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US Food and Drug Administration's Total Diet Study: Dietary intake of perchlorate and iodine

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The US Food and Drug Administration (FDA) has conducted the Total Diet Study (TDS) since 1961, which designed to monitor the US food supply for chemical contaminants, nutritional elements, and toxic elements. Recently, perchlorate was analyzed in TDS samples. Perchlorate is used as an oxidizing agent in rocket propellant, is found in other items (e.g., explosives, road flares, fireworks, and car airbags), occurs naturally in some fertilizers, and may be generated under certain climatic conditions. It has been detected in surface and groundwater and in food. Perchlorate at high (e.g., pharmacological) doses can interfere with iodide uptake into the thyroid gland, disrupting its function. The National Academy of Sciences (NAS) has identified that "the fetuses of pregnant women who might have hypothyroidism or iodide deficiency as the most sensitive population." This study reports on intake estimates of perchlorate and iodine, a precursor to iodide, using the analytical results from the TDS. Estimated average perchlorate and iodine daily intakes as well as the contribution of specific food groups to total intakes were estimated for 14 age/sex subgroups of the US population. The estimated smallest lower bound to the largest upper bound average perchlorate intakes by the 14 age/sex groups range from 0.08 to 0.39 micrograms per kilogram body weight per day (μ g/kg bw/day), compared with the US Environmental Protection Agency (EPA) reference dose (RfD) of 0.7 μ g/kg bw/day. Infants and children demonstrated the highest estimated intakes of perchlorate on a body weight basis. The estimated average iodine intakes by the 14 age/sex groups reveal a lower bound (ND = 0) and upper bound (ND = LOD) range of average intakes from 138 to 353 μ g/person/day. Estimated average requirement (EAR).

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Introduction

For the last 46 years, the Total Diet Study (TDS) has been an important monitoring program that provides the US Food and Drug Administration (FDA) with baseline information on the levels of pesticide residues, chemical contaminants, radionuclides, nutrient elements, and toxic elements in the US food supply. The study involves retail purchases of foods representative of the "total diet" of the average US population, which includes baby food, beverages including bottled water, dairy, eggs, fat, oil, fruits, grains, legumes, mixtures, meat, poultry, fish, sweets, and vegetables. The study also includes the analysis of the foods for levels of specific analytes and estimation of dietary intake of those analytes by selected age/sex groups.

concerns regarding the levels of radioactive contamination in foods from atmospheric nuclear testing. Initially, the study estimated dietary intakes of two radionuclides (strontium-90 and cesium-137), several organochlorine and organophosphate pesticides, and selected nutrients by 16- to 19-year old male subjects (Pennington and Gunderson, 1987). Since 1961, the TDS has undergone many changes and refinements — expansion of the sample collection sites and the number of foods analyzed, addition of many analytes, improvement of analytical methods, and addition of population subgroups for which intakes are estimated (Pennington and Gunderson, 1987; Pennington et al., 1996). For a complete listing of various TDS publications and a more in-depth description of the history, please go to the following website: http://www.cfsan.fda.gov/~comm/tds-toc.html.

FDA began the TDS mainly in response to public health

The present assessment focuses on perchlorate and iodine, two of the many analytes studied in the TDS. In recent years, perchlorate and iodine have received a fair amount of attention in the scientific literature. Perchlorate is a chemical that is found to occur naturally in Chilean nitrate fertilizer, which has been used in the United States (Dasgupta et al., 2006). Perchlorate is also synthesized in the United States

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and used as an oxidizing agent in solid rocket propellant and found in other items (e.g., explosives, road flares, fireworks, car airbags, herbicides, and so on). Since the mid 1990s, the US Environmental Protection Agency (EPA), along with other government agencies, has sought to understand and assess the potential health effects of perchlorate levels in soil, groundwater, and drinking water around the country. In 2002, EPA, along with other federal agencies asked the National Academy of Sciences (NAS) to review the relevant scientific literature and key findings underlying EPA's 2002 Toxicological Review (NAS, 2005). In 2005, the NAS (NAS, 2005) advised EPA that a reference dose (RfD) of 0.0007 milligram per kilogram body weight per day (mg/kg bw/day), based on a no-observed-effects level of 0.007 mg/kg bw/day from a study by Greer et al. (2002), with the application of an uncertainty factor of 10 would protect the most sensitive population — the fetuses of pregnant women who might have hypothyroidism or iodide deficiency. The EPA accepted the NAS recommendations for the RfD (http://www.epa.gov/ iris/subst/1007.htm).

Perchlorate at high pharmacological doses (0.02, 0.1, and 0.5 mg/kg bw/day) interferes with iodide uptake into the thyroid gland and, if the inhibition is severe enough, can disrupt thyroid function. Disruption of iodine uptake may cause the thyroid to become enlarged (goiter), and, if the disruption continues, it may cause hypothyroidism. The NAS (2005) reviewed findings in regards to iodine intake and thyroid function, and the committee stated that, "Generally, thyroid hormone production is normal even when iodide intake is quite low. Hypothyroidism occurs only if daily iodide intake is below about 10 to 20 µg (about one-fifth to one tenth of the average intake in the United States). However, for pregnant women, iodide deficiency of that severity can result in major neurodevelopmental deficits and goiter in their offspring. Lesser degrees of iodide deficiency may also cause important neurodevelopmental deficits in infants and children."

