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Exposure apportionment: Ranking food items by their contribution to dietary exposure†

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This paper identifies and ranks food items by estimating their contribution to the dietary exposure of the US population and 19 subpopulation groups. Contributions to dietary exposures to arsenic, cadmium, chromium, lead, nickel, benzene, chlorpyrifos, and diazinon are estimated using either the Dietary Exposure Potential Model (DEPM) approach, the National Human Exposure Assessment Survey Arizona (NHEXAS-AZ) approach or the combination of the two. The DEPM is a computer model that uses several national databases of food consumption and residue concentrations for estimating dietary. The DEPM approach ranks the contribution of food items to the total dietary exposure using two methods, the *direct method* that ranks contributions by population exposure magnitude and the *weighted method* that ranks by subpopulation exposure magnitude. The DEPM approach identifies highly exposed subpopulations and a relatively small number of food items contributing the most to dietary exposure. The NHEXAS-AZ approach uses the NHEXAS-AZ database containing food consumption data for each subject and chemical residues of a composite of food items consumed by each subject in 1 day during the sampling week. These data are then modeled to obtain estimates of dietary exposure to chemical residues. The third approach uses the NHEXAS-AZ consumption data with residue values from the national residue database. This approach also estimates percent contributions to exposure of each ranked food item for the Arizona population. Dietary exposures estimated using the three approaches are compared. The DEPM results indicate groups with highest dietary exposures include Nonnursing Infants, Children 1–6, Hispanic, Non-Hispanic White, Western, Northeast and Poverty 0–130%. The use of the Combined National Residue Database (CNRD) identifies 43 food items as primary contributors to total dietary exposure; they contribute a minimum of 68% of the total dietary exposure to each of the eight chemical residues. The percent contribution of ranked food items estimated using the NHEXAS samples is smaller than those obtained from the western US population *via* the DEPM. This indicates differences in consumption characteristics of the two groups with respect to the ranked food items. Six of 15 food items consumed by the NHEXAS-AZ subjects per day are ranked food items contributing between 56% and 70% of the estimated NHEXAS-AZ dietary exposure to each of the eight chemical residues. The difference between total dietary exposure estimates from the DEPM and NHEXAS-AZ approaches varies by chemical residue and is attributable to differences in sampling and analytical methods, and geographic areas represented by the data. Most metal exposures estimated using the NHEXAS consumption data with the CNRD have lower values than those estimated *via* the other approaches, possibly because the NHEXAS-AZ residue values are higher than the CNRD values. In addition, exposure estimates are seemingly affected by the difference in demographic characteristics and factors that affect types and amounts of food consumed. Efficient control strategies for reducing dietary exposure to chemical residues may be designed by focusing on the relatively small number of food items having similar ingredients that contribute substantively to the total ingestion exposure.
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Introduction

In the course of daily activities, people are routinely exposed to a variety of environmental chemicals through various media and pathways of exposure including air (inhalation), water and food (ingestion), and surfaces (dermal absorption). Food items, used here to denote both solid and liquid food items, may become contaminated through contact with chemical agents during growing, harvesting, processing, distribution, preparation, and eating. Sources of the chemical residues remaining on ingested foods include insecticide and herbicide applications in the

1. Abbreviations: NHEXAS, National Human Exposure Assessment Survey; DEPM, Dietary Exposure Potential Model; CSFII, Continuing Survey of Food Intake by Individual; CNRD, Combined National Residue Database; TDS, Total Diet Study; FDA, Food and Drug Administration; USDA, US Department of Agriculture; ECF, exposure core food

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field or garden, irrigation with contaminated water, and contact with soil or water that contains toxic compounds and industrial chemicals including toxic metals and volatile organic compounds (Berry, 1992).

The two components of estimating dietary exposure are frequency of consumption and chemical residue levels of food items. Several national surveys produce data useful for this purpose and those relevant to this work are discussed. Information on food consumption is available from the Continuing Survey of Food Intake by Individual (CSFII), a periodic national consumption survey sponsored by the US Department of Agriculture (USDA). Surveys on food residues are carried out routinely for regulatory monitoring of the nation's food supply. Data from the following seven national databases were used in this study:

(1) the 1982–1994 Total Diet Study (TDS), an ongoing monitoring program of normally 264 ready-to-eat foods conducted three times annually by the Food and Drug Administration (FDA);
(2) the 1987–1994 Pesticide Residue Information System, a database of residues primarily from state monitoring programs;
(3) the 1986–1993 California Pesticide Monitoring Database conducted by the state;
(4) FDA's Compliance and Surveillance Monitoring Program for 1992–1994;
(5) the 1995 monitoring data collected by the US Fish and Wildlife Service;
(6) USDA's 1990–1995 Microbiological and Residue Computer Information System; and
(7) the 1994 data from USDA's Pesticide Data Program, initiated in 1994 to collect information suitable for risk assessments on pesticides from foods.

