson et al. (2002) estimate is supported by the Clovis and Hargreaves (1988) data from a dietary record Canadian study covering six-grade students, average age ~12 years (range of 0.02 to 0.82 mg/day). |
| >14 | 0.59 | Singer et al., (1985); based on 5 market baskets from different areas of the country and excluding plain drinking water. The average water fluoride level was 0.14 mg/l ± 0.03. Based on data for 15-19 year-olds. |

There is no beverage intake estimate for the 0.5 to <1-year age group because this analysis focuses on the group most likely to be at risk for severe dental fluorosis, those infants who exclusively consume powdered formula prepared with tap water. The fluoride in the powdered formula is included in Table 6-1 and the fluoride in the tap water is in Table 6-3.

The Pang et al. (1992) data are used as representative for the 1-year through <11 year age groups. In this study the samples analyzed were selected from three-day diaries kept by or for 225 children. Plain drinking water and milk were excluded but fruit juices were included. Home-prepared beverages were reconstituted with deionized water. Accordingly, fluoride that would be introduced from preparing the beverages at home with tap water is included in Table 6-3 and not Table 6-2. The inclusion of fruit juice could bias the results to the high side, but, with the exception of grapes, most fruits are low in fluoride.

Table 2-46 includes other measures of fluoride intakes from beverages among children in the 4 to <7 year age group and the 7 to < 11 year age groups. Levy et al. (2003a) estimated an average fluoride intake of 0.2 mg/day from beverages, not including plain drinking water, for 3–6 year olds derived from questionnaires completed by the parents and historical data on fluoride concentrations in the beverages. The 90$^{th}$ percentile estimate was about 0.5 mg/day. The Levy study was a dietary record study. The Jackson et al. (2002) food frequency recall-based market basket for a town with 0.16 mg/L in the drinking water provided a beverage contribution of

0.12 mg/day for the 4 to < 7 year age group and a value of 0.22 mg/day for the 7 to < 11 year age group. The Pang et al. (1992) exposure estimate falls above the mean and 90$^{th}$ percentile levels in the Levy et al. (2003a) study and above the average values from Jackson et al. (1995). Accordingly it is a conservative value for fluoride intake from beverages.

The average male/female fluoride intake from beverages in the low fluoride town (0.16 mg/L; Connorsville, IN; Table 2-34) studied by Jackson et al. (2002) is used for the 11 to 14-year age group in the absence of other data. The Jackson data do not include fluoride from plain drinking water but do include fluoride from bottled water. This estimate may be slightly high since the beverage intakes apply to 12 to 19 year old adolescents and the local water was not totally free of fluoride. Inclusion of bottled water is expected to have a minimal impact on the fluoride intake. The USDA (2005) database indicates that commercial bottled waters are low in fluoride.

The beverage exposure estimate for adults is from Singer et al. (1985) for the 5 cities with the lowest drinking water fluoride levels (average 0.14 mg/L). It was selected as representative of intakes in communities with low fluoride in their drinking water and therefore in home prepared beverages. The Taves (1983) estimate of (1.38 mg/day) and that of San Fillipe and Battistone (1971; 1.34 mg/day) were higher. The San Fillipe and Battistone data were not selected because it included plain drinking water and used a colorimetric assay for fluoride analysis which is less accurate than the ion-specific electrode used by Singer et al. (1985). The Taves (1983) estimate was based on house diets for adults in a hospital setting. Taves (1983) attributed the high fluoride levels to the fact that the hospital diets included daily servings of orange juice, coffee, and two servings of tea, as well as other juices. The Taves data demonstrate that tea was the major contributor to the fluoride intake from beverages. All of the adult data are from studies where tap water was used for home beverage preparation.

## 6.2. Drinking Water

Table 6-3 provides the estimates for fluoride intakes from plain drinking water and the indirect water that is used in the home preparation of beverages and foods when it is part of a standard recipe. Following the RSC policy, the drinking water contribution is determined from the average fluoride concentration from public drinking water systems as reported to EPA through the ICR for the second six-year review of regulations combined with 90$^{th}$ percentile drinking water intakes. The data apply to consumers only at the 90$^{th}$ percentile intake level. The average water concentration (0.87 mg/L) was derived from 16 monitoring quarters covering the years 2002 through 2005 as described in Section 3.3. The drinking water intake data come from EPA (2004) rather than the EPA (2008) Child-Specific Exposure Factors Handbook because the age ranges in EPA (2004) match those used in the EPA (2010a) dose-response assessment for severe dental fluorosis while those in EPA (2008) do not. Both EPA (2004) and EPA (2008) are based in CSFII 1994–1998 water intake data.

| Table 6-3. Fluoride Intake from Consumption of Municipal Water (Direct and Indirect[a]) at the Average Concentration (0.87 mg/L) Determined from Monitoring Records for 2002 through 2005 | | |
|---|---|---|
| Group (yr) | Water Consumption[a] | Fluoride Intake[a] |
| | 90th Pecentile Intake Total mL | mg/day total |
| 0.5–0.9 | 971 | 0.84 |
| 1–3 | 723 | 0.63 |
| 4–6 | 943 | 0.82 |
| 7–10 | 993 | 0.86 |
| 11–14 | 1415 | 1.23 |
| 14+ | 2000[b] | 1.74[b] |

**SOURCE:** Adapted from U.S. EPA, 2004.
[a]Consumers only value.
[b]Value for the 14+ age group – EPA policy for adults.

## 6.3. Toothpaste

There are a number of studies that report on toothpaste use and resultant potential total exposure from fluoridated dentifrice. A more limited set of data are available from studies where the ingestion of toothpaste during tooth brushing was measured. In the toothpaste ingestion studies, the toothpaste placed on the toothbrush was measured and corrected for that left on the toothbrush after brushing and that expectorated during post-brushing rinsing of the mouth. The difference was assumed to be swallowed. The data from these studies are summarized in Table 6-4. Each estimate is highly uncertain since the confidence bounds around the mean values are indicative of high inter-individual variability (See Table 4-9). Estimates may be high because the studies were conducted before the recommendation became widely publicized for children to use only a pea-sized amount of toothpaste when brushing.

| Table 6-4. Age-Related Exposure Estimates for Fluoride from Toothpaste | | |
|---|---|---|
| Age (yr) | Fluoride Intake (mg/day) | Notes |
| 0.5 – <1 | 0.07 | Mean for 12-month old children, from Levy et al., 1995. The value for the six-month old child was 0.02 mg/day but few children brush at this age since children have few teeth at this age. For that reason the estimate for the 12-month olds was considered to be a better choice (see Table 4-9). |
| 1 – <4 | 0.34 | Average of estimates from Levy et al., 1995; Levy et al., 2000; Nacchache et al., 1992 and Rojas-Sanchez et al., 1999 (see Table 4-9). |
| 4 – <7 | 0.22 | Average of estimates for 4, 5, and 6 year-olds by Nacchache et al., 1992 (see Table 4-9). |
| 7 – <11 | 0.18 | Average for seven year olds by Nacchache et al., 1992 (see Table 4-9). |
| 11 – 14 | 0.2 | NRC estimate based on Levy et al., 1995 (see Table 4-9). |
| Adult | No data | No measured value for ingestion. Estimated as half that for 11 to 14 year olds. |

Data represent one brushing per day. Studies suggest high inter-individual variability in the amount swallowed.

Fluoride intakes represent one brushing per day, a value that is applicable to about half the population for children < 3 years old according to the data collected by Franzman et al. (2006), Levy et al. (1997), and Simard et al. (1991). The number of brushings appears to increase to twice a day for older children (Simard et al., 1989) but this estimate lacks confirmation from other studies. Increasing the number of brushings per day for children to 2 would double the intake estimates.

### 6.4. Soils

Although concentration varies with local geological conditions, 400 ppm was been identified as a reasonable estimate for an average fluoride concentration in soils (ATSDR, 2003). Based on this concentration and a combined soil and outdoor dust ingestion rate of 60 mg/day for children < 1 year old (U.S.EPA, 2008) the fluoride intake for an infant (<1 year) would be 0.02 mg/day. The comparable fluoride intake for the 0–14 year age groups would be 0.04 mg/day using the 100 mg/day estimate for intakes of soil and indoor dusts (U.S. EPA, 2008). The fluoride RSC assessment considers children older than 14 to be grouped with adults since they are no longer vulnerable to severe dental fluorosis. The estimated intake for adults in the EPA (1997) Exposure Factors Handbook is 50 mg/day and equivalent to a 0.02 mg/day intake from soils with an average concentration of 400 ppm. Lower fluoride concentrations in soil are likely the norms for areas of the country with minimal geological fluoride.

### 6.5. Uncertainty

There are many uncertainties in the estimates EPA selected for the RSC analysis related to analytical methods and study protocols. In addition, the food preferences and food intakes of the U.S. population shift as new products are introduced into the market-place and the dietary intakes change. The past thirty years have seen an increase in the use of pre-prepared commercial foods by the average consumer, increased imports of fresh produce from foreign countries, and more frequent eating of meals away from home at restaurants, schools and daycare facilities. Accordingly, the data from the selected studies (published between 1980 and 2002) are not necessarily representative of current food preferences and intakes.

Additional uncertainties in the exposure estimates are due to the lack of published studies that provide an exact match to the age ranges used in this analysis. Some of the data come from very localized areas whereas other studies collected food and beverage samples representing different geographical areas across the country. The concentrations of fluoride in the water used in food preparation were not always identified; in cases where the fluoride in the water was identified, the resultant concentrations in the finished foods did not always show a consistent relationship to the drinking water concentration. Each of these factors contributes to the uncertainty in the representative values chosen.

In recognition of the multiple uncertainties affecting the data, EPA has selected values that are representative of average to slightly above average fluoride intakes for the RSC analysis. EPA believes that these are reasonable estimates.

In addition to the methodological variables influencing the intake assessment, there are also uncertainties about the bioavailability of fluoride in the diet. The solubility product constants for calcium and magnesium fluoride are low and can limit fluoride absorption from foods that contain these cations. Spak et al. (1982) found that 72% of the fluoride in milk and 65% of the F in formula were bioavailable by measuring the fluoride levels in plasma after ingestion. Cury et al. (2005) found the gastrointestinal absorption of fluoride in an ingested toothpaste slurry was lower when slurry ingestion occurred directly after a meal than when it was consumed after fasting. Hydroxyfluoroapatite, the form of fluoride found in bone, has a low solubility product constant. Thus, when ashed for analysis, any meat, poultry, or fish products that may have contained bone fragments would contribute to an overestimate of the bioavailable fluoride in the product as consumed.

One major limitation with the food and beverage data reported in Tables 6-1 and 6-2 is the studies were all conducted before the approval of sulfuryl fluoride as a fumigant for food storage facilities and food processing plants. Accordingly, any fluoride currently in the food supply because of sulfuryl fluoride fumigation is not reflected in those data. The OPP (U.S. EPA, 2010b; see Appendix B) provided OW with estimated contributions of fluoride to the food supply from sulfuryl fluoride data (Table 6-5). As was the case for Tables 6-1 through 6-4, fluoride residues are reported to the hundredth of a mg/day.

**Table 6-5. Sulfuryl Fluoride Contributions to Dietary Fluoride Exposure.**

| Population Group | Exposure Estimates, mg/day | | |
|---|---|---|---|
| | Structural Fumigations | Food Fumigations | Total |
| 0.5– <1 year | <0.01 | 0.02 | 0.03 |
| Children 1 – <4 yrs | 0.01 | 0.03 | 0.05 |
| Children 4 – <7 yrs | 0.02 | 0.05 | 0.06 |
| Children 7 – <11 yrs | 0.02 | 0.05 | 0.07 |
| Youth 11– <14 yrs | 0.02 | 0.07 | 0.09 |
| Adults >14 | 0.02 | 0.06 | 0.08 |

**SOURCE:** U.S. EPA, 2010b

## 7. Relative Source Contribution (RSC)

The OW has followed the general principles for RSC determination outlined in the Ambient Water Quality Criteria Human Health Methodology (U.S. EPA, 2000b) when determining the RSC from drinking water intake for fluoride. According to OW policies, the subtraction approach to RSC determination is not appropriate because of the OPP registration of pesticides (cryolite, sulfuryl fluoride) that limit fluoride residues on treated food products (See Section 1.2 of this report). Accordingly, the percentage approach was applied.

The RSC for water from public systems is calculated using the following equation:

$$\mathbf{RSC} = \frac{\mathbf{DWI}}{\mathbf{DWI + FI + BI + DI + SI}} \times \mathbf{100}$$

where:

| | | |
|---|---|---|
| DWI | = | Intake from consuming water (direct and indirect) with an average of 0.87 mg/L F (see Section 3.3) by the 90$^{th}$ percentile consumer |
| FI | = | Average intake of F from dietary foods except for beverages |
| BI | = | Average intake of F from beverages (commercial and prepared with tap water) |
| DI | = | Average intake of F from toothpaste use |
| SI | = | Average intake from soils and outdoor dust |

Exposures from ambient air are not included in the RSC equation because they are a minor contributor (< 4 µg/day) to the total exposure estimate (Section 5.1.2). Based on the NRC estimated urban air concentration, the contribution of fluoride from air is ≤0.3% of the total exposure for a young child and < 0.1% of the total for an adult. Fluoride intakes from supplements are also not included because the average drinking water concentration falls within the recommended range for fluoridation of drinking water, and supplements are not recommended for those who receive fluoridated drinking water (see Section 5.2).

Table 7-1 provides the representative values for intakes of fluoride through each quantified medium for each age group of interest as well as the total fluoride intake and the percentage contributed by direct and indirect drinking water residential tap water. The 90$^{th}$ percentile drinking water intakes (consumers only) are used for all age groups as it is U.S. EPA policy to protect the majority of the population. The drinking water fluoride concentration is the average for all systems detecting fluoride. Average values are used for the fluoride contributions from the other media as required by (EPA, 2000b). Exposure estimates are presented at the one hundredth of a milligram intake level because of the analytical uncertainties surrounding the representative data selected.

| Table 7-1. Representative Values for Fluoride Intakes Used in Calculation of the Relative Source Contribution for Drinking Water | | | | | | | |
|---|---|---|---|---|---|---|---|
| Age Group (years) | DWI[a] (mg/day) | FI (mg/day) | BI (mg/day) | TI (mg/day) | SI (mg/day) | Total (mg/day) | RSC (%) |
| 0.5 – <1 | 0.84 | 0.25[b] | – | 0.07 | 0.02 | 1.19 | 71 |
| 1 – <4 | 0.63 | 0.16 | 0.36 | 0.34 | 0.04 | 1.53 | 41 |
| 4 – <7 | 0.82 | 0.35 | 0.54 | 0.22 | 0.04 | 1.97 | 42 |
| 7 – <11 | 0.86 | 0.41 | 0.60 | 0.18 | 0.04 | 2.09 | 41 |
| 11 – 14 | 1.23 | 0.47 | 0.38 | 0.20 | 0.04 | 2.32 | 53 |
| > 14 | 1.74 | 0.38 | 0.59 | 0.10[c] | 0.02 | 2.83 | 61 |

[a]Consumers only; 90$^{th}$ percentile intake except for >14 years. The > 14 year value is based on the OW policy of 2 L/day.
[b]Includes foods, F in powdered formula, and fruit juices; no allocation for other beverages.
[c]Assumed to be 50% of the value for the 11-14 year old age group.
DWI = Drinking Water Intake (see Table 6-3).
FI = Food Intake (Solid Foods) (see Table 6-1).
BI = Beverage intake (see Table 6-2).
TI = Toothpaste Intake (see Table 6-4).
SI = Soil Intake (see Section 6.4).

Table 7-1 does not include consideration of any residues from the use of sulfuryl fluoride, a fumigant that was approved for use on food products after all of the dietary data used for this report were collected (U.S. EPA, 2009b). A separate calculation that includes estimation of fluoride residues from sulfuryl fluoride (SuF) is provided in Table 7-2. Sulfuryl fluoride decomposes in the environment to produce sulfate and fluoride ions. The OPP (U.S. EPA, 2009; 2010b) has provided the OW with estimates of fluoride residues from the currently approved uses of this product which include fumigation of food storage facilities and processing plants, as well as direct fumigation of some foods for pest control purposes. Table 7-2 shows the results of the RSC calculation when sulfuryl fluoride residue is included in the RSC analysis.

| Table 7-2. Representative Values for Fluoride Intakes (Including Sulfuryl Fluoride) Used in Calculation of the Relative Source Contribution from Drinking Water | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Age Group (years) | DWI[a] (mg/day) | FI (mg/day) | SuF (mg/day) | BI (mg/day) | TI (mg/day) | SI (mg/day) | Total (mg/day) | RSC (%) |
| 0.5 – <1 | 0.84 | 0.25[b] | 0.03 | -- | 0.07 | 0.02 | 1.21 | 70 |
| 1 – <4 | 0.63 | 0.16 | 0.05 | 0.36 | 0.34 | 0.04 | 1.58 | 40 |
| 4 – <7 | 0.82 | 0.35 | 0.06 | 0.54 | 0.22 | 0.04 | 2.03 | 40 |
| 7 – <11 | 0.86 | 0.41 | 0.07 | 0.60 | 0.18 | 0.04 | 2.16 | 40 |
| 11 – 14 | 1.23 | 0.47 | 0.09 | 0.38 | 0.20 | 0.04 | 2.41 | 51 |
| > 14 | 1.74[b] | 0.38 | 0.08 | 0.59 | 0.10[c] | 0.02 | 2.91 | 60 |

[a]Consumers only; 90$^{th}$ percentile intake except for >14 years. The > 14 year value is based on the OW policy of 2 L/day.
[b]Includes foods, F in powdered formula, and fruit juices; no allocation for other beverages.
[c]Assumed. 50% of the 11-14 year old age group.
DWI = Drinking Water Intake (see Table 6-3).
FI = Food Intake (Solid Foods) (see Table 6-1).
SuF = Sulfuryl Fluoride Intake (see Table 6-5)
BI = Beverage Intake (see Table 6-2).
TI = Toothpaste Intake (see Table 6-4).
SI = Soil Intake (see Section 6.4).

Figure 7-1 illustrates the percentage contributed by each of the media in Table 7-2 to daily total fluoride intake. It is apparent that, for most individuals in the population, the contribution from drinking water is substantially less than the 100% assumed in the EPA 1986 derivation of the MCLG for crippling skeletal fluorosis. However, the contribution from drinking water for adults who are not at risk for dental fluorosis (60%) is greater than the limiting value for children (40%) who are susceptible to severe dental fluorosis.

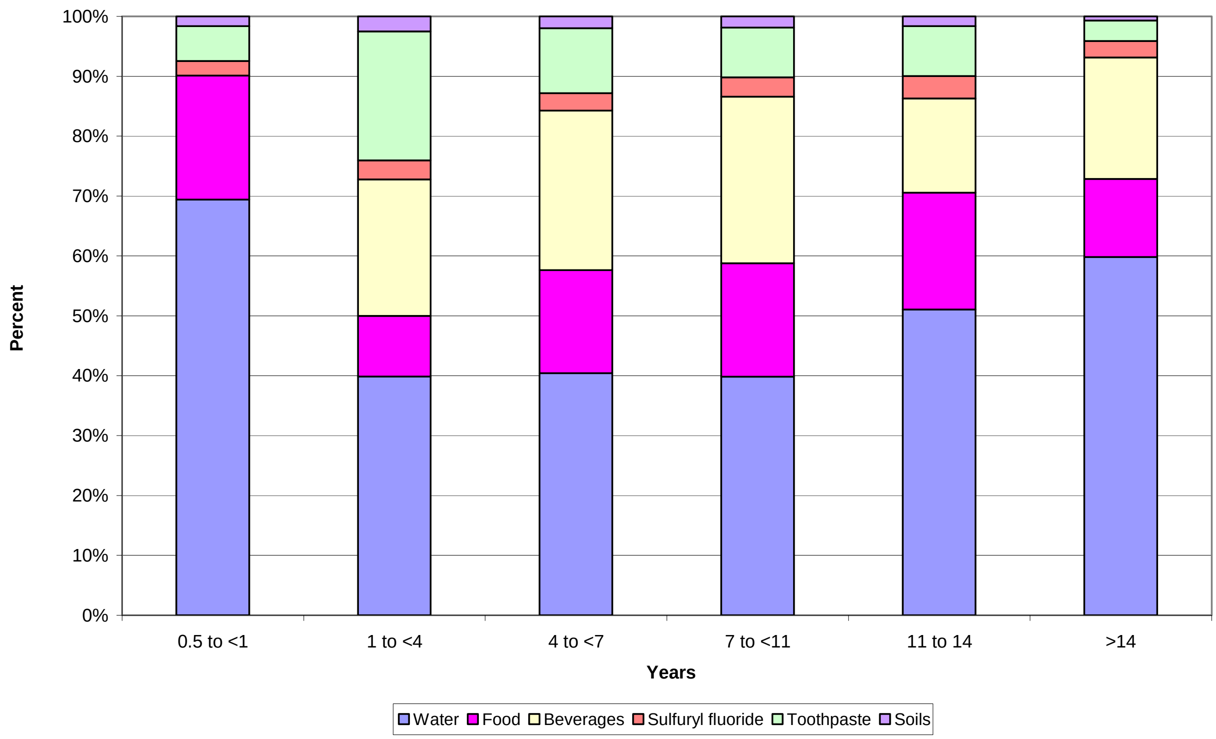


**Figure 7-1. Percentage Media Contribution to Total Daily Fluoride Intake: 90th Percentile Drinking Water Intakes for Consumers Only and a Fluoride Concentration of 0.87 mg/L**

Drinking water contributes the highest percentage of the total fluoride intake (70%) for infants six months to one year old. However, the high percentage contribution of drinking water for this age group is partially a consequence of the use of the intakes for infants fed exclusively with powdered formula reconstituted with tap water containing 0.87 mg/L fluoride for this analysis. The food intake data for the 0.5 to 1 year old age group came from a study of six-month old infants (Ophaug et al., 1985) as described in Table 6-1). This intake value also contributes to the high percent of the total coming from drinking water because, at this stage of development, the intake of formula is higher than that at one year when the typical infant's diet has expanded to include a variety of solid foods and juices.

The diet (solid foods, beverages, and sulfuryl fluoride) is another major contributor to total fluoride intake with sulfuryl fluoride making a minor contribution to the total. It is the largest contributor for children ages 4 to <11. However, dietary fluoride is indirectly impacted by the fluoride in drinking water because cooking and preparing foods in fluoride-containing water increases the fluoride content of the prepared food (Maier and Rose, 1966; Ophaug et al., 1985). Many food and beverage production facilities use fluoride-containing water in food preparation. When there is fluoride in the water supply, some of it will end up in the food supply. This is particularly true for beverages. The work by Ophaug et al. (1985) found that correlation coefficients between beverage fluoride and the drinking water fluoride concentration ranged from 0.72 to 0.98 for the four quadrants of the country. There was not a strong correlation between the drinking water fluoride and the fluoride content of solid foods (Ophaug et al., 1985) although cooking studies have shown uptake from the preparation water (Martin, 1951).

As discussed in Section 6, there are alternative estimates for the contribution of fluoride from beverages, excluding plain drinking water, for children in the 4 through <11 year age groups. The alternative estimates are lower than the values from the Pang et al. (1999) diary-based study selected by EPA. They are 0.2 mg/day from Levy (2003a) and 0.12 from Jackson et al (1995) for the 4 through <7 year group and 0.22 from Jackson et al. (1995) for the 7 to <11 year age group. When the RSCs for drinking water were calculated with these values in place of the Pang et al. (1999) data, the RSC values changed from 40 and 39% to 49/51 and 48%.

The relative source for drinking water would also be affected by the use of a soy-based powdered formula rather than a milk-based powdered formula by children in the 0.5 to 1 year old age group. Under this circumstance the drinking water RSC will decline from 70% to 64% if the soy-formula data from VanWinkle et al. (1995) are used.

Geologically, one-third to one-half of the U.S. has access to ground water containing less than 0.5 ppm fluoride (See Figure 3-1), while surface waters exhibit lower geochemical fluoride levels. However, currently, about 69% of U.S. population receives fluoridated water (CDC, 2008), where the natural fluoride level has been augmented through the addition of certified fluoridation chemicals to attain final fluoride concentrations that range between 0.7 and 1.2 mg/L. Consequently, the average fluoride concentration in the nation's drinking water has increased from what it was before systems began the practice of fluoridating drinking water on an experimental basis in 1945 as a public health measure to lower cavities for children and adults (CDC, 1999).

Figure 7-1 indicates that existing data and estimates regarding sulfuryl fluoride in food items support a determination that sulfuryl fluoride is a minor contributor to the diet at current use levels. Recent identification of sulfuryl fluoride as a greenhouse gas (Papadimitriou et al., 2008; Anderson et al., 2009; Mühle et al., 2009) could limit future projected increases in SuF use.

After drinking water and diet, third in contribution to total fluoride intake for humans is toothpaste (Figure 7-1). Most recently introduced in 1955 as a measure to increase protection against dental cavities (Procter and Gamble, 2009), fluoridation of toothpaste with sodium fluoride, monofluorophosphate, or stannous fluoride has grown so that by 1989 almost 95% of the toothpaste sold to the U.S. market was fluoridated (Newbrun, 1989). The relative contribution of fluoride in toothpaste to total intake is highest (21%) for children in the 1- to 3-year age group. This is a consequence of poor swallowing control by children in this age range (Levy et al., 2001). Ingestion of fluoride from toothpaste decreases linearly with increased age as control of swallowing and expectoration reflexes mature (Figure 7-1).

The estimates of average ingestion of fluoride from toothpaste are more uncertain than those for food and drinking water. There are several factors that contribute to the uncertainty including frequency of brushing, the amount of toothpaste used, and individual variability in use. The data in Figure 7-1 represent average values per brushing and a single brushing per day. In U.S. studies of the 1–3 year age group (Levy et al., 1997; Franzman et al., 2006) about 20 to 30 % of children brushed more frequently (Table 4-9). Estimates for population groups greater than 3-years also assume one brushing per day. Data on the frequency of brushing were not identified for school-aged children and adults but a substantial portion of those groups is likely to brush their teeth at least twice a day. Increased brushing frequency would increase intake contributed by toothpaste and its percent of the total. When the data were analyzed using estimates for two brushings per day for all age groups ≥7 years of age, the RSC values for drinking water decreased from 40, 40, 51 and 60% to 36, 37, 47 and 58%. None of these changes was substantial.

Another variable impacting the estimate is the amount of toothpaste placed on the toothbrush. The studies used to quantify the intake were conducted before the guidance (ADA, 1991) to reduce the toothpaste applied from a ribbon to a pea-sized portion was publicized and do not reflect the FDA (2009) recommendation that children younger than 2 years not use toothpaste when brushing their teeth. Decreasing the amounts of toothpaste applied to the toothbrush decreases the fluoride ingested. Finally, all of the dentifrice studies showed that there was high inter-individual variability among the subjects as indicated by the wide confidence bounds on the average values (see Table 4-8). Thus, there is considerable uncertainty in the toothpaste estimates.

Normalized daily intakes of fluoride from soils, indoor dust, ambient air, fluoride-containing pharmaceuticals, episodic dental treatments, and cigarette smoke are minor contributors to total exposures for the average children and adults. Use of fluoride-containing mouthwashes, particularly by children in the 1–7 year age group, is an unquantified exposure that could measurably increase the total estimates from Table 7-2. Mouthwash contributions were not quantified because of a lack of data. In 1983, the now dated National Health Interview Survey found that 5% of children under 5 used mouth rinses, as did 17% of children ages 5 to 17. However, this survey did not estimate fluoride intakes from such products and intakes from fluoridated mouthwashes are not included in the RSC analysis.

## 8. Relationship of Exposure Estimates to Dietary Guidelines

Although, the contributions of various individual media to total fluoride intakes are important, the total intake is even more important from a public health perspective. Fluoride is a nutritionally-active substance with beneficial properties for both teeth and bone (IOM, 1997). Accordingly, total intakes should provide adequate fluoride to meet dietary guidelines without leading to severe dental fluorosis in children and skeletal problems in adults.

### 8.1. Estimates of Daily Dietary Needs.

The National Academy of Sciences (NAS) provided dietary guidelines for fluoride beginning in 1989 (NRC, 1989). The most recent guidelines (Dietary Reference Intakes; IOM, 1997) established Adequate Intake (AI) recommendations for age groupings from infants through adults. The AI is the recommended average daily intake based on observed or experimentally determined approximations, or estimates of adequate nutrient intakes by a group (or groups) of apparently healthy people. The AI is used when a Recommended Dietary Allowance (RDA) cannot be determined because the data are not sufficient to establish average dietary needs based on a biological measure of a person's nutritional status. In the case of fluoride, an easily monitored biological measure of adequacy has not been established.

The AI for fluoride was based on the estimated dietary intakes that have been shown "to reduce the occurrence of dental cavities maximally in a population without causing unwanted side effects including moderate dental fluorosis." IOM (1997) determined that the role of fluoride in protecting tooth enamel, stimulating bone growth, and preventing calcification of soft tissues justified the development of dietary guidelines.

Table 8-1 provides the AI values for each age grouping targeted by IOM and compares the AI levels to the total dietary fluoride intake estimates from Table 7-2. The AI estimates for fluoride include drinking water and identify it as a major contributor to total fluoride (IOM, 1997). It is clear from these data that EPA estimates of current total F intakes meet the AI recommendations for infants, children through age 14, and females, but are below the AI recommendation for adult males.

IOM (1997) did not consider dental decay as a biomarker for low fluoride exposure because decay is associated with a variety of factors and cannot be attributed solely to low fluoride intakes. The AI for infants is based on the daily mean intake of a nutrient from human milk by exclusively breast-fed, healthy infants (IOM, 1997). Intakes from drinking water are included in the AI for fluoride for other age groups; in fact, the IOM (1997) considered ingested drinking water to be the major contributor to total dietary intake.

| Table 8-1. Comparison of Total Fluoride Intake Estimates to the Dietary Adequate Intake (AI). | | |
|---|---|---|
| **Age Range[a] (years)** | **AI [b] (mg/day)** | **F Intake Estimate (mg/day)[c]** |
| 0.5 – <1 | 0.5 | 1.21 |
| 1 – <4 | 0.7 | 1.58 |
| 4 – <7 | 1 | 2.03 |
| 7 – <11 | 2 | 2.16 |
| 11 to 14 | 2 | 2.41 |
| Adult females | 3 | 2.91 |
| Adult males | 4 | 2.91 |

[a]IOM age groups are not an exact match for those used by OW for intake assessments. OW used the best fit of the AI guideline to the age ranges used in this assessment.
[b]IOM, 1997.
[c]From Table 7.2.

## 8.2. Estimates of Tolerable Upper Limit Level

Avoiding intakes of fluoride at levels that could cause adverse effects is as important to public health as providing adequate fluoride for growth, development and maintenance. The IOM (1997) has established Tolerable Upper Intake Level (UL) recommendations for fluoride to protect against dental fluorosis in children and skeletal fluorosis in adults. A UL is the highest average daily nutrient intake level (including drinking water for fluoride) that is unlikely to pose a risk of adverse health effects to almost all individuals in the general population. The UL values established by IOM are based on protection of young children (up through age 8) from moderate dental fluorosis, and are based on a daily dose of 0.1 mg/kg derived from the Dean (1942) data and using a UF of 1. The UL values for children from 6 mo to ≤8 yr range from 0.9 to 2.2 mg/day. The age of concern for moderate dental fluorosis was capped at age 8 because the effects were classified as cosmetic and of greatest concern when the visible anterior teeth were impacted. The risk of cavities occurring on both the anterior and posterior teeth when dental fluorosis is severe, as identified by NRC (2006), was not considered by IOM (1997). The UL values for children older than age 8 and adults are based on skeletal fluorosis as a critical effect and a lack of related symptoms at daily intakes of 10 mg/day for 10 or more years (IOM, 1997). The IOM UL did not consider the data linking fluoride to a possible increase in the risk for bone fractures that were considered by NRC (2006).

The OW dose-response document for fluoride (U.S. EPA, 2010a) developed an estimated RfD of 0.08 mg/kg/day for protection of 99.5% of the vulnerable population against severe dental fluorosis and concluded that this value is also protective against fractures and skeletal effects in adults. The estimated RfD is lower than the equivalent value (UL of 0.1 mg/kg/day) used by IOM (1997). The OW estimated RfD was derived for the 95$^{th}$ percentile lower bound on the concentration of fluoride in drinking water associated with a 0.5% prevalence of severe dental fluorosis in the population studied by Dean (1942), equivalent to a fluoride dose of 0.07

mg/kg/day. A 0.01 mg/kg/day contribution from the diet, as derived from McClure (1943), was added to the drinking water component to yield the 0.08 mg/kg/day RfD. The RfD derivation can be found in the EPA (2010a) companion document, *Fluoride: Dose-Response Analysis for Non-cancer Effects*.

The RfD (mg/kg/day) was converted to age-specific oral exposure benchmarks (mg/day) that should be protective for severe dental fluorosis in most children and skeletal effects in most adults using mean bodyweights for each age group from (EPA, 2004) as reported in Table 8-2. They are compared in the table to both the IOM (1997) UL guidelines and the OW total daily intake estimates from this document.

**Table 8-2. Comparison of Total Fluoride Intake Estimates to the IOM (1997) Tolerable Upper Intake Level and the OW Age-Specific Benchmarks**

| Age Group (years) | OW Benchmark[a] (mg/day) | UL[b] (mg/day) | Intake Estimate (mg/day)[c] |
|---|---|---|---|
| 0.5 – <1 | 0.72 | 0.9 | 1.21 |
| 1– <4 | 1.12 | 1.3 | 1.58 |
| 4 – <7 | 1.68 | 2.2 | 2.03 |
| 7 – <11 | 2.56 | 2.2 for 8 year olds | 2.16 |
| | | 10 for >8 year olds (skeletal fluorosis)[d] | |
| 11 to 14 | 4.08 | 10 (skeletal fluorosis) | 2.41 |
| Adult females | 5.6 | 10 (skeletal fluorosis) | 2.91 |
| Adult males | 5.6 | 10 (skeletal fluorosis) | 2.91 |

[a]The OW benchmarks were established to protect against severe dental fluorosis in children up to age 14 and to protect against skeletal fractures and skeletal fluorosis in adults.

[b]IOM UL values were established to protect against dental fluorosis up to age 8.

[c]From Table 7-2.

[d]The IOM values for ages > 8 years were established to protect against skeletal fluorosis.