Blount et al. (2007), focused on perchlorate exposure of 2820 US residents 6 years of age and older from the National Health and Nutrition Examination Survey (NHANES) during 2001–2002. All the participants were found to have detectable levels of perchlorate in their urine. From this work, Blount et al. were able to estimate a total daily perchlorate dose for adults 20 years of age and older. The total daily perchlorate dose was based on urinary perchlorate, urinary creatinine concentration, and physiological parameters predictive of creatinine excretion rates, which resulted in a median estimate of $0.064 \,\mu\text{g/kg} \,\text{bw/day}$ and 95th percentile of $0.234 \,\mu\text{g/kg} \,\text{bw/day}$.

In another study, Blount et al. (2006) focused on urinary perchlorate and thyroid hormone levels in 2299 men and women participants who were 12 years of age and older from NHANES during 2001–2002. The investigators evaluated the potential relationship between urinary levels

of perchlorate and serum levels of thyroid stimulating hormone (TSH) and total thyroxine (T4). The subjects were categorized and analyzed based on a cutoff point of 100 µg/l urinary iodine level. This value was based on the World Health Organization (WHO) definition of sufficient iodine intake in populations (WHO, 2004). Blount et al. observed that perchlorate was not a significant predictor of hormone levels for men. For women with urinary iodine levels $< 100 \,\mu \text{g/l}$, perchlorate was a significant negative predictor for T4 and a positive predictor of TSH. For women with urinary iodine levels $\geq 100 \,\mu\text{g/l}$, perchlorate was a significant positive predictor of TSH, but not T4. Blount concluded that the associations of perchlorate with T4 and TSH are coherent in direction and independent of other variables known to affect thyroid function, but are present at perchlorate exposure levels that were unanticipated based on previous studies. Finally, Blount et al. concluded that additional research is needed to affirm these findings.

The FDA recognizes the potential for perchlorate contamination in food through the use of some fertilizers, contaminated irrigation water, processing water, and source waters for bottle water. During 2004-2005, the FDA conducted exploratory surveys to monitor perchlorate levels in 28 types of foods and beverages consisting of bottled water, milk, fruits and fruit juices, vegetables, grain products, and seafood. The results of these exploratory surveys are found at FDA (2007), http://www.cfsan.fda.gov/~dms/clo4data.html. Since the results of these exploratory surveys focused on selected foods, the data do not provide information on the presence of perchlorate in the US food supply representing the total diet of the US population and are not included in this estimate. In 2005, FDA began testing all samples from the TDS to determine whether perchlorate is found in a broader range of foods. The TDS was determined to be an appropriate tool, since it includes all major components of the average American diet. In addition, because iodine has been analyzed in all TDS foods since late 2003, estimates of daily intakes of both perchlorate and iodine by the US population could be derived from the TDS results.

This study reports the estimated average dietary intakes of iodine based on analytical results from TDS samples collected between 2003 and 2004 and of perchlorate based on analytical results from TDS samples collected between 2005 and 2006. The total estimated daily intakes were calculated for 14 age/sex population groups from infants through adults. Also, the contributions of major food groups to total estimated intakes of iodine and perchlorate are reported.

Methods

Dietary intakes of perchlorate and iodine were estimated by combining analytical results from the TDS with food



consumption estimates developed specifically for estimating dietary exposure from TDS results (referred to as TDS diets).

Development of the TDS Food List and Diets

The following is a brief discussion of the methodology for developing both the TDS food list and diets; a more exhaustive explanation of the methodology is provided by Egan et al. (2007). The current TDS food list and diets were compiled in 2003 from the results of the US Department of Agriculture's 1994-1996, 1998 Continuing Survey of Food Intakes by Individuals (94-98 CSFII). For this survey, the data collection in 1994–1996 included individuals of all ages, and data collected in 1998 included children from birth through 9 years of age; the survey design allowed for all years of data to be combined for analysis. During the 94–98 CSFII, survey participants reported detailed consumption information on about 6000 different foods and beverages. For compiling the TDS food list, all 6000 survey foods were grouped (or aggregated) according to the similarity of their primary ingredients. Then average per capita (all individuals - eaters and noneaters alike) daily consumption amounts were calculated for each survey food, and, from each group of aggregated food codes, the food consumed in greatest was selected as the representative TDS food. In all, 285 foods and beverages were selected for the current TDS food list.

For compiling the TDS diets, the consumption amounts of all survey foods assigned to each TDS food were subtotaled to derive a TDS diet consumption amount for each TDS food. The complete set of TDS consumption amounts for each of the 14 age/sex groups is referred to collectively as the TDS diets. This approach to estimating dietary intakes assumes that the analytical profiles of the survey foods would be similar to those of the TDS foods to which they are assigned and that the TDS diets could, therefore, provide a reasonable estimate of total dietary exposure to the analytes from all foods in the diet — not from the TDS foods alone. The TDS diets do not account for consumption of water other than that used in the

preparation of foods or beverages (i.e., the diets do not include drinking water from the tap although bottled water, consumed as a beverage, is included in calculations presented here). Additionally, the TDS diet for infants 6–11 months does not include consumption of breast milk, thus breastfed infants would have different exposure patterns from the estimates shown in Table 5.

TDS Sample Collection and Analyses

Total Diet Study samples are routinely collected four times a year, once in each of the four regions of the country (west, north central, south, and northeast). Each round of sample collections and analyses is referred to as a market basket. For each market basket, samples of each of the 285 foods are collected simultaneously in three cities within the region. The foods are purchased at retail from grocery stores and fast-food restaurants and are then shipped from the collecting locations to FDA's Kansas City District Laboratory in Lenexa (KS, USA). The foods are prepared table-ready prior to analyses, and salt is not added to any of TDS food prepared by the laboratory. Distilled water is used for all food preparation (e.g., washing, cooking, and beverage preparation). For each of the 285 foods, the products purchased in each of the three cities within the collection region are composited to form a single analytical sample for each regional market basket.