In addition to these national consumption and residue databases, consumption information obtained from questionnaires of the National Human Exposure Assessment Survey Arizona (NHEXAS-AZ) exposure study (Lebowitz et al., 1995) conducted from 1995 to 1997 is used in this paper. NHEXAS is an EPA-funded federal interagency effort. It is designed to document the occurrence, distribution, and determinants of exposure to classes of chemicals of the US population; to develop strategies for preventing or reducing such exposures and communicate information; and to provide data on population exposures to all involved in assessment and management of environmental concerns. The NHEXAS-AZ study is a population-based multistage survey based on a sample of 1225 residences. The NHEXAS-AZ study includes consumption information from a dietary survey component of 300 respondents, one respondent per residence, and the chemical residue content of a composite sample containing food items consumed during one sampling day by each respondent.

Prior studies have shown that different population groups have different dietary exposure and few food items contribute a high portion of the total food residue intake for example, Pennington and Gunderson (1987). The premise of this study is that the identification of highly exposed population groups and the ranking of food items will assist in reducing risks associated with dietary exposures. Specific objectives of this paper are as follows.

(1) To identify food items that contribute high portions to the dietary exposure to study subject chemical residues.
(2) To identify subpopulation groups most highly exposed to each of the eight target chemicals.
(3) To examine the difference among exposure values obtained using exclusively the extant national consumption and residue data, corresponding values obtained using solely the NHEXAS-AZ directly measured exposure data, and corresponding values obtained using the NHEXAS-AZ data with the national residue data, a combination of the two approaches.
(4) To estimate and compare contributions of ranked food items to dietary exposure using different estimation approaches.

Methods

In this study, dietary exposure is estimated for eight chemical residues: arsenic, cadmium, chromium, lead, nickel, benzene, chlorpyrifos, and diazinon. They are a subset of the target chemical residues of the NHEXAS-AZ study. Ingestion exposure was estimated using three approaches. The first approach uses the Dietary Exposure Potential Model (DEPM) (Berry and Tomerlin, 1996; Tomerlin et al., 1997), a computer software model and food database system, to estimate dietary exposure to chemical residues. Using pertinent information from the DEPM national databases mentioned above, this approach identifies subpopulations with the highest potential dietary exposure to each chemical residue. Within this approach, we developed two methods for ranking the contribution of individual food items to the dietary exposure. From the ranked food items, we selected a small number of food items that contribute a large portion of the dietary exposure to each of the eight target residues. The percent contribution of these top-ranked food items was calculated. The second approach uses a deterministic exposure model with the NHEXAS-AZ questionnaire and sampling data to estimate the dietary exposure to the same target residues and compare it with corresponding results from the first approach. The two approaches result in independent estimates of dietary exposure to the chemical residues.

The third approach is a combination of the first two. It uses the NHEXAS-AZ consumption data from questionnaires and an exposure model similar to that of the second approach, but uses residue values from the national residue database, as in the first approach.