## 8.3. Exposure Profiles

The data in Table 8-2 indicate that some children drinking water at the 90$^{th}$ percentile intake level up to about age 7 are being exposed to fluoride on a daily basis at levels at or higher than estimated acceptable intake levels when the concentration of fluoride in their drinking water is at or above 0.87 mg/L. Figure 8-1 shows the relationship between current intake estimates (drinking water intake at the 90$^{th}$ percentile level) and the OW RfD-derived benchmarks in units of mg/day. The RfD-derived benchmarks for each age group are shown as the solid black line in Figures 8-1 through 8-4. When examining Figure 8-1 it is important to remember that the RfD represents an exposure that is estimated to provide the anticaries benefits from fluoride without causing severe dental fluorosis in 99.5% of the children who drink water with 0.87 mg/L F at a 90$^{th}$ percentile intake level and have average intakes from other media during the period of secondary tooth formation. Based on the dose-response for severe dental fluorosis in EPA (2010a) only 0.5% or fewer of children consistently ingesting fluoride at a levels equivalent to the RfD for a several month period would be at risk of experiencing severe dental fluorosis in two or more teeth.

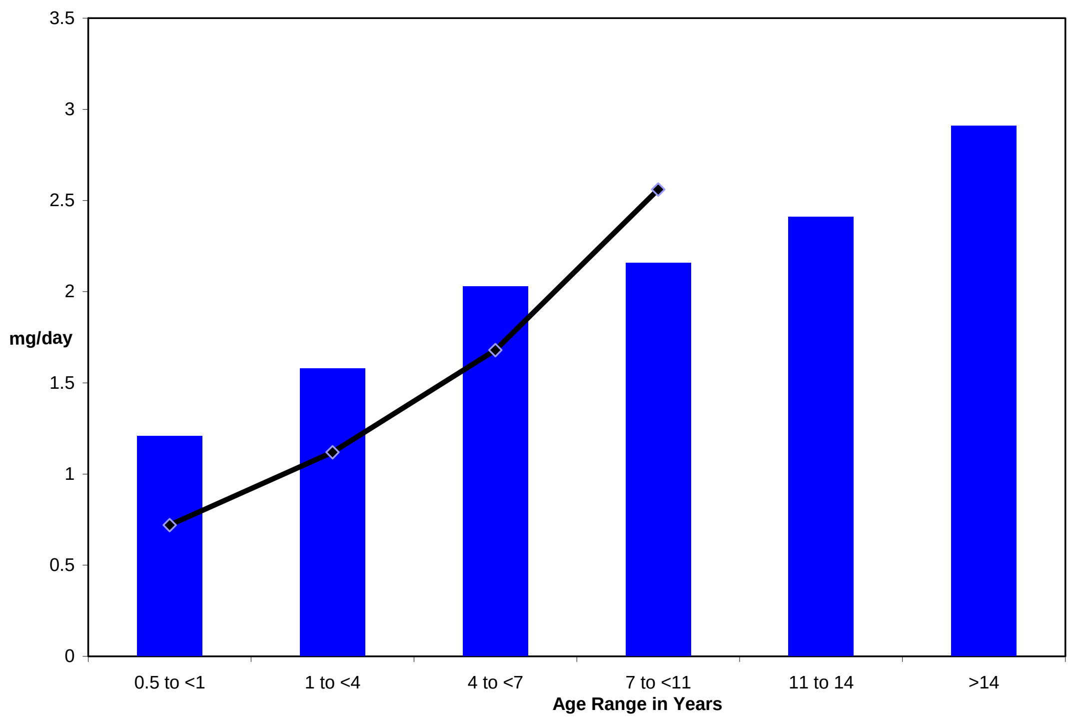


**Figure 8-1. Total Daily Fluoride Intake Estimates Relative to the Proposed RfD Using 90th Percentile Drinking Water Intake Data for Consumers Only and the Mean Drinking Water Fluoride Concentration (0.87 mg/L)**

If the drinking water intake level is adjusted to an average intake to match the average values used for the other exposure media, the relationship between exposure intakes and the RfD-equivalent intake changes (Figure 8.2). Children with average intake of all media in the younger age groups would still be slightly over exposed if the drinking water concentration were 0.87 mg/L. At higher concentrations in drinking water, the number of children at risk for severe dental fluorosis would likely increase. Risk would also increase if the fluoride from any other exposure media were greater than the values utilized by EPA in this assessment.

The OW RfD identifies a level of exposure that is considered to be acceptable for the general population. Levels above the RfD are not necessarily unacceptable but risk is considered to increase as the difference between the RfD-equivalent and the dose increases.

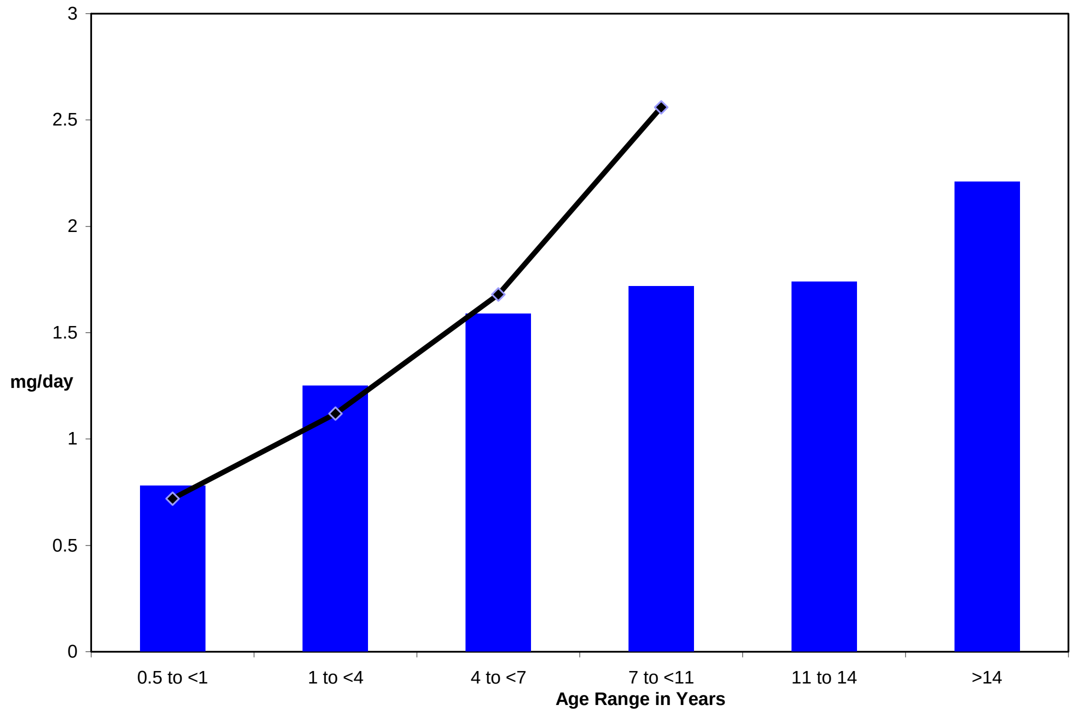


**Figure 8-2. Total Daily Fluoride Intake Estimates Relative to the Proposed RfD Using the Mean Drinking Water Intake Data for Consumers Only and the Mean Drinking Water Fluoride Concentration (0.87 mg/L)**

In any population, dietary intakes and food choices change from day to day. Each person's daily fluoride exposure will be influenced by what they eat each day and how they brush their teeth. No one is average on a continuous basis. Many people will consistently be exposed to higher levels of each fluoride-containing medium others will consistently be exposed to lower levels than depicted. Children in communities that routinely exceed the current SMCL for fluoride during the period when their teeth are forming will be particularly vulnerable to developing severe dental fluorosis. Figure 8-3 depicts the impact of an average drinking water intake for consumers only and the 90th percentile fluoride concentration for all systems reporting detections of fluoride. Figure 8-4 depicts the age-specific intakes for populations where drinking water intakes are at the 90$^{th}$ percentile level and the fluoride concentration (1.76 mg/L) is the average for those systems that reached or exceeded the SMCL of 2 mg/L at least once during the last 4 years of the ICR monitoring period for the second six-year review of regulations (Table 3-2). Children in areas of the country with high geological levels of fluoride and resultant higher levels in their drinking water who are also at the high end of the drinking water intake distribution are those with the greatest risk for severe dental fluorosis.

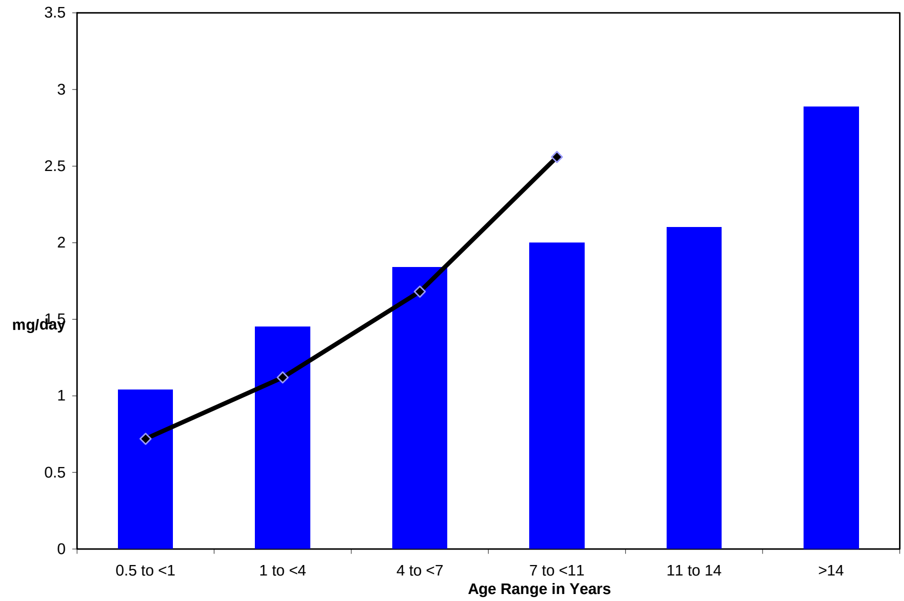


**Figure 8-3. Total Daily Fluoride Intake Estimates Relative to the Proposed RfD using Mean Drinking Water Intakes for Consumers Only and the 90th percentile Fluoride Concentration for all Systems Reporting Detections of Fluoride.**

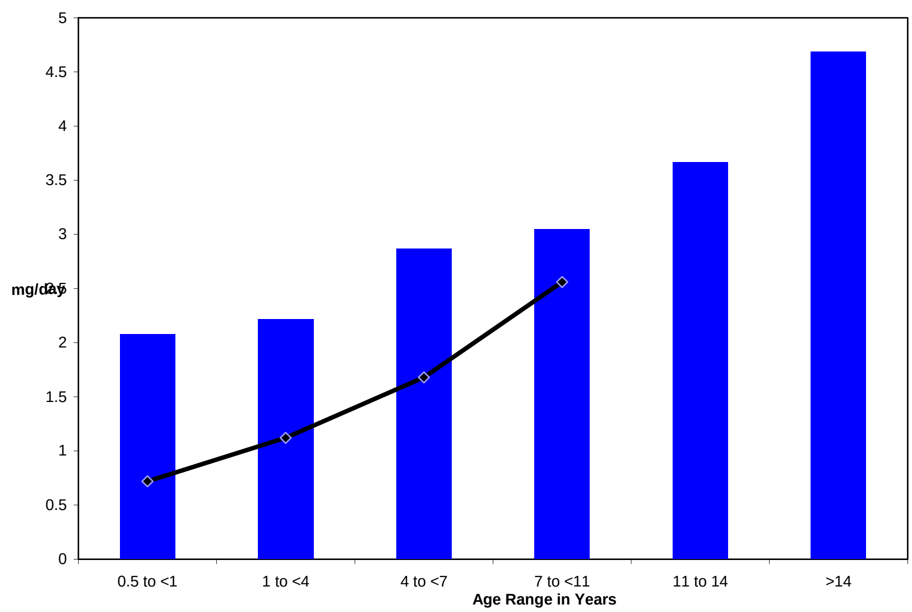


**Figure 8-4. Total Daily Fluoride Intake Estimates Relative to the Proposed RfD using 90$^{th}$ Percentile Drinking Water Intakes for Consumers Only and Average Concentration (1.76 mg/L) for those Systems that Reached or Exceeded the SMCL of 2 mg/L at Least Once During the ICR Monitoring Period for the Second Six-year Review.**

In the case of fluoride, there are data on prevalence of dental fluorosis to support a conclusion that fluoride exposure levels among the population have increased in the last 40 to 50 years resulting in an increase in dental fluorosis (Iida and Kumar, 2009; CDC, 2005). The prevalence of dental fluorosis has increased from 10–12% in the areas with about 1 mg/L in drinking water at the time of Dean (NRC, 1993) to 23 % in 1986/87 (NRC, 1993; Iida and Kumar, 2009) and to 32% in the 1999-2002 NHANES survey (CDC, 2005). The 1986/1987 data come from the National Survey of Oral Health of U.S. School Children, which examined 40,693 subjects. The NHANES survey included a smaller set of subjects (17,092) at ages greater than 2 years (CDC, 2005). Comparable data are not available for severe dental fluorosis.

The CDC (2005) report found that the prevalence of fluorosis was higher in the 12–15 and 16–19 year age groups during the 1999–2002 survey than in the 20–39 year old age groups, which may be a reflection of recent increases in total fluoride exposure. The data also indicated that posterior teeth were impacted to a greater extent than the visible anterior teeth and that there was a higher prevalence among the Non-Hispanic African Americans than Non-Hispanic Caucasian Population. Most of the fluorosis reported in the CDC (2005) report was very-mild or mild, conditions that are associated with decreases in tooth decay. However, there were cases of moderate/severe dental fluorosis combined and the percentages reported were higher for the age groups younger than 20 years old than for older individuals indicating that increases in total fluoride intakes may be relatively recent.

## 8.4. Summary of findings

The OW conducted the Exposure and Relative Source Contribution assessment in order to determine the relationship of total fluoride intakes to the inorganic fluoride RfD from the companion dose-response assessment (U.S. EPA, 2010a). The relative contribution of ingested drinking water from public drinking water systems to total exposures was also examined.

The EPA MCLG/MCL for fluoride was established in 1986 and determined to be protective for Stage III (crippling) skeletal fluorosis. The determination of the MCLG/MCL included an assumption that drinking water contributed 100% of the exposure because the data used for quantification were derived from measures of the fluoride in the drinking water among the cases of Stage III skeletal fluorosis that provided the point of departure for the calculation.

The NRC (2006) examination of the MCL/MCLG for fluoride was an outgrowth of the first six-year review of the 1986 fluoride drinking water regulation as mandated by the 1996 SDWA and recognition by EPA of the number of scientific studies on the bone and dental effects of fluoride that were published after the regulation (U.S. EPA, 2003). The NRC published the report of their effort in 2006 as: *Fluoride in Drinking Water: A Scientific Review of EPA's Standards.* The NRC committee concluded that EPA's current MCLG of 4 mg/L for fluoride should be lowered to reduce the risk of severe enamel fluorosis and minimize the risk for bone fractures and skeletal fluorosis in adults. It charged the OW with conducting a dose-response assessment for the critical noncancer effects of fluoride on teeth and bone (U.S. EPA, 2010a) and the exposure and relative source assessment presented in this report. Through this effort, EPA has concluded that:

- Some young children are being exposed to fluoride up to about age 7 at levels that increase the risk for severe dental fluorosis.
- The contribution of residential tap water to total ingested fluoride is lower that it was in the past.

- Use of fluoridated water for commercial beverage production has likely resulted in increased dietary fluoride in purchased beverages, adding to the risk for over-exposure.
- The increase of fluoride in solid foods because of fluoridated commercial process water is more variable than that for beverages.
- Incidental toothpaste ingestion is an important source of fluoride exposure in children up to about 4 years of age. However, use of fluoridated toothpaste is not recommended for children under age 2 according to FDA guidance and package labeling suggesting the need for greater parental awareness of the FDA (2009) recommendations.
- Ambient air, soils, and sulfuryl fluoride residues in foods are minor contributions to total fluoride exposure.

Based on the data collected and evaluated by the OW, it is likely that most children, even those that live in fluoridated communities, can be over-exposed to fluoride at least occasionally. Children who live in communities where the fluoride concentration routinely falls between 2 mg/L and 4 mg/L have an even greater opportunity for over-exposure unless parents follow the EPA public notification advice not to allow their children to routinely drink the tap water until they are nine years of age (the upper age limit for the current public notification), and consult with their dental professions regarding use of fluoridated dental products. The impact of the elevated intakes on the risk for severe dental fluorosis in one or more teeth depends on the timing, frequency and duration of the over-exposures.

The data from this report and its companion dose-response assessment (U.S. EPA, 2010a) will be used by the EPA in order to determine whether lowering the MCLG and/or MCL for fluoride will provide a meaningful opportunity to reduce the risk for severe dental fluorosis and skeletal effects among populations served by public drinking water systems. The EPA is required to consider whether the costs of reducing fluoride in public water supplies are justified by the health benefits accrued through such a change. Regulatory decisions related to the MCLG and MCL are separate from the assessment of hazard in the NRC (2006) report, the OW dose-response report (U.S. EPA, 2010a), and this exposure and RSC document.

The OW's exposure and relative source contribution analysis accomplished each of its desired objectives within the limitations of the data provided in the published literature and the monitoring information from the second six-year review ICR data. The output of the analysis is age-group specific for children at risk of developing severe dental fluorosis and is presented in a format that will aid the OGWDW in characterizing opportunities for reducing population risk from fluoride in public drinking water systems. The data are intended as a resource for facilitating any necessary adjustment in the regulatory nonenforceable MCLG and enforceable MCL.

It is important to remember, however, that the exposure quantification provided follows the policy guidelines from EPA (2000b) using average exposure estimates for all media other than drinking water, the average drinking water concentration for systems that detect fluoride, and $90^{th}$ percentile drinking water intakes. Thus, the intake estimates are more representative of average consumers than they are for individuals residing in areas of the country where the average drinking water concentration falls between the 2 mg/L and 4 mg/L concentrations allowed by the NPDWR rather than the 0.87 mg/L that is representative of the country as a whole and for those where the drinking water has very low fluoride concentrations (< 0.1 mg/L). For children residing in areas where the fluoride levels are close to the MCL (4 mg/L) the risk for severe dental fluorosis is considerably higher. Some adolescents and adults receiving drinking water that is consistently close to the MCL can easily exceed the 6 mg/day where the risk for effects on bone are considered to be a concern (WHO, 2001).

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# APPENDICES

**Appendix A: Fluoride Chronic Dietary Exposure Analysis**

**Appendic B: Sulfuryl Fluoride: Estimates of Fluoride Exposure from Pesticidal Sources – Customized Age Groups**

**UNITED STATES ENVIRONMENTAL PROTECTION AGENCY**
**WASHINGTON, D.C. 20460**

**OFFICE OF PREVENTION, PESTICIDE AND TOXIC SUBSTANCES**

MEMORANDUM

Date: 6 May 2009

SUBJECT: Fluoride Chronic Dietary Exposure Analysis

| | |
|---|---|
| PC Code: None | DP Barcode: 362183 |
| MRID No.: 47594701 | Registration No.: None |
| Petition No.: None | Regulatory Action: None |
| Assessment Type: Single Chemical | Registration Case No.: None |
| TXR No.: None | CAS No.: 16984-48-8 |

FROM: Michael A. Doherty, Ph.D., Senior Chemist
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**Background**

The Agency's Office of Water (OW) is currently examining its drinking water standard for fluoride (F). As part of that examination, they are determining relative source contributions for fluoride exposure (*i.e.*, how much fluoride comes from various sources such as toothpaste, natural residues in foods, etc.). The Office of Pesticide Programs (OPP) has been asked to supply OW with estimates of dietary exposure to fluoride that results from the use of pesticides. OPP has identified two pesticides, cryolite and sulfuryl fluoride (SF), whose use results in fluoride levels which may be elevated above background levels in treated foods. In 2006, OPP completed an aggregate human health risk assessment for fluoride that addressed dietary exposure to F from these two sources (M. Doherty, D312659, 18 January 2006). Since that time, the registrant (Dow AgroSciences; DAS) for sulfuryl fluoride has provided the Agency with information that will support a more refined estimate of F exposure attributable to the use of sulfuryl fluoride. This assessment incorporates refinements to fluoride residue levels, taking into account the information from DAS, and examines the contribution of various crops and crop groups to pesticidal F exposure. Note that because of the purpose of this assessment and the on-going work by OW, this document presents exposure estimates for fluoride and not risk estimates.

**Executive Summary**

Chronic dietary (food only) exposure assessments were conducted using the Dietary Exposure Evaluation Model DEEM-FCID™, Version 2.03 which use food consumption data from the U.S. Department of Agriculture's Continuing Surveys of Food Intakes by Individuals (CSFII) from 1994-1996 and 1998. Two analyses have been completed for sulfuryl fluoride, the first to address the structural fumigation uses, wherein foods may receive inadvertent treatment, and the second to obtain estimates for the food uses of the product, wherein foods are intentionally fumigated to treat pest problems. An additional, third analysis was conducted to determine the food-specific contributions to F exposure due to the use of cryolite.

All analyses include the use of percent crop treated (% CT) information and incorporate anticipated residues. The % CT and anticipated-residue values represent a range of refinements in which some conservatism remains. As more data become available and are validated by the Agency, further refinement to the exposure estimates may be possible. On the other hand, this assessment departs to some extent from regular HED practice by using very low percent crop treated estimates and anticipated residues predicted from data bearing on historical application rates. Regulatory measures, such as frequent mandatory reporting, may be appropriate to insure that usage and application rates do not change. Fluoride exposure estimates from pesticidal sources range from 0.0015 to 0.0063 mg/kg/day, depending on the population subgroup.

## I. Residue Information

*Fluoride from Cryolite*. Residue and % CT estimates in the cryolite assessment are identical to those used in the previous assessment (M. Doherty, D309013, 12 October 2004). That assessment is based on average residue values from field trials and incorporates %CT estimates for the majority of the foods in the analysis. The resulting exposure estimates are considered to be moderately refined. OPP notes that the analytical method used to obtain the residue estimates in the cryolite field trials reports total fluoride and does not differentiate between the various aluminum-fluoride species that may be present in the treated commodities. Therefore, the residue estimates associated with the use of cryolite likely represent an overestimate of fluoride anion residues resulting from cryolite use.

*Fluoride from Sulfuryl Fluoride*. As in previous assessments (*e.g.*, M. Doherty, D317731, 18 January 2006), average residue values from fumigation trials conducted at the maximum total application rate of sulfuryl fluoride (1500 mg·h/L) were used to assess dietary exposure, except as noted below. OPP has received a significant amount of data depicting F residues in foods at various treatment rates, and for many foods there is a relationship between treatment rate and terminal F residues. Dow AgroSciences maintains a database which tracks sites where sulfuryl fluoride is used as well as various parameters associated with each fumigation, including the actual treatment rates. For foods with demonstrable rate/residue relationships, the average residues from trials at the maximum rate have been adjusted to their average-fumigation-rate equivalent using linear regression (Table 1). The regression analysis, as described in the submission, is as follows and is in line with OPP guidance for deriving anticipated residues (USEPA/OPP, 15 June 2000):

> [Regressions] were based on relative concentrations and relative dose levels. Specifically, within each commodity, one or more samples treated at dose levels closest to the maximum label rate (1500 oz-hr) were designated as "reference samples". Reference dose levels and reference concentrations were derived as the average dose levels and concentrations associated with these reference samples. The dose levels and concentrations associated with all samples were then expressed as relative dose level and relative concentrations (percent of the reference dose level and of the reference concentration), and these relative values were used in the linear regression models of the form:
>
> Relative Concentration = a + b × Relative dose,
>
> where a and b are the intercept and slope. The regression models described above were used to estimate the anticipated concentration at the average (historical) dose levels summarized above. In addition, predicted levels at the maximum dose rate of 1,500 oz-hr were also derived. Specifically, the concentration at dose level D was derived as:
>
> Concentration at Dose D = Reference concentration × [a + ( b × Dose D/Reference dose)],
>
> where a and b are the intercept and slope from the corresponding regression model. Note that regression models were used only for commodities with more than two data points which spanned a range of at least 600 oz-hr and for which the regression p-value was 20% or lower. [The p-value of 20% was footnoted as follows: *The regression models for four commodities (corn flour, figs, raisins and white rice) had p-values lower than 20% but higher than the 5% level typically used to represent statistical significance. Nevertheless, the regression models were used to predict concentration levels for these four commodities because a visual examination of the data indicated a linear relationship between [dose] and residues. The regression models produced conservative estimates of anticipated residues at the average [dose], since the estimates were comparable to, if not higher than, the observed residues at the maximum [dose]*.]

For all foods in the analysis, DAS has used a correction factor to account for the inability of the analytical method to measure total fluoride. The correction factor is commodity-specific and the values range from 0.37 to 1.0. Residue estimates are divided by the correction factor to obtain an estimated total fluoride concentration. The data used to obtain the correction factors are not available to OPP at this time and therefore the factors cannot be verified. The factors, being ≤ 1, result in residue estimates that are greater than or equal to prior OPP assessments; therefore, OPP is accepting them at this time without further review. The study should be submitted for review by the Agency. EPA will revise residue estimates, as needed, following review of these data.

HED has verified the regression parameters and analysis presented by DAS and, except for hazelnuts (filberts), concurs that the anticipated residues are not likely to underestimate actual residues resulting from the use of SF at the average dose levels reported to date. Hazelnuts are reported as having a regression with a negative intercept. Conceptually, the intercept represents the background residue in the untreated matrix; therefore, a negative intercept does not make sense from a residue perspective and is considered to be an artifact of the regression process. Therefore, for hazelnuts, OPP has recalculated the anticipated residues assuming an intercept of zero (the slope was not recalculated).

Sulfuryl fluoride currently has two use strategies (1) pest control in structures via structural fumigations and (2) control of pests in foods via direct fumigation of foods. During structural fumigation, facilities are to be emptied of foods to the extent possible. Nevertheless, there will be some foods that remain in the structure and that will be inadvertently treated with SF. OPP is assessing these two uses separately. Residue data are not available for a number of commodities that may be treated with SF. For those commodities surrogate data have been used (*e.g.*, the residue estimate for figs is used for a number of other dried fruits). Residue estimates are summarized in Table 5 for both the structural and food fumigation uses. As previously noted, the residue estimates for fluoride coming from cryolite are identical to those used in the previous assessment. A complete listing of the residue inputs is included in Attachments 1-3.

Table 1. Reference Values and Regression Parameters for Determination of Fluoride Anticipated Residues.

| Food | Reference Dose, mg·hr/L | Residue at Reference Dose, ppm | Slope | Intercept | Average Rate, mg·hr/L | | Residue at Average Rate, ppm | |
|---|---|---|---|---|---|---|---|---|
| | | | | | Structural | Food | Structural | Food |
| Barley | 1628 | 2.95 | 1.149 | 0.611 | 590 | 610 | 3.03 | 3.07 |
| Cocoa Beans | 1483 | 5.12 | 1.082 | 0.034 | 790 | 790 | 3.12 | 3.12 |
| Corn | 1549 | 1.78 | 0.868 | 0.383 | 670 | 390 | 1.35 | 1.07 |
| Corn Flour | 1573 | 21.73 | 0.340 | 0.694 | 670 | 390 | 18.22 | 16.91 |
| Corn, Popcorn | 1505 | 0.95 | 0.801 | 0.513 | 1340 | 1340 | 1.16 | 1.16 |
| Dates | 1484 | 0.70 | 0.333 | 0.748 | 380 | 380 | 0.58 | 0.58 |
| Dried Plums | 1543 | 0.85 | 0.470 | 0.537 | 380 | 380 | 0.56 | 0.56 |
| Figs | 1524 | 1.14 | 0.239 | 0.657 | 380 | 380 | 0.81 | 0.81 |
| Oats | 1534 | 7.90 | 0.859 | 0.408 | 560 | 560 | 5.70 | 5.70 |
| Pistachios | 1517 | 4.10 | 1.248 | 0.094 | 350 | 540 | 1.56 | 2.21 |
| Raisin | 1545 | 0.05 | 1.033 | 14.612 | 380 | 380 | 0.74 | 0.74 |
| Rice, Brown | 1558 | 5.68 | 0.892 | 0.250 | 620 | 960 | 3.43 | 4.54 |
| Wheat Flour | 1533 | 25.93 | 0.770 | 0.550 | 590 | 610 | 21.96 | 22.22 |
| Wheat Germ | 1512 | 67.95 | 0.700 | 0.220 | 590 | 610 | 33.50 | 34.13 |
| Wheat Grain | 1539 | 2.92 | 0.382 | 0.669 | 590 | 610 | 2.38 | 2.39 |
| Rice, White | 1509 | 1.90 | 1.290 | 0.987 | 620 | 960 | 2.88 | 3.44 |
| Almonds | 1539 | 4.70 | 0.470 | 0.610 | 350 | 540 | 3.37 | 3.64 |
| Pecans | 1533 | 8.55 | 0.831 | 0.091 | 350 | 540 | 2.40 | 3.28 |
| Walnuts | 2460 | 2.25 | 1.939 | 0.336 | 350 | 540 | 1.38 | 1.71 |
| Hazelnuts† (Space)* | 1576 | 2.32 | 2.014 | -0.179 | 350 | -- | 1.03 | -- |
| Hazelnuts† (Food) | 1576 | 1.81 | 2.749 | -0.861 | -- | 540 | -- | 1.70 |

† The calculated residues assume the intercept is zero.
* Data are from hazelnuts without shell

## II. Use Information

*Food Fumigations*. Information regarding % $CT_F$ (% CT for food fumigations) was submitted to the Agency by DAS. OPP's Biological and Economic Analysis Division (BEAD) has examined the information submitted by DAS and has derived recommended %$CT_F$ values for use in the dietary exposure analysis for food fumigations (C. Cook and E. Rim, D361041, 30 April 2009). The recommendations from BEAD are summarized in Table 2.

Table 2. Summary of Revised Estimates of Percent Commodity Directly Treated with Sulfuryl Fluoride.

| BEAD Commodity Grouping | Commodity | Percent Commodity Treated | | |
|---|---|---|---|---|
| | | DAS | BEAD | |
| | | | Estimate | Recommended |
| Meats and Cheese | Cheese[1] | 0.0 % | 0.0 % | 0.0 % |
| | Ham[1] | 0.0 % | 0.0 % | 0.0 % |

Table 2. Summary of Revised Estimates of Percent Commodity Directly Treated with Sulfuryl Fluoride.

| BEAD Commodity Grouping | Commodity | Percent Commodity Treated | | |
|---|---|---|---|---|
| | | DAS | BEAD | |
| | | | Estimate | Recommended |
| | Beef (Dried) | 0.0 % | 0.0 % | 0.0 % |
| Quarantined Uses[2] | Coconut | 0.1 % | 0.0 % | 0.1 % |
| | Coffee Bean | 0.1 % | 0.0 % | 0.1 % |
| | Macadamia Nut | 0.1 % | 0.0 % | 0.1 % |
| | Ginger | 0.1 % | 0.0 % | 0.1 % |
| Coarse Grains | Barley[3] | 0.1 % | 0.1 % | 0.1 % |
| | Corn[6] | 0.1 % | 0.1 % | 0.1 % |
| | Cottonseed[3] | 0.1 % | 0.1 % | 0.1 % |
| | Millet[3] | 0.0 % | 0.1 % | 0.1 % |
| | Oats[6] | 0.1 % | 0.1 % | 0.1 % |
| | Rice Hulls[3] | 0.0 % | 0.1 % | 0.1 % |
| | Sorghum[6] | 0.1 % | 0.1 % | 0.1 % |
| | Triticale[3] | 0.0 % | 0.1 % | 0.1 % |
| Processed Commodities | Corn – Flour, Grits, Meal | 0.1 % | 0.0 % | 0.1 % |
| | Herbs And Spices | 0.1 % | <0.1 % | 0.1 % |
| | Popcorn | 0.1 % | <0.1 % | 0.1 % |
| | Rice – Flour, Bran | 3.0 % | 0.0 % | 3.0 % |
| | Wheat – Flour, Germ, Bran, Shorts, Milled Byproducts | 0.1 % | 0.0 % | 0.1 % |
| Stored Commodities | Peanut[6] | 0.1 % | 0.6 % | 0.6 % |
| | Wheat[6] | 0.1 % | 0.4 % | 0.4 % |
| | Rice[6] | 3.0 % | 0.9 % | 3.0 % |
| | Wild Rice | | 0.9 % | 3.0 % |
| Nuts[4] | Almonds | 10.0 % | 2.2 % | 10.0 % |
| | Beechnut | 0.0 % | 2.2 % | 10.0 % |
| | Brazil Nut | 0.1 % | 2.2 % | 10.0 % |
| | Butternut | 0.0 % | 2.2 % | 10.0 % |
| | Cashew | 0.1 % | 2.2 % | 10.0 % |
| | Chestnut | 0.1 % | 2.2 % | 10.0 % |
| | Chinquapin | 0.0 % | 2.2 % | 10.0 % |
| | Filbert | 0.1 % | 2.2 % | 10.0 % |
| | Hickory Nut | 0.1 % | 2.2 % | 10.0 % |
| | Pecans | 0.1 % | 2.2 % | 10.0 % |
| | Pine Nut | 0.1 % | 2.2 % | 10.0 % |
| Methyl Bromide Critical Use Exemption Commodities | Pistachio[1] | 0.1 % | 27.0 % | 27.0 % |
| | Walnuts[1] | 20.0 % | 99.0 % | 99.0 % |
| | Dates[1] | 40.0 % | 42.0 % | 42.0 % |
| | Prunes, Raisins, Figs[1] | 40.0 % | 69.0 % | 69.0 % |
| | Other Dried Fruit[5, 7] | 0.1 % | 69.0 % | 69.0 % |
| | Dried Beans1 | 100.0 % | 92.0 % | 100.0 % |
| | Legumes (Dried, except Chickpea & Cowpea) [5, 7] | 0.1 % | 92.0 % | 100.0 % |
| | Cocoa Beans[1] | 100.0 % | 100.0 % | 100.0 % |

1. Based on BEAD calculations from comparative methyl bromide usage.
2. Currently fumigated with methyl bromide to fulfill federal or state quarantine requirements.
3. Estimates based on PCT for sorghum and oats. BEAD assumes similar categorization of small coarse grains.
4. This group did not request a methyl bromide CUE and BEAD is anticipating sulfuryl fluoride to replace methyl bromide. BEAD estimates PCT to be no more than the DAS estimate for almonds. Based on the pest spectrum, nuts are primarily treated with phosphine, with some treated with Propylene Oxide.
5. Based on estimates from similar methyl bromide critical use exemption commodities
6. Based on reports of methyl bromide usage by USDA NASS
7. BEAD's estimate is based on a commodity with a similar use pattern; therefore BEAD defaults to the higher of the two estimates of the original commodity.