The estimated intakes reported in this study are based on analytical results for TDS samples collected between 2003 and 2006. Iodine was analyzed in all TDS foods from five market baskets conducted in late 2003 through 2004. For perchlorate, 54 of 57 baby foods were analyzed in four market baskets conducted in 2005; the remaining three baby foods were analyzed in only three market baskets because they were not available in the fourth market basket for 2005. The other 228 TDS foods were analyzed in 2006; of those, 128 were analyzed in four market baskets and 100 were analyzed in two market baskets. The dates and locations of each market basket are listed in Table 1.

Table 1. Dates and locations of sample collections for iodine and perchlorate results.

Market basket	Sample collection dates	Collection region and locations	
2003-4	July 2003	North (Monmouth-Ocean City, NJ; Rochester, NY; Philadelphia, PA)	
2004-1	October 2003	Central (Chicago, IL; Youngstown-Warren, OH; Detroit, MI)	
2004-2	January 2004	West (Salt Lake City/Ogden, UT; Phoenix-Mesa, AZ; Las Vegas, NV)	
2004-3	April 2004	South (Atlanta, GA; San Antonia, TX; Shreveport-Bossier City, LA)	
2004-4	July 2004	North (Boston, MA; Syracuse, NY; Pittsburgh, PA)	
2005-1	October 2004	Central (Kalamazoo-Battle Creek, MI; Omaha, NE; St. Cloud, MN)	
2005-2	January 2005	West (Pueblo, CO; San Jose, CA; Boise City, ID)	
2005-3	April 2005	South (Roanoke, VA; West Palm Beach-Boca Raton, FL; New Orleans, LA)	
2005-4	July 2005	North (Hartford, CT; Bergen-Passaic, NJ; Binghamton, NY)	
2006-1	October 2005	Central (Rockford, IL; Cincinnati, OH; Fargo-Moorhead, ND)	
2006-2	January 2006	West (Los Angeles-Long Beach, CA; Santa Clara, CA; Seattle-Everett, WA)	
2006-3	April 2006	South (Raleigh, NC; Norfolk-Virginia Beach, VA; Tulsa, OK)	
2006-4	July 2006	North (Portland, ME; Nassau-Suffolk, NY; Scranton Wilkes-Barre, PA)	



Table 2. FDA analytical techniques and limits for iodine and perchlorate.

Chemical name	Analytical technique	Nominal analytical limits			
		Limit of detection	Limit of quantitation		
Iodine Perchlorate	UV-Vis IC-TMS	0.03 p.p.m.; for some up to 0.06 p.p.m. 1.00 p.p.b.	0.3 p.p.m., for some up to 0.6 p.p.m. 3.00 p.p.b.		

IC-TMS, ion chromatography-tandem mass spectrometry; UV-Vis, ultraviolet-visible spectrometry.

Table 3. Description of food groups contributing to intakes.

Food groups	Includes
Baby food	All baby foods and infant formulas (excluding adult foods consumed by children). Infant formulas were samples of ready-to-eat products
Beverages	Beverages, including bottled water, except for fruit/vegetable juices
Dairy	All dairy products (e.g., butter, milk, cheese, and ice cream)
Eggs	Boiled egg, scrambled egg, omelet, and egg salad
Fat/oil	Vegetable fats and oils, and salad dressings
Fruits	Fruits and fruit juices
Grains	Items that are primarily grains, including cookies and pastries
Legumes	Legumes, nuts, and seeds
Mixtures	Primarily entrée items containing mixtures of meat/poultry/fish, grains, and vegetables (no predominant ingredient)
Meat, poultry,	Items that are primarily meat, poultry, or fish (e.g., roasts, fried chicken, fish filets, and luncheon meats)
fish (MPF)	
Sweets	Sugars, sweeteners, syrups, candy, jelly, and gelatin
Vegetables	Vegetables and vegetable juices

Iodine was measured by FDA's Kansas City District Laboratory using a method adapted from Fischer et al. (1986). The method consists of a ternary acid digestion with a determination of iodine by UV-VIS spectrophotometry through the catalysis of the Ce + 4/As + 3 reaction. The method for perchlorate was developed by FDA in a collaborative effort among the Center for Food Safety and Applied Nutrition, the Southeastern Regional Laboratory, and the Total Diet Research Center; the method was published by Krynitsky et al. (2006). Table 2 reports the analytical techniques, the nominal limit of detection (LOD), and limit of quantitation (LOQ). Cases in which perchlorate and iodine were found to be present in concentrations greater than or equal to the LOD but less than the LOO were considered "trace" amounts. The LOD for perchlorate was $1.00 \,\mu\text{g/kg}$, while the LOD for iodine ranged from 0.03 to $0.06\,mg/kg$.