Exposure Estimation Using DEPM

The DEPM is used in this study as the first approach for estimating dietary exposure. The main attribute of the DEPM is its use of exposure principles to link consumption survey data with the residue information. To calculate exposure, DEPM allows user selection of a national food consumption survey, a national residue database and chemical residue, and defined target populations (Tomerlin et al., 1997). The food items in DEPM are based on 11 food groups: (1) Beverages, (2) Candies/Sugar Products, (3) Dairy/Egg Products, (4) Fruits, (5) Grains/Grain Products, (6) Infant Foods, (7) Legumes/Nuts, (8) Meat/Poultry, (9) Fish/Seafood, (10) Vegetables, and (11) Miscellaneous. These groups contain approximately 800 exposure core foods (ECFs), established from over 6500 common food items identified in the CSFII. A core food consists of one or more common foods with similar major ingredients. For example, the ECF "Wheat Flour with Apples, Pies, and Cobbler (G119)" consists of apple pies, cobblers, crisps, fritters, strudels, dumplings, and coffee cakes—foods that have apples, sugar, wheat flour, and water as major ingredients. Recipe files for each core food item link food consumption and chemical residue information. Consumption characteristics of 19 subpopulation groups of the US population are structured in the DEPM into four demographic categories: age/gender, ethnicity, geographic region, and family income. However, the 19 population groups are not inclusive; e.g., selecting Nonnursing Infants and Hispanic results in two distinct groups, not a single group of Hispanic Nonnursing Infants. In addition, it is important to note that chemical residue values are not specific to demographic characteristics, i.e., demographic data are specific to consumption only. Using the selected consumption and chemical residue database, the DEPM can calculate dietary exposure to selected chemicals for each population group. DEPM exposure estimates are estimated in micrograms of chemical residue per kilogram body weight per day. Mean values of consumption and chemical residues for the ECFs are used in the estimates of exposure. Residue data reported as below detection limit can be assigned a value of zero or one-half the detection limit for exposure estimates. In this study, values reported below the detection limit were assigned a value of zero. There are three levels of DEPM dietary exposure estimation. Level I estimates total exposure to each selected chemical residue from consumption of all selected food items included in the model. Any or all food groups and items may be used in the estimation. Level II exposure estimates signify a total exposure estimate to one target residue from consumption of each of the 11 core food groups. Level III exposure values compute exposures to one target chemical residue from consumption of individual ECFs.

There are several consumption databases; the one selected for evaluation in this paper is the CSFII of 1994–1996, the latest available consumption database included in the DEPM version 3.3.2 (DEPM, 2000) used in this study. DEPM also contains several residue databases that may be used independently or collectively to estimate exposures to target chemical residues. For this analysis, a residue database, termed the Combined National Residue Database (CNRD), was used to estimate dietary exposures. The CNRD is a database formulated from within DEPM by combining the relevant residue data from all the DEPM databases. The combination of residue databases leads to a more complete coverage of residue values for ECFs than would be obtained from any single database. A single database is typically sparse relative to all food items. Mean residue values in CNRD for each ECF were weighted based on the number of detectable values reported in each of the seven DEPM databases. For example, if residue values of a food item equal to 0.1 mg/kg based on 10 detectable values in database 1, 0.2 mg/kg based on 30 detectable values in database 2, and there is no detectable value in the other five databases, then the mean residue value in CNRD would be $(0.1\times(1/4))+(0.2\times(3/4))=0.175$ mg/kg. The CNRD thus contains a variety of residue concentrations from national sources for agricultural commodities or basic food ingredients.

Food Item Ranking Using DEPM

Two methods were developed and used to rank food items as a function of estimated dietary exposure: a direct, by magnitude, ranking method and a weighted ranking method. The food items ranked by these methods constitute a list of food items that either are consumed in great quantities or contain high chemical residue levels, or a combination of the two, resulting in high exposure.

Method I is a *direct* ranking method. For each of the eight selected chemical residues, the following steps are carried out using the DEPM with Level III values: (1) estimate the US general population exposure to the subject chemical residue for consuming each food item; and (2) identify and list the 10 food items with highest magnitudes of exposure to the residue.

Method II is a *weighted* ranking method; it applies the principles used in the first method to each of the 19 subpopulation groups included in the DEPM. The following two-step process is executed for the weighting of the ranked food items. Step 1: An integer N, from 0 to 10, is the exposure rank of a food item in a subpopulation group. For example, $N=10$ signifies that the food item has the maximum exposure value among the 10 ranked food items for certain subpopulation group; $N=9$ represents the second highest food item, and so on, to $N=1$, which represents the 10th ranked food item. $N=0$ represents a food item that is not ranked among the top 10 in the subpopulation group. Each food item has an N value for each subpopulation group, but the N value may be different among different subpopulation groups. Step 2: An R value is calculated as the sum of N values of a food item within the same population category (i.e., Age/Gender, Ethnicity, *etc.*) divided by number of total selected food items for this population category:

$$R = \sum (N_i)/(10 \times n) \qquad (1)$$

where n is the number of subpopulation groups within a population category (For example, when the US population is grouped by ethnicity in the DEPM, n equals 4 because there are four ethnicity subpopulation groups.); N_i is the N value of a food item for the ith subpopulation group; and "10" is an indication of the selection of 10 food items with highest dietary exposure values for each subpopulation group.