*Structural Fumigations*. As in previous assessments, information regarding the percentage of facilities treated, the number of days the facilities are in operation, and the amount of material onsite during fumigation has been used to obtain % CT estimates associated with structural fumigations (% $CT_S$). The estimate is calculated as follows:

% $CT_S$ = % facilities treated × number of days production held during fumigation × number of fumigations per year ÷ number of operating days per year,

where the percent of both the grain mills and processing facilities treated equals 40%, the number of days production held in the facility during a fumigation is 2 days for grain mills and 1 day for processing facilities, the number of fumigations per year is 3 for grain mills and 2.5 for processing facilities, and both grain mills and processing facilities are in operation for 300 days per year. These values give a % $CT_S$ of 0.8 for grain mills and a % $CT_S$ of 0.4% for processing facilities. Given knowledge of industry practices, EPA believes this to be a conservative manner of estimating residues resulting from structural fumigation; however, with the current label directions, further refinement is not appropriate.

There is the potential for “sequential” treatment of certain foods. For example, wheat grain could be inadvertently during a structural fumigation, that grain milled into flour, and then a portion of that same flour could be inadvertently treated during a mill fumigation. Past assessments have taken the extremely conservative assumption that the probability of sequential treatment occurring is 100%. This assessment uses % $CT_S$ information to derive a more realistic picture of the likelihood of sequential treatments. There are four scenarios that describe the sequential treatment possibilities associated with structural fumigations:

1. Flour is incidentally treated, source grain is incidentally treated
2. Flour is incidentally treated, source grain is not incidentally treated
3. Flour is not incidentally treated, source grain is incidentally treated
4. Flour is not incidentally treated, source grain is not incidentally treated.

The likelihood of each scenario can be estimated by multiplying the % $CT_S$ estimates for the various combinations (Table 3).

Table 3. Likelihood of Sequential Treatment with Sulfuryl Fluoride from Structural Fumigations.

| | Flour Treated (0.4%)* | Flour Not Treated (99.6%) |
|---|---|---|
| Grain Treated (0.8%) | 0.0032% (Scenario 1) | 0.797% (Scenario 3) |
| Grain Not Treated (99.2%) | 0.397% (Scenario 2) | 98.8% (Scenario 4) |

* Parenthetical values are % of facilities treated. Values in the table are obtained by multiplying the % of facilities treated for each scenario (*e.g.*, % of flour bearing residues from both mill fumigation and grain fumigation = 0.004 × 0.008 = 0.000032 = 0.0032%).

Combining the scenario likelihood values, residue estimates for flours, and empirical factors for processing grains into flours (0.38 for wheat, 0.73 for other grains) gives the weighted average residue values presented in Table 4. These values are used to estimate the exposure from grain flour as a result of structural fumigations. Flour residue estimates for food fumigations are based on the regression analyses discussed above. Exposure estimates from the inadvertent treatments and food treatments are added together in a separate step in the assessment process to provide estimates of overall dietary exposure from the uses of SF.

Table 4. Weighted average residue estimates for grain flours resulting from structural treatment.

| Flour Source | Treated Grain Residue, ppm | Analytical Correction Factor | Corrected Treated Grain Residue, ppm | Flour Residue, ppm | | | Weighted Average, ppm[b] |
|---|---|---|---|---|---|---|---|
| | | | | Processed from Treated Grain[a] (0.797%)[*] | Treated Flour (0.397%) | Processed from Treated Grain + Treated Flour (0.0032%) | |
| Barley | 3.03 | 0.83 | 3.65 | 2.66 | 33.70 | 36.36 | 0.156 |
| Corn | 1.35 | 1.00 | 1.35 | 0.99 | 18.22 | 19.21 | 0.081 |
| Oats | 5.70 | 0.70 | 8.14 | 5.94 | 72.28 | 78.22 | 0.337 |
| Rice | 2.88 | 0.56 | 5.14 | 3.75 | 32.50 | 36.25 | 0.160 |
| Wheat | 2.38 | 0.83 | 2.90 | 1.10 | 31.40 | 32.50 | 0.134 |

[a] Grain residue × processing factor (0.38 for wheat, 0.73 for others)
[b] Weighted Average = Σ (Flour Residue × % Likelihood from Table 3 ÷ 100). The contribution from Scenario 4 to the weighted average is zero since no treatments were involved (flour residue = 0 ppm).
[*] Parenthetical values are % likelihood estimates from Table 3.

Table 5 summarizes the inputs used for the dietary exposure assessment for fluoride coming from the uses of sulfuryl fluoride. Where appropriate, the regression parameters (Table 1) were applied to the available residue data to obtain residue estimates based on actual use patterns. The %CT information (Table 2) is also summarized in Table 5, and was used to derive weighted averages (Tables 3 and 4) for residues associated with structural fumigations and processed grain commodities.

Table 5. Summary of Analytical Correction Factors, Percent Crop Treated, and Residue Estimates for the Dietary Exposure Analysis of Fluoride from Use of Sulfuryl Fluoride.

| Food | Analytical Correction Factor | Structural | | | Food | | |
|---|---|---|---|---|---|---|---|
| | | Residue Value Data Source | % $CT_S$[*] | Residue, ppm[†] | Residue Value Data Source | % $CT_F$[*] | Residue, ppm[†] |
| Alfalfa, seed | 0.51 | See sorghum | 0.4 | 20.4 | | | |
| Almond | 0.37 | Regression[‡] | 0.4 | 9.2 | Regression | 10 | 9.7 |
| Almond, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Amaranth, grain | 0.51 | See sorghum | 0.4 | 20.4 | | | |
| Apple, dried | 0.79 | See figs | 0.4 | 1.0 | See figs | 69 | 1.0 |
| Apricot, dried | 0.79 | See figs | 0.4 | 1.0 | See figs | 69 | 1.0 |
| Arrowroot, flour | 0.70 | Non-mixed wheat flour | 0.4 | 31.4 | | | |
| Banana, dried | 0.79 | See figs | 0.4 | 1.0 | See figs | 69 | 1.0 |
| Barley, pearled barley | 0.83 | Regression[‡] | 0.8 | 3.65 | Regression[‡] | 0.1 | 3.7 |
| Barley, flour | 0.83 | Wtd avg (see text) | ** | 0.156 | Regression[‡] with 0.73X factor | 0.1 | 3.7 |
| Barley, bran | 0.83 | See Barley, pearled | 0.8 | 3.65 | Regression[‡] with 2.56X factor | 0.1 | 3.7 |
| Basil, fresh leaves | 0.69 | Avg @ 1569-1596 rate[§] | 0.4 | 67.1 | | | |
| Basil, dried leaves | 0.69 | Avg @ 1569-1596 rate | 0.4 | 67.1 | Avg @ 1569-1596 rate | 0.1 | 67.1 |
| Bean, black, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Bean, broad, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Bean, cowpea, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Bean, great northern, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Bean, kidney, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Bean, lima, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Bean, mung, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Bean, navy, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Bean, pink, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |

Table 5. Summary of Analytical Correction Factors, Percent Crop Treated, and Residue Estimates for the Dietary Exposure Analysis of Fluoride from Use of Sulfuryl Fluoride.

| Food | Analytical Correction Factor | Structural | | | Food | | |
|---|---|---|---|---|---|---|---|
| | | Residue Value Data Source | % $CT_S$ * | Residue, ppm† | Residue Value Data Source | % $CT_F$ * | Residue, ppm† |
| Bean, pinto, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Beef, meat, dried | 0.69 | Avg @ 1573-1658 rate | 0.4 | 58.4 | | | |
| Beet, sugar, molasses | 0.69 | Avg @ 1454-1523 rate | 0.4 | 1.2 | | | |
| Brazil nut | 0.62 | See pecans | 0.4 | 3.9 | See pecans | 10 | 5.3 |
| Buckwheat | 0.83 | See wheat grain | 0.8 | 2.9 | | | |
| Buckwheat, flour | 0.70 | See wheat flour | ** | 0.112 | | | |
| Butternut | 0.62 | See pecans | 0.4 | 3.9 | See pecans | 10 | 5.3 |
| Cashew | 0.62 | See pecans | 0.4 | 3.9 | See pecans | 10 | 5.3 |
| Chestnut | 0.62 | See pecans | 0.4 | 3.9 | See pecans | 10 | 5.3 |
| Chickpea, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Chickpea, flour | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Chicory, roots | 0.69 | Avg @ 1616-1658 rate | 0.4 | 13.9 | | | |
| Chive | 0.69 | See dried parsley | 0.4 | 63.5 | See dried parsley | 0.1 | 63.5 |
| Chrysanthemum, garland | 0.69 | See dried parsley | 0.4 | 63.5 | | | |
| Cinnamon | 0.69 | Avg @ 1573-1580 rate | 0.4 | 73.5 | Avg @ 1573-1580 rate | 0.1 | 73.5 |
| Cocoa bean, chocolate | 0.69 | Cocoa beans @ 1500 rate | 0.4 | 8.4 | Cocoa beans @ 1500 rate | 100 | 8.4 |
| Cocoa bean, powder | 0.69 | Cocoa beans @ 1500 rate | 0.4 | 8.4 | Cocoa beans @ 1500 rate | 100 | 8.4 |
| Coconut, meat | 0.69 | Avg @ 1596-1607 rate | 0.4 | 49.1 | Avg @ 1596-1607 rate | 0.1 | 49.1 |
| Coconut, dried | 0.69 | Avg @ 1596-1607 rate | 0.4 | 49.1 | Avg @ 1596-1607 rate | 0.1 | 49.1 |
| Coconut, milk | 0.69 | Avg @ 1596-1607 rate | 0.4 | 49.1 | Avg @ 1596-1607 rate | 0.1 | 49.1 |
| Coconut, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Coffee, roasted bean | 0.69 | Avg @ 1573-1580 rate | 0.4 | 7.1 | Avg @ 1573-1580 rate | 0.1 | 7.1 |
| Coffee, instant | 0.69 | Avg @ 1610-1658 rate | 0.4 | 13.9 | Avg @ 1610-1658 rate | 0.1 | 13.9 |
| Coriander, leaves | 0.69 | See dried parsley | 0.4 | 63.5 | See dried parsley | 0.1 | 63.5 |
| Coriander, seed | 0.69 | See pepper, black/white | 0.4 | 7.1 | See pepper, black/white | 0.1 | 7.1 |
| Corn, field, flour | 1.00 | Wtd avg (see text) | ** | 0.081 | Regression | 0.1 | 16.9 |
| Corn, field, meal | 1.00 | Avg @ 1573-1590 rate | 0.8 | 14.0 | Corn grain × 0.78 | 0.1 | 2.8 |
| Corn, field, bran | 1.00 | Avg @ 1573-1590 rate | 0.8 | 14.0 | Corn grain × 0.78 | 0.1 | 2.8 |
| Corn, field, starch | 0.80 | Avg @ 1534-1573 rate | 0.8 | 6.6 | Corn grain × 0.17 | 0.1 | 0.6 |
| Corn, field, syrup | 0.78 | Corn grain × 0.17 | 0.8 | 0.6 | Corn grain × 0.17 | 0.1 | 0.6 |
| Corn, field, oil | 0.78 | Avg @ 1540-1580 rate | 0.8 | 0.4 | | | |
| Corn, pop | 0.69 | Regression | 0.8 | 1.7 | Regression | 0.1 | 1.7 |
| Cottonseed, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Cranberry, dried | 0.79 | See figs | 0.4 | 1.0 | See figs | 10 | 1.0 |
| Currant, dried | 0.79 | See figs | 0.4 | 1.0 | See figs | 10 | 1.0 |
| Date | 0.69 | Regression | 0.4 | 0.9 | Regression | 42 | 0.9 |
| Dill, seed | 0.69 | See pepper, black/white | 0.4 | 7.1 | See pepper, black/white | 0.1 | 7.1 |
| Dillweed | 0.69 | See dried parsley | 0.4 | 63.5 | See dried parsley | 0.1 | 63.5 |
| Egg (dried) | 0.69 | Avg @ 1414-1580 rate | 0.4 | 402.5 | | | |
| Fig, dried | 0.79 | Regression | 0.4 | 1.0 | Regression | 69 | 1.0 |
| Filbert | 0.69 | Regression | 0.4 | 1.5 | Regression | 10 | 2.5 |
| Filbert, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Flaxseed, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Garlic, dried | 0.69 | Avg @ 1414-1446 rate | 0.4 | 10.9 | Avg @ 1414-1446 rate | 0.1 | 10.9 |
| Ginger | 0.69 | | | | See garlic | 0.1 | 10.9 |
| Ginger, dried | 0.69 | See garlic | 0.4 | 10.9 | See garlic | 0.1 | 10.9 |
| Ginseng, dried | 0.69 | See garlic | 0.4 | 10.9 | | | |
| Grape, raisin | 0.72 | Regression | 0.4 | 1.0 | Regression | 69 | 1.0 |
| Guar, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Herbs, other | 0.69 | See dried parsley | 0.4 | 63.5 | See dried parsley | 0.1 | 63.5 |
| Hickory nut | 0.62 | See pecans | 0.4 | 3.9 | See pecans | 10 | 5.3 |
| Lemongrass | 0.69 | See dried parsley | 0.4 | 63.5 | See dried parsley | 0.1 | 63.5 |
| Lentil, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Lychee, dried | 0.79 | See figs | 0.4 | 1.0 | See figs | 69 | 1.0 |
| Macadamia nut | 0.62 | See pecans | 0.4 | 3.9 | See pecans | 0.1 | 5.3 |
| Mango, dried | 0.79 | See figs | 0.4 | 1.0 | See figs | 69 | 1.0 |

| Table 5. Summary of Analytical Correction Factors, Percent Crop Treated, and Residue Estimates for the Dietary Exposure Analysis of Fluoride from Use of Sulfuryl Fluoride. | | | | | | | |
|---|---|---|---|---|---|---|---|
| Food | Analytical Correction Factor | Structural | | | Food | | |
| | | Residue Value Data Source | % $CT_S$ * | Residue, ppm† | Residue Value Data Source | % $CT_F$ * | Residue, ppm† |
| Maple, sugar | 0.69 | Avg @ 1454-1523 rate | 0.4 | 1.2 | | | |
| Maple syrup | 0.69 | Avg @ 1454-1523 rate | 0.4 | 1.2 | | | |
| Marjoram | 0.69 | See basil | 0.4 | 67.1 | See basil | 0.1 | 67.1 |
| Milk (powdered) | 0.69 | Avg @ 1414-1580 rate | 0.4 | 5.4 | | | |
| Milk (cured; cheese) | 0.69 | Avg @ 1414-1446 rate | 0.4 | 3.9 | | | |
| Millet, grain | 0.83 | See wheat grain | 0.8 | 2.9 | See wheat grain | 0.1 | 2.9 |
| Oat, bran | 0.52 | See wheat bran | 0.8 | 74.2 | Barley with 2.56X factor | 0.1 | 18.5 |
| Oat, flour | 0.70 | Wtd avg (see text) | ** | 0.337 | Barley with 0.73X factor | 0.1 | 18.5 |
| Oat, groats/rolled oats | 0.83 | See pearled barley | 0.8 | 18.5 | Barley | 0.1 | 18.5 |
| Olive, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Onion, dry bulb, dried | 0.69 | Control value; treated samples <LOQ | 0.4 | 1.7 | | | |
| Palm, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Papaya, dried | 0.79 | See figs | 0.4 | 1.0 | See figs | 69 | 1.0 |
| Parsley, dried leaves | 0.69 | Avg @ 1454-1523 rate | 0.4 | 63.5 | Avg @ 1454-1523 rate | 0.1 | 63.5 |
| Pea, dry | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Pea, pigeon, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | Cocoa beans @ avg rate | 100 | 4.5 |
| Peach, dried | 0.79 | See figs | 0.4 | 1.0 | See figs | 69 | 1.0 |
| Peanut | 0.69 | Avg @ 1569-1596 rate | 0.4 | 16.4 | Avg @ 1569-1596 rate | 0.6 | 16.4 |
| Peanut, butter | 0.69 | Avg @ 1569-1596 rate | 0.4 | 16.4 | Avg @ 1569-1596 rate | 0.6 | 16.4 |
| Peanut, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Pear, dried | 0.79 | See figs | 0.4 | 1.0 | See figs | 69 | 1.0 |
| Pecan | 0.62 | Regression | 0.4 | 3.9 | Regression | 10 | 5.3 |
| Pepper, bell, dried | 0.69 | Avg @ 1569-1596 rate | 0.4 | 36.1 | | | |
| Pepper, nonbell, dried | 0.69 | Avg @ 1569-1596 rate | 0.4 | 36.1 | | | |
| Pepper, black/white | 0.69 | Avg @ 1454-1523 rate | 0.4 | 7.1 | Avg @ 1454-1523 rate | 0.1 | 7.1 |
| Peppermint, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Pine nut | 0.69 | Avg @ 1573-1580 rate | 0.4 | 8.8 | Avg @ 1573-1580 rate | 10 | 8.8 |
| Pineapple, dried | 0.79 | See figs | 0.4 | 1.0 | See figs | 69 | 1.0 |
| Pistachio | 0.69 | Regression | 0.4 | 2.3 | Regression | 27 | 3.2 |
| Plantain, dried | 0.79 | See figs | 0.4 | 1.0 | See figs | 69 | 1.0 |
| Plum, prune, dried | 0.82 | Regression | 0.4 | 0.7 | Regression | 69 | 0.7 |
| Potato, chips | 0.69 | Avg @ 1725-1734 rate | 0.4 | 7.1 | | | |
| Potato, dry | 0.69 | From egg noodles | 0.4 | 25.6 | | | |
| Potato, flour | 0.70 | Non-mixed wheat flour | 0.4 | 31.4 | | | |
| Psyllium, seed | 0.69 | See pepper, black/white | 0.4 | 7.1 | | | |
| Pumpkin, seed | 0.69 | See pine nut | 0.4 | 8.8 | | | |
| Quinoa, grain | 0.51 | See sorghum grain | 0.4 | 20.4 | | | |
| Rapeseed, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Rice, white | 0.75 | Regression | 0.8 | 3.9 | Regression | 3 | 4.5 |
| Rice, brown | 0.36 | Regression | 0.8 | 9.4 | Regression | 3 | 12.5 |
| Rice, flour | 0.56 | Wtd avg (see text) | ** | 0.160 | Avg @ 1573-1580 rate | 3 | 32.5 |
| Rice, bran | 0.69 | Avg @ 1573 rate | 0.8 | 37.5 | Avg @ 1573 rate | 3 | 37.5 |
| Rye, grain | 0.83 | See wheat grain | 0.8 | 2.9 | | | |
| Rye, flour | 0.70 | See wheat flour | 0.8 | 0.112 | | | |
| Safflower, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Savory | 0.69 | See dried parsley | 0.4 | 63.5 | See dried parsley | 0.1 | 63.5 |
| Sesame, seed | 0.69 | See pepper, black/white | 0.4 | 7.1 | | | |
| Sesame, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Sorghum, grain | 0.51 | Avg @ 1573-1580 rate | 0.8 | 20.4 | Avg @ 1573-1580 rate | 0.1 | 20.4 |
| Sorghum, syrup | 0.78 | Corn syrup | 0.8 | 0.6 | | | |
| Soybean, flour | 0.70 | Corn flour | ** | 0.081 | | | |
| Soybean, soy milk | 0.69 | Powdered milk | 0.4 | 2.4 | | | |
| Soybean, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Spearmint, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Spices, other | 0.69 | See pepper, black/white | 0.4 | 7.1 | See pepper, black/white | 0.1 | 7.1 |

| Table 5. Summary of Analytical Correction Factors, Percent Crop Treated, and Residue Estimates for the Dietary Exposure Analysis of Fluoride from Use of Sulfuryl Fluoride. | | | | | | | |
|---|---|---|---|---|---|---|---|
| Food | Analytical Correction Factor | Structural | | | Food | | |
| | | Residue Value Data Source | % $CT_S$* | Residue, ppm† | Residue Value Data Source | % $CT_F$* | Residue, ppm† |
| Sugarcane, sugar | 0.69 | Avg @ 1454-1523 rate | 0.4 | 1.2 | | | |
| Sugarcane, molasses | 0.69 | Avg @ 1454-1523 rate | 0.4 | 1.2 | | | |
| Sunflower, seed | 0.69 | Cocoa beans @ avg rate | 0.4 | 4.5 | | | |
| Sunflower, oil | 0.78 | ½ LOQ | 0.4 | 1.5 | | | |
| Tea, dried | 0.69 | See basil | 0.4 | 67.1 | | | |
| Tea, instant | 0.69 | See basil | 0.4 | 67.1 | | | |
| Tomato, dried | 0.79 | See figs | 0.4 | 1.0 | | | |
| Triticale, flour | 0.70 | See wheat flour | ** | 0.134 | Wheat with 0.38 factor | 0.1 | 2.9 |
| Turmeric | 0.69 | See pepper, black/white | 0.4 | 7.1 | See pepper, black/white | 0.1 | 7.1 |
| Walnut | 0.70 | Regression | 0.4 | 2.0 | Regression | 99 | 2.4 |
| Wheat, grain | 0.83 | Regression | 0.8 | 2.9 | Regression | 0.4 | 2.9 |
| Wheat, flour | 0.70 | Wtd avg (see text) | ** | 0.134 | Regression | 0.1 | 31.4 |
| Wheat, germ | 0.62 | Regression | 0.8 | 54.0 | Wheat grain × 4.8 | 0.1 | 13.9 |
| Wheat, bran | 0.52 | Avg @ 1573-1717 rate | 0.8 | 74.2 | Avg @ 1573-1717 rate | 0.1 | 74.2 |
| Wild rice | 0.36 | See rice, brown | 0.8 | 9.4 | See rice, brown | 3 | 12.5 |

* % $CT_S$ = percent crop treated for structural fumigations. % $CT_F$ = percent crop treated for food fumigations.
† Residue values include the analytical correction factor.
‡ Residue values are from Table 1, after application of the analytical correction factor.
§ Avg @ rate = the average residue value from the listed application rate
** % $CT_S$ estimates associated with grain flours are incorporated into the residue estimate directly and are, therefore, not used as a modifying factor for these commodities. For the DEEM input file, the value is set at 1.00.

For grains, the current label for sulfuryl fluoride allows for fumigation of corn, rice, and wheat processed commodities. Fumigation of processed commodities of other grains (*e.g.*, barley, oats, and triticale) is not permitted. The entries in Table 4 associated with the food fumigation of the processed commodities for these other grains include factors of 0.38 for flour and 2.56 for bran. These factors are from a study (MRID 45396301) in which wheat was fumigated with sulfuryl fluoride and then processed into flour, bran, germ, etc. using simulated commercial practices. A processing factor for chickpea flour is not available. HED has assumed that there is no concentration of fluoride residue during the processing of chickpeas into flour, and believes that this is a conservative assumption given the processing factors for wheat flour (0.38) and corn flour (0.73).

## III. DEEM-FCID™ Program and Consumption Information

These dietary exposure assessments were conducted using the Dietary Exposure Evaluation Model software with the Food Commodity Intake Database DEEM-FCID™, Version 2.03 which incorporates consumption data from USDA's Continuing Surveys of Food Intakes by Individuals (CSFII), 1994-1996 and 1998. The 1994-96, 98 data are based on the reported consumption of more than 20,000 individuals over two non-consecutive survey days. Foods "as consumed" (e.g., apple pie) are linked to EPA-defined food commodities (e.g. apples, peeled fruit - cooked; fresh or N/S; baked; or wheat flour - cooked; fresh or N/S, baked) using publicly available recipe translation files developed jointly by USDA/ARS and EPA. For chronic exposure assessment, consumption data are averaged for the entire U.S. population and within population subgroups, but for acute exposure assessment are retained as individual consumption events. Based on analysis of the 1994-96, 98 CSFII consumption data, which took into account dietary patterns and survey respondents, HED concluded that it is most appropriate to report risk for the following population subgroups: the general U.S. population, all infants (<1 year old), children

1-2, children 3-5, children 6-12, youth 13-19, adults 20-49, females 13-49, and adults 50+ years old.

For chronic dietary exposure assessment, an estimate of the residue level in each food or food-form (e.g., orange or orange juice) on the food commodity residue list is multiplied by the average daily consumption estimate for that food/food form to produce a residue intake estimate. The resulting residue intake estimate for each food/food form is summed with the residue intake estimates for all other food/food forms on the commodity residue list to arrive at the total average estimated exposure. Exposure is expressed in mg/kg body weight/day. This procedure is performed for each population subgroup.

## IV. Results/Discussion

Chronic dietary exposure estimates are summarized in Table 6 for each source and each population subgroup noted above. The estimated contributions from the various crop subgroups to total fluoride exposure from the currently registered uses of cryolite and sulfuryl fluoride are provided in Table 7. The results of the commodity contribution analysis for each source are summarized in Attachments 7 through 9. The complete commodity contribution reports have not been included in this document due to their excessive length. These reports are available upon request. Overall exposure is estimated to be greatest for the age group consisting of 1-2 year olds; however, 3-5 year olds have higher exposure for certain food groups (*e.g.*, leafy vegetables, cucurbit vegetables, citrus fruits, pome fruits, and tree nuts; Table 7).

Table 6. Summary of Pesticidal Fluoride Contributions to Dietary Fluoride Exposure.

| Population Group | Exposure Estimates, mg/kg/day | | | |
|---|---|---|---|---|
| | Cryolite | SF Structural Fumigations | SF Food Fumigations | Total |
| U.S. Population (total) | 0.000682 | 0.000336 | 0.001023 | 0.002041 |
| All infants (< 1 year) | 0.000956 | 0.000505 | 0.001071 | 0.002532 |
| Children 1-2 yrs | 0.003275 | 0.000827 | 0.002169 | 0.006271 |
| Children 3-5 yrs | 0.002112 | 0.000800 | 0.002293 | 0.005205 |
| Children 6-12 yrs | 0.000885 | 0.000543 | 0.001743 | 0.003171 |
| Youth 13-19 yrs | 0.000346 | 0.000320 | 0.001032 | 0.001698 |
| Adults 20-49 yrs | 0.000445 | 0.000272 | 0.000814 | 0.001531 |
| Adults 50+ yrs | 0.000547 | 0.000215 | 0.000719 | 0.001481 |
| Females 13-49 yrs | 0.000473 | 0.000249 | 0.000799 | 0.001521 |

## V. Characterization of Inputs/Outputs

The residue estimates for most of the commodities in these analyses are moderately to highly refined. Data reflecting residues of F at various fumigation rates could be used to further refine residue estimates for a number of commodities. However, such data are not expected to result in significant changes to the exposure estimates presented in Section IV. Percent CT estimates have been used for both the structural and food fumigation uses. The % CT values used by OPP are considered to be highly refined, although certain conservatism remains in the values in that where there are discrepancies between the estimates from BEAD and Dow AgroSciences, the higher value was used.

| Table 7. Summary of Fluoride Exposure Estimates by Age Group and Crop/Food Group. | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| Group* | Exposure, mg/kg/day | | | | | | | | |
| | U.S. Pop. | Grouping by Age (Years) | | | | | | | |
| | | <1 | 1 – 2 | 3 – 5 | 6 – 12 | 13 – 19 | 20 – 50 | >50 | Females 13-49 |
| (O) Other† | 0.0009222 | 0.0009517 | 0.0037942 | 0.0028948 | 0.0016121 | 0.0006471 | 0.0005932 | 0.0006092 | 0.0006484 |
| (M) Meat | 0.0000004 | 0.0000000 | 0.0000009 | 0.0000002 | 0.0000006 | 0.0000011 | 0.0000004 | 0.0000002 | 0.0000004 |
| (P) Poultry | 0.0000103 | 0.0000055 | 0.0000296 | 0.0000210 | 0.0000118 | 0.0000079 | 0.0000102 | 0.0000068 | 0.0000110 |
| (D) Dairy Products | 0.0000032 | 0.0000684 | 0.0000135 | 0.0000096 | 0.0000031 | 0.0000017 | 0.0000011 | 0.0000014 | 0.0000011 |
| (1) Root and Tuber Vegetables | 0.0000218 | 0.0000216 | 0.0000509 | 0.0000470 | 0.0000310 | 0.0000224 | 0.0000181 | 0.0000164 | 0.0000171 |
| (3) Bulb Vegetables | 0.0000002 | 0.0000002 | 0.0000007 | 0.0000006 | 0.0000004 | 0.0000002 | 0.0000002 | 0.0000002 | 0.0000002 |
| (4) Leafy Vegetables (except Brassica) | 0.0000077 | 0.0000000 | 0.0000040 | 0.0000061 | 0.0000063 | 0.0000075 | 0.0000088 | 0.0000077 | 0.0000094 |
| (5) Brassica Leafy Vegetables | 0.0000148 | 0.0000082 | 0.0000313 | 0.0000230 | 0.0000167 | 0.0000094 | 0.0000131 | 0.0000161 | 0.0000134 |
| (6) Legume Veg. (Succulent or Dried) | 0.0005292 | 0.0003465 | 0.0010570 | 0.0009845 | 0.0006567 | 0.0005222 | 0.0004703 | 0.0004453 | 0.0004313 |
| (8) Fruiting Vegetables | 0.0000153 | 0.0000062 | 0.0000284 | 0.0000260 | 0.0000188 | 0.0000140 | 0.0000145 | 0.0000129 | 0.0000134 |
| (9) Curcurbit Vegetables | 0.0000091 | 0.0000110 | 0.0000162 | 0.0000186 | 0.0000121 | 0.0000074 | 0.0000068 | 0.0000098 | 0.0000074 |
| (10) Citrus | 0.0000462 | 0.0000159 | 0.0001014 | 0.0001027 | 0.0000544 | 0.0000298 | 0.0000305 | 0.0000614 | 0.0000322 |
| (11) Pome Fruits | 0.0000013 | 0.0000017 | 0.0000027 | 0.0000030 | 0.0000023 | 0.0000012 | 0.0000009 | 0.0000011 | 0.0000010 |
| (12) Stone Fruits | 0.0000091 | 0.0000503 | 0.0000401 | 0.0000179 | 0.0000114 | 0.0000035 | 0.0000049 | 0.0000100 | 0.0000055 |
| (13) Berries | 0.0000061 | 0.0000117 | 0.0000147 | 0.0000147 | 0.0000093 | 0.0000049 | 0.0000042 | 0.0000056 | 0.0000046 |
| (14) Tree Nuts | 0.0000163 | 0.0000013 | 0.0000257 | 0.0000262 | 0.0000190 | 0.0000113 | 0.0000150 | 0.0000171 | 0.0000150 |
| (15) Cereal Grains | 0.0004246 | 0.0010255 | 0.0010501 | 0.0010019 | 0.0007000 | 0.0004035 | 0.0003361 | 0.0002590 | 0.0003077 |
| (19) Herbs and Spices | 0.0000026 | 0.0000016 | 0.0000092 | 0.0000072 | 0.0000044 | 0.0000026 | 0.0000020 | 0.0000014 | 0.0000020 |
| (20) Oilseeds | 0.0000003 | 0.0000039 | 0.0000004 | 0.0000004 | 0.0000004 | 0.0000003 | 0.0000002 | 0.0000002 | 0.0000002 |
| Fruit Juices‡ | 0.0002769 | 0.0007996 | 0.0022384 | 0.0012761 | 0.0004614 | 0.0001593 | 0.0001053 | 0.0000941 | 0.0001126 |
| Total | 0.0020407 | 0.0025312 | 0.0062710 | 0.0052054 | 0.0031708 | 0.0016980 | 0.0015305 | 0.0014818 | 0.0015213 |

* For crops, the groups correspond to OPP crop groupings. Groups 7, 16, 17, and 18 do not consist of commodities for human consumption and are not included in this table.

† Foods not captured in one of the listed groups, including, but not limited to, cocoa beans, coconut, cranberry, grape, and grape juice. Use of cryolite is the predominant source of fluoride for this group.

‡ The exposure contributions from fruit juices are included in the overall total via the crop groups; therefore, the values listed as total do not include the specific exposure estimate from fruit juices.