Calculation of TDS Dietary Intakes

In calculating estimated intakes, the average iodine concentration per food was calculated from results of five market baskets. For perchlorate, the average concentration per food was calculated from results of either two or four market baskets, as mentioned above. To account for uncertainties associated with samples with no detectable concentrations of perchlorate or iodine (non-detects or NDs), three average concentrations were calculated for

each TDS food assuming values of zero, half the LOD, and the LOD for non-detects. The three average concentrations in each food were then multiplied by the average daily consumption amount of the food for the given subpopulation group as compiled for the TDS diets to provide a range from lower bound (ND = 0) to upper bound (ND=LOD) estimated average intakes from each TDS food. Finally, estimated intakes from all TDS foods were summed to estimate the range of average total estimated daily intakes of iodine and perchlorate for each age/sex group. The estimated perchlorate intakes were compared with the EPA's RfD for perchlorate, and estimated iodine intakes were compared with the appropriate US Dietary Reference Intakes that represent average daily intake requirements (NAS, 2000). For the TDS age/sex groups other than infants, estimated iodine intakes were compared with the relevant estimated average requirements (EARs), which are defined by NAS as the nutrient intake levels estimated to meet the requirements of half the healthy individuals within a particular age/sex group. The estimated iodine intake by the TDS group of infants 6-11 months was compared with the adequate intake (AI) of 130 µg/person/ day (NAS, 2000); an AI is set by NAS when there is insufficient scientific evidence to determine an EAR and is defined as the recommended average daily intake level of a nutrient that is assumed to be adequate for a group of apparently health individuals.



The contributions of major food groups to total estimated intakes of perchlorate and iodine were also calculated. TDS foods were assigned to 1 of 12 major food groups; descriptions of these food groups are provided in Table 3, and a further rationale for TDS food assignment into the 12 major food groups are explained by Egan et al. (2007). The contributions of food groups to total estimated intake were calculated from the intake estimates based on average concentrations assuming values of half the LOD for non-detects. Contributions by food groups were determined by summing the estimated intakes from all TDS foods in each of the 12 food groups, and calculating the percentage of total intake for each food group.

Results

Perchlorate

From the TDS analytical results, it is evident that perchlorate is found in a wide range of foods. Detectable levels of perchlorate were found in 625 of 1065 (59%) of the total samples analyzed and 440 of 1065 (41%) of the samples had

no detectable levels of perchlorate. Of the 625 samples with detectable levels of perchlorate, 231 contained "trace" amounts (i.e., concentrations between the LOD and LOQ). As for findings in specific foods, detectable levels of perchlorate were found in at least one sample in 74% (211 of 285) of TDS foods. In contrast, perchlorate was not detected in any sample of 74 of 285 (26%) of TDS foods.

Estimated dietary intakes of perchlorate are reported in Tables 4 and 5. The percentage contributions to total estimated daily intake by food group are presented in Table 4. The majority (81%) of the estimated perchlorate intake by infants 6–11 months comes from baby foods, which includes infant formula, and dairy foods. Dairy foods contribute about half of the total estimated daily intake of perchlorate by children 2, 6, and 10 years of age. Vegetables and dairy foods combined account for between 46% and 59% of the total estimated intake of perchlorate by teenagers and adults.

Table 5 presents the lower and upper bound estimated average total daily intakes as well as intakes by food group on a per person basis. Total estimated daily intakes are also presented per kg of body weight to compare with EPA's RfD of $0.7 \,\mu\text{g/kg} \,\text{bw/day}$. Average body weights for each

Table 4. Contribution (%) by food groups to total estimated daily intake of perchlorate for 2005–2006.

Food group	Intake (% of total)								
	Infants 6–11 months	Children 2 years	Children 6 years	Children 10 years	Teenage girls 14–16 years	Teenage boys 14–16 years	Women 25–30 years		
Baby food	49	0	0	0	0	0	0		
Beverage	1	3	3	4	7	7	12		
Dairy	32	51	50	47	29	37	20		
Egg	0	0	0	0	0	0	0		
Fat/oil	0	0	0	0	0	0	0		
Fruit	4	15	11	9	11	7	8		
Grain	2	6	8	8	8	9	8		
Legume	0	0	0	0	0	0	0		
Mixture	6	8	9	10	14	12	14		
MPF	1	4	6	5	7	7	11		
Sweets	0	1	1	1	1	1	1		
Vegetable	5	12	12	16	23	20	26		
	Men 25–30 years	Women 40–45 years	Men 40–45 years	Women 60–65 years	Men 60–65 years	Women 70 + years	Men 70+ years		
Baby food	0	0	0	0	0	0	0		
Beverage	12	12	11	9	9	6	7		
Dairy	20	17	21	17	19	23	22		
Egg	0	0	0	0	0	0	0		
Fat/oil	0	0	0	0	0	0	0		
Fruit	5	11	8	12	9	12	12		
Grain	8	8	9	8	8	8	9		
Legume	0	0	0	0	0	0	0		
Mixture	16	13	13	9	10	10	10		
MPF	9	7	8	7	8	5	7		
Sweets	0	1	1	0	0	0	0		
Vegetable	30	31	29	38	37	36	33		

MPF, meat, poultry, fish.



Table 5. Range of estimated lower and upper bound average perchlorate intakes for 2005–2006.