The R value is an estimate of the contribution of a food item to dietary exposure of the US population weighted by its importance to a specific subpopulation group. The range of R values is from 0 to 1. A large R value of a food item indicates a contribution of this food item to the dietary exposure of the US population that is larger than the contribution of a food item with a small R value. Method II of ranking food items gives priority to food items of importance to certain subpopulations and may be ignored by the nonweighted Method I of food item ranking. For example, the exposure contribution to arsenic of whole milk for ethnic subpopulations in the US is estimated by the R value of whole milk within the ethnicity category. The range of i is from 1 to 4 because the DEPM has four ethnic subpopulations available for selection. Based on the DEPM calculation, whole milk is the third food item when ranked as a function of dietary exposure to arsenic for Hispanics (N_1 is equal to 8); the sixth for Non-Hispanic Whites ($N_2=5$); the fourth for Non-Hispanic Blacks ($N_3=7$); and not within the list of 10 food items for Non-Hispanic Others ($N_4=0$). Hence, the R value of whole milk within ethnicity category is the sum of N_i ($i=1,2,3,4$) divided by 40, or $R=0.5$.

Exposure Estimation Using the NHEXAS Data

The second estimation approach for dietary exposure uses a deterministic exposure model with the NHEXAS-AZ data from diet diary questionnaire and composite food sampling. The NHEXAS-AZ study formulated a "diet diary" using a food checklist approach. Respondents reported daily food consumption by type, amount, and date. The diary was divided into 10 food categories: (1) Dairy; (2) Breads, Cereals, Grains, and Pasta; (3) Fruits; (4) Vegetables and Beans; (5) Eggs, Fish, and Meat; (6) Main Dishes/Prepared Meals; (7) Condiments, Dressings, Oils, and Sauces; (8) Snacks; (9) Desserts/Sweets; and (10) Beverages (excluding dairy). These categories are slightly different from those included in the DEPM, but the comparisons performed in this study focus on individual food items and not categories. These categories contained 279 precoded Food/Beverage items derived from FDA's TDS and a few predominantly Hispanic food items selected from a border-wide (US–Mexico border) minimarket basket collection undertaken by EPA and FDA (1995, unpublished). For each food item, the respondent recorded the number of size-specific servings consumed each day by providing on the food diary serving sizes adjacent to the food item. In each category, respondents itemized nonlisted food items by type and amount consumed in the additional space provided in the diary. These nonlisted items are assigned one specific code in each category. Respondents completed the diet diary for four consecutive days coinciding with the environmental sampling week of the study.

Compositing of food items took place on 1 of 4 days that respondents completed the diet diary, usually the fourth day. A duplicate of each item consumed was collected in one of two containers, one for solid and the other for liquid food items. The solid and liquid food groups were composited and later analyzed for chemical residues in the composites. Drinking and tap water were separately collected and analyzed. Consumption of each food item was converted from serving units recorded by the respondent into grams of the food item consumed using standard CSFII serving-to-gram conversion factors. Subsequently, dietary ingestion exposure to a chemical residue for each subject was estimated by:

$$E_{\mathrm{T},i} = \frac{\sum_{F} (C_{F,i} \times W_{F,i})}{\mathrm{BW}_i} \times 10^6 \qquad (2)$$

where $E_{\mathrm{T},i}$ is the total dietary ingestion exposure to a chemical residue from all composite food item types F consumed by subject i during the day of measurement [ng/kg BW/day]; $C_{F,i}$ is the concentration of the chemical residue in the composited food items F consumed by subject i during the day of measurement [mg/kg]; $W_{F,i}$ is the weight of composited food items consumed by subject i during the day of measurement [kg/day]; BW_i is the body weight of subject i [kg]; F is the type of composited food items. There are three types: solid food, liquid food, and water.

Ingestion of benzene is a rare event for most people (USDHHS, 1997). Exceptions occur in areas where groundwater has been contaminated by leakage from underground storage tanks, landfills, or hazardous waste sites. For this reason, benzene contamination was measured in water samples throughout the NHEXAS-AZ survey. Since volatile organic compounds were a tertiary analyte class, we did not measure the benzene content of either liquid or solid food. As a result, we have not estimated benzene exposure for the NHEXAS-AZ component of this study.