**Attachments**

1. Inputs for the chronic dietary exposure analysis of fluoride from cryolite

2. Inputs for the chronic dietary exposure analysis of fluoride from structural fumigation with sulfuryl fluoride

3. Inputs for the chronic dietary exposure analysis of fluoride from food fumigation with sulfuryl fluoride

4. Results of the chronic dietary exposure analysis of fluoride from cryolite

5. Results of the chronic dietary exposure analysis of fluoride from structural fumigation with sulfuryl fluoride

6. Results of the chronic dietary exposure analysis of fluoride from food fumigation with sulfuryl fluoride

7. Commodity contribution summary for the chronic dietary exposure analysis of fluoride from cryolite

8. Commodity contribution summary for the chronic dietary exposure analysis of fluoride from structural fumigation with sulfuryl fluoride

9. Commodity contribution summary for the chronic dietary exposure analysis of fluoride from food fumigation with sulfuryl fluoride

Attachment 1. Inputs for the chronic dietary exposure analysis of fluoride from cryolite

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U.S. Environmental Protection Agency                                 Ver. 2.00
DEEM-FCID Chronic analysis for CRYOLITE                             1994-98 data
Residue file: C:\Documents and Settings\mdoherty\My Documents\Chemistry Reviews\!DEEM
Runs\Sulfuryl Fluoride\Cryolite-AR-CT new raisin factor.R98
                                                               Adjust. #2 used
Analysis Date 02-19-2009            Residue file dated: 06-24-2004/10:05:08/8
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| Food Crop EPA Code | Grp | Food Name | Residue (ppm) | Adj.Factors #1 | #2 | Comment |
|---|---|---|---|---|---|---|
| 12000120 | 12 | Apricot | 4.500000 | 1.000 | 0.010 | |
| 12000121 | 12 | Apricot-babyfood | 4.500000 | 1.000 | 0.010 | |
| 12000130 | 12 | Apricot, dried | 4.500000 | 6.000 | 0.010 | |
| 12000140 | 12 | Apricot, juice | 4.500000 | 1.000 | 0.010 | |
| 12000141 | 12 | Apricot, juice-babyfood | 4.500000 | 1.000 | 0.010 | |
| 13010550 | 13A | Blackberry | 0.250000 | 1.000 | 1.000 | |
| 13010560 | 13A | Blackberry, juice | 0.250000 | 1.000 | 1.000 | |
| 13010561 | 13A | Blackberry, juice-babyfood | 0.250000 | 1.000 | 1.000 | |
| 13020570 | 13B | Blueberry | 0.110000 | 1.000 | 1.000 | |
| 13020571 | 13B | Blueberry-babyfood | 0.110000 | 1.000 | 1.000 | |
| 13010580 | 13A | Boysenberry | 0.250000 | 1.000 | 1.000 | |
| 05010610 | 5A | Broccoli | 5.000000 | 1.000 | 0.020 | |
| 05010611 | 5A | Broccoli-babyfood | 5.000000 | 1.000 | 0.020 | |
| 05010640 | 5A | Brussels sprouts | 4.000000 | 1.000 | 0.020 | |
| 05010690 | 5A | Cabbage | 1.500000 | 1.000 | 0.010 | |
| 05020700 | 5B | Cabbage, Chinese, bok choy | 4.000000 | 1.000 | 0.010 | |
| 09010750 | 9A | Cantaloupe | 2.160000 | 1.000 | 0.010 | |
| 09010800 | 9A | Casaba | 2.160000 | 1.000 | 0.010 | |
| 05010830 | 5A | Cauliflower | 3.000000 | 1.000 | 0.020 | |
| 10001060 | 10 | Citrus citron | 8.000000 | 1.000 | 0.040 | |
| 05021170 | 5B | Collards | 4.000000 | 1.000 | 0.020 | |
| 95001300 | O | Cranberry | 0.500000 | 1.000 | 1.000 | |
| 95001301 | O | Cranberry-babyfood | 0.500000 | 1.000 | 1.000 | |
| 95001310 | O | Cranberry, dried | 0.500000 | 1.000 | 1.000 | |
| 95001320 | O | Cranberry, juice | 0.500000 | 1.100 | 1.000 | |
| 95001321 | O | Cranberry, juice-babyfood | 0.500000 | 1.100 | 1.000 | |
| 09021350 | 9B | Cucumber | 2.500000 | 1.000 | 0.010 | |
| 13021360 | 13B | Currant | 0.110000 | 1.000 | 1.000 | |
| 13021370 | 13B | Currant, dried | 0.110000 | 1.000 | 1.000 | |
| 13011420 | 13A | Dewberry | 0.250000 | 1.000 | 1.000 | |
| 08001480 | 8 | Eggplant | 1.500000 | 1.000 | 0.010 | |
| 13021490 | 13B | Elderberry | 0.110000 | 1.000 | 1.000 | |
| 13021740 | 13B | Gooseberry | 0.110000 | 1.000 | 1.000 | |
| 95001750 | O | Grape | 3.500000 | 1.000 | 0.330 | |
| 95001760 | O | Grape, juice | 3.500000 | 0.830 | 0.330 | |
| 95001761 | O | Grape, juice-babyfood | 3.500000 | 0.830 | 0.330 | |
| 95001770 | O | Grape, leaves | 3.500000 | 1.000 | 0.330 | |
| 95001780 | O | Grape, raisin | 3.500000 | 1.350 | 0.330 | |
| 95001790 | O | Grape, wine and sherry | 3.500000 | 0.830 | 0.330 | |
| 10001800 | 10 | Grapefruit | 9.000000 | 1.000 | 0.040 | |
| 10001810 | 10 | Grapefruit, juice | 9.000000 | 0.026 | 0.040 | |
| 09011870 | 9A | Honeydew melon | 2.160000 | 1.000 | 0.010 | |
| 13021910 | 13B | Huckleberry | 0.110000 | 1.000 | 1.000 | |
| 05021940 | 5B | Kale | 4.000000 | 1.000 | 0.020 | |
| 95001950 | O | Kiwifruit | 4.500000 | 1.000 | 0.140 | |
| 05011960 | 5A | Kohlrabi | 5.000000 | 1.000 | 0.020 | |
| 10001970 | 10 | Kumquat | 8.000000 | 1.000 | 0.040 | |
| 10001990 | 10 | Lemon | 13.500000 | 1.000 | 0.020 | |
| 10002000 | 10 | Lemon, juice | 13.500000 | 0.024 | 0.020 | |
| 10002001 | 10 | Lemon, juice-babyfood | 13.500000 | 0.024 | 0.020 | |
| 10002010 | 10 | Lemon, peel | 13.500000 | 0.280 | 0.020 | |
| 04012040 | 4A | Lettuce, head | 2.500000 | 1.000 | 0.010 | |
| 04012050 | 4A | Lettuce, leaf | 15.000000 | 1.000 | 0.010 | |
| 10002060 | 10 | Lime | 13.500000 | 1.000 | 0.040 | |
| 10002070 | 10 | Lime, juice | 13.500000 | 0.024 | 0.040 | |
| 10002071 | 10 | Lime, juice-babyfood | 13.500000 | 0.024 | 0.040 | |
| 13012080 | 13A | Loganberry | 0.250000 | 1.000 | 1.000 | |
| 12002300 | 12 | Nectarine | 4.500000 | 1.000 | 0.010 | |
| 10002400 | 10 | Orange | 8.000000 | 1.000 | 0.020 | |

| | | | | | |
|---|---|---|---|---|---|
| 10002410 | 10 | Orange, juice | 8.000000 | 0.022 | 0.020 |
| 10002411 | 10 | Orange, juice-babyfood | 8.000000 | 0.022 | 0.020 |
| 10002420 | 10 | Orange, peel | 8.000000 | 0.280 | 0.020 |
| 12002600 | 12 | Peach | 4.500000 | 1.000 | 0.010 |
| 12002601 | 12 | Peach-babyfood | 4.500000 | 1.000 | 0.010 |
| 12002610 | 12 | Peach, dried | 4.500000 | 7.000 | 0.010 |
| 12002611 | 12 | Peach, dried-babyfood | 4.500000 | 7.000 | 0.010 |
| 12002620 | 12 | Peach, juice | 4.500000 | 1.000 | 0.010 |
| 12002621 | 12 | Peach, juice-babyfood | 4.500000 | 1.000 | 0.010 |
| 08002700 | 8 | Pepper, bell | 3.500000 | 1.000 | 0.010 |
| 08002701 | 8 | Pepper, bell-babyfood | 3.500000 | 1.000 | 0.010 |
| 08002710 | 8 | Pepper, bell, dried | 3.500000 | 1.000 | 0.010 |
| 08002711 | 8 | Pepper, bell, dried-babyfood | 3.500000 | 1.000 | 0.010 |
| 08002720 | 8 | Pepper, nonbell | 3.500000 | 1.000 | 0.010 |
| 08002721 | 8 | Pepper, nonbell-babyfood | 3.500000 | 1.000 | 0.010 |
| 08002730 | 8 | Pepper, nonbell, dried | 3.500000 | 1.000 | 0.010 |
| 95002750 | O | Peppermint | 19.500000 | 1.000 | 1.000 |
| 95002760 | O | Peppermint, oil | 19.500000 | 0.026 | 1.000 |
| 12002850 | 12 | Plum | 0.500000 | 1.000 | 0.010 |
| 12002851 | 12 | Plum-babyfood | 0.500000 | 1.000 | 0.010 |
| 12002860 | 12 | Plum, prune, fresh | 0.500000 | 1.000 | 0.010 |
| 12002861 | 12 | Plum, prune, fresh-babyfood | 2.000000 | 1.000 | 0.010 |
| 12002870 | 12 | Plum, prune, dried | 2.000000 | 5.000 | 0.010 |
| 12002871 | 12 | Plum, prune, dried-babyfood | 2.000000 | 5.000 | 0.010 |
| 12002880 | 12 | Plum, prune, juice | 2.000000 | 1.400 | 0.010 |
| 12002881 | 12 | Plum, prune, juice-babyfood | 2.000000 | 1.400 | 0.010 |
| 01032960 | 1C | Potato, chips | 0.650000 | 1.000 | 0.030 |
| 01032970 | 1C | Potato, dry (granules/ flakes) | 0.650000 | 6.500 | 0.030 |
| 01032971 | 1C | Potato, dry (granules/ flakes)-b | 0.650000 | 6.500 | 0.030 |
| 01032980 | 1C | Potato, flour | 0.650000 | 6.500 | 0.030 |
| 01032981 | 1C | Potato, flour-babyfood | 0.650000 | 6.500 | 0.030 |
| 01032990 | 1C | Potato, tuber, w/peel | 0.650000 | 1.000 | 0.030 |
| 01032991 | 1C | Potato, tuber, w/peel-babyfood | 0.650000 | 1.000 | 0.030 |
| 01033000 | 1C | Potato, tuber, w/o peel | 0.650000 | 1.000 | 0.030 |
| 01033001 | 1C | Potato, tuber, w/o peel-babyfood | 0.650000 | 1.000 | 0.030 |
| 10003070 | 10 | Pummelo | 9.000000 | 1.000 | 0.040 |
| 09023080 | 9B | Pumpkin | 2.500000 | 1.000 | 0.010 |
| 09023090 | 9B | Pumpkin, seed | 2.500000 | 1.000 | 0.010 |
| 13013200 | 13A | Raspberry | 0.250000 | 1.000 | 1.000 |
| 13013201 | 13A | Raspberry-babyfood | 0.250000 | 1.000 | 1.000 |
| 13013210 | 13A | Raspberry, juice | 0.250000 | 1.000 | 1.000 |
| 13013211 | 13A | Raspberry, juice-babyfood | 0.250000 | 1.000 | 1.000 |
| 95003520 | O | Spearmint | 19.500000 | 1.000 | 1.000 |
| 95003530 | O | Spearmint, oil | 19.500000 | 0.026 | 1.000 |
| 09023560 | 9B | Squash, summer | 2.500000 | 1.000 | 0.010 |
| 09023561 | 9B | Squash, summer-babyfood | 2.500000 | 1.000 | 0.010 |
| 09023570 | 9B | Squash, winter | 2.500000 | 1.000 | 0.010 |
| 09023571 | 9B | Squash, winter-babyfood | 2.500000 | 1.000 | 0.010 |
| 95003590 | O | Strawberry | 1.000000 | 1.000 | 0.020 |
| 95003591 | O | Strawberry-babyfood | 1.000000 | 1.000 | 0.020 |
| 95003600 | O | Strawberry, juice | 1.000000 | 1.000 | 0.020 |
| 95003601 | O | Strawberry, juice-babyfood | 1.000000 | 1.000 | 0.020 |
| 10003690 | 10 | Tangerine | 8.000000 | 1.000 | 0.040 |
| 10003700 | 10 | Tangerine, juice | 8.000000 | 0.028 | 0.040 |
| 08003750 | 8 | Tomato | 1.500000 | 1.000 | 0.010 |
| 08003751 | 8 | Tomato-babyfood | 1.500000 | 1.000 | 0.010 |
| 08003760 | 8 | Tomato, paste | 1.500000 | 1.500 | 0.010 |
| 08003761 | 8 | Tomato, paste-babyfood | 1.500000 | 1.500 | 0.010 |
| 08003770 | 8 | Tomato, puree | 1.500000 | 1.000 | 0.010 |
| 08003771 | 8 | Tomato, puree-babyfood | 1.500000 | 1.000 | 0.010 |
| 08003780 | 8 | Tomato, dried | 1.500000 | 14.300 | 0.010 |
| 08003781 | 8 | Tomato, dried-babyfood | 1.500000 | 14.300 | 0.010 |
| 08003790 | 8 | Tomato, juice | 1.500000 | 1.500 | 0.010 |
| 09013990 | 9A | Watermelon | 2.160000 | 1.000 | 0.010 |
| 09014000 | 9A | Watermelon, juice | 2.160000 | 1.000 | 0.010 |

Attachment 2. Inputs for the chronic dietary exposure analysis of fluoride from structural fumigation with sulfuryl fluoride

U.S. Environmental Protection Agency Ver. 2.00
DEEM-FCID Chronic analysis for FLUORIDE 1994-98 data
Residue file: C:\Documents and Settings\mdoherty\My Documents\Chemistry Reviews\!DEEM Runs\Sulfuryl Fluoride\F Space Fumigation - 2009 - 5-1.R98
Adjust. #2 used
Analysis Date 05-06-2009 Residue file dated: 05-06-2009/13:52:45/8
Reference dose (RfD) = 0.114 mg/kg bw/day

| Food EPA Code | Crop Grp | Food Name | Residue (ppm) | Adj.Factors #1 | Adj.Factors #2 | Comment |
|---|---|---|---|---|---|---|
| 18000020 | 18 | Alfalfa, seed | 20.400000 | 1.000 | 0.004 | |
| 14000030 | 14 | Almond | 9.200000 | 1.000 | 0.004 | |
| 14000031 | 14 | Almond-babyfood | 9.200000 | 1.000 | 0.004 | |
| 14000040 | 14 | Almond, oil | 1.500000 | 1.000 | 0.004 | |
| 14000041 | 14 | Almond, oil-babyfood | 1.500000 | 1.000 | 0.004 | |
| 95000060 | O | Amaranth, grain | 20.400000 | 1.000 | 0.004 | |
| 11000090 | 11 | Apple, dried | 1.000000 | 1.000 | 0.004 | |
| 11000091 | 11 | Apple, dried-babyfood | 1.000000 | 1.000 | 0.004 | |
| 12000130 | 12 | Apricot, dried | 1.000000 | 1.000 | 0.004 | |
| 01030150 | 1CD | Arrowroot, flour | 31.400000 | 1.000 | 0.004 | |
| 01030151 | 1CD | Arrowroot, flour-babyfood | 31.400000 | 1.000 | 0.004 | |
| 95000240 | O | Banana, dried | 1.000000 | 1.000 | 0.004 | |
| 95000241 | O | Banana, dried-babyfood | 1.000000 | 1.000 | 0.004 | |
| 15000250 | 15 | Barley, pearled barley | 3.650000 | 1.000 | 0.008 | |
| 15000251 | 15 | Barley, pearled barley-babyfood | 3.650000 | 1.000 | 0.008 | |
| 15000260 | 15 | Barley, flour | 0.156000 | 1.000 | 1.000 | |
| 15000261 | 15 | Barley, flour-babyfood | 0.156000 | 1.000 | 1.000 | |
| 15000270 | 15 | Barley, bran | 3.650000 | 1.000 | 0.008 | |
| 19010280 | 19A | Basil, fresh leaves | 67.100000 | 1.000 | 0.004 | |
| 19010281 | 19A | Basil, fresh leaves-babyfood | 67.100000 | 1.000 | 0.004 | |
| 19010290 | 19A | Basil, dried leaves | 67.100000 | 1.000 | 0.004 | |
| 19010291 | 19A | Basil, dried leaves-babyfood | 67.100000 | 1.000 | 0.004 | |
| 06030300 | 6C | Bean, black, seed | 4.500000 | 1.000 | 0.004 | |
| 06030320 | 6C | Bean, broad, seed | 4.500000 | 1.000 | 0.004 | |
| 06030340 | 6C | Bean, cowpea, seed | 4.500000 | 1.000 | 0.004 | |
| 06030350 | 6C | Bean, great northern, seed | 4.500000 | 1.000 | 0.004 | |
| 06030360 | 6C | Bean, kidney, seed | 4.500000 | 1.000 | 0.004 | |
| 06030380 | 6C | Bean, lima, seed | 4.500000 | 1.000 | 0.004 | |
| 06030390 | 6C | Bean, mung, seed | 4.500000 | 1.000 | 0.004 | |
| 06030400 | 6C | Bean, navy, seed | 4.500000 | 1.000 | 0.004 | |
| 06030410 | 6C | Bean, pink, seed | 4.500000 | 1.000 | 0.004 | |
| 06030420 | 6C | Bean, pinto, seed | 4.500000 | 1.000 | 0.004 | |
| 21000450 | M | Beef, meat, dried | 58.400000 | 1.000 | 0.004 | |
| 01010530 | 1A | Beet, sugar, molasses | 1.200000 | 1.000 | 0.004 | |
| 01010531 | 1A | Beet, sugar, molasses-babyfood | 1.200000 | 1.000 | 0.004 | |
| 14000590 | 14 | Brazil nut | 3.900000 | 1.000 | 0.004 | |
| 15000650 | 15 | Buckwheat | 2.900000 | 1.000 | 0.008 | |
| 15000660 | 15 | Buckwheat, flour | 0.134000 | 1.000 | 1.000 | |
| 14000680 | 14 | Butternut | 3.900000 | 1.000 | 0.004 | |
| 14000810 | 14 | Cashew | 3.900000 | 1.000 | 0.004 | |
| 14000920 | 14 | Chestnut | 3.900000 | 1.000 | 0.004 | |
| 06030980 | 6C | Chickpea, seed | 4.500000 | 1.000 | 0.004 | |
| 06030981 | 6C | Chickpea, seed-babyfood | 4.500000 | 1.000 | 0.004 | |
| 06030990 | 6C | Chickpea, flour | 4.500000 | 1.000 | 0.004 | |
| 01011000 | 1AB | Chicory, roots | 13.900000 | 1.000 | 0.004 | |
| 19011030 | 19A | Chive | 63.500000 | 1.000 | 0.004 | |
| 04011040 | 4A | Chrysanthemum, garland | 63.500000 | 1.000 | 0.004 | |
| 19021050 | 19B | Cinnamon | 73.500000 | 1.000 | 0.004 | |
| 19021051 | 19B | Cinnamon-babyfood | 73.500000 | 1.000 | 0.004 | |
| 95001090 | O | Cocoa bean, chocolate | 8.400000 | 1.000 | 0.004 | |
| 95001100 | O | Cocoa bean, powder | 8.400000 | 1.000 | 0.004 | |
| 95001110 | O | Coconut, meat | 49.100000 | 1.000 | 0.004 | |
| 95001111 | O | Coconut- meat-babyfood | 49.100000 | 1.000 | 0.004 | |
| 95001120 | O | Coconut, dried | 49.100000 | 1.000 | 0.004 | |
| 95001130 | O | Coconut, milk | 49.100000 | 1.000 | 0.004 | |
| 95001140 | O | Coconut, oil | 1.500000 | 1.000 | 0.004 | |

| | | | | | |
|---|---|---|---|---|---|
| 95001141 | O | Coconut, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 95001150 | O | Coffee, roasted bean | 7.100000 | 1.000 | 0.004 |
| 95001160 | O | Coffee, instant | 13.900000 | 1.000 | 0.004 |
| 19011180 | 19A | Coriander, leaves | 63.500000 | 1.000 | 0.004 |
| 19011181 | 19A | Coriander, leaves-babyfood | 63.500000 | 1.000 | 0.004 |
| 19021190 | 19B | Coriander, seed | 7.100000 | 1.000 | 0.004 |
| 19021191 | 19B | Coriander, seed-babyfood | 7.100000 | 1.000 | 0.004 |
| 15001200 | 15 | Corn, field, flour | 0.081000 | 1.000 | 1.000 |
| 15001201 | 15 | Corn, field, flour-babyfood | 0.081000 | 1.000 | 1.000 |
| 15001210 | 15 | Corn, field, meal | 14.000000 | 1.000 | 0.008 |
| 15001211 | 15 | Corn, field, meal-babyfood | 14.000000 | 1.000 | 0.008 |
| 15001220 | 15 | Corn, field, bran | 14.000000 | 1.000 | 0.008 |
| 15001230 | 15 | Corn, field, starch | 6.600000 | 1.000 | 0.008 |
| 15001231 | 15 | Corn, field, starch-babyfood | 6.600000 | 1.000 | 0.008 |
| 15001240 | 15 | Corn, field, syrup | 0.600000 | 1.000 | 0.008 |
| 15001241 | 15 | Corn, field, syrup-babyfood | 0.600000 | 1.000 | 0.008 |
| 15001250 | 15 | Corn, field, oil | 0.400000 | 1.000 | 0.008 |
| 15001251 | 15 | Corn, field, oil-babyfood | 0.400000 | 1.000 | 0.008 |
| 15001260 | 15 | Corn, pop | 1.700000 | 1.000 | 0.008 |
| 95001280 | O | Cottonseed, oil | 1.500000 | 1.000 | 0.004 |
| 95001281 | O | Cottonseed, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 95001310 | O | Cranberry, dried | 1.000000 | 1.000 | 0.004 |
| 13021370 | 13B | Currant, dried | 1.000000 | 1.000 | 0.004 |
| 95001410 | O | Date | 0.900000 | 1.000 | 0.004 |
| 19021430 | 19B | Dill, seed | 7.100000 | 1.000 | 0.004 |
| 19011440 | 19A | Dillweed | 63.500000 | 1.000 | 0.004 |
| 70001450 | P | Egg, whole | | | |
| | | 110-Uncooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 120-Uncooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 210-Cooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 211-Cooked; Fresh or N/S; Baked | 0.000000 | 1.000 | 0.004 |
| | | 212-Cooked; Fresh or N/S; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 213-Cooked; Fresh or N/S; Fried | 0.000000 | 1.000 | 0.004 |
| | | 214-Cooked; Fresh or N/S; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | | 215-Cooked; Fresh or N/S; Boiled/baked | 0.000000 | 1.000 | 0.004 |
| | | 221-Cooked; Frozen; Baked | 0.000000 | 1.000 | 0.004 |
| | | 223-Cooked; Frozen; Fried | 0.000000 | 1.000 | 0.004 |
| | | 224-Cooked; Frozen; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | | 230-Cooked; Dried; Cook Meth N/S | 402.500000 | 1.000 | 0.004 |
| | | 232-Cooked; Dried; Boiled | 402.500000 | 1.000 | 0.004 |
| | | 233-Cooked; Dried; Fried | 402.500000 | 1.000 | 0.004 |
| | | 240-Cooked; Canned; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 242-Cooked; Canned; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 252-Cooked; Cured etc; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 253-Cooked; Cured etc; Fried | 0.000000 | 1.000 | 0.004 |
| 70001460 | P | Egg, white | | | |
| | | 110-Uncooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 120-Uncooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 130-Uncooked; Dried; Cook Meth N/S | 402.500000 | 1.000 | 0.004 |
| | | 210-Cooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 211-Cooked; Fresh or N/S; Baked | 0.000000 | 1.000 | 0.004 |
| | | 212-Cooked; Fresh or N/S; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 213-Cooked; Fresh or N/S; Fried | 0.000000 | 1.000 | 0.004 |
| | | 214-Cooked; Fresh or N/S; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | | 221-Cooked; Frozen; Baked | 0.000000 | 1.000 | 0.004 |
| | | 223-Cooked; Frozen; Fried | 0.000000 | 1.000 | 0.004 |
| | | 230-Cooked; Dried; Cook Meth N/S | 402.500000 | 1.000 | 0.004 |
| | | 232-Cooked; Dried; Boiled | 402.500000 | 1.000 | 0.004 |

| | | | | | |
|---|---|---|---|---|---|
| | | 233-Cooked; Dried; Fried | 402.500000 | 1.000 | 0.004 |
| | | 240-Cooked; Canned; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 242-Cooked; Canned; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 250-Cooked; Cured etc; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| 70001461 | P | Egg, white (solids)-babyfood | 402.500000 | 1.000 | 0.004 |
| 95001540 | O | Fig, dried | 1.000000 | 1.000 | 0.004 |
| 14001550 | 14 | Filbert | 1.500000 | 1.000 | 0.004 |
| 14001560 | 14 | Filbert, oil | 1.500000 | 1.000 | 0.004 |
| 20001630 | 20 | Flaxseed, oil | 1.500000 | 1.000 | 0.004 |
| 03001650 | 3 | Garlic, dried | 10.900000 | 1.000 | 0.004 |
| 03001651 | 3 | Garlic, dried-babyfood | 10.900000 | 1.000 | 0.004 |
| 01031670 | 1CD | Ginger, dried | 10.900000 | 1.000 | 0.004 |
| 01011680 | 1AB | Ginseng, dried | 10.900000 | 1.000 | 0.004 |
| 95001780 | O | Grape, raisin | 1.000000 | 1.000 | 0.004 |
| 06031820 | 6C | Guar, seed | 4.500000 | 1.000 | 0.004 |
| 06031821 | 6C | Guar, seed-babyfood | 4.500000 | 1.000 | 0.004 |
| 19011840 | 19A | Herbs, other | 63.500000 | 1.000 | 0.004 |
| 19011841 | 19A | Herbs, other-babyfood | 63.500000 | 1.000 | 0.004 |
| 14001850 | 14 | Hickory nut | 3.900000 | 1.000 | 0.004 |
| 19012020 | 19A | Lemongrass | 63.500000 | 1.000 | 0.004 |
| 06032030 | 6C | Lentil, seed | 4.500000 | 1.000 | 0.004 |
| 95002120 | O | Lychee, dried | 1.000000 | 1.000 | 0.004 |
| 14002130 | 14 | Macadamia nut | 3.900000 | 1.000 | 0.004 |
| 95002160 | O | Mango, dried | 1.000000 | 1.000 | 0.004 |
| 95002180 | O | Maple, sugar | 1.200000 | 1.000 | 0.004 |
| 95002190 | O | Maple syrup | 1.200000 | 1.000 | 0.004 |
| 19012200 | 19A | Marjoram | 67.100000 | 1.000 | 0.004 |
| 19012201 | 19A | Marjoram-babyfood | 67.100000 | 1.000 | 0.004 |
| 27002220 | D | Milk, fat | | | |
| | | 110-Uncooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 120-Uncooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 130-Uncooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | | 150-Uncooked; Cured etc; Cook Meth N/S | 3.900000 | 1.000 | 0.004 |
| | | 210-Cooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 211-Cooked; Fresh or N/S; Baked | 0.000000 | 1.000 | 0.004 |
| | | 212-Cooked; Fresh or N/S; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 213-Cooked; Fresh or N/S; Fried | 0.000000 | 1.000 | 0.004 |
| | | 214-Cooked; Fresh or N/S; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | | 215-Cooked; Fresh or N/S; Boiled/baked | 0.000000 | 1.000 | 0.004 |
| | | 220-Cooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 221-Cooked; Frozen; Baked | 0.000000 | 1.000 | 0.004 |
| | | 222-Cooked; Frozen; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 223-Cooked; Frozen; Fried | 0.000000 | 1.000 | 0.004 |
| | | 224-Cooked; Frozen; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | | 230-Cooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | | 231-Cooked; Dried; Baked | 5.400000 | 1.000 | 0.004 |
| | | 232-Cooked; Dried; Boiled | 5.400000 | 1.000 | 0.004 |
| | | 233-Cooked; Dried; Fried | 5.400000 | 1.000 | 0.004 |
| | | 240-Cooked; Canned; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 242-Cooked; Canned; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 250-Cooked; Cured etc; Cook Meth N/S | 3.900000 | 1.000 | 0.004 |
| | | 253-Cooked; Cured etc; Fried | 3.900000 | 1.000 | 0.004 |
| | | 255-Cooked; Cured etc; Boiled/baked | 3.900000 | 1.000 | 0.004 |
| 27012230 | D | Milk, nonfat solids | | | |
| | | 110-Uncooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 120-Uncooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |

| | | | | |
|---|---|---|---|---|
| | 130-Uncooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | 150-Uncooked; Cured etc; Cook Meth N/S | 3.900000 | 1.000 | 0.004 |
| | 210-Cooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 211-Cooked; Fresh or N/S; Baked | 0.000000 | 1.000 | 0.004 |
| | 212-Cooked; Fresh or N/S; Boiled | 0.000000 | 1.000 | 0.004 |
| | 213-Cooked; Fresh or N/S; Fried | 0.000000 | 1.000 | 0.004 |
| | 214-Cooked; Fresh or N/S; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | 215-Cooked; Fresh or N/S; Boiled/baked | 0.000000 | 1.000 | 0.004 |
| | 220-Cooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 221-Cooked; Frozen; Baked | 0.000000 | 1.000 | 0.004 |
| | 222-Cooked; Frozen; Boiled | 0.000000 | 1.000 | 0.004 |
| | 223-Cooked; Frozen; Fried | 0.000000 | 1.000 | 0.004 |
| | 224-Cooked; Frozen; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | 230-Cooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | 231-Cooked; Dried; Baked | 5.400000 | 1.000 | 0.004 |
| | 232-Cooked; Dried; Boiled | 5.400000 | 1.000 | 0.004 |
| | 233-Cooked; Dried; Fried | 5.400000 | 1.000 | 0.004 |
| | 240-Cooked; Canned; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 242-Cooked; Canned; Boiled | 0.000000 | 1.000 | 0.004 |
| | 245-Cooked; Canned; Boiled/baked | 0.000000 | 1.000 | 0.004 |
| | 250-Cooked; Cured etc; Cook Meth N/S | 3.900000 | 1.000 | 0.004 |
| | 253-Cooked; Cured etc; Fried | 3.900000 | 1.000 | 0.004 |
| | 255-Cooked; Cured etc; Boiled/baked | 3.900000 | 1.000 | 0.004 |
| 27012231 D | Milk, nonfat solids-baby food/infant | | | |
| | 110-Uncooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 130-Uncooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | 211-Cooked; Fresh or N/S; Baked | 0.000000 | 1.000 | 0.004 |
| | 240-Cooked; Canned; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| 27022240 D | Milk, water | | | |
| | 110-Uncooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 120-Uncooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 130-Uncooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | 150-Uncooked; Cured etc; Cook Meth N/S | 3.900000 | 1.000 | 0.004 |
| | 210-Cooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 211-Cooked; Fresh or N/S; Baked | 0.000000 | 1.000 | 0.004 |
| | 212-Cooked; Fresh or N/S; Boiled | 0.000000 | 1.000 | 0.004 |
| | 213-Cooked; Fresh or N/S; Fried | 0.000000 | 1.000 | 0.004 |
| | 214-Cooked; Fresh or N/S; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | 215-Cooked; Fresh or N/S; Boiled/baked | 0.000000 | 1.000 | 0.004 |
| | 220-Cooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 221-Cooked; Frozen; Baked | 0.000000 | 1.000 | 0.004 |
| | 222-Cooked; Frozen; Boiled | 0.000000 | 1.000 | 0.004 |
| | 223-Cooked; Frozen; Fried | 0.000000 | 1.000 | 0.004 |
| | 224-Cooked; Frozen; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | 230-Cooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | 231-Cooked; Dried; Baked | 5.400000 | 1.000 | 0.004 |
| | 232-Cooked; Dried; Boiled | 5.400000 | 1.000 | 0.004 |
| | 233-Cooked; Dried; Fried | 5.400000 | 1.000 | 0.004 |

| | | | | | |
|---|---|---|---|---|---|
| | | 240-Cooked; Canned; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 242-Cooked; Canned; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 250-Cooked; Cured etc; Cook Meth N/S | 3.900000 | 1.000 | 0.004 |
| | | 253-Cooked; Cured etc; Fried | 3.900000 | 1.000 | 0.004 |
| | | 255-Cooked; Cured etc; Boiled/baked | 3.900000 | 1.000 | 0.004 |
| 27032251 | D | Milk, sugar (lactose)-baby food/infa | | | |
| | | 110-Uncooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 130-Uncooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | | 210-Cooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 212-Cooked; Fresh or N/S; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 230-Cooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | | 240-Cooked; Canned; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| 15002260 | 15 | Millet, grain | 2.900000 | 1.000 | 0.008 |
| 15002310 | 15 | Oat, bran | 74.200000 | 1.000 | 0.008 |
| 15002320 | 15 | Oat, flour | 0.337000 | 1.000 | 1.000 |
| 15002321 | 15 | Oat, flour-babyfood | 0.337000 | 1.000 | 1.000 |
| 15002330 | 15 | Oat, groats/rolled oats | 18.500000 | 1.000 | 0.008 |
| 15002331 | 15 | Oat, groats/rolled oats-babyfood | 18.500000 | 1.000 | 0.008 |
| 95002360 | O | Olive, oil | 1.500000 | 1.000 | 0.004 |
| 03002380 | 3 | Onion, dry bulb, dried | 1.700000 | 1.000 | 0.004 |
| 03002381 | 3 | Onion, dry bulb, dried-babyfood | 1.700000 | 1.000 | 0.004 |
| 95002440 | O | Palm, oil | 1.500000 | 1.000 | 0.004 |
| 95002441 | O | Palm, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 95002460 | O | Papaya, dried | 1.000000 | 1.000 | 0.004 |
| 19012490 | 19A | Parsley, dried leaves | 63.500000 | 1.000 | 0.004 |
| 19012491 | 19A | Parsley, dried leaves-babyfood | 63.500000 | 1.000 | 0.004 |
| 06032560 | 6C | Pea, dry | 4.500000 | 1.000 | 0.004 |
| 06032561 | 6C | Pea, dry-babyfood | 4.500000 | 1.000 | 0.004 |
| 06032580 | 6C | Pea, pigeon, seed | 4.500000 | 1.000 | 0.004 |
| 12002610 | 12 | Peach, dried | 1.000000 | 1.000 | 0.004 |
| 12002611 | 12 | Peach, dried-babyfood | 1.000000 | 1.000 | 0.004 |
| 95002630 | O | Peanut | 16.400000 | 1.000 | 0.004 |
| 95002640 | O | Peanut, butter | 16.400000 | 1.000 | 0.004 |
| 95002650 | O | Peanut, oil | 1.500000 | 1.000 | 0.004 |
| 11002670 | 11 | Pear, dried | 1.000000 | 1.000 | 0.004 |
| 14002690 | 14 | Pecan | 3.900000 | 1.000 | 0.004 |
| 08002710 | 8 | Pepper, bell, dried | 36.100000 | 1.000 | 0.004 |
| 08002711 | 8 | Pepper, bell, dried-babyfood | 36.100000 | 1.000 | 0.004 |
| 08002730 | 8 | Pepper, nonbell, dried | 36.100000 | 1.000 | 0.004 |
| 19022740 | 19B | Pepper, black and white | 7.100000 | 1.000 | 0.004 |
| 19022741 | 19B | Pepper, black and white-babyfood | 7.100000 | 1.000 | 0.004 |
| 95002760 | O | Peppermint, oil | 1.500000 | 1.000 | 0.004 |
| 95002780 | O | Pine nut | 8.800000 | 1.000 | 0.004 |
| 95002800 | O | Pineapple, dried | 1.000000 | 1.000 | 0.004 |
| 14002820 | 14 | Pistachio | 2.300000 | 1.000 | 0.004 |
| 95002840 | O | Plantain, dried | 1.000000 | 1.000 | 0.004 |
| 12002870 | 12 | Plum, prune, dried | 0.700000 | 1.000 | 0.004 |
| 12002871 | 12 | Plum, prune, dried-babyfood | 0.700000 | 1.000 | 0.004 |
| 01032960 | 1C | Potato, chips | 7.100000 | 1.000 | 0.004 |
| 01032970 | 1C | Potato, dry (granules/ flakes) | 25.600000 | 1.000 | 0.004 |
| 01032971 | 1C | Potato, dry (granules/ flakes)-b | 25.600000 | 1.000 | 0.004 |
| 01032980 | 1C | Potato, flour | 31.400000 | 1.000 | 0.004 |
| 01032981 | 1C | Potato, flour-babyfood | 31.400000 | 1.000 | 0.004 |
| 95003060 | O | Psyllium, seed | 7.100000 | 1.000 | 0.004 |
| 09023090 | 9B | Pumpkin, seed | 8.800000 | 1.000 | 0.004 |
| 95003110 | O | Quinoa, grain | 20.400000 | 1.000 | 0.004 |
| 20003190 | 20 | Rapeseed, oil | 1.500000 | 1.000 | 0.004 |
| 20003191 | 20 | Rapeseed, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 15003230 | 15 | Rice, white | 3.900000 | 1.000 | 0.008 |
| 15003231 | 15 | Rice, white-babyfood | 3.900000 | 1.000 | 0.008 |
| 15003240 | 15 | Rice, brown | 9.400000 | 1.000 | 0.008 |
| 15003241 | 15 | Rice, brown-babyfood | 9.400000 | 1.000 | 0.008 |
| 15003250 | 15 | Rice, flour | 0.160000 | 1.000 | 1.000 |
| 15003251 | 15 | Rice, flour-babyfood | 0.160000 | 1.000 | 1.000 |

| | | | | | |
|---|---|---|---|---|---|
| 15003260 | 15 | Rice, bran | 37.500000 | 1.000 | 0.008 |
| 15003261 | 15 | Rice, bran-babyfood | 37.500000 | 1.000 | 0.008 |
| 15003280 | 15 | Rye, grain | 2.900000 | 1.000 | 0.008 |
| 15003290 | 15 | Rye, flour | 0.134000 | 1.000 | 1.000 |
| 20003300 | 20 | Safflower, oil | 1.500000 | 1.000 | 0.004 |
| 20003301 | 20 | Safflower, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 19013340 | 19A | Savory | 63.500000 | 1.000 | 0.004 |
| 95003360 | O | Sesame, seed | 7.100000 | 1.000 | 0.004 |
| 95003361 | O | Sesame, seed-babyfood | 7.100000 | 1.000 | 0.004 |
| 95003370 | O | Sesame, oil | 1.500000 | 1.000 | 0.004 |
| 95003371 | O | Sesame, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 15003440 | 15 | Sorghum, grain | 20.400000 | 1.000 | 0.008 |
| 15003450 | 15 | Sorghum, syrup | 0.600000 | 1.000 | 0.008 |
| 06003480 | 6 | Soybean, flour | 0.081000 | 1.000 | 1.000 |
| 06003481 | 6 | Soybean, flour-babyfood | 0.081000 | 1.000 | 1.000 |
| 06003490 | 6 | Soybean, soy milk | 2.400000 | 1.000 | 0.004 |
| 06003491 | 6 | Soybean, soy milk-babyfood or in | 2.400000 | 1.000 | 0.004 |
| 06003500 | 6 | Soybean, oil | 1.500000 | 1.000 | 0.004 |
| 06003501 | 6 | Soybean, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 95003530 | O | Spearmint, oil | 1.500000 | 1.000 | 0.004 |
| 19023540 | 19B | Spices, other | 7.100000 | 1.000 | 0.004 |
| 19023541 | 19B | Spices, other-babyfood | 7.100000 | 1.000 | 0.004 |
| 95003620 | O | Sugarcane, sugar | 1.200000 | 1.000 | 0.004 |
| 95003621 | O | Sugarcane, sugar-babyfood | 1.200000 | 1.000 | 0.004 |
| 95003630 | O | Sugarcane, molasses | 1.200000 | 1.000 | 0.004 |
| 95003631 | O | Sugarcane, molasses-babyfood | 1.200000 | 1.000 | 0.004 |
| 20003640 | 20 | Sunflower, seed | 4.500000 | 1.000 | 0.004 |
| 20003650 | 20 | Sunflower, oil | 1.500000 | 1.000 | 0.004 |
| 20003651 | 20 | Sunflower, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 95003720 | O | Tea, dried | 67.100000 | 1.000 | 0.004 |
| 95003730 | O | Tea, instant | 67.100000 | 1.000 | 0.004 |
| 08003780 | 8 | Tomato, dried | 1.000000 | 1.000 | 0.004 |
| 08003781 | 8 | Tomato, dried-babyfood | 1.000000 | 1.000 | 0.004 |
| 15003810 | 15 | Triticale, flour | 0.134000 | 1.000 | 1.000 |
| 15003811 | 15 | Triticale, flour-babyfood | 0.134000 | 1.000 | 1.000 |
| 01033870 | 1CD | Turmeric | 7.100000 | 1.000 | 0.004 |
| 14003910 | 14 | Walnut | 2.000000 | 1.000 | 0.004 |
| 15004010 | 15 | Wheat, grain | 2.900000 | 1.000 | 0.008 |
| 15004011 | 15 | Wheat, grain-babyfood | 2.900000 | 1.000 | 0.008 |
| 15004020 | 15 | Wheat, flour | 0.134000 | 1.000 | 1.000 |
| 15004021 | 15 | Wheat, flour-babyfood | 0.134000 | 1.000 | 1.000 |
| 15004030 | 15 | Wheat, germ | 54.000000 | 1.000 | 0.008 |
| 15004040 | 15 | Wheat, bran | 74.200000 | 1.000 | 0.008 |
| 15004050 | 15 | Wild rice | 9.400000 | 1.000 | 0.008 |