Food group			Iı	ntake (µg/person/o	lay)		
	Infants 6–11 month	Children 2 years	Children 6 years	Children 10 years	Teenage girls 14–16 years	Teenage boys 14–16 years	Women 25–30 years
Baby food	1.1–1.3	0.0-0.0	0.0-0.0	0.0-0.0	0.0-0.0	0.0-0.0	0.0-0.0
Beverage	0.00-0.1	0.0-0.3	0.0 – 0.4	0.0-0.5	0.02 - 0.8	0.0 - 1.1	0.2 - 1.2
Dairy	0.8 – 0.8	2.6-2.6	2.9-2.9	3.1-3.1	1.6-1.6	3.1-3.1	1.2-1.2
Egg	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0
Fat/oil	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0
Fruit	0.1 - 0.1	0.7 - 0.9	0.6-0.7	0.5-0.6	0.6 - 0.7	0.5-0.6	0.5 - 0.6
Grain	0.0-0.1	0.3 - 0.3	0.4-0.5	0.5-0.5	0.4-0.5	0.7 - 0.8	0.4-0.5
Legume	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0
Mixture	0.1 - 0.1	0.4-0.5	0.5-0.6	0.6 - 0.7	0.8 – 0.8	1.0-1.1	0.9-0.9
MPF	0.0 – 0.0	0.2 - 0.2	0.3 - 0.3	0.3 - 0.4	0.3-0.4	0.5 - 0.6	0.7 - 0.7
Sweets	0.0 – 0.0	0.0 – 0.0	0.0-0.1	0.0-0.1	0.0-0.1	0.0-0.1	0.0-0.1
Vegetable	0.1 - 0.1	0.6 – 0.6	0.7 - 0.7	1.0-1.0	1.2-1.3	1.7 - 1.7	1.5-1.5
Total intake	2.4-2.7	4.9-5.5	5.4-6.1	6.1-6.9	5.1-6.1	7.7–9.1	5.4-6.8
Total intake (µg/kg bw/day)	0.26-0.29	0.35-0.39	0.25-0.28	0.17-0.20	0.09-0.11	0.12-0.14	0.09-0.11
	Men 25–30 years	Women 40–45 years	Men 40–45 years	Women 60–65 years	Men 60–65 years	Women 70 + years	Men 70+ years
Baby food	0.0-0.0	0.0-0.0	0.0-0.0	0.0-0.0	0.0-0.0	0.0-0.0	0.0-0.0
Beverage	0.2 - 1.6	0.3 - 1.3	0.2 - 1.7	0.2 - 1.0	0.2 - 1.3	0.1 - 0.7	0.1 - 0.9
Dairy	1.5-1.5	1.1 - 1.1	1.8-1.8	1.1 - 1.1	1.5-1.5	1.4-1.4	1.7 - 1.7
Egg	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0
Fat/oil	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0
Fruit	0.3 - 0.4	0.7 - 0.8	0.6 – 0.7	0.7 - 0.8	0.6 – 0.8	0.7 - 0.8	0.8 - 1.0
Grain	0.6 – 0.7	0.5 - 0.6	0.7 - 0.8	0.5 - 0.5	0.6 - 0.7	0.5 - 0.6	0.6 - 0.7
Legume	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0
Mixture	1.2-1.3	0.8 – 0.9	1.1-1.1	0.6 – 0.6	0.8 – 0.9	0.6 - 0.6	0.7 - 0.8
MPF	0.7 – 0.7	0.5-0.5	0.6 – 0.7	0.4-0.5	0.6 - 0.7	0.3 - 0.4	0.5 - 0.6
Sweets	0.0 – 0.0	0.0 – 0.0	0.1 - 0.1	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0	0.0 – 0.0
Vegetable	2.2 - 2.2	1.9-2.0	2.4-2.4	2.4-2.4	2.8 - 2.9	2.2 - 2.2	2.5-2.5
Total intake	6.7-8.6	5.9-7.3	7.4–9.4	5.9-7.1	7.2-8.8	5.8-6.9	7.1-8.3
Total intake (µg/kg bw/day)	0.08-0.11	0.09-0.11	0.09-0.11	0.09-0.10	0.09-0.11	0.09-0.11	0.11-0.12

MPF, meat, poultry, fish.

The total intake for a specific age/sex group are provided in bold.

population group were based on self-reported body weights from respondents in the 94–98 CSFII (Egan et al., 2007). Estimated perchlorate intakes by all age/sex groups are below the RfD. Children 2 years of age, with estimated lower and upper bound average intakes ranging from 0.35 to 0.39 μ g/kg bw/day, have the highest total perchlorate intake per kg body weight per day. Total lower- and upper bound average intake ranges for infants 6–11 months, and children 6–10 years of age are estimated to be 0.26 to 0.29 μ g/kg bw/day, 0.25 to 0.28 μ g/kg bw/day, and 0.17 to 0.20 μ g/kg bw/day, respectively. The estimated smallest lower bound and the highest upper bound average intakes by the other age/sex groups ranged from 0.08 to 0.14 μ g/kg bw/day.

Iodine

From the TDS analytical results, it is evident that iodine is found in more than half the foods in the TDS. Detectable levels of iodine were found in at least one sample of 169 of

285 (59%) of the TDS foods, while iodine was not detected in 116 of 285 or 41% of TDS foods.

The percentage contributions by food group to total estimated daily intake of iodine are reported in Table 6. As with perchlorate, baby foods and dairy products account for nearly all (90%) of the estimated iodine intake by infants. Dairy products account for 70% or more of total estimated daily intake of iodine by children 2, 6, and 10 years of age, and 63% of total estimated iodine intake by teenage boys. For all other age/sex groups, dairy foods contribute about 50% of total estimated iodine intake. For children 2, 6, and 10 years of age, grains account for 10%, 14%, and 15%, respectively, of the total estimated daily iodine intake. Grain products contribute between 16% and 23% of total estimated iodine intake for teenagers and adults.

Table 7 reports the lower bound (ND=0) and upper bound (ND=LOD) estimates of average iodine intakes as well as intakes by food group on a per person basis.

Table 6. Contribution (%) by food group to total estimated daily intake of iodine for 2003–2004.