Chemical analysts designated several residue concentrations at below detection limit values. Such values are usually referred to as censored values. Dietary exposure estimates calculated by DEPM used a value of zero for residue concentrations reported in the DEPM databases as below detection limit. Information on censored residue concentrations from the NHEXAS-AZ database is shown in Table 1. Rather than use the zero value approach, censored values from the NHEXAS-AZ database were treated using the robust method developed by Helsel (1990). The robust method assumes that all residue concentrations follow one distribution — the one that best fits the above detection limit values. The above detection limit values were fit using Crystal Ball, a commercial software that fits the data points to several distribution types. The Chi-square test, the Kolmogorov–Smirnov test, and the Anderson–Darling test were used to assess the goodness of fit. At least one of these tests must consider the fit acceptable. A modification of the robust method was used to incorporate into the method the portion of the censored data. Based on extrapolation of the fitted distribution, this step creates "fill-in" values for samples that are below the detection limit. Such values were assigned once and were used for all subsequent analyses in the study.

As shown in Table 1, almost all of chlorpyrifos concentration values in liquid foods, drinking water, or tap water are censored values. Diazinon concentration values also include 99% censored values in liquid foods. Water samples were not analyzed for diazinon. Therefore, the censored value treatment was not performed for pesticide data and the exposure to pesticide was not estimated in this approach.

Exposure Estimation Using the NHEXAS Data with the National Residue Database

In the third estimation approach, a deterministic exposure model is used with the NHEXAS-AZ consumption data from questionnaires, and chemical residue values from the national residue database, the CNRD. Since the list of food items in the NHEXAS study is somewhat different from the one in the DEPM, a two-step process is used to estimate the NHEXAS-AZ dietary exposure and food item contribution *via* this approach. The first step identifies the NHEXAS food items that correspond to the food items in the DEPM. The NHEXAS food items use the TDS food codes (except for the supplemental Hispanic food items) and the DEPM food items use the ECF codes. TDS and ECF codes were matched using the DEPM conversion codes. For certain NHEXAS food items without a matching code, a comparable food item with similar name, classification, use, or major ingredients was selected from the DEPM food list and used for subsequent analysis. Thus, each NHEXAS food item had a matching DEPM food item. The second step identifies the ranked food items in NHEXAS corresponding to those in the DEPM. The number of ranked items in NHEXAS was higher than the number of ranked items in the DEPM because some DEPM items correspond to more than one NHEXAS item, i.e., the matching was not one-to-one. Next, the NHEXAS food items were matched with food items in the CNRD residue database. Food items in the DEPM resident database use ECF codes, so they were matched with the corresponding ECF codes of the NHEXAS items established from the first matching procedure. After this matching process, each of the NHEXAS food items was assigned residue values based on the CNRD database, and was identified as a ranked food item. One food item in each NHEXAS food category represents all nonlisted foods of that category. For each chemical residue, this item was assigned a median residue value of all items in the category. These nonlisted items were conservatively considered as ranked food items.

Consequently, the dietary ingestion exposures of subjects in the NHEXAS study were calculated using the following model:

$$E_{\mathrm{T},i} = \frac{\sum_f (C_f \times W_{f,i})}{\mathrm{BW}_i} \times 10^6 \qquad (3)$$

where $E_{\mathrm{T},i}$ is the total dietary ingestion exposure to a chemical residue from the food items consumed by subject i [ng/kg BW/day]; C_f is the concentration of a chemical residue, obtained from the CNRD, in food item f consumed by subject i [mg/kg]; $W_{f,i}$ is the weight of food item f consumed by subject i [kg/day]; BW_i is the body weight of subject i [kg].

Estimation of the NHEXAS Ranked Food Contribution

For each subject and for each of the four days, Eq. (3) was used to calculate the exposure from only ranked food items and the exposure from all food items. The ratio of the two values was multiplied by 100 to get the percent contribution

Table 1. Censored residue concentrations from the NHEXAS-AZ database.