Attachment 3. Inputs for the chronic dietary exposure analysis of fluoride from food fumigation with sulfuryl fluoride

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U.S. Environmental Protection Agency                              Ver. 2.00
DEEM-FCID Chronic analysis for FLUORIDE                          1994-98 data
Residue file: C:\Documents and Settings\mdoherty\My Documents\Chemistry Reviews\!DEEM
Runs\Sulfuryl Fluoride\F Food Fumigation - 2009  RevisedCT - 4-28 Strict Label.R98
                                                               Adjust. #2 used
Analysis Date 05-06-2009            Residue file dated: 05-06-2009/13:54:06/8
Reference dose (RfD) = 0.114 mg/kg bw/day
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| Food Crop EPA Code | Grp | Food Name | Residue (ppm) | Adj.Factors #1 | Adj.Factors #2 | Comment |
|---|---|---|---|---|---|---|
| 14000030 | 14 | Almond | 9.700000 | 1.000 | 0.100 | |
| 14000031 | 14 | Almond-babyfood | 9.700000 | 1.000 | 0.100 | |
| 11000090 | 11 | Apple, dried | 1.000000 | 1.000 | 0.690 | |
| 11000091 | 11 | Apple, dried-babyfood | 1.000000 | 1.000 | 0.690 | |
| 12000130 | 12 | Apricot, dried | 1.000000 | 1.000 | 0.690 | |
| 95000240 | O | Banana, dried | 1.000000 | 1.000 | 0.690 | |
| 95000241 | O | Banana, dried-babyfood | 1.000000 | 1.000 | 0.690 | |
| 15000250 | 15 | Barley, pearled barley | 3.700000 | 1.000 | 0.001 | |
| 15000251 | 15 | Barley, pearled barley-babyfood | 3.700000 | 1.000 | 0.001 | |
| 15000260 | 15 | Barley, flour | 3.700000 | 0.730 | 0.001 | |
| 15000261 | 15 | Barley, flour-babyfood | 3.700000 | 0.730 | 0.001 | |
| 15000270 | 15 | Barley, bran | 3.700000 | 2.560 | 0.001 | |
| 19010290 | 19A | Basil, dried leaves | 67.100000 | 1.000 | 0.001 | |
| 19010291 | 19A | Basil, dried leaves-babyfood | 67.100000 | 1.000 | 0.001 | |
| 06030300 | 6C | Bean, black, seed | 4.500000 | 1.000 | 1.000 | |
| 06030320 | 6C | Bean, broad, seed | 4.500000 | 1.000 | 1.000 | |
| 06030340 | 6C | Bean, cowpea, seed | 4.500000 | 1.000 | 1.000 | |
| 06030350 | 6C | Bean, great northern, seed | 4.500000 | 1.000 | 1.000 | |
| 06030360 | 6C | Bean, kidney, seed | 4.500000 | 1.000 | 1.000 | |
| 06030380 | 6C | Bean, lima, seed | 4.500000 | 1.000 | 1.000 | |
| 06030390 | 6C | Bean, mung, seed | 4.500000 | 1.000 | 1.000 | |
| 06030400 | 6C | Bean, navy, seed | 4.500000 | 1.000 | 1.000 | |
| 06030410 | 6C | Bean, pink, seed | 4.500000 | 1.000 | 1.000 | |
| 06030420 | 6C | Bean, pinto, seed | 4.500000 | 1.000 | 1.000 | |
| 14000590 | 14 | Brazil nut | 5.300000 | 1.000 | 0.100 | |
| 14000680 | 14 | Butternut | 5.300000 | 1.000 | 0.100 | |
| 14000810 | 14 | Cashew | 5.300000 | 1.000 | 0.100 | |
| 14000920 | 14 | Chestnut | 5.300000 | 1.000 | 0.100 | |
| 06030980 | 6C | Chickpea, seed | 4.500000 | 1.000 | 1.000 | |
| 06030981 | 6C | Chickpea, seed-babyfood | 4.500000 | 1.000 | 1.000 | |
| 06030990 | 6C | Chickpea, flour | 4.500000 | 1.000 | 1.000 | |
| 19011030 | 19A | Chive | 63.500000 | 1.000 | 0.001 | |
| 19021050 | 19B | Cinnamon | 73.500000 | 1.000 | 0.001 | |
| 19021051 | 19B | Cinnamon-babyfood | 73.500000 | 1.000 | 0.001 | |
| 95001090 | O | Cocoa bean, chocolate | 8.400000 | 1.000 | 1.000 | |
| 95001100 | O | Cocoa bean, powder | 8.400000 | 1.000 | 1.000 | |
| 95001110 | O | Coconut, meat | 49.100000 | 1.000 | 0.001 | |
| 95001111 | O | Coconut- meat-babyfood | 49.100000 | 1.000 | 0.001 | |
| 95001120 | O | Coconut, dried | 49.100000 | 1.000 | 0.001 | |
| 95001130 | O | Coconut, milk | 49.100000 | 1.000 | 0.001 | |
| 95001150 | O | Coffee, roasted bean | 7.100000 | 1.000 | 0.001 | |
| 95001160 | O | Coffee, instant | 13.900000 | 1.000 | 0.001 | |
| 19011180 | 19A | Coriander, leaves | 63.500000 | 1.000 | 0.001 | |
| 19011181 | 19A | Coriander, leaves-babyfood | 63.500000 | 1.000 | 0.001 | |
| 19021190 | 19B | Coriander, seed | 7.100000 | 1.000 | 0.001 | |
| 19021191 | 19B | Coriander, seed-babyfood | 7.100000 | 1.000 | 0.001 | |
| 15001200 | 15 | Corn, field, flour | 16.900000 | 1.000 | 0.001 | |
| 15001201 | 15 | Corn, field, flour-babyfood | 16.900000 | 1.000 | 0.001 | |
| 15001210 | 15 | Corn, field, meal | 2.800000 | 1.000 | 0.001 | |
| 15001211 | 15 | Corn, field, meal-babyfood | 2.800000 | 1.000 | 0.001 | |
| 15001220 | 15 | Corn, field, bran | 2.800000 | 1.000 | 0.001 | |
| 15001230 | 15 | Corn, field, starch | 0.600000 | 1.000 | 0.001 | |
| 15001231 | 15 | Corn, field, starch-babyfood | 0.600000 | 1.000 | 0.001 | |
| 15001240 | 15 | Corn, field, syrup | 0.600000 | 1.000 | 0.001 | |
| 15001241 | 15 | Corn, field, syrup-babyfood | 0.600000 | 1.000 | 0.001 | |
| 15001260 | 15 | Corn, pop | 1.700000 | 1.000 | 0.001 | |

| | | | | | |
|---|---|---|---|---|---|
| 95001310 | O | Cranberry, dried | 1.000000 | 1.000 | 0.100 |
| 13021370 | 13B | Currant, dried | 1.000000 | 1.000 | 0.100 |
| 95001410 | O | Date | | | |
| | | 130-Uncooked; Dried; Cook Meth N/S | 0.900000 | 1.000 | 0.420 |
| | | 210-Cooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.000 |
| | | 211-Cooked; Fresh or N/S; Baked | 0.000000 | 1.000 | 0.000 |
| | | 212-Cooked; Fresh or N/S; Boiled | 0.000000 | 1.000 | 0.000 |
| | | 230-Cooked; Dried; Cook Meth N/S | 0.900000 | 1.000 | 0.420 |
| 19021430 | 19B | Dill, seed | 7.100000 | 1.000 | 0.001 |
| 19011440 | 19A | Dillweed | 63.500000 | 1.000 | 0.001 |
| 95001540 | O | Fig, dried | 1.000000 | 1.000 | 0.690 |
| 14001550 | 14 | Filbert | 2.500000 | 1.000 | 0.100 |
| 03001650 | 3 | Garlic, dried | 10.900000 | 1.000 | 0.001 |
| 01031660 | 1CD | Ginger | 10.900000 | 1.000 | 0.001 |
| 01031661 | 1CD | Ginger-babyfood | 10.900000 | 1.000 | 0.001 |
| 01031670 | 1CD | Ginger, dried | 10.900000 | 1.000 | 0.001 |
| 95001780 | O | Grape, raisin | 1.000000 | 1.000 | 0.690 |
| 06031820 | 6C | Guar, seed | 4.500000 | 1.000 | 1.000 |
| 06031821 | 6C | Guar, seed-babyfood | 4.500000 | 1.000 | 1.000 |
| 19011840 | 19A | Herbs, other | 63.500000 | 1.000 | 0.001 |
| 19011841 | 19A | Herbs, other-babyfood | 63.500000 | 1.000 | 0.001 |
| 14001850 | 14 | Hickory nut | 5.300000 | 1.000 | 0.100 |
| 19012020 | 19A | Lemongrass | 63.500000 | 1.000 | 0.001 |
| 06032030 | 6C | Lentil, seed | 4.500000 | 1.000 | 1.000 |
| 95002120 | O | Lychee, dried | 1.000000 | 1.000 | 0.690 |
| 14002130 | 14 | Macadamia nut | 5.300000 | 1.000 | 0.001 |
| 95002160 | O | Mango, dried | 1.000000 | 1.000 | 0.690 |
| 19012200 | 19A | Marjoram | 67.100000 | 1.000 | 0.001 |
| 19012201 | 19A | Marjoram-babyfood | 67.100000 | 1.000 | 0.001 |
| 15002260 | 15 | Millet, grain | 2.900000 | 1.000 | 0.001 |
| 15002310 | 15 | Oat, bran | 18.500000 | 2.560 | 0.001 |
| 15002320 | 15 | Oat, flour | 18.500000 | 0.730 | 0.001 |
| 15002321 | 15 | Oat, flour-babyfood | 18.500000 | 0.730 | 0.001 |
| 15002330 | 15 | Oat, groats/rolled oats | 18.500000 | 1.000 | 0.001 |
| 15002331 | 15 | Oat, groats/rolled oats-babyfood | 18.500000 | 1.000 | 0.001 |
| 95002460 | O | Papaya, dried | 1.000000 | 1.000 | 0.690 |
| 19012490 | 19A | Parsley, dried leaves | 63.500000 | 1.000 | 0.001 |
| 19012491 | 19A | Parsley, dried leaves-babyfood | 63.500000 | 1.000 | 0.001 |
| 06032560 | 6C | Pea, dry | 4.500000 | 1.000 | 1.000 |
| 06032561 | 6C | Pea, dry-babyfood | 4.500000 | 1.000 | 1.000 |
| 06032580 | 6C | Pea, pigeon, seed | 4.500000 | 1.000 | 1.000 |
| 12002610 | 12 | Peach, dried | 1.000000 | 1.000 | 0.690 |
| 12002611 | 12 | Peach, dried-babyfood | 1.000000 | 1.000 | 0.690 |
| 95002630 | O | Peanut | 16.400000 | 1.000 | 0.006 |
| 95002640 | O | Peanut, butter | 16.400000 | 1.000 | 0.006 |
| 11002670 | 11 | Pear, dried | 1.000000 | 1.000 | 0.690 |
| 14002690 | 14 | Pecan | 5.300000 | 1.000 | 0.100 |
| 19022740 | 19B | Pepper, black and white | 7.100000 | 1.000 | 0.001 |
| 19022741 | 19B | Pepper, black and white-babyfood | 7.100000 | 1.000 | 0.001 |
| 95002780 | O | Pine nut | 8.800000 | 1.000 | 0.100 |
| 95002800 | O | Pineapple, dried | 1.000000 | 1.000 | 0.690 |
| 14002820 | 14 | Pistachio | 3.200000 | 1.000 | 0.270 |
| 95002840 | O | Plantain, dried | 1.000000 | 1.000 | 0.690 |
| 12002870 | 12 | Plum, prune, dried | 0.700000 | 1.000 | 0.690 |
| 12002871 | 12 | Plum, prune, dried-babyfood | 0.700000 | 1.000 | 0.690 |
| 15003230 | 15 | Rice, white | 4.500000 | 1.000 | 0.030 |
| 15003231 | 15 | Rice, white-babyfood | 4.500000 | 1.000 | 0.030 |
| 15003240 | 15 | Rice, brown | 12.500000 | 1.000 | 0.030 |
| 15003241 | 15 | Rice, brown-babyfood | 12.500000 | 1.000 | 0.030 |
| 15003250 | 15 | Rice, flour | 32.500000 | 1.000 | 0.030 |
| 15003251 | 15 | Rice, flour-babyfood | 32.500000 | 1.000 | 0.030 |
| 15003260 | 15 | Rice, bran | 37.500000 | 1.000 | 0.030 |
| 15003261 | 15 | Rice, bran-babyfood | 37.500000 | 1.000 | 0.030 |
| 19013340 | 19A | Savory | 63.500000 | 1.000 | 0.001 |
| 15003440 | 15 | Sorghum, grain | 20.400000 | 1.000 | 0.001 |
| 19023540 | 19B | Spices, other | 7.100000 | 1.000 | 0.001 |
| 19023541 | 19B | Spices, other-babyfood | 7.100000 | 1.000 | 0.001 |
| 15003810 | 15 | Triticale, flour | 2.900000 | 0.380 | 0.001 |
| 15003811 | 15 | Triticale, flour-babyfood | 2.900000 | 0.380 | 0.001 |

| | | | | | |
|---|---|---|---|---|---|
| 01033870 | 1CD | Turmeric | 7.100000 | 1.000 | 0.001 |
| 14003910 | 14 | Walnut | 2.400000 | 1.000 | 0.990 |
| 15004010 | 15 | Wheat, grain | 2.900000 | 1.000 | 0.004 |
| 15004011 | 15 | Wheat, grain-babyfood | 2.900000 | 1.000 | 0.004 |
| 15004020 | 15 | Wheat, flour | 31.400000 | 1.000 | 0.001 |
| 15004021 | 15 | Wheat, flour-babyfood | 31.400000 | 1.000 | 0.001 |
| 15004030 | 15 | Wheat, germ | 13.900000 | 1.000 | 0.001 |
| 15004040 | 15 | Wheat, bran | 74.200000 | 1.000 | 0.001 |
| 15004050 | 15 | Wild rice | 12.500000 | 1.000 | 0.030 |

Attachment 4. Results of the chronic dietary exposure analysis of fluoride from cryolite

```
U.S. Environmental Protection Agency                              Ver. 2.00
DEEM-FCID Chronic analysis for CRYOLITE                       (1994-98 data)
Residue file name: C:\Documents and Settings\mdoherty\My Documents\Chemistry Reviews\!DEEM
Runs\Sulfuryl Fluoride\Cryolite-AR-CT new raisin factor.R98
                                                     Adjustment factor #2 used.
Analysis Date 02-19-2009/14:16:56     Residue file dated: 06-24-2004/10:05:08/8
===============================================================================
                    Total exposure by population subgroup
-------------------------------------------------------------------------------

                                                 Total Exposure
                                         -----------------------------------
          Population                        mg/kg
           Subgroup                       body wt/day
 -------------------------------------   -------------
U.S. Population (total)                    0.000682

U.S. Population (spring season)            0.000655
U.S. Population (summer season)            0.000735
U.S. Population (autumn season)            0.000637
U.S. Population (winter season)            0.000702

Northeast region                           0.000807
Midwest region                             0.000670
Southern region                            0.000579
Western region                             0.000745

Hispanics                                  0.000605
Non-hispanic whites                        0.000716
Non-hispanic blacks                        0.000590
Non-hisp/non-white/non-black               0.000563

All infants (< 1 year)                     0.000956
Nursing infants                            0.000401
Non-nursing infants                        0.001167
Children 1-6  yrs                          0.002334
Children 7-12 yrs                          0.000842

Females 13-19 (not preg or nursing)        0.000390
Females 20+ (not preg or nursing)          0.000530
Females 13-50 yrs                          0.000499
Females 13+ (preg/not nursing)             0.000342
Females 13+ (nursing)                      0.000471

Males 13-19 yrs                            0.000304
Males 20+ yrs                              0.000434
Seniors 55+                                0.000563

Children 1-2 yrs                           0.003275
Children 3-5 yrs                           0.002112
Children 6-12 yrs                          0.000885
Youth 13-19 yrs                            0.000346
Adults 20-49 yrs                           0.000445
Adults 50+ yrs                             0.000547
Females 13-49 yrs                          0.000473

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Attachment 5. Results of the chronic dietary exposure analysis of fluoride from structural fumigation with sulfuryl fluoride

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U.S. Environmental Protection Agency                               Ver. 2.00
DEEM-FCID Chronic analysis for FLUORIDE                       (1994-98 data)
Residue file name: C:\Documents and Settings\mdoherty\My Documents\Chemistry Reviews\!DEEM
Runs\Sulfuryl Fluoride\F Space Fumigation - 2009 - 5-1.R98
                                                      Adjustment factor #2 used.
Analysis Date 05-06-2009/13:56:30     Residue file dated: 05-06-2009/13:52:45/8
Reference dose (RfD, Chronic) = .114 mg/kg bw/day
===============================================================================
                    Total exposure by population subgroup
-------------------------------------------------------------------------------
```

| Population Subgroup | Total Exposure mg/kg body wt/day |
|---|---|
| U.S. Population (total) | 0.000336 |
| U.S. Population (spring season) | 0.000343 |
| U.S. Population (summer season) | 0.000328 |
| U.S. Population (autumn season) | 0.000333 |
| U.S. Population (winter season) | 0.000341 |
| Northeast region | 0.000354 |
| Midwest region | 0.000348 |
| Southern region | 0.000314 |
| Western region | 0.000341 |
| Hispanics | 0.000336 |
| Non-hispanic whites | 0.000337 |
| Non-hispanic blacks | 0.000317 |
| Non-hisp/non-white/non-black | 0.000364 |
| All infants (< 1 year) | 0.000505 |
| Nursing infants | 0.000272 |
| Non-nursing infants | 0.000593 |
| Children 1-6 yrs | 0.000792 |
| Children 7-12 yrs | 0.000515 |
| Females 13-19 (not preg or nursing) | 0.000282 |
| Females 20+ (not preg or nursing) | 0.000225 |
| Females 13-50 yrs | 0.000264 |
| Females 13+ (preg/not nursing) | 0.000256 |
| Females 13+ (nursing) | 0.000306 |
| Males 13-19 yrs | 0.000355 |
| Males 20+ yrs | 0.000278 |
| Seniors 55+ | 0.000213 |
| Children 1-2 yrs | 0.000827 |
| Children 3-5 yrs | 0.000800 |
| Children 6-12 yrs | 0.000543 |
| Youth 13-19 yrs | 0.000320 |
| Adults 20-49 yrs | 0.000272 |
| Adults 50+ yrs | 0.000215 |
| Females 13-49 yrs | 0.000249 |

Attachment 6. Results of the chronic dietary exposure analysis of fluoride from food fumigation with sulfuryl fluoride

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U.S. Environmental Protection Agency                                 Ver. 2.00
DEEM-FCID Chronic analysis for FLUORIDE                         (1994-98 data)
Residue file name: C:\Documents and Settings\mdoherty\My Documents\Chemistry Reviews\!DEEM
Runs\Sulfuryl Fluoride\F Food Fumigation - 2009  RevisedCT - 4-28 Strict Label.R98
                                                     Adjustment factor #2 used.
Analysis Date 05-06-2009/13:57:26     Residue file dated: 05-06-2009/13:54:06/8
Reference dose (RfD, Chronic) = .114 mg/kg bw/day
===============================================================================
                    Total exposure by population subgroup
-------------------------------------------------------------------------------

                                                Total Exposure
                                         ----------------------------------
          Population                        mg/kg
           Subgroup                       body wt/day
--------------------------------------   -------------
U.S. Population (total)                    0.001023

U.S. Population (spring season)            0.000989
U.S. Population (summer season)            0.000931
U.S. Population (autumn season)            0.001055
U.S. Population (winter season)            0.001123

Northeast region                           0.000925
Midwest region                             0.000941
Southern region                            0.000972
Western region                             0.00128

Hispanics                                  0.001799
Non-hispanic whites                        0.000925
Non-hispanic blacks                        0.000869
Non-hisp/non-white/non-black               0.00122

All infants (< 1 year)                     0.001071
Nursing infants                            0.00061
Non-nursing infants                        0.001246
Children 1-6  yrs                          0.00224
Children 7-12 yrs                          0.001657

Females 13-19 (not preg or nursing)        0.000961
Females 20+ (not preg or nursing)          0.000702
Females 13-50 yrs                          0.000975
Females 13+ (preg/not nursing)             0.001258
Females 13+ (nursing)                      0.001155

Males 13-19 yrs                            0.001099
Males 20+ yrs                              0.000847
Seniors 55+                                0.000695

Children 1-2 yrs                           0.002169
Children 3-5 yrs                           0.002293
Children 6-12 yrs                          0.001743
Youth 13-19 yrs                            0.001032
Adults 20-49 yrs                           0.000814
Adults 50+ yrs                             0.000719
Females 13-49 yrs                          0.000799

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Attachment 7.  Commodity contribution summary for the chronic dietary exposure analysis of fluoride from cryolite

| | Exposure, mg/kg/day | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | U.S. | Infants | K1-2 | K3-5 | K6-12 | Y13-19 | A20-50 | A50+ | Fem13-49 |
| Crop Group = (O)  Other | 0.0005567 | 0.0008359 | 0.0030082 | 0.0018675 | 0.0007324 | 0.0002507 | 0.0003483 | 0.0004115 | 0.0003740 |
| Crop Group = (1)  Root and Tuber Vegetables | 0.0000189 | 0.0000169 | 0.0000430 | 0.0000388 | 0.0000260 | 0.0000195 | 0.0000157 | 0.0000150 | 0.0000147 |
| Crop Group = (1C) Tuberous and Corm Vegetables | 0.0000189 | 0.0000169 | 0.0000430 | 0.0000388 | 0.0000260 | 0.0000195 | 0.0000157 | 0.0000150 | 0.0000147 |
| Crop Group = (4)  Leafy Vegetables (except Brassica) | 0.0000077 | 0.0000000 | 0.0000040 | 0.0000061 | 0.0000063 | 0.0000075 | 0.0000088 | 0.0000077 | 0.0000094 |
| Crop Group = (4A) Leafy Greens | 0.0000077 | 0.0000000 | 0.0000040 | 0.0000061 | 0.0000063 | 0.0000075 | 0.0000088 | 0.0000077 | 0.0000094 |
| Crop Group = (5)  Brassica (Cole) Leafy Vegetables | 0.0000148 | 0.0000082 | 0.0000313 | 0.0000230 | 0.0000167 | 0.0000094 | 0.0000131 | 0.0000161 | 0.0000134 |
| Crop Group = (5A) Brassica: Head and Stem | 0.0000135 | 0.0000080 | 0.0000287 | 0.0000201 | 0.0000157 | 0.0000073 | 0.0000122 | 0.0000146 | 0.0000123 |
| Crop Group = (5B) Brassica: Leafy Greens | 0.0000013 | 0.0000002 | 0.0000026 | 0.0000029 | 0.0000011 | 0.0000021 | 0.0000009 | 0.0000015 | 0.0000010 |
| Crop Group = (8)  Fruiting Vegetables | 0.0000149 | 0.0000059 | 0.0000274 | 0.0000251 | 0.0000182 | 0.0000136 | 0.0000141 | 0.0000127 | 0.0000131 |
| Crop Group = (9)  Curcurbit Vegetables | 0.0000091 | 0.0000110 | 0.0000162 | 0.0000186 | 0.0000121 | 0.0000074 | 0.0000068 | 0.0000098 | 0.0000074 |
| Crop Group = (9A) Melons | 0.0000052 | 0.0000017 | 0.0000109 | 0.0000133 | 0.0000080 | 0.0000043 | 0.0000034 | 0.0000057 | 0.0000041 |
| Crop Group = (9B) Squash/Cucumbers | 0.0000039 | 0.0000094 | 0.0000052 | 0.0000054 | 0.0000041 | 0.0000031 | 0.0000034 | 0.0000041 | 0.0000033 |
| Crop Group = (10) Citrus Fruits | 0.0000462 | 0.0000159 | 0.0001014 | 0.0001027 | 0.0000544 | 0.0000298 | 0.0000305 | 0.0000614 | 0.0000322 |
| Crop Group = (12) Stone Fruits | 0.0000073 | 0.0000502 | 0.0000294 | 0.0000159 | 0.0000092 | 0.0000033 | 0.0000039 | 0.0000073 | 0.0000046 |
| Crop Group = (13) Berries | 0.0000061 | 0.0000117 | 0.0000147 | 0.0000147 | 0.0000093 | 0.0000049 | 0.0000042 | 0.0000056 | 0.0000046 |
| Crop Group = (13A)Berries: Caneberry Group | 0.0000041 | 0.0000050 | 0.0000088 | 0.0000096 | 0.0000062 | 0.0000035 | 0.0000027 | 0.0000043 | 0.0000028 |
| Crop Group = (13B)Berries: Bushberry Group | 0.0000020 | 0.0000067 | 0.0000059 | 0.0000051 | 0.0000030 | 0.0000014 | 0.0000015 | 0.0000013 | 0.0000017 |

Attachment 8. Commodity contribution summary for the chronic dietary exposure analysis of fluoride from structural fumigation with sulfuryl fluoride

| | Exposure, mg/kg/day | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | U.S. | Infants | K1-2 | K3-5 | K6-12 | Y13-19 | A20-50 | A50+ | Fem13-49 |
| Crop Group = (O) Other | 0.0000126 | 0.0000145 | 0.0000255 | 0.0000293 | 0.0000196 | 0.0000098 | 0.0000101 | 0.0000103 | 0.0000095 |
| Crop Group = (M) Meat | 0.0000004 | 0.0000000 | 0.0000009 | 0.0000002 | 0.0000006 | 0.0000011 | 0.0000004 | 0.0000002 | 0.0000004 |
| Crop Group = (P) Poultry | 0.0000103 | 0.0000055 | 0.0000296 | 0.0000210 | 0.0000118 | 0.0000079 | 0.0000102 | 0.0000068 | 0.0000110 |
| Crop Group = (D) Dairy Products | 0.0000032 | 0.0000684 | 0.0000135 | 0.0000096 | 0.0000031 | 0.0000017 | 0.0000011 | 0.0000014 | 0.0000011 |
| Crop Group = (1) Root and Tuber Vegetables | 0.0000029 | 0.0000047 | 0.0000079 | 0.0000082 | 0.0000050 | 0.0000029 | 0.0000024 | 0.0000014 | 0.0000024 |
| Crop Group = (1A) Root Vegetables | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (1B) Root Vegetables (exc sugar beet) subgroup | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (1C) Tuberous and Corm Vegetables | 0.0000029 | 0.0000047 | 0.0000079 | 0.0000082 | 0.0000050 | 0.0000029 | 0.0000024 | 0.0000014 | 0.0000023 |
| Crop Group = (1D) Tuberous/Corm Vegetables (exc sugar beet) | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (3) Bulb Vegetables | 0.0000002 | 0.0000002 | 0.0000006 | 0.0000005 | 0.0000003 | 0.0000002 | 0.0000002 | 0.0000002 | 0.0000002 |
| Crop Group = (4) Leafy Vegetables (except Brassica) | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (4A) Leafy Greens | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (6) Legume Vegetables (Succulent or Dried) | 0.0000063 | 0.0000318 | 0.0000141 | 0.0000126 | 0.0000083 | 0.0000056 | 0.0000050 | 0.0000044 | 0.0000046 |
| Crop Group = (6C) Dried Shelled Pea/Bean (exc Soybean) | 0.0000021 | 0.0000013 | 0.0000042 | 0.0000039 | 0.0000026 | 0.0000021 | 0.0000019 | 0.0000018 | 0.0000017 |
| Crop Group = (8) Fruiting Vegetables | 0.0000004 | 0.0000003 | 0.0000010 | 0.0000009 | 0.0000006 | 0.0000004 | 0.0000004 | 0.0000002 | 0.0000003 |
| Crop Group = (9) Curcurbit Vegetables | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (9B) Squash/Cucumbers | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (11) Pome Fruits | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (12) Stone Fruits | 0.0000000 | 0.0000000 | 0.0000001 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (13) Berries | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (13B)Berries: Bushberry Group | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (14) Tree Nuts | 0.0000003 | 0.0000000 | 0.0000003 | 0.0000004 | 0.0000003 | 0.0000002 | 0.0000003 | 0.0000003 | 0.0000003 |
| Crop Group = (15) Cereal Grains | 0.0002967 | 0.0003739 | 0.0007256 | 0.0007112 | 0.0004892 | 0.0002876 | 0.0002397 | 0.0001887 | 0.0002171 |
| Crop Group = (19) Herbs and Spices | 0.0000021 | 0.0000013 | 0.0000074 | 0.0000058 | 0.0000035 | 0.0000021 | 0.0000016 | 0.0000011 | 0.0000016 |
| Crop Group = (19A)Herbs | 0.0000017 | 0.0000011 | 0.0000067 | 0.0000049 | 0.0000027 | 0.0000017 | 0.0000013 | 0.0000007 | 0.0000012 |
| Crop Group = (19B)Spices | 0.0000004 | 0.0000002 | 0.0000007 | 0.0000009 | 0.0000008 | 0.0000005 | 0.0000004 | 0.0000003 | 0.0000003 |
| Crop Group = (20) Oilseeds | 0.0000003 | 0.0000039 | 0.0000004 | 0.0000004 | 0.0000004 | 0.0000003 | 0.0000002 | 0.0000002 | 0.0000002 |

Attachment 9. Commodity contribution summary for the chronic dietary exposure analysis of fluoride from food fumigation with sulfuryl fluoride

| | Exposure, mg/kg/day | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | U.S. | Infants | K1-2 | K3-5 | K6-12 | Y13-19 | A20-50 | A50+ | Fem13-49 |
| Crop Group = (O) Other | 0.0003529 | 0.0001013 | 0.0007605 | 0.0009980 | 0.0008601 | 0.0003866 | 0.0002348 | 0.0001874 | 0.0002649 |
| Crop Group = (1) Root and Tuber Vegetables | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (1C) Tuberous and Corm Vegetables | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (1D) Tuberous/Corm Vegetables (exc sugar beet) | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (3) Bulb Vegetables | 0.0000000 | 0.0000000 | 0.0000001 | 0.0000001 | 0.0000001 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (6) Legume Vegetables (Succulent or Dried) | 0.0005229 | 0.0003147 | 0.0010429 | 0.0009719 | 0.0006484 | 0.0005166 | 0.0004653 | 0.0004409 | 0.0004267 |
| Crop Group = (6C) Dried Shelled Pea/Bean (exc Soybean) | 0.0005229 | 0.0003147 | 0.0010429 | 0.0009719 | 0.0006484 | 0.0005166 | 0.0004653 | 0.0004409 | 0.0004267 |
| Crop Group = (11) Pome Fruits | 0.0000013 | 0.0000017 | 0.0000027 | 0.0000030 | 0.0000023 | 0.0000012 | 0.0000009 | 0.0000011 | 0.0000010 |
| Crop Group = (12) Stone Fruits | 0.0000018 | 0.0000001 | 0.0000106 | 0.0000020 | 0.0000022 | 0.0000002 | 0.0000010 | 0.0000027 | 0.0000009 |
| Crop Group = (13) Berries | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (13B)Berries: Bushberry Group | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 | 0.0000000 |
| Crop Group = (14) Tree Nuts | 0.0000160 | 0.0000013 | 0.0000254 | 0.0000258 | 0.0000187 | 0.0000111 | 0.0000147 | 0.0000168 | 0.0000147 |
| Crop Group = (15) Cereal Grains | 0.0001279 | 0.0006516 | 0.0003245 | 0.0002907 | 0.0002108 | 0.0001159 | 0.0000964 | 0.0000703 | 0.0000906 |
| Crop Group = (19) Herbs and Spices | 0.0000005 | 0.0000003 | 0.0000018 | 0.0000014 | 0.0000009 | 0.0000005 | 0.0000004 | 0.0000003 | 0.0000004 |
| Crop Group = (19A)Herbs | 0.0000004 | 0.0000003 | 0.0000017 | 0.0000012 | 0.0000007 | 0.0000004 | 0.0000003 | 0.0000002 | 0.0000003 |
| Crop Group = (19B)Spices | 0.0000001 | 0.0000001 | 0.0000002 | 0.0000002 | 0.0000002 | 0.0000001 | 0.0000001 | 0.0000001 | 0.0000001 |

**UNITED STATES ENVIRONMENTAL PROTECTION AGENCY**
**WASHINGTON, D.C. 20460**

**OFFICE OF CHEMICAL SAFETY AND POLLUTION PREVENTION**

MEMORANDUM

Date: 1 July 2010

SUBJECT: Sulfuryl Fluoride: Estimates of Fluoride Exposure from Pesticidal Sources – Customized Age Groups

| | |
|---|---|
| PC Code: 078003 (Sulfuryl Fluoride) | DP Barcode: D379854 |
| MRID No.: None | Registration No.: None |
| Petition No.: None | Regulatory Action: None |
| Assessment Type: Single Chemical | Registration Case No.: None |
| TXR No.: None | CAS No.: 16984-48-8 |

FROM: Michael A. Doherty, Ph.D., Senior Chemist
Risk Assessment Branch II
Health Effects Division (7509P)

THROUGH: Christina Swartz, Branch Chief
Douglas A. Dotson, Ph.D., Senior Chemist
Risk Assessment Branch II
Office of Pesticide Programs
Health Effects Division (7509P)

TO: Elizabeth Doyle, Branch Chief
Human Health Risk Assessment Branch
Office of Water
Office of Science and Technology (4304T)

In May 2009, the EPA's Office of Pesticide Programs (OPP) provided the Office of Water (OW) with estimates of exposure to fluoride from use of the pesticides cryolite and sulfuryl fluoride (Memorandum from M. Doherty to E. Doyle, DP Number D362184, 6 May 2009). Those estimates were for the populations and sub-populations typically addressed by OPP dietary risk assessments; namely the general U.S. population, all infants (< 1 year), children 1-2 years, children 3-5 years, children 6-12 years, youth 13-19 years, adults 20-49 years, adults 50+ years, and females 13-49 years. Exposure estimates were presented in units of mg/kg body weight/day.