Food group	Intake (% of total)									
	Infants 6–11 months	Children 2 years	Children 6 years	Children 10 years	Girls 14–16 years	Boys 14–16 years	Women 25–30 years			
Baby food	56	0	0	0	0	0	0			
Beverage	1	2	2	3	6	5	9			
Dairy	34	73	70	70	53	63	49			
Egg	2	3	2	2	2	2	4			
Fat/oil	0	0	0	0	0	0	0			
Fruit	2	5	3	2	4	3	3			
Grain	3	10	14	15	20	16	20			
Legume	0	0	0	0	0	0	0			
Mixture	1	4	5	5	8	7	8			
MPF	0	1	2	1	3	2	3			
Sweets	0	1	1	1	2	1	2			
Vegetable	1	1	1	1	2	1	2			
	Men 25–30 years	Women 40–45 years	Men 40–45 years	Women 60–65 years	Men 60–65 years	Women 70 + years	Men 70+ years			
Baby food	0	0	0	0	0	0	0			
Beverage	10	10	9	9	8	6	6			
Dairy	45	47	51	48	48	57	57			
Egg	4	3	3	4	5	4	4			
Fat/oil	0	0	0	0	0	0	0			
Fruit	3	3	2	4	3	4	3			
Grain	21	23	21	21	21	18	18			
Legume	0	0	0	0	0	0	0			
Mixture	11	8	7	6	7	5	5			
MPF	3	3	3	4	4	3	4			
Sweets	1	1	2	1	1	1	1			
Vegetable	2	2	2	3	3	2	2			

MPF, meat, poultry, fish.

Estimated intakes are compared to the AI or EAR relevant to the TDS population group. The lower bound (ND=0)total estimated iodine intake by infants of $144 \mu g/person/day$ exceeds their AI for iodine (130 µg/person/day). The lower bound (ND = 0) daily estimated intakes of iodine by children are as follows: 225 µg/person/day for children 2 years, 255 µg/person/day for children 6 years, and 276 µg/person/day for children 10 years. These estimated intakes exceed the relevant EARs of 65 μg/person/day for children 1 through 8 years of age and 73 μ g/person/day for children 9 through 13 years of age.

For teenage boys and girls aged 14-16 years, dairy and grain provide the highest sources of dietary iodine. These two food groups contribute 73% of total estimated intake by teenage girls and 79% of total estimated intake by teenage boys (Table 6). Teenage boys have the highest total daily estimated intake of iodine (304 to 353 µg/person/day) in comparison with the all other age/sex groups in the TDS (Table 7). Their lower bound (ND=0) estimated iodine intake is three times their EAR of 95 μ g/person/day. Like the teenage boys, the teenage girls' estimated dietary intake of iodine of 178 to 214 µg/person/day exceeds their EAR, which is also $95 \,\mu g/person/day$.

For adults, dairy and grain provided the most significant sources of dietary iodine for all groups of adults (Table 6). The total estimated lower and upper bound average intakes by women 25–30 years of age range from 148 to $196 \,\mu g$ / person/day; for women 40–45 years of age, estimated intakes range from 145 to $197 \mu g/person/day$ (Table 7). For adult men 25-30 and 40-45 years of age, estimated iodine intakes range from 203 μ g/person/day at the lower bound to 284 μ g/ person/day at the upper bound.

Finally, for older (60–65 and 70+ years of age) women and men, their main sources of dietary iodine are dairy and grains (Table 6). These foods account for between 69% and 75% of their total estimated daily intake. Total estimated lower and upper bound average intakes by women 60-65 years of age range from 138 to $182 \mu g/person/day$ (Table 7). Women 70 + years of age have an estimated iodine intake ranging from 154 to 192 μg/person/day. Estimated lower and upper bound average iodine intakes by both groups of older men range from 192 to 249 µg/person/day for men



Table 7. Range of estimated lower and upper bound average iodine intakes for 2003–2004.