The 2009 exposure estimates were developed for use by the OW in their relative source contribution (RSC) analysis for fluoride. A draft RSC analysis document was provided by the OW to the OPP, at which time it became apparent that the OW is focusing on different population subgroups and that exposure estimates are being presented in units of mg/day. The

OPP is providing new exposure estimates in order to match the population groups and exposure units of primary interest to the OW.

The OPP has used the same input files that were used to estimate the exposures reported in the May 2009 memorandum (included in Attachments 1-3 for reference). Rather than use the chronic dietary exposure module in the Dietary Exposure Evaluation Model (DEEM), the OPP has used the acute dietary exposure module, which provides a mechanism to (1) specify customized population groupings and (2) leave body weight out of the exposure estimates to give exposure in units of mg/day. Average exposure estimates from the acute module are identical to the exposure estimates given by the chronic module. Average exposure estimates for the customized population groups are summarized in Table 1. Complete outputs of the three analyses, including error estimates, are included as Attachments 4, 5, and 6.

Table 1. Summary of Cryolite and Sulfuryl Fluoride (SF) Contributions to Dietary Fluoride Exposure.

| Age Range, years | Estimated Average Exposure, mg/day | | | | |
|---|---|---|---|---|---|
| | Cryolite | SF Structural Fumigations | SF Food Fumigations | Total SF | Total |
| 0.5 - <1 | 0.0308 | 0.0087 | 0.0213 | 0.0300 | 0.0608 |
| 1 - <4 | 0.0404 | 0.0121 | 0.0329 | 0.0450 | 0.0854 |
| 4 - <7 | 0.0313 | 0.0153 | 0.0466 | 0.0619 | 0.0932 |
| 7 - <11 | 0.0285 | 0.0170 | 0.0544 | 0.0714 | 0.0999 |
| 11 - <14 | 0.0248 | 0.0182 | 0.0675 | 0.0857 | 0.1105 |
| 14+ | 0.0327 | 0.0187 | 0.0576 | 0.0763 | 0.1090 |

These values do not have the uncertainty associated with translating the exposure values to different age group definitions or with converting the units from mg/kg body weight/day to mg/day, which had to rely on assumptions regarding body weights. The OPP notes that the residue inputs used in the analysis are point estimates; therefore, the error estimates reported in Attachments 4-6 reflect the variability due to consumption only and not variability in fluoride residues.

For most age groups, the exposure estimates in Table 1 are very similar to those used by the OW (Draft RSC Analysis Document, Table 6-5). The exception to this appears to be the 0.5-1 year old group for which the values in Table 1 are 2-3 times greater than those reported by the OW in Table 6-5. Examination of the "percent of person-days that are user-days" (Table 2) indicates that a higher proportion of the survey respondents in the 0.5-1 year group actually ate the foods associated with the various analyses compared to the previously reported group which includes children < 0.5 years old (Table 2).

Table 2. Percent of Person-Days that are User-Days in the Fluoride Dietary Exposure Analyses.

| Age Range, years | Cryolite | SF Structural Fumigations | SF Food Fumigations |
|---|---|---|---|
| < 1 year | 51.0 | 89.1 | 71.7 |
| 0.5 - <1 | 93.1 | 99.4 | 98.1 |
| 1 - <4 | 98.6 | 99.9 | 99.8 |
| 4 - <7 | 99.3 | 100 | 100 |
| 7 - <11 | 99.2 | 100 | 100 |
| 11 - <14 | 98.9 | 100 | 100 |
| 14+ | 98.2 | 99.8 | 99.7 |

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Attachments:
1. Inputs for cryolite fluoride exposure estimates
2. Inputs for sulfuryl fluoride food fumigation exposure estimates
3. Inputs for sulfuryl fluoride structural fumigation exposure estimates
4. Results of cryolite fluoride exposure analysis
5. Results of sulfuryl fluoride food fumigation exposure analysis
6. Results of sulfuryl fluoride structural fumigation exposure analysis

## Attachment 1. Inputs for cryolite fluoride exposure estimates

```
Filename: C:\Documents and Settings\MDOHERTY\My Documents\Chemistry Reviews\DEEM Runs\Sulfuryl
Fluoride\Cryolite-AR-CT new raisin factor.R98
Chemical: Cryolite
RfD(Chronic): .114 mg/kg bw/day  NOEL(Chronic): 0 mg/kg bw/day
RfD(Acute): 0 mg/kg bw/day  NOEL(Acute):  0 mg/kg bw/day
Date created/last modified: 06-24-2004/10:05:08/8          Program ver. 2.03
```

| EPA Code | Crop Grp | Commodity Name | Def Res (ppm) | Adj.Factors #1 | Adj.Factors #2 | Comment |
|---|---|---|---|---|---|---|
| 12000120 | 12 | Apricot | 4.500000 | 1.000 | 0.010 | |
| 12000121 | 12 | Apricot-babyfood | 4.500000 | 1.000 | 0.010 | |
| 12000130 | 12 | Apricot, dried | 4.500000 | 6.000 | 0.010 | |
| 12000140 | 12 | Apricot, juice | 4.500000 | 1.000 | 0.010 | |
| 12000141 | 12 | Apricot, juice-babyfood | 4.500000 | 1.000 | 0.010 | |
| 13010550 | 13A | Blackberry | 0.250000 | 1.000 | 1.000 | |
| 13010560 | 13A | Blackberry, juice | 0.250000 | 1.000 | 1.000 | |
| 13010561 | 13A | Blackberry, juice-babyfood | 0.250000 | 1.000 | 1.000 | |
| 13020570 | 13B | Blueberry | 0.110000 | 1.000 | 1.000 | |
| 13020571 | 13B | Blueberry-babyfood | 0.110000 | 1.000 | 1.000 | |
| 13010580 | 13A | Boysenberry | 0.250000 | 1.000 | 1.000 | |
| 05010610 | 5A | Broccoli | 5.000000 | 1.000 | 0.020 | |
| 05010611 | 5A | Broccoli-babyfood | 5.000000 | 1.000 | 0.020 | |
| 05010640 | 5A | Brussels sprouts | 4.000000 | 1.000 | 0.020 | |
| 05010690 | 5A | Cabbage | 1.500000 | 1.000 | 0.010 | |
| 05020700 | 5B | Cabbage, Chinese, bok choy | 4.000000 | 1.000 | 0.010 | |
| 09010750 | 9A | Cantaloupe | 2.160000 | 1.000 | 0.010 | |
| 09010800 | 9A | Casaba | 2.160000 | 1.000 | 0.010 | |
| 05010830 | 5A | Cauliflower | 3.000000 | 1.000 | 0.020 | |
| 10001060 | 10 | Citrus citron | 8.000000 | 1.000 | 0.040 | |
| 05021170 | 5B | Collards | 4.000000 | 1.000 | 0.020 | |
| 95001300 | O | Cranberry | 0.500000 | 1.000 | 1.000 | |
| 95001301 | O | Cranberry-babyfood | 0.500000 | 1.000 | 1.000 | |
| 95001310 | O | Cranberry, dried | 0.500000 | 1.000 | 1.000 | |
| 95001320 | O | Cranberry, juice | 0.500000 | 1.100 | 1.000 | |
| 95001321 | O | Cranberry, juice-babyfood | 0.500000 | 1.100 | 1.000 | |
| 09021350 | 9B | Cucumber | 2.500000 | 1.000 | 0.010 | |
| 13021360 | 13B | Currant | 0.110000 | 1.000 | 1.000 | |
| 13021370 | 13B | Currant, dried | 0.110000 | 1.000 | 1.000 | |
| 13011420 | 13A | Dewberry | 0.250000 | 1.000 | 1.000 | |
| 08001480 | 8 | Eggplant | 1.500000 | 1.000 | 0.010 | |
| 13021490 | 13B | Elderberry | 0.110000 | 1.000 | 1.000 | |
| 13021740 | 13B | Gooseberry | 0.110000 | 1.000 | 1.000 | |
| 95001750 | O | Grape | 3.500000 | 1.000 | 0.330 | |
| 95001760 | O | Grape, juice | 3.500000 | 0.830 | 0.330 | |
| 95001761 | O | Grape, juice-babyfood | 3.500000 | 0.830 | 0.330 | |
| 95001770 | O | Grape, leaves | 3.500000 | 1.000 | 0.330 | |
| 95001780 | O | Grape, raisin | 3.500000 | 1.350 | 0.330 | |
| 95001790 | O | Grape, wine and sherry | 3.500000 | 0.830 | 0.330 | |
| 10001800 | 10 | Grapefruit | 9.000000 | 1.000 | 0.040 | |
| 10001810 | 10 | Grapefruit, juice | 9.000000 | 0.026 | 0.040 | |
| 09011870 | 9A | Honeydew melon | 2.160000 | 1.000 | 0.010 | |
| 13021910 | 13B | Huckleberry | 0.110000 | 1.000 | 1.000 | |
| 05021940 | 5B | Kale | 4.000000 | 1.000 | 0.020 | |
| 95001950 | O | Kiwifruit | 4.500000 | 1.000 | 0.140 | |
| 05011960 | 5A | Kohlrabi | 5.000000 | 1.000 | 0.020 | |
| 10001970 | 10 | Kumquat | 8.000000 | 1.000 | 0.040 | |
| 10001990 | 10 | Lemon | 13.500000 | 1.000 | 0.020 | |
| 10002000 | 10 | Lemon, juice | 13.500000 | 0.024 | 0.020 | |
| 10002001 | 10 | Lemon, juice-babyfood | 13.500000 | 0.024 | 0.020 | |
| 10002010 | 10 | Lemon, peel | 13.500000 | 0.280 | 0.020 | |
| 04012040 | 4A | Lettuce, head | 2.500000 | 1.000 | 0.010 | |
| 04012050 | 4A | Lettuce, leaf | 15.000000 | 1.000 | 0.010 | |
| 10002060 | 10 | Lime | 13.500000 | 1.000 | 0.040 | |
| 10002070 | 10 | Lime, juice | 13.500000 | 0.024 | 0.040 | |
| 10002071 | 10 | Lime, juice-babyfood | 13.500000 | 0.024 | 0.040 | |
| 13012080 | 13A | Loganberry | 0.250000 | 1.000 | 1.000 | |
| 12002300 | 12 | Nectarine | 4.500000 | 1.000 | 0.010 | |
| 10002400 | 10 | Orange | 8.000000 | 1.000 | 0.020 | |
| 10002410 | 10 | Orange, juice | 8.000000 | 0.022 | 0.020 | |

| | | | | | |
|---|---|---|---|---|---|
| 10002411 | 10 | Orange, juice-babyfood | 8.000000 | 0.022 | 0.020 |
| 10002420 | 10 | Orange, peel | 8.000000 | 0.280 | 0.020 |
| 12002600 | 12 | Peach | 4.500000 | 1.000 | 0.010 |
| 12002601 | 12 | Peach-babyfood | 4.500000 | 1.000 | 0.010 |
| 12002610 | 12 | Peach, dried | 4.500000 | 7.000 | 0.010 |
| 12002611 | 12 | Peach, dried-babyfood | 4.500000 | 7.000 | 0.010 |
| 12002620 | 12 | Peach, juice | 4.500000 | 1.000 | 0.010 |
| 12002621 | 12 | Peach, juice-babyfood | 4.500000 | 1.000 | 0.010 |
| 08002700 | 8 | Pepper, bell | 3.500000 | 1.000 | 0.010 |
| 08002701 | 8 | Pepper, bell-babyfood | 3.500000 | 1.000 | 0.010 |
| 08002710 | 8 | Pepper, bell, dried | 3.500000 | 1.000 | 0.010 |
| 08002711 | 8 | Pepper, bell, dried-babyfood | 3.500000 | 1.000 | 0.010 |
| 08002720 | 8 | Pepper, nonbell | 3.500000 | 1.000 | 0.010 |
| 08002721 | 8 | Pepper, nonbell-babyfood | 3.500000 | 1.000 | 0.010 |
| 08002730 | 8 | Pepper, nonbell, dried | 3.500000 | 1.000 | 0.010 |
| 95002750 | O | Peppermint | 19.500000 | 1.000 | 1.000 |
| 95002760 | O | Peppermint, oil | 19.500000 | 0.026 | 1.000 |
| 12002850 | 12 | Plum | 0.500000 | 1.000 | 0.010 |
| 12002851 | 12 | Plum-babyfood | 0.500000 | 1.000 | 0.010 |
| 12002860 | 12 | Plum, prune, fresh | 0.500000 | 1.000 | 0.010 |
| 12002861 | 12 | Plum, prune, fresh-babyfood | 2.000000 | 1.000 | 0.010 |
| 12002870 | 12 | Plum, prune, dried | 2.000000 | 5.000 | 0.010 |
| 12002871 | 12 | Plum, prune, dried-babyfood | 2.000000 | 5.000 | 0.010 |
| 12002880 | 12 | Plum, prune, juice | 2.000000 | 1.400 | 0.010 |
| 12002881 | 12 | Plum, prune, juice-babyfood | 2.000000 | 1.400 | 0.010 |
| 01032960 | 1C | Potato, chips | 0.650000 | 1.000 | 0.030 |
| 01032970 | 1C | Potato, dry (granules/ flakes) | 0.650000 | 6.500 | 0.030 |
| 01032971 | 1C | Potato, dry (granules/ flakes)-b | 0.650000 | 6.500 | 0.030 |
| 01032980 | 1C | Potato, flour | 0.650000 | 6.500 | 0.030 |
| 01032981 | 1C | Potato, flour-babyfood | 0.650000 | 6.500 | 0.030 |
| 01032990 | 1C | Potato, tuber, w/peel | 0.650000 | 1.000 | 0.030 |
| 01032991 | 1C | Potato, tuber, w/peel-babyfood | 0.650000 | 1.000 | 0.030 |
| 01033000 | 1C | Potato, tuber, w/o peel | 0.650000 | 1.000 | 0.030 |
| 01033001 | 1C | Potato, tuber, w/o peel-babyfood | 0.650000 | 1.000 | 0.030 |
| 10003070 | 10 | Pummelo | 9.000000 | 1.000 | 0.040 |
| 09023080 | 9B | Pumpkin | 2.500000 | 1.000 | 0.010 |
| 09023090 | 9B | Pumpkin, seed | 2.500000 | 1.000 | 0.010 |
| 13013200 | 13A | Raspberry | 0.250000 | 1.000 | 1.000 |
| 13013201 | 13A | Raspberry-babyfood | 0.250000 | 1.000 | 1.000 |
| 13013210 | 13A | Raspberry, juice | 0.250000 | 1.000 | 1.000 |
| 13013211 | 13A | Raspberry, juice-babyfood | 0.250000 | 1.000 | 1.000 |
| 95003520 | O | Spearmint | 19.500000 | 1.000 | 1.000 |
| 95003530 | O | Spearmint, oil | 19.500000 | 0.026 | 1.000 |
| 09023560 | 9B | Squash, summer | 2.500000 | 1.000 | 0.010 |
| 09023561 | 9B | Squash, summer-babyfood | 2.500000 | 1.000 | 0.010 |
| 09023570 | 9B | Squash, winter | 2.500000 | 1.000 | 0.010 |
| 09023571 | 9B | Squash, winter-babyfood | 2.500000 | 1.000 | 0.010 |
| 95003590 | O | Strawberry | 1.000000 | 1.000 | 0.020 |
| 95003591 | O | Strawberry-babyfood | 1.000000 | 1.000 | 0.020 |
| 95003600 | O | Strawberry, juice | 1.000000 | 1.000 | 0.020 |
| 95003601 | O | Strawberry, juice-babyfood | 1.000000 | 1.000 | 0.020 |
| 10003690 | 10 | Tangerine | 8.000000 | 1.000 | 0.040 |
| 10003700 | 10 | Tangerine, juice | 8.000000 | 0.028 | 0.040 |
| 08003750 | 8 | Tomato | 1.500000 | 1.000 | 0.010 |
| 08003751 | 8 | Tomato-babyfood | 1.500000 | 1.000 | 0.010 |
| 08003760 | 8 | Tomato, paste | 1.500000 | 1.500 | 0.010 |
| 08003761 | 8 | Tomato, paste-babyfood | 1.500000 | 1.500 | 0.010 |
| 08003770 | 8 | Tomato, puree | 1.500000 | 1.000 | 0.010 |
| 08003771 | 8 | Tomato, puree-babyfood | 1.500000 | 1.000 | 0.010 |
| 08003780 | 8 | Tomato, dried | 1.500000 | 14.300 | 0.010 |
| 08003781 | 8 | Tomato, dried-babyfood | 1.500000 | 14.300 | 0.010 |
| 08003790 | 8 | Tomato, juice | 1.500000 | 1.500 | 0.010 |
| 09013990 | 9A | Watermelon | 2.160000 | 1.000 | 0.010 |
| 09014000 | 9A | Watermelon, juice | 2.160000 | 1.000 | 0.010 |

Attachment 2. Inputs for sulfuryl fluoride food fumigation exposure estimates

```
U.S. Environmental Protection Agency                              Ver. 2.00
DEEM-FCID Chronic analysis for FLUORIDE                        1994-98 data
Residue file: C:\Documents and Settings\mdoherty\My Documents\Chemistry Reviews\!DEEM
Runs\Sulfuryl Fluoride\F Food Fumigation - 2009  RevisedCT - 4-28 Strict Label.R98
                                                                Adjust. #2 used
Analysis Date 05-06-2009          Residue file dated: 05-06-2009/13:54:06/8
Reference dose (RfD) = 0.114 mg/kg bw/day
```

| Food Crop EPA Code | Grp | Food Name | Residue (ppm) | Adj.Factors #1 | Adj.Factors #2 | Comment |
|---|---|---|---|---|---|---|
| 14000030 | 14 | Almond | 9.700000 | 1.000 | 0.100 | |
| 14000031 | 14 | Almond-babyfood | 9.700000 | 1.000 | 0.100 | |
| 11000090 | 11 | Apple, dried | 1.000000 | 1.000 | 0.690 | |
| 11000091 | 11 | Apple, dried-babyfood | 1.000000 | 1.000 | 0.690 | |
| 12000130 | 12 | Apricot, dried | 1.000000 | 1.000 | 0.690 | |
| 95000240 | O | Banana, dried | 1.000000 | 1.000 | 0.690 | |
| 95000241 | O | Banana, dried-babyfood | 1.000000 | 1.000 | 0.690 | |
| 15000250 | 15 | Barley, pearled barley | 3.700000 | 1.000 | 0.001 | |
| 15000251 | 15 | Barley, pearled barley-babyfood | 3.700000 | 1.000 | 0.001 | |
| 15000260 | 15 | Barley, flour | 3.700000 | 0.730 | 0.001 | |
| 15000261 | 15 | Barley, flour-babyfood | 3.700000 | 0.730 | 0.001 | |
| 15000270 | 15 | Barley, bran | 3.700000 | 2.560 | 0.001 | |
| 19010290 | 19A | Basil, dried leaves | 67.100000 | 1.000 | 0.001 | |
| 19010291 | 19A | Basil, dried leaves-babyfood | 67.100000 | 1.000 | 0.001 | |
| 06030300 | 6C | Bean, black, seed | 4.500000 | 1.000 | 1.000 | |
| 06030320 | 6C | Bean, broad, seed | 4.500000 | 1.000 | 1.000 | |
| 06030340 | 6C | Bean, cowpea, seed | 4.500000 | 1.000 | 1.000 | |
| 06030350 | 6C | Bean, great northern, seed | 4.500000 | 1.000 | 1.000 | |
| 06030360 | 6C | Bean, kidney, seed | 4.500000 | 1.000 | 1.000 | |
| 06030380 | 6C | Bean, lima, seed | 4.500000 | 1.000 | 1.000 | |
| 06030390 | 6C | Bean, mung, seed | 4.500000 | 1.000 | 1.000 | |
| 06030400 | 6C | Bean, navy, seed | 4.500000 | 1.000 | 1.000 | |
| 06030410 | 6C | Bean, pink, seed | 4.500000 | 1.000 | 1.000 | |
| 06030420 | 6C | Bean, pinto, seed | 4.500000 | 1.000 | 1.000 | |
| 14000590 | 14 | Brazil nut | 5.300000 | 1.000 | 0.100 | |
| 14000680 | 14 | Butternut | 5.300000 | 1.000 | 0.100 | |
| 14000810 | 14 | Cashew | 5.300000 | 1.000 | 0.100 | |
| 14000920 | 14 | Chestnut | 5.300000 | 1.000 | 0.100 | |
| 06030980 | 6C | Chickpea, seed | 4.500000 | 1.000 | 1.000 | |
| 06030981 | 6C | Chickpea, seed-babyfood | 4.500000 | 1.000 | 1.000 | |
| 06030990 | 6C | Chickpea, flour | 4.500000 | 1.000 | 1.000 | |
| 19011030 | 19A | Chive | 63.500000 | 1.000 | 0.001 | |
| 19021050 | 19B | Cinnamon | 73.500000 | 1.000 | 0.001 | |
| 19021051 | 19B | Cinnamon-babyfood | 73.500000 | 1.000 | 0.001 | |
| 95001090 | O | Cocoa bean, chocolate | 8.400000 | 1.000 | 1.000 | |
| 95001100 | O | Cocoa bean, powder | 8.400000 | 1.000 | 1.000 | |
| 95001110 | O | Coconut, meat | 49.100000 | 1.000 | 0.001 | |
| 95001111 | O | Coconut- meat-babyfood | 49.100000 | 1.000 | 0.001 | |
| 95001120 | O | Coconut, dried | 49.100000 | 1.000 | 0.001 | |
| 95001130 | O | Coconut, milk | 49.100000 | 1.000 | 0.001 | |
| 95001150 | O | Coffee, roasted bean | 7.100000 | 1.000 | 0.001 | |
| 95001160 | O | Coffee, instant | 13.900000 | 1.000 | 0.001 | |
| 19011180 | 19A | Coriander, leaves | 63.500000 | 1.000 | 0.001 | |
| 19011181 | 19A | Coriander, leaves-babyfood | 63.500000 | 1.000 | 0.001 | |
| 19021190 | 19B | Coriander, seed | 7.100000 | 1.000 | 0.001 | |
| 19021191 | 19B | Coriander, seed-babyfood | 7.100000 | 1.000 | 0.001 | |
| 15001200 | 15 | Corn, field, flour | 16.900000 | 1.000 | 0.001 | |
| 15001201 | 15 | Corn, field, flour-babyfood | 16.900000 | 1.000 | 0.001 | |
| 15001210 | 15 | Corn, field, meal | 2.800000 | 1.000 | 0.001 | |
| 15001211 | 15 | Corn, field, meal-babyfood | 2.800000 | 1.000 | 0.001 | |
| 15001220 | 15 | Corn, field, bran | 2.800000 | 1.000 | 0.001 | |
| 15001230 | 15 | Corn, field, starch | 0.600000 | 1.000 | 0.001 | |
| 15001231 | 15 | Corn, field, starch-babyfood | 0.600000 | 1.000 | 0.001 | |
| 15001240 | 15 | Corn, field, syrup | 0.600000 | 1.000 | 0.001 | |
| 15001241 | 15 | Corn, field, syrup-babyfood | 0.600000 | 1.000 | 0.001 | |
| 15001260 | 15 | Corn, pop | 1.700000 | 1.000 | 0.001 | |
| 95001310 | O | Cranberry, dried | 1.000000 | 1.000 | 0.100 | |

| | | | | | |
|---|---|---|---|---|---|
| 13021370 | 13B | Currant, dried | 1.000000 | 1.000 | 0.100 |
| 95001410 | O | Date | | | |
| | | 130-Uncooked; Dried; Cook Meth N/S | 0.900000 | 1.000 | 0.420 |
| | | 210-Cooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.000 |
| | | 211-Cooked; Fresh or N/S; Baked | 0.000000 | 1.000 | 0.000 |
| | | 212-Cooked; Fresh or N/S; Boiled | 0.000000 | 1.000 | 0.000 |
| | | 230-Cooked; Dried; Cook Meth N/S | 0.900000 | 1.000 | 0.420 |
| 19021430 | 19B | Dill, seed | 7.100000 | 1.000 | 0.001 |
| 19011440 | 19A | Dillweed | 63.500000 | 1.000 | 0.001 |
| 95001540 | O | Fig, dried | 1.000000 | 1.000 | 0.690 |
| 14001550 | 14 | Filbert | 2.500000 | 1.000 | 0.100 |
| 03001650 | 3 | Garlic, dried | 10.900000 | 1.000 | 0.001 |
| 01031660 | 1CD | Ginger | 10.900000 | 1.000 | 0.001 |
| 01031661 | 1CD | Ginger-babyfood | 10.900000 | 1.000 | 0.001 |
| 01031670 | 1CD | Ginger, dried | 10.900000 | 1.000 | 0.001 |
| 95001780 | O | Grape, raisin | 1.000000 | 1.000 | 0.690 |
| 06031820 | 6C | Guar, seed | 4.500000 | 1.000 | 1.000 |
| 06031821 | 6C | Guar, seed-babyfood | 4.500000 | 1.000 | 1.000 |
| 19011840 | 19A | Herbs, other | 63.500000 | 1.000 | 0.001 |
| 19011841 | 19A | Herbs, other-babyfood | 63.500000 | 1.000 | 0.001 |
| 14001850 | 14 | Hickory nut | 5.300000 | 1.000 | 0.100 |
| 19012020 | 19A | Lemongrass | 63.500000 | 1.000 | 0.001 |
| 06032030 | 6C | Lentil, seed | 4.500000 | 1.000 | 1.000 |
| 95002120 | O | Lychee, dried | 1.000000 | 1.000 | 0.690 |
| 14002130 | 14 | Macadamia nut | 5.300000 | 1.000 | 0.001 |
| 95002160 | O | Mango, dried | 1.000000 | 1.000 | 0.690 |
| 19012200 | 19A | Marjoram | 67.100000 | 1.000 | 0.001 |
| 19012201 | 19A | Marjoram-babyfood | 67.100000 | 1.000 | 0.001 |
| 15002260 | 15 | Millet, grain | 2.900000 | 1.000 | 0.001 |
| 15002310 | 15 | Oat, bran | 18.500000 | 2.560 | 0.001 |
| 15002320 | 15 | Oat, flour | 18.500000 | 0.730 | 0.001 |
| 15002321 | 15 | Oat, flour-babyfood | 18.500000 | 0.730 | 0.001 |
| 15002330 | 15 | Oat, groats/rolled oats | 18.500000 | 1.000 | 0.001 |
| 15002331 | 15 | Oat, groats/rolled oats-babyfood | 18.500000 | 1.000 | 0.001 |
| 95002460 | O | Papaya, dried | 1.000000 | 1.000 | 0.690 |
| 19012490 | 19A | Parsley, dried leaves | 63.500000 | 1.000 | 0.001 |
| 19012491 | 19A | Parsley, dried leaves-babyfood | 63.500000 | 1.000 | 0.001 |
| 06032560 | 6C | Pea, dry | 4.500000 | 1.000 | 1.000 |
| 06032561 | 6C | Pea, dry-babyfood | 4.500000 | 1.000 | 1.000 |
| 06032580 | 6C | Pea, pigeon, seed | 4.500000 | 1.000 | 1.000 |
| 12002610 | 12 | Peach, dried | 1.000000 | 1.000 | 0.690 |
| 12002611 | 12 | Peach, dried-babyfood | 1.000000 | 1.000 | 0.690 |
| 95002630 | O | Peanut | 16.400000 | 1.000 | 0.006 |
| 95002640 | O | Peanut, butter | 16.400000 | 1.000 | 0.006 |
| 11002670 | 11 | Pear, dried | 1.000000 | 1.000 | 0.690 |
| 14002690 | 14 | Pecan | 5.300000 | 1.000 | 0.100 |
| 19022740 | 19B | Pepper, black and white | 7.100000 | 1.000 | 0.001 |
| 19022741 | 19B | Pepper, black and white-babyfood | 7.100000 | 1.000 | 0.001 |
| 95002780 | O | Pine nut | 8.800000 | 1.000 | 0.100 |
| 95002800 | O | Pineapple, dried | 1.000000 | 1.000 | 0.690 |
| 14002820 | 14 | Pistachio | 3.200000 | 1.000 | 0.270 |
| 95002840 | O | Plantain, dried | 1.000000 | 1.000 | 0.690 |
| 12002870 | 12 | Plum, prune, dried | 0.700000 | 1.000 | 0.690 |
| 12002871 | 12 | Plum, prune, dried-babyfood | 0.700000 | 1.000 | 0.690 |
| 15003230 | 15 | Rice, white | 4.500000 | 1.000 | 0.030 |
| 15003231 | 15 | Rice, white-babyfood | 4.500000 | 1.000 | 0.030 |
| 15003240 | 15 | Rice, brown | 12.500000 | 1.000 | 0.030 |
| 15003241 | 15 | Rice, brown-babyfood | 12.500000 | 1.000 | 0.030 |
| 15003250 | 15 | Rice, flour | 32.500000 | 1.000 | 0.030 |
| 15003251 | 15 | Rice, flour-babyfood | 32.500000 | 1.000 | 0.030 |
| 15003260 | 15 | Rice, bran | 37.500000 | 1.000 | 0.030 |
| 15003261 | 15 | Rice, bran-babyfood | 37.500000 | 1.000 | 0.030 |
| 19013340 | 19A | Savory | 63.500000 | 1.000 | 0.001 |
| 15003440 | 15 | Sorghum, grain | 20.400000 | 1.000 | 0.001 |
| 19023540 | 19B | Spices, other | 7.100000 | 1.000 | 0.001 |
| 19023541 | 19B | Spices, other-babyfood | 7.100000 | 1.000 | 0.001 |
| 15003810 | 15 | Triticale, flour | 2.900000 | 0.380 | 0.001 |
| 15003811 | 15 | Triticale, flour-babyfood | 2.900000 | 0.380 | 0.001 |
| 01033870 | 1CD | Turmeric | 7.100000 | 1.000 | 0.001 |

| | | | | | |
|---|---|---|---|---|---|
| 14003910 | 14 | Walnut | 2.400000 | 1.000 | 0.990 |
| 15004010 | 15 | Wheat, grain | 2.900000 | 1.000 | 0.004 |
| 15004011 | 15 | Wheat, grain-babyfood | 2.900000 | 1.000 | 0.004 |
| 15004020 | 15 | Wheat, flour | 31.400000 | 1.000 | 0.001 |
| 15004021 | 15 | Wheat, flour-babyfood | 31.400000 | 1.000 | 0.001 |
| 15004030 | 15 | Wheat, germ | 13.900000 | 1.000 | 0.001 |
| 15004040 | 15 | Wheat, bran | 74.200000 | 1.000 | 0.001 |
| 15004050 | 15 | Wild rice | 12.500000 | 1.000 | 0.030 |

Attachment 3. Inputs for sulfuryl fluoride structural fumigation exposure estimates

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U.S. Environmental Protection Agency                                    Ver. 2.00
DEEM-FCID Chronic analysis for FLUORIDE                              1994-98 data
Residue file: C:\Documents and Settings\mdoherty\My Documents\Chemistry Reviews\!DEEM
Runs\Sulfuryl Fluoride\F Space Fumigation - 2009 - 5-1.R98
                                                                  Adjust. #2 used
Analysis Date 05-06-2009          Residue file dated: 05-06-2009/13:52:45/8
Reference dose (RfD) = 0.114 mg/kg bw/day
```

| Food Crop EPA Code | Grp | Food Name | Residue (ppm) | Adj.Factors #1 | Adj.Factors #2 | Comment |
|---|---|---|---|---|---|---|
| 18000020 | 18 | Alfalfa, seed | 20.400000 | 1.000 | 0.004 | |
| 14000030 | 14 | Almond | 9.200000 | 1.000 | 0.004 | |
| 14000031 | 14 | Almond-babyfood | 9.200000 | 1.000 | 0.004 | |
| 14000040 | 14 | Almond, oil | 1.500000 | 1.000 | 0.004 | |
| 14000041 | 14 | Almond, oil-babyfood | 1.500000 | 1.000 | 0.004 | |
| 95000060 | O | Amaranth, grain | 20.400000 | 1.000 | 0.004 | |
| 11000090 | 11 | Apple, dried | 1.000000 | 1.000 | 0.004 | |
| 11000091 | 11 | Apple, dried-babyfood | 1.000000 | 1.000 | 0.004 | |
| 12000130 | 12 | Apricot, dried | 1.000000 | 1.000 | 0.004 | |
| 01030150 | 1CD | Arrowroot, flour | 31.400000 | 1.000 | 0.004 | |
| 01030151 | 1CD | Arrowroot, flour-babyfood | 31.400000 | 1.000 | 0.004 | |
| 95000240 | O | Banana, dried | 1.000000 | 1.000 | 0.004 | |
| 95000241 | O | Banana, dried-babyfood | 1.000000 | 1.000 | 0.004 | |
| 15000250 | 15 | Barley, pearled barley | 3.650000 | 1.000 | 0.008 | |
| 15000251 | 15 | Barley, pearled barley-babyfood | 3.650000 | 1.000 | 0.008 | |
| 15000260 | 15 | Barley, flour | 0.156000 | 1.000 | 1.000 | |
| 15000261 | 15 | Barley, flour-babyfood | 0.156000 | 1.000 | 1.000 | |
| 15000270 | 15 | Barley, bran | 3.650000 | 1.000 | 0.008 | |
| 19010280 | 19A | Basil, fresh leaves | 67.100000 | 1.000 | 0.004 | |
| 19010281 | 19A | Basil, fresh leaves-babyfood | 67.100000 | 1.000 | 0.004 | |
| 19010290 | 19A | Basil, dried leaves | 67.100000 | 1.000 | 0.004 | |
| 19010291 | 19A | Basil, dried leaves-babyfood | 67.100000 | 1.000 | 0.004 | |
| 06030300 | 6C | Bean, black, seed | 4.500000 | 1.000 | 0.004 | |
| 06030320 | 6C | Bean, broad, seed | 4.500000 | 1.000 | 0.004 | |
| 06030340 | 6C | Bean, cowpea, seed | 4.500000 | 1.000 | 0.004 | |
| 06030350 | 6C | Bean, great northern, seed | 4.500000 | 1.000 | 0.004 | |
| 06030360 | 6C | Bean, kidney, seed | 4.500000 | 1.000 | 0.004 | |
| 06030380 | 6C | Bean, lima, seed | 4.500000 | 1.000 | 0.004 | |
| 06030390 | 6C | Bean, mung, seed | 4.500000 | 1.000 | 0.004 | |
| 06030400 | 6C | Bean, navy, seed | 4.500000 | 1.000 | 0.004 | |
| 06030410 | 6C | Bean, pink, seed | 4.500000 | 1.000 | 0.004 | |
| 06030420 | 6C | Bean, pinto, seed | 4.500000 | 1.000 | 0.004 | |
| 21000450 | M | Beef, meat, dried | 58.400000 | 1.000 | 0.004 | |
| 01010530 | 1A | Beet, sugar, molasses | 1.200000 | 1.000 | 0.004 | |
| 01010531 | 1A | Beet, sugar, molasses-babyfood | 1.200000 | 1.000 | 0.004 | |
| 14000590 | 14 | Brazil nut | 3.900000 | 1.000 | 0.004 | |
| 15000650 | 15 | Buckwheat | 2.900000 | 1.000 | 0.008 | |
| 15000660 | 15 | Buckwheat, flour | 0.134000 | 1.000 | 1.000 | |
| 14000680 | 14 | Butternut | 3.900000 | 1.000 | 0.004 | |
| 14000810 | 14 | Cashew | 3.900000 | 1.000 | 0.004 | |
| 14000920 | 14 | Chestnut | 3.900000 | 1.000 | 0.004 | |
| 06030980 | 6C | Chickpea, seed | 4.500000 | 1.000 | 0.004 | |
| 06030981 | 6C | Chickpea, seed-babyfood | 4.500000 | 1.000 | 0.004 | |
| 06030990 | 6C | Chickpea, flour | 4.500000 | 1.000 | 0.004 | |
| 01011000 | 1AB | Chicory, roots | 13.900000 | 1.000 | 0.004 | |
| 19011030 | 19A | Chive | 63.500000 | 1.000 | 0.004 | |
| 04011040 | 4A | Chrysanthemum, garland | 63.500000 | 1.000 | 0.004 | |
| 19021050 | 19B | Cinnamon | 73.500000 | 1.000 | 0.004 | |
| 19021051 | 19B | Cinnamon-babyfood | 73.500000 | 1.000 | 0.004 | |
| 95001090 | O | Cocoa bean, chocolate | 8.400000 | 1.000 | 0.004 | |
| 95001100 | O | Cocoa bean, powder | 8.400000 | 1.000 | 0.004 | |
| 95001110 | O | Coconut, meat | 49.100000 | 1.000 | 0.004 | |
| 95001111 | O | Coconut- meat-babyfood | 49.100000 | 1.000 | 0.004 | |
| 95001120 | O | Coconut, dried | 49.100000 | 1.000 | 0.004 | |
| 95001130 | O | Coconut, milk | 49.100000 | 1.000 | 0.004 | |
| 95001140 | O | Coconut, oil | 1.500000 | 1.000 | 0.004 | |
| 95001141 | O | Coconut, oil-babyfood | 1.500000 | 1.000 | 0.004 | |

| | | | | | |
|---|---|---|---|---|---|
| 95001150 | O | Coffee, roasted bean | 7.100000 | 1.000 | 0.004 |
| 95001160 | O | Coffee, instant | 13.900000 | 1.000 | 0.004 |
| 19011180 | 19A | Coriander, leaves | 63.500000 | 1.000 | 0.004 |
| 19011181 | 19A | Coriander, leaves-babyfood | 63.500000 | 1.000 | 0.004 |
| 19021190 | 19B | Coriander, seed | 7.100000 | 1.000 | 0.004 |
| 19021191 | 19B | Coriander, seed-babyfood | 7.100000 | 1.000 | 0.004 |
| 15001200 | 15 | Corn, field, flour | 0.081000 | 1.000 | 1.000 |
| 15001201 | 15 | Corn, field, flour-babyfood | 0.081000 | 1.000 | 1.000 |
| 15001210 | 15 | Corn, field, meal | 14.000000 | 1.000 | 0.008 |
| 15001211 | 15 | Corn, field, meal-babyfood | 14.000000 | 1.000 | 0.008 |
| 15001220 | 15 | Corn, field, bran | 14.000000 | 1.000 | 0.008 |
| 15001230 | 15 | Corn, field, starch | 6.600000 | 1.000 | 0.008 |
| 15001231 | 15 | Corn, field, starch-babyfood | 6.600000 | 1.000 | 0.008 |
| 15001240 | 15 | Corn, field, syrup | 0.600000 | 1.000 | 0.008 |
| 15001241 | 15 | Corn, field, syrup-babyfood | 0.600000 | 1.000 | 0.008 |
| 15001250 | 15 | Corn, field, oil | 0.400000 | 1.000 | 0.008 |
| 15001251 | 15 | Corn, field, oil-babyfood | 0.400000 | 1.000 | 0.008 |
| 15001260 | 15 | Corn, pop | 1.700000 | 1.000 | 0.008 |
| 95001280 | O | Cottonseed, oil | 1.500000 | 1.000 | 0.004 |
| 95001281 | O | Cottonseed, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 95001310 | O | Cranberry, dried | 1.000000 | 1.000 | 0.004 |
| 13021370 | 13B | Currant, dried | 1.000000 | 1.000 | 0.004 |
| 95001410 | O | Date | 0.900000 | 1.000 | 0.004 |
| 19021430 | 19B | Dill, seed | 7.100000 | 1.000 | 0.004 |
| 19011440 | 19A | Dillweed | 63.500000 | 1.000 | 0.004 |
| 70001450 | P | Egg, whole | | | |
| | | 110-Uncooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 120-Uncooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 210-Cooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 211-Cooked; Fresh or N/S; Baked | 0.000000 | 1.000 | 0.004 |
| | | 212-Cooked; Fresh or N/S; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 213-Cooked; Fresh or N/S; Fried | 0.000000 | 1.000 | 0.004 |
| | | 214-Cooked; Fresh or N/S; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | | 215-Cooked; Fresh or N/S; Boiled/baked | 0.000000 | 1.000 | 0.004 |
| | | 221-Cooked; Frozen; Baked | 0.000000 | 1.000 | 0.004 |
| | | 223-Cooked; Frozen; Fried | 0.000000 | 1.000 | 0.004 |
| | | 224-Cooked; Frozen; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | | 230-Cooked; Dried; Cook Meth N/S | 402.500000 | 1.000 | 0.004 |
| | | 232-Cooked; Dried; Boiled | 402.500000 | 1.000 | 0.004 |
| | | 233-Cooked; Dried; Fried | 402.500000 | 1.000 | 0.004 |
| | | 240-Cooked; Canned; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 242-Cooked; Canned; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 252-Cooked; Cured etc; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 253-Cooked; Cured etc; Fried | 0.000000 | 1.000 | 0.004 |
| 70001460 | P | Egg, white | | | |
| | | 110-Uncooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 120-Uncooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 130-Uncooked; Dried; Cook Meth N/S | 402.500000 | 1.000 | 0.004 |
| | | 210-Cooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 211-Cooked; Fresh or N/S; Baked | 0.000000 | 1.000 | 0.004 |
| | | 212-Cooked; Fresh or N/S; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 213-Cooked; Fresh or N/S; Fried | 0.000000 | 1.000 | 0.004 |
| | | 214-Cooked; Fresh or N/S; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | | 221-Cooked; Frozen; Baked | 0.000000 | 1.000 | 0.004 |
| | | 223-Cooked; Frozen; Fried | 0.000000 | 1.000 | 0.004 |
| | | 230-Cooked; Dried; Cook Meth N/S | 402.500000 | 1.000 | 0.004 |
| | | 232-Cooked; Dried; Boiled | 402.500000 | 1.000 | 0.004 |
| | | 233-Cooked; Dried; Fried | 402.500000 | 1.000 | 0.004 |

| | | | | |
|---|---|---|---|---|
| | 240-Cooked; Canned; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 242-Cooked; Canned; Boiled | 0.000000 | 1.000 | 0.004 |
| | 250-Cooked; Cured etc; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| 70001461 P | Egg, white (solids)-babyfood | 402.500000 | 1.000 | 0.004 |
| 95001540 O | Fig, dried | 1.000000 | 1.000 | 0.004 |
| 14001550 14 | Filbert | 1.500000 | 1.000 | 0.004 |
| 14001560 14 | Filbert, oil | 1.500000 | 1.000 | 0.004 |
| 20001630 20 | Flaxseed, oil | 1.500000 | 1.000 | 0.004 |
| 03001650 3 | Garlic, dried | 10.900000 | 1.000 | 0.004 |
| 03001651 3 | Garlic, dried-babyfood | 10.900000 | 1.000 | 0.004 |
| 01031670 1CD | Ginger, dried | 10.900000 | 1.000 | 0.004 |
| 01011680 1AB | Ginseng, dried | 10.900000 | 1.000 | 0.004 |
| 95001780 O | Grape, raisin | 1.000000 | 1.000 | 0.004 |
| 06031820 6C | Guar, seed | 4.500000 | 1.000 | 0.004 |
| 06031821 6C | Guar, seed-babyfood | 4.500000 | 1.000 | 0.004 |
| 19011840 19A | Herbs, other | 63.500000 | 1.000 | 0.004 |
| 19011841 19A | Herbs, other-babyfood | 63.500000 | 1.000 | 0.004 |
| 14001850 14 | Hickory nut | 3.900000 | 1.000 | 0.004 |
| 19012020 19A | Lemongrass | 63.500000 | 1.000 | 0.004 |
| 06032030 6C | Lentil, seed | 4.500000 | 1.000 | 0.004 |
| 95002120 O | Lychee, dried | 1.000000 | 1.000 | 0.004 |
| 14002130 14 | Macadamia nut | 3.900000 | 1.000 | 0.004 |
| 95002160 O | Mango, dried | 1.000000 | 1.000 | 0.004 |
| 95002180 O | Maple, sugar | 1.200000 | 1.000 | 0.004 |
| 95002190 O | Maple syrup | 1.200000 | 1.000 | 0.004 |
| 19012200 19A | Marjoram | 67.100000 | 1.000 | 0.004 |
| 19012201 19A | Marjoram-babyfood | 67.100000 | 1.000 | 0.004 |
| 27002220 D | Milk, fat | | | |
| | 110-Uncooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 120-Uncooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 130-Uncooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | 150-Uncooked; Cured etc; Cook Meth N/S | 3.900000 | 1.000 | 0.004 |
| | 210-Cooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 211-Cooked; Fresh or N/S; Baked | 0.000000 | 1.000 | 0.004 |
| | 212-Cooked; Fresh or N/S; Boiled | 0.000000 | 1.000 | 0.004 |
| | 213-Cooked; Fresh or N/S; Fried | 0.000000 | 1.000 | 0.004 |
| | 214-Cooked; Fresh or N/S; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | 215-Cooked; Fresh or N/S; Boiled/baked | 0.000000 | 1.000 | 0.004 |
| | 220-Cooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 221-Cooked; Frozen; Baked | 0.000000 | 1.000 | 0.004 |
| | 222-Cooked; Frozen; Boiled | 0.000000 | 1.000 | 0.004 |
| | 223-Cooked; Frozen; Fried | 0.000000 | 1.000 | 0.004 |
| | 224-Cooked; Frozen; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | 230-Cooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | 231-Cooked; Dried; Baked | 5.400000 | 1.000 | 0.004 |
| | 232-Cooked; Dried; Boiled | 5.400000 | 1.000 | 0.004 |
| | 233-Cooked; Dried; Fried | 5.400000 | 1.000 | 0.004 |
| | 240-Cooked; Canned; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 242-Cooked; Canned; Boiled | 0.000000 | 1.000 | 0.004 |
| | 250-Cooked; Cured etc; Cook Meth N/S | 3.900000 | 1.000 | 0.004 |
| | 253-Cooked; Cured etc; Fried | 3.900000 | 1.000 | 0.004 |
| | 255-Cooked; Cured etc; Boiled/baked | 3.900000 | 1.000 | 0.004 |
| 27012230 D | Milk, nonfat solids | | | |
| | 110-Uncooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 120-Uncooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 130-Uncooked; Dried; Cook Meth N/S | | | |

| | | | | |
|---|---|---|---|---|
| | | 5.400000 | 1.000 | 0.004 |
| | 150-Uncooked; Cured etc; Cook Meth N/S | 3.900000 | 1.000 | 0.004 |
| | 210-Cooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 211-Cooked; Fresh or N/S; Baked | 0.000000 | 1.000 | 0.004 |
| | 212-Cooked; Fresh or N/S; Boiled | 0.000000 | 1.000 | 0.004 |
| | 213-Cooked; Fresh or N/S; Fried | 0.000000 | 1.000 | 0.004 |
| | 214-Cooked; Fresh or N/S; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | 215-Cooked; Fresh or N/S; Boiled/baked | 0.000000 | 1.000 | 0.004 |
| | 220-Cooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 221-Cooked; Frozen; Baked | 0.000000 | 1.000 | 0.004 |
| | 222-Cooked; Frozen; Boiled | 0.000000 | 1.000 | 0.004 |
| | 223-Cooked; Frozen; Fried | 0.000000 | 1.000 | 0.004 |
| | 224-Cooked; Frozen; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | 230-Cooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | 231-Cooked; Dried; Baked | 5.400000 | 1.000 | 0.004 |
| | 232-Cooked; Dried; Boiled | 5.400000 | 1.000 | 0.004 |
| | 233-Cooked; Dried; Fried | 5.400000 | 1.000 | 0.004 |
| | 240-Cooked; Canned; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 242-Cooked; Canned; Boiled | 0.000000 | 1.000 | 0.004 |
| | 245-Cooked; Canned; Boiled/baked | 0.000000 | 1.000 | 0.004 |
| | 250-Cooked; Cured etc; Cook Meth N/S | 3.900000 | 1.000 | 0.004 |
| | 253-Cooked; Cured etc; Fried | 3.900000 | 1.000 | 0.004 |
| | 255-Cooked; Cured etc; Boiled/baked | 3.900000 | 1.000 | 0.004 |
| 27012231 D | Milk, nonfat solids-baby food/infant | | | |
| | 110-Uncooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 130-Uncooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | 211-Cooked; Fresh or N/S; Baked | 0.000000 | 1.000 | 0.004 |
| | 240-Cooked; Canned; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| 27022240 D | Milk, water | | | |
| | 110-Uncooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 120-Uncooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 130-Uncooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | 150-Uncooked; Cured etc; Cook Meth N/S | 3.900000 | 1.000 | 0.004 |
| | 210-Cooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 211-Cooked; Fresh or N/S; Baked | 0.000000 | 1.000 | 0.004 |
| | 212-Cooked; Fresh or N/S; Boiled | 0.000000 | 1.000 | 0.004 |
| | 213-Cooked; Fresh or N/S; Fried | 0.000000 | 1.000 | 0.004 |
| | 214-Cooked; Fresh or N/S; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | 215-Cooked; Fresh or N/S; Boiled/baked | 0.000000 | 1.000 | 0.004 |
| | 220-Cooked; Frozen; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | 221-Cooked; Frozen; Baked | 0.000000 | 1.000 | 0.004 |
| | 222-Cooked; Frozen; Boiled | 0.000000 | 1.000 | 0.004 |
| | 223-Cooked; Frozen; Fried | 0.000000 | 1.000 | 0.004 |
| | 224-Cooked; Frozen; Fried/baked | 0.000000 | 1.000 | 0.004 |
| | 230-Cooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | 231-Cooked; Dried; Baked | 5.400000 | 1.000 | 0.004 |
| | 232-Cooked; Dried; Boiled | 5.400000 | 1.000 | 0.004 |
| | 233-Cooked; Dried; Fried | 5.400000 | 1.000 | 0.004 |
| | 240-Cooked; Canned; Cook Meth N/S | | | |

| | | | | | |
|---|---|---|---|---|---|
| | | | 0.000000 | 1.000 | 0.004 |
| | | 242-Cooked; Canned; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 250-Cooked; Cured etc; Cook Meth N/S | 3.900000 | 1.000 | 0.004 |
| | | 253-Cooked; Cured etc; Fried | 3.900000 | 1.000 | 0.004 |
| | | 255-Cooked; Cured etc; Boiled/baked | 3.900000 | 1.000 | 0.004 |
| 27032251 | D | Milk, sugar (lactose)-baby food/infa | | | |
| | | 110-Uncooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 130-Uncooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | | 210-Cooked; Fresh or N/S; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| | | 212-Cooked; Fresh or N/S; Boiled | 0.000000 | 1.000 | 0.004 |
| | | 230-Cooked; Dried; Cook Meth N/S | 5.400000 | 1.000 | 0.004 |
| | | 240-Cooked; Canned; Cook Meth N/S | 0.000000 | 1.000 | 0.004 |
| 15002260 | 15 | Millet, grain | 2.900000 | 1.000 | 0.008 |
| 15002310 | 15 | Oat, bran | 74.200000 | 1.000 | 0.008 |
| 15002320 | 15 | Oat, flour | 0.337000 | 1.000 | 1.000 |
| 15002321 | 15 | Oat, flour-babyfood | 0.337000 | 1.000 | 1.000 |
| 15002330 | 15 | Oat, groats/rolled oats | 18.500000 | 1.000 | 0.008 |
| 15002331 | 15 | Oat, groats/rolled oats-babyfood | 18.500000 | 1.000 | 0.008 |
| 95002360 | O | Olive, oil | 1.500000 | 1.000 | 0.004 |
| 03002380 | 3 | Onion, dry bulb, dried | 1.700000 | 1.000 | 0.004 |
| 03002381 | 3 | Onion, dry bulb, dried-babyfood | 1.700000 | 1.000 | 0.004 |
| 95002440 | O | Palm, oil | 1.500000 | 1.000 | 0.004 |
| 95002441 | O | Palm, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 95002460 | O | Papaya, dried | 1.000000 | 1.000 | 0.004 |
| 19012490 | 19A | Parsley, dried leaves | 63.500000 | 1.000 | 0.004 |
| 19012491 | 19A | Parsley, dried leaves-babyfood | 63.500000 | 1.000 | 0.004 |
| 06032560 | 6C | Pea, dry | 4.500000 | 1.000 | 0.004 |
| 06032561 | 6C | Pea, dry-babyfood | 4.500000 | 1.000 | 0.004 |
| 06032580 | 6C | Pea, pigeon, seed | 4.500000 | 1.000 | 0.004 |
| 12002610 | 12 | Peach, dried | 1.000000 | 1.000 | 0.004 |
| 12002611 | 12 | Peach, dried-babyfood | 1.000000 | 1.000 | 0.004 |
| 95002630 | O | Peanut | 16.400000 | 1.000 | 0.004 |
| 95002640 | O | Peanut, butter | 16.400000 | 1.000 | 0.004 |
| 95002650 | O | Peanut, oil | 1.500000 | 1.000 | 0.004 |
| 11002670 | 11 | Pear, dried | 1.000000 | 1.000 | 0.004 |
| 14002690 | 14 | Pecan | 3.900000 | 1.000 | 0.004 |
| 08002710 | 8 | Pepper, bell, dried | 36.100000 | 1.000 | 0.004 |
| 08002711 | 8 | Pepper, bell, dried-babyfood | 36.100000 | 1.000 | 0.004 |
| 08002730 | 8 | Pepper, nonbell, dried | 36.100000 | 1.000 | 0.004 |
| 19022740 | 19B | Pepper, black and white | 7.100000 | 1.000 | 0.004 |
| 19022741 | 19B | Pepper, black and white-babyfood | 7.100000 | 1.000 | 0.004 |
| 95002760 | O | Peppermint, oil | 1.500000 | 1.000 | 0.004 |
| 95002780 | O | Pine nut | 8.800000 | 1.000 | 0.004 |
| 95002800 | O | Pineapple, dried | 1.000000 | 1.000 | 0.004 |
| 14002820 | 14 | Pistachio | 2.300000 | 1.000 | 0.004 |
| 95002840 | O | Plantain, dried | 1.000000 | 1.000 | 0.004 |
| 12002870 | 12 | Plum, prune, dried | 0.700000 | 1.000 | 0.004 |
| 12002871 | 12 | Plum, prune, dried-babyfood | 0.700000 | 1.000 | 0.004 |
| 01032960 | 1C | Potato, chips | 7.100000 | 1.000 | 0.004 |
| 01032970 | 1C | Potato, dry (granules/ flakes) | 25.600000 | 1.000 | 0.004 |
| 01032971 | 1C | Potato, dry (granules/ flakes)-b | 25.600000 | 1.000 | 0.004 |
| 01032980 | 1C | Potato, flour | 31.400000 | 1.000 | 0.004 |
| 01032981 | 1C | Potato, flour-babyfood | 31.400000 | 1.000 | 0.004 |
| 95003060 | O | Psyllium, seed | 7.100000 | 1.000 | 0.004 |
| 09023090 | 9B | Pumpkin, seed | 8.800000 | 1.000 | 0.004 |
| 95003110 | O | Quinoa, grain | 20.400000 | 1.000 | 0.004 |
| 20003190 | 20 | Rapeseed, oil | 1.500000 | 1.000 | 0.004 |
| 20003191 | 20 | Rapeseed, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 15003230 | 15 | Rice, white | 3.900000 | 1.000 | 0.008 |
| 15003231 | 15 | Rice, white-babyfood | 3.900000 | 1.000 | 0.008 |
| 15003240 | 15 | Rice, brown | 9.400000 | 1.000 | 0.008 |
| 15003241 | 15 | Rice, brown-babyfood | 9.400000 | 1.000 | 0.008 |
| 15003250 | 15 | Rice, flour | 0.160000 | 1.000 | 1.000 |
| 15003251 | 15 | Rice, flour-babyfood | 0.160000 | 1.000 | 1.000 |
| 15003260 | 15 | Rice, bran | 37.500000 | 1.000 | 0.008 |

| | | | | | |
|---|---|---|---|---|---|
| 15003261 | 15 | Rice, bran-babyfood | 37.500000 | 1.000 | 0.008 |
| 15003280 | 15 | Rye, grain | 2.900000 | 1.000 | 0.008 |
| 15003290 | 15 | Rye, flour | 0.134000 | 1.000 | 1.000 |
| 20003300 | 20 | Safflower, oil | 1.500000 | 1.000 | 0.004 |
| 20003301 | 20 | Safflower, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 19013340 | 19A | Savory | 63.500000 | 1.000 | 0.004 |
| 95003360 | O | Sesame, seed | 7.100000 | 1.000 | 0.004 |
| 95003361 | O | Sesame, seed-babyfood | 7.100000 | 1.000 | 0.004 |
| 95003370 | O | Sesame, oil | 1.500000 | 1.000 | 0.004 |
| 95003371 | O | Sesame, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 15003440 | 15 | Sorghum, grain | 20.400000 | 1.000 | 0.008 |
| 15003450 | 15 | Sorghum, syrup | 0.600000 | 1.000 | 0.008 |
| 06003480 | 6 | Soybean, flour | 0.081000 | 1.000 | 1.000 |
| 06003481 | 6 | Soybean, flour-babyfood | 0.081000 | 1.000 | 1.000 |
| 06003490 | 6 | Soybean, soy milk | 2.400000 | 1.000 | 0.004 |
| 06003491 | 6 | Soybean, soy milk-babyfood or in | 2.400000 | 1.000 | 0.004 |
| 06003500 | 6 | Soybean, oil | 1.500000 | 1.000 | 0.004 |
| 06003501 | 6 | Soybean, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 95003530 | O | Spearmint, oil | 1.500000 | 1.000 | 0.004 |
| 19023540 | 19B | Spices, other | 7.100000 | 1.000 | 0.004 |
| 19023541 | 19B | Spices, other-babyfood | 7.100000 | 1.000 | 0.004 |
| 95003620 | O | Sugarcane, sugar | 1.200000 | 1.000 | 0.004 |
| 95003621 | O | Sugarcane, sugar-babyfood | 1.200000 | 1.000 | 0.004 |
| 95003630 | O | Sugarcane, molasses | 1.200000 | 1.000 | 0.004 |
| 95003631 | O | Sugarcane, molasses-babyfood | 1.200000 | 1.000 | 0.004 |
| 20003640 | 20 | Sunflower, seed | 4.500000 | 1.000 | 0.004 |
| 20003650 | 20 | Sunflower, oil | 1.500000 | 1.000 | 0.004 |
| 20003651 | 20 | Sunflower, oil-babyfood | 1.500000 | 1.000 | 0.004 |
| 95003720 | O | Tea, dried | 67.100000 | 1.000 | 0.004 |
| 95003730 | O | Tea, instant | 67.100000 | 1.000 | 0.004 |
| 08003780 | 8 | Tomato, dried | 1.000000 | 1.000 | 0.004 |
| 08003781 | 8 | Tomato, dried-babyfood | 1.000000 | 1.000 | 0.004 |
| 15003810 | 15 | Triticale, flour | 0.134000 | 1.000 | 1.000 |
| 15003811 | 15 | Triticale, flour-babyfood | 0.134000 | 1.000 | 1.000 |
| 01033870 | 1CD | Turmeric | 7.100000 | 1.000 | 0.004 |
| 14003910 | 14 | Walnut | 2.000000 | 1.000 | 0.004 |
| 15004010 | 15 | Wheat, grain | 2.900000 | 1.000 | 0.008 |
| 15004011 | 15 | Wheat, grain-babyfood | 2.900000 | 1.000 | 0.008 |
| 15004020 | 15 | Wheat, flour | 0.134000 | 1.000 | 1.000 |
| 15004021 | 15 | Wheat, flour-babyfood | 0.134000 | 1.000 | 1.000 |
| 15004030 | 15 | Wheat, germ | 54.000000 | 1.000 | 0.008 |
| 15004040 | 15 | Wheat, bran | 74.200000 | 1.000 | 0.008 |
| 15004050 | 15 | Wild rice | 9.400000 | 1.000 | 0.008 |

## Attachment 4. Results of cryolite fluoride exposure analysis

```
U.S. Environmental Protection Agency                                 Ver. 2.02
DEEM-FCID ACUTE Analysis for CRYOLITE                            (1994-98 data)
Residue file: Cryolite-AR-CT new raisin factor.R98
    Adjustment factor #2 used.
Analysis Date: 06-03-2010/13:01:11    Residue file dated: 06-24-2004/10:05:08/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: ""
===============================================================================

Summary calculations (per capita):