Food group			Inta	ake (µg/person/d	ay)		
	Infants 6–11 months	Children 2 years	Children 6 years	Children 10 years	Girls 14–16 years	Boys 14–16 years	Women 25–30 years
Baby food	82.8-88.3	1.1–1.2	0.0-0.0	0.0-0.0	0.0-0.0	0.0-0.0	0.0-0.0
Beverage	0.0 - 1.8	0.0 - 7.6	0.1 - 11.3	0.0 - 14.6	0.1 - 22.9	0.0 - 31.2	0.2 - 32.1
Dairy	50.8-50.8	173.9-173.9	187.9-188.0	202.5-202.6	106.0-106.0	207.9-207.9	83.2-83.2
Egg	2.5-2.5	7.1 - 7.1	5.1-5.1	5.5-5.5	4.4-4.4	5.9-5.9	6.0-6.0
Fat/oil	0.0 – 0.0	0.1 - 0.2	0.2 - 0.3	0.3 - 0.4	0.5 - 0.6	0.5 - 0.7	0.6 - 0.7
Fruit	1.6-3.1	7.9-13.8	5.8-9.7	5.4-8.7	6.3-9.3	7.1 - 10.1	4.2 - 7.4
Grain	3.6-4.1	21.5-23.5	37.1-39.5	41.6-43.9	37.5-39.7	49.6-52.6	32.3-34.8
Legume	0.0-0.1	0.1 - 0.4	0.2 - 0.5	0.2 - 0.5	0.1 - 0.6	0.2 - 0.6	0.2 - 0.6
Mixture	1.9-2.5	8.1 - 10.0	11.2-13.3	12.3-14.4	14.6-16.9	22.5-25.8	12.6-15.8
MPF	0.5-0.6	2.9-4.0	4.0-5.3	3.4-5.1	4.2-6.0	5.2-7.6	4.6-6.4
Sweets	0.0-0.1	1.6-2.0	2.7-3.4	3.1-4.0	2.7-3.3	2.7-3.5	2.8 - 3.2
Vegetable	0.6-1.1	1.0-3.1	1.2-3.9	1.3-4.6	1.2-4.7	2.3-7.1	1.4-5.8
Total intake	144-155	225-247	255-280	276-304	178-214	304-353	148-196
Estimated average requirement (EAR) ^a	130 (AI)	65	65	73	95	95	95
	Men	Women	Men	Women	Men	Women	Men
	25-30 years	40-45 years	40-45 years	60–65 years	60–65 years	70 + years	70 + years
Baby food	0.0-0.0	0.0-0.0	0.0-0.0	0.0-0.0	0.0-0.0	0.0-0.0	0.0-0.0
Beverage	0.1 - 45.5	0.2 - 35.5	0.1 - 45.7	0.4 - 27.7	0.1 - 36.2	0.2 - 20.9	0.1 - 24.9
Dairy	105.2-105.3	79.1 - 79.2	125.0-125.1	76.9-77.0	105.7-105.7	96.9-97.0	123.7-123.8
Egg	9.9–9.9	5.5-5.5	8.2 - 8.2	7.1 - 7.1	11.4-11.4	6.7 - 6.7	8.6-8.6
Fat/oil	0.7 – 0.9	0.7 - 1.0	1.0-1.3	0.6 – 0.9	0.7 - 1.0	0.5 - 0.7	0.6 – 0.8
Fruit	6.6-9.2	2.9-6.3	4.0 - 7.5	3.6-7.7	3.9-8.1	4.5-9.0	4.5-9.8
Grain	47.2-50.2	36.2-38.6	50.9-54.0	33.3-35.5	45.2-48.2	29.9-32.4	37.6-40.8
Legume	0.4 - 1.1	0.2 - 0.6	0.3 - 0.9	0.1 - 0.5	0.3 - 0.9	0.1 - 0.5	0.2 - 0.8
Mixture	22.9-26.7	12.8-15.5	15.9-19.7	7.8 - 10.4	13.3-16.9	7.8 - 10.6	9.7 - 13.0
MPF	6.1 - 9.0	4.0-6.0	5.6-8.7	5.6-7.5	7.2-9.9	4.6-6.2	7.0-9.0
Sweets	1.9-2.2	2.0-2.5	3.7-4.3	0.9 - 1.5	1.6-2.2	0.9 - 1.4	1.4-2.0
Vegetable	2.0-8.0	1.4-6.3	2.3-8.5	1.6-6.6	2.4-8.7	1.8-6.6	2.2 - 7.9
Total intake	203-268	145-197	217-284	138-182	192-249	154-192	196-241
Estimated average requirement (EAR) ^a	95	95	95	95	95	95	95

AI, adequate intake; MPF, meat, poultry, fish.

60–65 years of age and 196 to $241 \,\mu\text{g/person/day}$ for men 70 + years of age. Estimated lower bound (ND = 0) average intakes by all groups of men and women exceed the EAR for adults of $95 \,\mu\text{g/person/day}$.

Discussion

This assessment provides information on major dietary sources and estimated average dietary intakes of perchlorate and iodine in the United States. Intakes estimated from the TDS diets are based on average per capita food consumption; that is, the TDS diets reflect the average amounts of foods consumed by all individuals (eater and noneaters alike) within each of the 14 age/sex groups. However, the TDS as currently designed does not allow for estimating intakes at

the extremes (i.e., upper or lower percentiles of food consumption) or for population subgroups within the 14 age/sex groups that may have specific nutritional needs (e.g., the subgroups of pregnant or lactating women within the groups of women of childbearing age). Given the increased caloric needs of these two groups of women, their perchlorate and iodine intakes are likely to be somewhat higher than those of women of childbearing age as a whole as represented by the TDS population groups. We also note that children 2 years of age are estimated to consume iodine at levels that exceed the tolerable upper limit. Nevertheless, the results of this estimated dietary intake assessment of iodine and perchlorate provides a general estimation of the average iodine and perchlorate intakes by specific age/sex groups in the United States.

^aTaken from National Academy of Sciences, Dietary Reference Intake for Vitamin A, Vitamin K, Arsenic, Boron, Chromium, Copper, Iodine, Iron, Manganese, Molybdenum, Nickel, Silicon, Vanadium, and Zinc, National Academies Press, Washington, D.C., 2000.

The total intake for a specific age/sex group are provided in bold.



The perchlorate intake estimates reveal that infants and children (2, 6, and 10 years) have the highest estimated intake on a body weight basis in comparison to other TDS age/sex groups, because they consume more food per their body weight and they have different food consumption patterns. Children 2 years of age have the highest estimated average perchlorate intake ranging from 0.35 to 0.39 μ g/kg bw/day, which is between 50% and 56% of the EPA RfD, with dairy foods providing about 51% of perchlorate in their diet. The estimated lower and upper bound average perchlorate intakes by infants 6-11 months and children 6 years of age range from 0.26 to 0.29 and 0.25 to 0.28 μ g/kg bw/day, respectively. The infants' estimated perchlorate intake range is 37% to 41% of EPA's RfD of $0.7 \mu g/kg$ body weight per day, with dairy foods providing 32% of their total estimated intake of perchlorate. For children 6 years of age, the estimated average range of perchlorate intake is between 36% and 40% of the EPA's RfD. Children 10 years of age had estimated lower and upper bound average perchlorate intakes of 0.17 to $0.20 \,\mu g/kg \,bw/day$, which is between 24% and 29% of the RfD.