                    95th Percentile      99th Percentile     99.9th Percentile
                       Exposure             Exposure             Exposure
                      ----------           ----------           ----------
All infants:
                        0.048374             0.180153             0.437825
Custom demographics 1: 0.5 - <1:
                        0.175850             0.367189             0.703934
Custom demographics 2: 1 - <4:
                        0.202992             0.443950             0.921210
Custom demographics 3: 4 - <7:
                        0.159838             0.313532             0.737197
Custom demographics 4: 7 - <11:
                        0.126163             0.332575             0.741806
Custom demographics 5: 11 - <14:
                        0.113618             0.324475             0.733375
Custom demographics 6: 14+:
                        0.179623             0.425648             0.861668
```

U.S. Environmental Protection Agency — Ver. 2.02
DEEM-FCID ACUTE Analysis for CRYOLITE — (1994-98 data)
Residue file: Cryolite-AR-CT new raisin factor.R98
Adjustment factor #2 used.
Analysis Date: 06-03-2010/13:01:11 Residue file dated: 06-24-2004/10:05:08/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: ""

All infants

Daily Exposure Analysis /a (mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.008410 | 0.016502 |
| Standard Deviation | 0.035543 | 0.048427 |
| Standard Error of mean | 0.000652 | 0.001252 |

Percent of Person-Days that are User-Days = 50.97%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000113 | 90.00 | 0.047259 |
| 20.00 | 0.000331 | 95.00 | 0.102587 |
| 30.00 | 0.000635 | 97.50 | 0.126726 |
| 40.00 | 0.001133 | 99.00 | 0.243049 |
| 50.00 | 0.001882 | 99.50 | 0.289194 |
| 60.00 | 0.002814 | 99.75 | 0.366479 |
| 70.00 | 0.004828 | 99.90 | 0.481071 |
| 80.00 | 0.009149 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000000 | 90.00 | 0.009432 |
| 20.00 | 0.000000 | 95.00 | 0.048374 |
| 30.00 | 0.000000 | 97.50 | 0.104997 |
| 40.00 | 0.000000 | 99.00 | 0.180153 |
| 50.00 | 0.000021 | 99.50 | 0.243227 |
| 60.00 | 0.000370 | 99.75 | 0.289300 |
| 70.00 | 0.001204 | 99.90 | 0.437825 |
| 80.00 | 0.002981 | | |

a/ Analysis based on all two-day participant records in CSFII 1994-98 survey.

1

U.S. Environmental Protection Agency                                Ver. 2.02
DEEM-FCID ACUTE Analysis for CRYOLITE                              (1994-98 data)
Residue file: Cryolite-AR-CT new raisin factor.R98
    Adjustment factor #2 used.
Analysis Date: 06-03-2010/13:01:11    Residue file dated: 06-24-2004/10:05:08/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: ""

===============================================================================

Custom demographics 1: 0.5 - <1
All Seasons
All Regions
Sex: M/F-all/
All Races
Nursing and Non-Nursing (Ages <= 3)
Age-Low: 6 m  High: 1 yrs

-------------------------------

Daily Exposure Analysis (mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.030833 | 0.033133 |
| Standard Deviation | 0.073530 | 0.075721 |
| Standard Error of mean | 0.001241 | 0.001334 |

Percent of Person-Days that are User-Days = 93.06%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000308 | 90.00 | 0.100236 |
| 20.00 | 0.000788 | 95.00 | 0.183991 |
| 30.00 | 0.001443 | 97.50 | 0.248128 |
| 40.00 | 0.002305 | 99.00 | 0.369782 |
| 50.00 | 0.004039 | 99.50 | 0.485080 |
| 60.00 | 0.007185 | 99.75 | 0.577116 |
| 70.00 | 0.015028 | 99.90 | 0.705472 |
| 80.00 | 0.041860 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000101 | 90.00 | 0.094318 |
| 20.00 | 0.000481 | 95.00 | 0.175850 |
| 30.00 | 0.001091 | 97.50 | 0.246802 |
| 40.00 | 0.001901 | 99.00 | 0.367189 |
| 50.00 | 0.003216 | 99.50 | 0.483932 |
| 60.00 | 0.005960 | 99.75 | 0.525181 |
| 70.00 | 0.012055 | 99.90 | 0.703934 |
| 80.00 | 0.034430 | | |

U.S. Environmental Protection Agency Ver. 2.02
DEEM-FCID ACUTE Analysis for CRYOLITE (1994-98 data)
Residue file: Cryolite-AR-CT new raisin factor.R98
Adjustment factor #2 used.
Analysis Date: 06-03-2010/13:01:11 Residue file dated: 06-24-2004/10:05:08/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: ""

Custom demographics 2: 1 - <4
All Seasons
All Regions
Sex: M/F-all/
All Races
Nursing and Non-Nursing (Ages <= 3)
Age-Low: 1 yrs High: 4 yrs

Daily Exposure Analysis (mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.040443 | 0.041017 |
| Standard Deviation | 0.090977 | 0.091492 |
| Standard Error of mean | 0.000857 | 0.000867 |

Percent of Person-Days that are User-Days = 98.60%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000553 | 90.00 | 0.121346 |
| 20.00 | 0.001268 | 95.00 | 0.205310 |
| 30.00 | 0.002092 | 97.50 | 0.287423 |
| 40.00 | 0.003407 | 99.00 | 0.445745 |
| 50.00 | 0.006150 | 99.50 | 0.527179 |
| 60.00 | 0.012014 | 99.75 | 0.634370 |
| 70.00 | 0.023498 | 99.90 | 0.924251 |
| 80.00 | 0.050975 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000463 | 90.00 | 0.120416 |
| 20.00 | 0.001185 | 95.00 | 0.202992 |
| 30.00 | 0.001996 | 97.50 | 0.284998 |
| 40.00 | 0.003255 | 99.00 | 0.443950 |
| 50.00 | 0.005910 | 99.50 | 0.523800 |
| 60.00 | 0.011640 | 99.75 | 0.632941 |
| 70.00 | 0.023114 | 99.90 | 0.921210 |
| 80.00 | 0.050184 | | |

Residue file: Cryolite-AR-CT new raisin factor.R98
Adjustment factor #2 used.
Analysis Date: 06-03-2010/13:01:11    Residue file dated: 06-24-2004/10:05:08/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: ""

===============================================================================

Custom demographics 3: 4 - <7
All Seasons
All Regions
Sex: M/F-all/
All Races
Age-Low: 4 yrs   High: 7 yrs

-----------------------------

Daily Exposure Analysis
(mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.031330 | 0.031550 |
| Standard Deviation | 0.070270 | 0.070467 |
| Standard Error of mean | 0.000841 | 0.000846 |

Percent of Person-Days that are User-Days = 99.30%

Estimated percentile of user-days falling below calculated exposure
in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000698 | 90.00 | 0.090283 |
| 20.00 | 0.001435 | 95.00 | 0.160747 |
| 30.00 | 0.002307 | 97.50 | 0.231215 |
| 40.00 | 0.003723 | 99.00 | 0.313832 |
| 50.00 | 0.006322 | 99.50 | 0.440487 |
| 60.00 | 0.011540 | 99.75 | 0.577595 |
| 70.00 | 0.021241 | 99.90 | 0.737251 |
| 80.00 | 0.037074 | | |

Estimated percentile of per-capita days falling below calculated exposure
in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000656 | 90.00 | 0.089739 |
| 20.00 | 0.001389 | 95.00 | 0.159838 |
| 30.00 | 0.002246 | 97.50 | 0.229675 |
| 40.00 | 0.003648 | 99.00 | 0.313532 |
| 50.00 | 0.006191 | 99.50 | 0.439401 |
| 60.00 | 0.011397 | 99.75 | 0.576810 |
| 70.00 | 0.021063 | 99.90 | 0.737197 |
| 80.00 | 0.036609 | | |

U.S. Environmental Protection Agency Ver. 2.02
DEEM-FCID ACUTE Analysis for CRYOLITE (1994-98 data)
Residue file: Cryolite-AR-CT new raisin factor.R98
Adjustment factor #2 used.
Analysis Date: 06-03-2010/13:01:11 Residue file dated: 06-24-2004/10:05:08/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: ""

Custom demographics 4: 7 - <11
All Seasons
All Regions
Sex: M/F-all/
All Races
Age-Low: 7 yrs High: 11 yrs

Daily Exposure Analysis (mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.028471 | 0.028704 |
| Standard Deviation | 0.069277 | 0.069512 |
| Standard Error of mean | 0.001337 | 0.001347 |

Percent of Person-Days that are User-Days = 99.19%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000759 | 90.00 | 0.074546 |
| 20.00 | 0.001569 | 95.00 | 0.126652 |
| 30.00 | 0.002533 | 97.50 | 0.225153 |
| 40.00 | 0.003744 | 99.00 | 0.333141 |
| 50.00 | 0.005884 | 99.50 | 0.485657 |
| 60.00 | 0.010417 | 99.75 | 0.671156 |
| 70.00 | 0.017264 | 99.90 | 0.741857 |
| 80.00 | 0.028363 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000703 | 90.00 | 0.074291 |
| 20.00 | 0.001513 | 95.00 | 0.126163 |
| 30.00 | 0.002456 | 97.50 | 0.224248 |
| 40.00 | 0.003685 | 99.00 | 0.332575 |
| 50.00 | 0.005711 | 99.50 | 0.484766 |
| 60.00 | 0.010173 | 99.75 | 0.671024 |
| 70.00 | 0.017039 | 99.90 | 0.741806 |
| 80.00 | 0.028166 | | |

U.S. Environmental Protection Agency Ver. 2.02
DEEM-FCID ACUTE Analysis for CRYOLITE (1994-98 data)
Residue file: Cryolite-AR-CT new raisin factor.R98
Adjustment factor #2 used.
Analysis Date: 06-03-2010/13:01:11 Residue file dated: 06-24-2004/10:05:08/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: ""

===============================================================================

Custom demographics 5: 11 - <14
All Seasons
All Regions
Sex: M/F-all/
All Races
Age-Low: 11 yrs High: 14 yrs

------------------------------

Daily Exposure Analysis
(mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.024786 | 0.025056 |
| Standard Deviation | 0.061571 | 0.061850 |
| Standard Error of mean | 0.001524 | 0.001540 |

Percent of Person-Days that are User-Days = 98.92%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000871 | 90.00 | 0.059909 |
| 20.00 | 0.001695 | 95.00 | 0.115049 |
| 30.00 | 0.002571 | 97.50 | 0.188393 |
| 40.00 | 0.003597 | 99.00 | 0.324694 |
| 50.00 | 0.005329 | 99.50 | 0.425857 |
| 60.00 | 0.008325 | 99.75 | 0.489522 |
| 70.00 | 0.014132 | 99.90 | 0.733597 |
| 80.00 | 0.025251 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000773 | 90.00 | 0.059621 |
| 20.00 | 0.001634 | 95.00 | 0.113618 |
| 30.00 | 0.002497 | 97.50 | 0.188208 |
| 40.00 | 0.003503 | 99.00 | 0.324475 |
| 50.00 | 0.005215 | 99.50 | 0.425786 |
| 60.00 | 0.008098 | 99.75 | 0.489044 |
| 70.00 | 0.013948 | 99.90 | 0.733375 |
| 80.00 | 0.025070 | | |

U.S. Environmental Protection Agency                                    Ver. 2.02
DEEM-FCID ACUTE Analysis for CRYOLITE                              (1994-98 data)
Residue file: Cryolite-AR-CT new raisin factor.R98
    Adjustment factor #2 used.
Analysis Date: 06-03-2010/13:01:11    Residue file dated: 06-24-2004/10:05:08/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: ""

===============================================================================

Custom demographics 6: 14+
All Seasons
All Regions
Sex: M/F-all/
All Races
Age-Low: 14 yrs   High: 99 yrs

--------------------------

Daily Exposure Analysis
(mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.032704 | 0.033291 |
| Standard Deviation | 0.084056 | 0.084692 |
| Standard Error of mean | 0.000584 | 0.000594 |

Percent of Person-Days that are User-Days = 98.24%

Estimated percentile of user-days falling below calculated exposure
in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000863 | 90.00 | 0.078174 |
| 20.00 | 0.001938 | 95.00 | 0.182131 |
| 30.00 | 0.003068 | 97.50 | 0.278982 |
| 40.00 | 0.004460 | 99.00 | 0.427729 |
| 50.00 | 0.006449 | 99.50 | 0.533164 |
| 60.00 | 0.009821 | 99.75 | 0.681739 |
| 70.00 | 0.016997 | 99.90 | 0.872898 |
| 80.00 | 0.029959 | | |

Estimated percentile of per-capita days falling below calculated exposure
in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000684 | 90.00 | 0.076275 |
| 20.00 | 0.001796 | 95.00 | 0.179623 |
| 30.00 | 0.002912 | 97.50 | 0.276623 |
| 40.00 | 0.004277 | 99.00 | 0.425648 |
| 50.00 | 0.006224 | 99.50 | 0.530005 |
| 60.00 | 0.009528 | 99.75 | 0.681273 |
| 70.00 | 0.016530 | 99.90 | 0.861668 |
| 80.00 | 0.029249 | | |

# Attachment 5. Results of sulfuryl fluoride food fumigation exposure analysis

U.S. Environmental Protection Agency Ver. 2.02
DEEM-FCID ACUTE Analysis for FLUORIDE (1994-98 data)
Residue file: F Food Fumigation - 2009 RevisedCT - 4-28 Strict Label.R98
Adjustment factor #2 used.
Analysis Date: 06-03-2010/12:59:31 Residue file dated: 05-06-2009/13:54:06/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: "RfD is converted Fluoride MCL and for adult pops only."

===============================================================================

Summary calculations (per capita):

| | 95th Percentile Exposure | 99th Percentile Exposure | 99.9th Percentile Exposure |
|---|---|---|---|
| All infants: | 0.039579 | 0.098237 | 0.230985 |
| Custom demographics 1: 0.5 - <1: | 0.096990 | 0.226422 | 0.429346 |
| Custom demographics 2: 1 - <4: | 0.140401 | 0.282132 | 0.528114 |
| Custom demographics 3: 4 - <7: | 0.176854 | 0.355497 | 0.663533 |
| Custom demographics 4: 7 - <11: | 0.204775 | 0.406162 | 0.744262 |
| Custom demographics 5: 11 - <14: | 0.291818 | 0.602765 | 0.981154 |
| Custom demographics 6: 14+: | 0.273986 | 0.563283 | 1.122529 |

U.S. Environmental Protection Agency Ver. 2.02
DEEM-FCID ACUTE Analysis for FLUORIDE (1994-98 data)
Residue file: F Food Fumigation - 2009 RevisedCT - 4-28 Strict Label.R98
Adjustment factor #2 used.
Analysis Date: 06-03-2010/12:59:31 Residue file dated: 05-06-2009/13:54:06/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: "RfD is converted Fluoride MCL and for adult pops only."

All infants

Daily Exposure Analysis /a (mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.009074 | 0.012660 |
| Standard Deviation | 0.020369 | 0.023098 |
| Standard Error of mean | 0.000374 | 0.000500 |

Percent of Person-Days that are User-Days = 71.67%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000096 | 90.00 | 0.033116 |
| 20.00 | 0.000323 | 95.00 | 0.048078 |
| 30.00 | 0.000933 | 97.50 | 0.073765 |
| 40.00 | 0.002440 | 99.00 | 0.108894 |
| 50.00 | 0.004827 | 99.50 | 0.135625 |
| 60.00 | 0.007798 | 99.75 | 0.197544 |
| 70.00 | 0.011819 | 99.90 | 0.274808 |
| 80.00 | 0.019633 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000000 | 90.00 | 0.027144 |
| 20.00 | 0.000000 | 95.00 | 0.039579 |
| 30.00 | 0.000022 | 97.50 | 0.057927 |
| 40.00 | 0.000214 | 99.00 | 0.098237 |
| 50.00 | 0.000957 | 99.50 | 0.124019 |
| 60.00 | 0.003358 | 99.75 | 0.159292 |
| 70.00 | 0.007314 | 99.90 | 0.230985 |
| 80.00 | 0.013155 | | |

a/ Analysis based on all two-day participant records in CSFII 1994-98 survey.
1

```
U.S. Environmental Protection Agency                                    Ver. 2.02
DEEM-FCID ACUTE Analysis for FLUORIDE                              (1994-98 data)
Residue file: F Food Fumigation - 2009  RevisedCT - 4-28 Strict Label.R98
    Adjustment factor #2 used.
Analysis Date: 06-03-2010/12:59:31    Residue file dated: 05-06-2009/13:54:06/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: "RfD is converted Fluoride MCL and for adult pops only."
===============================================================================

Custom demographics 1: 0.5 - <1
All Seasons
All Regions
Sex: M/F-all/
All Races
Nursing and Non-Nursing (Ages <= 3)
Age-Low: 6 m  High: 1 yrs
-------------------------------
```

Daily Exposure Analysis (mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.021276 | 0.021691 |
| Standard Deviation | 0.041194 | 0.041486 |
| Standard Error of mean | 0.000696 | 0.000709 |

Percent of Person-Days that are User-Days = 98.08%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000713 | 90.00 | 0.053524 |
| 20.00 | 0.001437 | 95.00 | 0.097690 |
| 30.00 | 0.002363 | 97.50 | 0.132565 |
| 40.00 | 0.003805 | 99.00 | 0.227504 |
| 50.00 | 0.006416 | 99.50 | 0.273936 |
| 60.00 | 0.010962 | 99.75 | 0.324460 |
| 70.00 | 0.018673 | 99.90 | 0.429503 |
| 80.00 | 0.029818 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.000592 | 90.00 | 0.052671 |
| 20.00 | 0.001332 | 95.00 | 0.096990 |
| 30.00 | 0.002232 | 97.50 | 0.131812 |
| 40.00 | 0.003620 | 99.00 | 0.226422 |
| 50.00 | 0.005965 | 99.50 | 0.273629 |
| 60.00 | 0.010310 | 99.75 | 0.323589 |
| 70.00 | 0.018080 | 99.90 | 0.429346 |
| 80.00 | 0.029392 | | |

```
U.S. Environmental Protection Agency                                    Ver. 2.02
DEEM-FCID ACUTE Analysis for FLUORIDE                               (1994-98 data)
Residue file: F Food Fumigation - 2009  RevisedCT - 4-28 Strict Label.R98
    Adjustment factor #2 used.
Analysis Date: 06-03-2010/12:59:31    Residue file dated: 05-06-2009/13:54:06/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: "RfD is converted Fluoride MCL and for adult pops only."
===============================================================================

Custom demographics 2: 1 - <4
All Seasons
All Regions
Sex: M/F-all/
All Races
Nursing and Non-Nursing (Ages <= 3)
Age-Low: 1 yrs   High: 4 yrs
-----------------------------
```

Daily Exposure Analysis (mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.032891 | 0.032958 |
| Standard Deviation | 0.059247 | 0.059289 |
| Standard Error of mean | 0.000558 | 0.000559 |

Percent of Person-Days that are User-Days = 99.79%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.001493 | 90.00 | 0.088963 |
| 20.00 | 0.002474 | 95.00 | 0.140511 |
| 30.00 | 0.003756 | 97.50 | 0.198017 |
| 40.00 | 0.005811 | 99.00 | 0.282227 |
| 50.00 | 0.010068 | 99.50 | 0.359262 |
| 60.00 | 0.017265 | 99.75 | 0.443445 |
| 70.00 | 0.028945 | 99.90 | 0.528153 |
| 80.00 | 0.045984 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.001475 | 90.00 | 0.088844 |
| 20.00 | 0.002457 | 95.00 | 0.140401 |
| 30.00 | 0.003733 | 97.50 | 0.197707 |
| 40.00 | 0.005780 | 99.00 | 0.282132 |
| 50.00 | 0.009990 | 99.50 | 0.358994 |
| 60.00 | 0.017173 | 99.75 | 0.443314 |
| 70.00 | 0.028867 | 99.90 | 0.528114 |
| 80.00 | 0.045864 | | |

```
U.S. Environmental Protection Agency                                Ver. 2.02
DEEM-FCID ACUTE Analysis for FLUORIDE                            (1994-98 data)
Residue file: F Food Fumigation - 2009  RevisedCT - 4-28 Strict Label.R98
    Adjustment factor #2 used.
Analysis Date: 06-03-2010/12:59:31    Residue file dated: 05-06-2009/13:54:06/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: "RfD is converted Fluoride MCL and for adult pops only."
===============================================================================

Custom demographics 3: 4 - <7
All Seasons
All Regions
Sex: M/F-all/
All Races
Age-Low: 4 yrs   High: 7 yrs
-----------------------------
```

Daily Exposure Analysis (mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.046643 | 0.046649 |
| Standard Deviation | 0.073659 | 0.073662 |
| Standard Error of mean | 0.000881 | 0.000881 |

Percent of Person-Days that are User-Days = 99.99%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.002394 | 90.00 | 0.121850 |
| 20.00 | 0.004068 | 95.00 | 0.176863 |
| 30.00 | 0.006915 | 97.50 | 0.266527 |
| 40.00 | 0.012303 | 99.00 | 0.355504 |
| 50.00 | 0.021403 | 99.50 | 0.487238 |
| 60.00 | 0.032227 | 99.75 | 0.534215 |
| 70.00 | 0.044555 | 99.90 | 0.663534 |
| 80.00 | 0.067720 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.002392 | 90.00 | 0.121841 |
| 20.00 | 0.004066 | 95.00 | 0.176854 |
| 30.00 | 0.006911 | 97.50 | 0.266522 |
| 40.00 | 0.012297 | 99.00 | 0.355497 |
| 50.00 | 0.021396 | 99.50 | 0.487227 |
| 60.00 | 0.032222 | 99.75 | 0.534211 |
| 70.00 | 0.044549 | 99.90 | 0.663533 |
| 80.00 | 0.067714 | | |

U.S. Environmental Protection Agency Ver. 2.02
DEEM-FCID ACUTE Analysis for FLUORIDE (1994-98 data)
Residue file: F Food Fumigation - 2009 RevisedCT - 4-28 Strict Label.R98
Adjustment factor #2 used.
Analysis Date: 06-03-2010/12:59:31 Residue file dated: 05-06-2009/13:54:06/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: "RfD is converted Fluoride MCL and for adult pops only."

===============================================================================

Custom demographics 4: 7 - <11
All Seasons
All Regions
Sex: M/F-all/
All Races
Age-Low: 7 yrs High: 11 yrs

------------------------------

Daily Exposure Analysis (mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.054355 | 0.054355 |
| Standard Deviation | 0.089280 | 0.089280 |
| Standard Error of mean | 0.001723 | 0.001723 |

Percent of Person-Days that are User-Days = 100.00%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.002781 | 90.00 | 0.130309 |
| 20.00 | 0.005067 | 95.00 | 0.204775 |
| 30.00 | 0.009619 | 97.50 | 0.297045 |
| 40.00 | 0.017785 | 99.00 | 0.406162 |
| 50.00 | 0.028257 | 99.50 | 0.539413 |
| 60.00 | 0.037462 | 99.75 | 0.639542 |
| 70.00 | 0.050645 | 99.90 | 0.744262 |
| 80.00 | 0.076801 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.002781 | 90.00 | 0.130309 |
| 20.00 | 0.005067 | 95.00 | 0.204775 |
| 30.00 | 0.009619 | 97.50 | 0.297045 |
| 40.00 | 0.017785 | 99.00 | 0.406162 |
| 50.00 | 0.028257 | 99.50 | 0.539413 |
| 60.00 | 0.037462 | 99.75 | 0.639542 |
| 70.00 | 0.050645 | 99.90 | 0.744262 |
| 80.00 | 0.076801 | | |

U.S. Environmental Protection Agency Ver. 2.02
DEEM-FCID ACUTE Analysis for FLUORIDE (1994-98 data)
Residue file: F Food Fumigation - 2009 RevisedCT - 4-28 Strict Label.R98
Adjustment factor #2 used.
Analysis Date: 06-03-2010/12:59:31 Residue file dated: 05-06-2009/13:54:06/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: "RfD is converted Fluoride MCL and for adult pops only."

===============================================================================

Custom demographics 5: 11 - <14
All Seasons
All Regions
Sex: M/F-all/
All Races
Age-Low: 11 yrs High: 14 yrs

Daily Exposure Analysis
(mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.067522 | 0.067522 |
| Standard Deviation | 0.127399 | 0.127399 |
| Standard Error of mean | 0.003154 | 0.003154 |

Percent of Person-Days that are User-Days = 100.00%

Estimated percentile of user-days falling below calculated exposure
in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.002472 | 90.00 | 0.179495 |
| 20.00 | 0.004212 | 95.00 | 0.291818 |
| 30.00 | 0.006674 | 97.50 | 0.400124 |
| 40.00 | 0.014134 | 99.00 | 0.602765 |
| 50.00 | 0.027275 | 99.50 | 0.792960 |
| 60.00 | 0.040233 | 99.75 | 0.827384 |
| 70.00 | 0.057641 | 99.90 | 0.981154 |
| 80.00 | 0.092775 | | |

Estimated percentile of per-capita days falling below calculated exposure
in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.002472 | 90.00 | 0.179495 |
| 20.00 | 0.004212 | 95.00 | 0.291818 |
| 30.00 | 0.006674 | 97.50 | 0.400124 |
| 40.00 | 0.014134 | 99.00 | 0.602765 |
| 50.00 | 0.027275 | 99.50 | 0.792960 |
| 60.00 | 0.040233 | 99.75 | 0.827384 |
| 70.00 | 0.057641 | 99.90 | 0.981154 |
| 80.00 | 0.092775 | | |

U.S. Environmental Protection Agency Ver. 2.02
DEEM-FCID ACUTE Analysis for FLUORIDE (1994-98 data)
Residue file: F Food Fumigation - 2009 RevisedCT - 4-28 Strict Label.R98
Adjustment factor #2 used.
Analysis Date: 06-03-2010/12:59:31 Residue file dated: 05-06-2009/13:54:06/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: "RfD is converted Fluoride MCL and for adult pops only."

===============================================================================

Custom demographics 6: 14+
All Seasons
All Regions
Sex: M/F-all/
All Races
Age-Low: 14 yrs High: 99 yrs

Daily Exposure Analysis (mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.057620 | 0.057768 |
| Standard Deviation | 0.117804 | 0.117919 |
| Standard Error of mean | 0.000819 | 0.000821 |

Percent of Person-Days that are User-Days = 99.74%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.001770 | 90.00 | 0.168094 |
| 20.00 | 0.002968 | 95.00 | 0.274313 |
| 30.00 | 0.004572 | 97.50 | 0.395651 |
| 40.00 | 0.007237 | 99.00 | 0.563477 |
| 50.00 | 0.012067 | 99.50 | 0.747002 |
| 60.00 | 0.021400 | 99.75 | 0.883366 |
| 70.00 | 0.039452 | 99.90 | 1.122607 |
| 80.00 | 0.074577 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.001744 | 90.00 | 0.167643 |
| 20.00 | 0.002939 | 95.00 | 0.273986 |
| 30.00 | 0.004537 | 97.50 | 0.395310 |
| 40.00 | 0.007181 | 99.00 | 0.563283 |
| 50.00 | 0.011979 | 99.50 | 0.746855 |
| 60.00 | 0.021271 | 99.75 | 0.883065 |
| 70.00 | 0.039263 | 99.90 | 1.122529 |
| 80.00 | 0.074327 | | |

## Attachment 6. Results of sulfuryl fluoride structural fumigation exposure analysis

U.S. Environmental Protection Agency Ver. 2.02
DEEM-FCID ACUTE Analysis for FLUORIDE (1994-98 data)
Residue file: F Space Fumigation - 2009 - 5-1.R98 Adjustment factor #2 used.
Analysis Date: 06-03-2010/12:57:19 Residue file dated: 05-06-2009/13:52:45/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: "RfD is converted Fluoride MCL and is valid for adult pops. only"

===============================================================================

Summary calculations (per capita):

| | 95th Percentile Exposure | 99th Percentile Exposure | 99.9th Percentile Exposure |
|---|---|---|---|
| All infants: | 0.012880 | 0.021374 | 0.046940 |
| Custom demographics 1: 0.5 - <1: | 0.020257 | 0.032080 | 0.050413 |
| Custom demographics 2: 1 - <4: | 0.025853 | 0.039290 | 0.062731 |
| Custom demographics 3: 4 - <7: | 0.030100 | 0.045022 | 0.076355 |
| Custom demographics 4: 7 - <11: | 0.034938 | 0.050785 | 0.074763 |
| Custom demographics 5: 11 - <14: | 0.037161 | 0.051720 | 0.089794 |
| Custom demographics 6: 14+: | 0.045247 | 0.074354 | 0.135312 |

```
U.S. Environmental Protection Agency                                   Ver. 2.02
DEEM-FCID ACUTE Analysis for FLUORIDE                               (1994-98 data)
Residue file: F Space Fumigation - 2009 - 5-1.R98     Adjustment factor #2 used.
Analysis Date: 06-03-2010/12:57:19    Residue file dated: 05-06-2009/13:52:45/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: "RfD is converted Fluoride MCL and is valid for adult pops. only"
===============================================================================

All infants                        Daily Exposure Analysis  /a
-----------                              (mg/day)
                                   per Capita    per User
                                   -----------  -----------
            Mean                     0.004162     0.004671
            Standard Deviation       0.005018     0.005087
            Standard Error of mean   0.000092     0.000099

       Percent of Person-Days that are User-Days =  89.10%


  Estimated percentile of user-days falling below calculated exposure
                     in mg/day

      Percentile   Exposure                  Percentile   Exposure
      ----------  ----------                 ----------  ----------
         10.00     0.000287                     90.00     0.010182
         20.00     0.001141                     95.00     0.013552
         30.00     0.001796                     97.50     0.017857
         40.00     0.002352                     99.00     0.022398
         50.00     0.003240                     99.50     0.032183
         60.00     0.004202                     99.75     0.041136
         70.00     0.005457                     99.90     0.047989
         80.00     0.007331


  Estimated percentile of per-capita days falling below calculated exposure
                          in mg/day

      Percentile   Exposure                  Percentile   Exposure
      ----------  ----------                 ----------  ----------
         10.00     0.000000                     90.00     0.009705
         20.00     0.000296                     95.00     0.012880
         30.00     0.001279                     97.50     0.017653
         40.00     0.001912                     99.00     0.021374
         50.00     0.002668                     99.50     0.032032
         60.00     0.003716                     99.75     0.040955
         70.00     0.004955                     99.90     0.046940
         80.00     0.006913
-------------------------------------------------------------------------------
a/ Analysis based on all two-day participant records in CSFII 1994-98 survey.
                                   1
```

U.S. Environmental Protection Agency                                    Ver. 2.02
DEEM-FCID ACUTE Analysis for FLUORIDE                               (1994-98 data)
Residue file: F Space Fumigation - 2009 - 5-1.R98     Adjustment factor #2 used.
Analysis Date: 06-03-2010/12:57:19     Residue file dated: 05-06-2009/13:52:45/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: "RfD is converted Fluoride MCL and is valid for adult pops. only"

===============================================================================

Custom demographics 1: 0.5 - <1
All Seasons
All Regions
Sex: M/F-all/
All Races
Nursing and Non-Nursing (Ages <= 3)
Age-Low: 6 m  High: 1 yrs

-------------------------------

Daily Exposure Analysis
(mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.008687 | 0.008742 |
| Standard Deviation | 0.006490 | 0.006473 |
| Standard Error of mean | 0.000110 | 0.000110 |

Percent of Person-Days that are User-Days = 99.37%

Estimated percentile of user-days falling below calculated exposure
in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.002317 | 90.00 | 0.016759 |
| 20.00 | 0.003754 | 95.00 | 0.020288 |
| 30.00 | 0.004835 | 97.50 | 0.024883 |
| 40.00 | 0.006085 | 99.00 | 0.032100 |
| 50.00 | 0.007295 | 99.50 | 0.038552 |
| 60.00 | 0.008688 | 99.75 | 0.041749 |
| 70.00 | 0.010478 | 99.90 | 0.050424 |
| 80.00 | 0.012723 | | |

Estimated percentile of per-capita days falling below calculated exposure
in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.002228 | 90.00 | 0.016715 |
| 20.00 | 0.003685 | 95.00 | 0.020257 |
| 30.00 | 0.004790 | 97.50 | 0.024758 |
| 40.00 | 0.006034 | 99.00 | 0.032080 |
| 50.00 | 0.007254 | 99.50 | 0.038501 |
| 60.00 | 0.008650 | 99.75 | 0.041724 |
| 70.00 | 0.010442 | 99.90 | 0.050413 |
| 80.00 | 0.012683 | | |

U.S. Environmental Protection Agency Ver. 2.02
DEEM-FCID ACUTE Analysis for FLUORIDE (1994-98 data)
Residue file: F Space Fumigation - 2009 - 5-1.R98 Adjustment factor #2 used.
Analysis Date: 06-03-2010/12:57:19 Residue file dated: 05-06-2009/13:52:45/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: "RfD is converted Fluoride MCL and is valid for adult pops. only"

===============================================================================

Custom demographics 2: 1 - <4
All Seasons
All Regions
Sex: M/F-all/
All Races
Nursing and Non-Nursing (Ages <= 3)
Age-Low: 1 yrs High: 4 yrs

Daily Exposure Analysis (mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.012080 | 0.012093 |
| Standard Deviation | 0.007781 | 0.007776 |
| Standard Error of mean | 0.000073 | 0.000073 |

Percent of Person-Days that are User-Days = 99.89%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.004254 | 90.00 | 0.021199 |
| 20.00 | 0.006188 | 95.00 | 0.025862 |
| 30.00 | 0.007792 | 97.50 | 0.030621 |
| 40.00 | 0.009186 | 99.00 | 0.039299 |
| 50.00 | 0.010638 | 99.50 | 0.047837 |
| 60.00 | 0.012244 | 99.75 | 0.054314 |
| 70.00 | 0.014101 | 99.90 | 0.062737 |
| 80.00 | 0.016786 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.004233 | 90.00 | 0.021193 |
| 20.00 | 0.006169 | 95.00 | 0.025853 |
| 30.00 | 0.007783 | 97.50 | 0.030615 |
| 40.00 | 0.009178 | 99.00 | 0.039290 |
| 50.00 | 0.010629 | 99.50 | 0.047830 |
| 60.00 | 0.012237 | 99.75 | 0.054306 |
| 70.00 | 0.014095 | 99.90 | 0.062731 |
| 80.00 | 0.016780 | | |

U.S. Environmental Protection Agency Ver. 2.02
DEEM-FCID ACUTE Analysis for FLUORIDE (1994-98 data)
Residue file: F Space Fumigation - 2009 - 5-1.R98 Adjustment factor #2 used.
Analysis Date: 06-03-2010/12:57:19 Residue file dated: 05-06-2009/13:52:45/8
No body weight adjustment; Toxicology endpoints not used
Daily totals for food and foodform consumption used.
Run Comment: "RfD is converted Fluoride MCL and is valid for adult pops. only"

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Custom demographics 3: 4 - <7
All Seasons
All Regions
Sex: M/F-all/
All Races
Age-Low: 4 yrs High: 7 yrs

Daily Exposure Analysis
(mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.015334 | 0.015335 |
| Standard Deviation | 0.008582 | 0.008582 |
| Standard Error of mean | 0.000103 | 0.000103 |

Percent of Person-Days that are User-Days = 100.00%

Estimated percentile of user-days falling below calculated exposure
in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.006783 | 90.00 | 0.025067 |
| 20.00 | 0.008838 | 95.00 | 0.030101 |
| 30.00 | 0.010475 | 97.50 | 0.036002 |
| 40.00 | 0.012192 | 99.00 | 0.045022 |
| 50.00 | 0.013877 | 99.50 | 0.054278 |
| 60.00 | 0.015620 | 99.75 | 0.062688 |
| 70.00 | 0.017752 | 99.90 | 0.076356 |
| 80.00 | 0.020477 | | |

Estimated percentile of per-capita days falling below calculated exposure
in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.006782 | 90.00 | 0.025067 |
| 20.00 | 0.008837 | 95.00 | 0.030100 |
| 30.00 | 0.010475 | 97.50 | 0.036002 |
| 40.00 | 0.012192 | 99.00 | 0.045022 |
| 50.00 | 0.013876 | 99.50 | 0.054278 |
| 60.00 | 0.015620 | 99.75 | 0.062687 |
| 70.00 | 0.017752 | 99.90 | 0.076355 |
| 80.00 | 0.020476 | | |

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Custom demographics 4: 7 - <11
All Seasons
All Regions
Sex: M/F-all/
All Races
Age-Low: 7 yrs High: 11 yrs

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Daily Exposure Analysis (mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.016963 | 0.016963 |
| Standard Deviation | 0.009573 | 0.009573 |
| Standard Error of mean | 0.000185 | 0.000185 |

Percent of Person-Days that are User-Days = 100.00%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.007496 | 90.00 | 0.027494 |
| 20.00 | 0.009838 | 95.00 | 0.034938 |
| 30.00 | 0.011950 | 97.50 | 0.041059 |
| 40.00 | 0.013499 | 99.00 | 0.050785 |
| 50.00 | 0.015090 | 99.50 | 0.064499 |
| 60.00 | 0.016983 | 99.75 | 0.071295 |
| 70.00 | 0.019627 | 99.90 | 0.074763 |
| 80.00 | 0.022463 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.007496 | 90.00 | 0.027494 |
| 20.00 | 0.009838 | 95.00 | 0.034938 |
| 30.00 | 0.011950 | 97.50 | 0.041059 |
| 40.00 | 0.013499 | 99.00 | 0.050785 |
| 50.00 | 0.015090 | 99.50 | 0.064499 |
| 60.00 | 0.016983 | 99.75 | 0.071295 |
| 70.00 | 0.019627 | 99.90 | 0.074763 |
| 80.00 | 0.022463 | | |

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Custom demographics 5: 11 - <14
All Seasons
All Regions
Sex: M/F-all/
All Races
Age-Low: 11 yrs   High: 14 yrs
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Daily Exposure Analysis (mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.018195 | 0.018195 |
| Standard Deviation | 0.010600 | 0.010600 |
| Standard Error of mean | 0.000262 | 0.000262 |

Percent of Person-Days that are User-Days = 100.00%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.007430 | 90.00 | 0.031142 |
| 20.00 | 0.009973 | 95.00 | 0.037161 |
| 30.00 | 0.011986 | 97.50 | 0.044191 |
| 40.00 | 0.014242 | 99.00 | 0.051720 |
| 50.00 | 0.016293 | 99.50 | 0.065023 |
| 60.00 | 0.018556 | 99.75 | 0.074234 |
| 70.00 | 0.021368 | 99.90 | 0.089794 |
| 80.00 | 0.024821 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.007430 | 90.00 | 0.031142 |
| 20.00 | 0.009973 | 95.00 | 0.037161 |
| 30.00 | 0.011986 | 97.50 | 0.044191 |
| 40.00 | 0.014242 | 99.00 | 0.051720 |
| 50.00 | 0.016293 | 99.50 | 0.065023 |
| 60.00 | 0.018556 | 99.75 | 0.074234 |
| 70.00 | 0.021368 | 99.90 | 0.089794 |
| 80.00 | 0.024821 | | |

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Custom demographics 6: 14+
All Seasons
All Regions
Sex: M/F-all/
All Races
Age-Low: 14 yrs High: 99 yrs

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Daily Exposure Analysis (mg/day)

| | per Capita | per User |
|---|---|---|
| Mean | 0.018672 | 0.018711 |
| Standard Deviation | 0.014970 | 0.014961 |
| Standard Error of mean | 0.000104 | 0.000104 |

Percent of Person-Days that are User-Days = 99.79%

Estimated percentile of user-days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.005648 | 90.00 | 0.035058 |
| 20.00 | 0.008329 | 95.00 | 0.045280 |
| 30.00 | 0.010533 | 97.50 | 0.057802 |
| 40.00 | 0.012727 | 99.00 | 0.074405 |
| 50.00 | 0.015106 | 99.50 | 0.087706 |
| 60.00 | 0.017760 | 99.75 | 0.110445 |
| 70.00 | 0.021331 | 99.90 | 0.135358 |
| 80.00 | 0.026257 | | |

Estimated percentile of per-capita days falling below calculated exposure in mg/day

| Percentile | Exposure | Percentile | Exposure |
|---|---|---|---|
| 10.00 | 0.005592 | 90.00 | 0.035028 |
| 20.00 | 0.008289 | 95.00 | 0.045247 |
| 30.00 | 0.010499 | 97.50 | 0.057763 |
| 40.00 | 0.012698 | 99.00 | 0.074354 |
| 50.00 | 0.015084 | 99.50 | 0.087587 |
| 60.00 | 0.017733 | 99.75 | 0.110395 |
| 70.00 | 0.021308 | 99.90 | 0.135312 |
| 80.00 | 0.026235 | | |