For teenage girls 14–16 years, women 25–30 years of age, and women 40–45 years of age had the same estimated average perchlorate intake ranges of 0.09 to $0.11\,\mu\mathrm{g/kg}\,\mathrm{bw/day}$, respectively. For these three age groups (teenage girls 14–16 years of age, women 25–30 years of age, and women 40–45 years) had estimated average range of perchlorate intakes between 13% and 16% of the EPA's RfD.

The remaining seven age/sex groups displayed estimated perchlorate intakes from the smallest lower bound of 0.08 to the highest upper bound of 0.14 μ g/kg bw/day, which is between 11% and 20% of the EPA's RfD. The lower bound (ND=0) range of estimated average perchlorate intakes for eight age/sex group that consist of men and women over 20 years of age (0.08 to 0.11 μ g/kg bw/day) show relative agreement with Blount et al. (2007) median estimated perchlorate dose of 0.064 μ g/kg bw/day.

It could be assumed that perchlorate would be found mainly in foods with high moisture content (e.g., milk and vegetables) because of its affinity for water, but results of the TDS analyses appear to indicate that perchlorate is more widely distributed in the food supply. As noted, detectable levels of perchlorate were found in 74% of the 285 TDS food. Since this assessment is based on a small number of composite samples (two or four) per TDS food, FDA plans to continue analyzing the full range of TDS foods for perchlorate in the future to develop a more robust data set on perchlorate levels in foods.

Perchlorate and iodine levels in selected foods have been reported previously in the literature (Pearce et al., 2004; Jackson et al., 2005; Kirk et al., 2005; Sanchez et al., 2005a, b; Sanchez et al., 2006). In addition, FDA conducted exploratory surveys in 2004 and 2005 to determine perchlorate levels in selected foods. Table 8 compares the

perchlorate concentrations in 10 commodities reported elsewhere with the levels found in similar TDS foods. Perchlorate results show fairly good agreement for 5 of the 10 commodities (milk, iceberg lettuce, green leaf lettuce, oranges, and grapefruit). For the other commodities (spinach, collards, cucumbers, tomatoes, and cantaloupe), perchlorate results varied considerably. Table 9 compares iodine concentrations for three foods as reported in the literature to findings in similar TDS foods. The iodine

Table 8. Perchlorate levels in selected foods.

Perchlorate levels		oncentration–we weight (μg/kg)	et
Commodity	n samples	Mean ^a	Source
Milk	47	2	Kirk et al. (2005)
	125	5.8	FDA exploratory samples
	8	7	FDA TDS
Lettuce, iceberg	63	7.4	Sanchez et al. (2005a)
	24	8	Sanchez et al. (2005b)
	43	8.1	FDA exploratory samples
	4	2.1	FDA TDS
Lettuce, green leaf	f 69	16.5	Sanchez et al. (2005a)
	24	33	Sanchez et al. (2005b)
	26	10.6	FDA exploratory samples
	2	4.4	FDA TDS
Spinach	10	85.1	Sanchez et al. (2005a)
	36	115	FDA exploratory samples
	4	40	FDA TDS
Collards	1	5	Sanchez et al. (2005a)
	13	95.1	FDA exploratory samples
	4	17.7	FDA TDS
Cucumbers	1	40	Jackson et al. (2005)
	1	770	Jackson et al. (2005)
	20	6.6	FDA exploratory samples
	4	19.1	FDA TDS
Tomatoes	1	42	Jackson et al. (2005)
	1	220	Jackson et al. (2005)
	73	13.6	FDA exploratory samples
	4	78	FDA TDS
Cantaloupe	1	1600	Jackson et al. (2005)
	48	28.6	FDA exploratory samples
	4	24.4	FDA TDS
Oranges	28	7.4	Sanchez et al. (2006)
	10	3.4	FDA exploratory samples
	4	2.7	FDA TDS
Grapefruit	15	3.3	Sanchez et al. (2006)
	4	0.5	FDA TDS

LOD, limit of detection; ND, non-detect.

^aMean for FDA samples are based on ND = 1/2LOD.



Table 9. Iodine levels in selected foods.

Iodine levels		Concentration—wet weight $(\mu g/kg)$		
Commodity	n samples	Mean	Source	
Milk	47	89.2	Kirk et al. (2005)	
	18	464	Pearce et al. (2004)	
	20	417	FDA TDS	
Infant formula	8	159	Pearce et al. (2004)	
	15	136	FDA TDS	
Bread	17	334	Pearce et al. (2004)	
	25	312	FDA TDS	

concentrations in milk reported by Kirk et al. (2005) were considerably lower than either the TDS samples or those reported by Pearce et al. (2004), but TDS iodine levels in infant formula and bread were consistent with those reported in the literature.

These TDS results increase substantially the available data for characterizing dietary exposure to perchlorate and provide a useful basis for the beginning to evaluate overall perchlorate and iodine estimated dietary intakes in the US population. The next major step is to analyze future TDS market baskets for perchlorate and iodine. More robust data sets will provide a clearer picture of estimated perchlorate and iodine intakes using not only the TDS approach to estimating intakes but also by using the analytical results from the TDS with detailed consumption data from the CSFII or NHANES surveys. Targeting the food consumption patterns based upon results from these surveys could provide an estimate of the distribution of iodine and perchlorate intakes by women of childbearing age who are pregnant and/or lactating. Data from these surveys could also be combined to develop an estimate of iodine and perchlorate intakes specifically for pregnant and lactating women, which could provide more information about the potential for perchlorate inhibition of iodide uptake by the thyroid to occur in this population subgroup.

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