Chemical residue	Medium			
	Solid food	Liquid food	Drinking water[a]	Tap water
Arsenic	0/152[b]	10/147	0/63	0/72
Cadmium	0/152	43/147	57/64	0/72
Chromium	47/152	59/147	25/64	17/72
Lead	1/152	7/147	33/64	4/72
Nickel	0/152	3/147	27/64	2/72
Chlorpyrifos	122/157	155/155	113/113	173/174
Diazinon	144/157	153/155	–[c]	–

[a]Drinking water refers to water from sources other than kitchen tap, e.g., bottled water, filtered tap, *etc*.
[b]Number of censored samples/Number of all samples.
[c]Analysis was not done.

of ranked food items for each subject. In essence, this procedure allows a ranking of exposures from foods actually consumed by the NHEXAS-AZ respondents for comparisons to rankings based on the national consumption values, using the same residue database. Comparing the two ranking approaches allows an independent evaluation of the importance of consumption data on dietary exposure.

Results and discussion

The estimated total dietary exposure by the DEPM approach of selected US subpopulation groups to the target chemicals using the CNRD database is listed in Table 2. Underlined values in the table indicate the maximum exposure estimate for each chemical residue and each subpopulation group. The difference between these maximal values and other exposure values within a subpopulation group is frequently small. The notable exceptions are for Nonnursing Infants and Children 1–6 years of age caused by their high consumption-to-body weight ratio and, to a lesser extent, the types of foods consumed by them. In the Family Income and Geographic Region categories, one group is identified with the highest exposure for most chemical residues. The Poverty 0–130% group is the highest exposed group in the Family Income category for all chemical residues. The Western group is the highest exposed group in the Geographic Region category for seven chemical residues; one exception for chlorpyrifos, the Northeast group, is the highest exposed group. For the Age/Gender and Ethnicity categories, two young children groups are identified with the highest exposure for the eight chemical residues. For the Age/Gender category, the Nonnursing Infants group has the highest exposure to arsenic, lead, nickel, benzene, and diazinon. The Children 1–6 group has the highest exposure to the other three chemical residues: cadmium, chromium, and chlorpyrifos. For the Ethnicity category, two groups, the Hispanic group and the Non-Hispanic White group, have highest exposures that are similar and predominantly higher than the other groups. The Hispanic group has the highest exposure to cadmium, lead, benzene, and diazinon; the Non-Hispanic White group to arsenic, chromium, nickel, and chlorpyrifos.

The Reference Dose (RfD; mg/kg BW/day) is a threshold value for adverse health effects from the daily

Table 2. Exposure estimates of US subpopulations using the DEPM (μg/kg BW/day).

Subpopulation	Arsenic	Benzene	Cadmium	Chlorpyrifos	Chromium	Diazinon	Lead	Nickel
US population	0.653	0.007	0.103	0.108	0.505	0.123	1.009	0.374
Age/gender								
Nonnursing infants	**1.741**[a]	**0.026**	0.160	0.124	0.673	**0.434**	**3.117**	**0.870**
Children 1–6	1.188	0.014	**0.195**	**0.318**	**0.867**	0.240	1.952	0.669
Children 7–12	0.700	0.008	0.121	0.178	0.583	0.143	1.164	0.425
Females 13–19	0.485	0.006	0.083	0.105	0.371	0.101	0.824	0.281
Females 20+	0.644	0.007	0.090	0.070	0.464	0.113	0.920	0.350
Females 55+	0.659	0.007	0.081	0.064	0.482	0.118	0.946	0.368
Males 13–19	0.545	0.006	0.103	0.125	0.460	0.105	0.890	0.324
Males 20+	0.579	0.006	0.093	0.072	0.475	0.108	0.895	0.342
Males 55+	0.629	0.007	0.087	0.062	0.520	0.112	0.918	0.369
Ethnicity								
Hispanic	0.710	**0.008**	**0.124**	0.129	0.542	**0.143**	**1.177**	0.407
Non-Hispanic white	**0.746**	0.008	0.142	**0.156**	**0.583**	0.131	1.095	**0.424**
Non-Hispanic black	0.581	0.005	0.113	0.140	0.380	0.096	0.797	0.295
Non-Hispanic other	0.498	0.006	0.075	0.144	0.293	0.109	0.871	0.258
Geographic region[b]								
North central	0.361	0.004	0.080	0.109	0.389	0.067	0.611	0.238
Northeast	0.621	0.007	0.106	**0.129**	0.529	0.116	0.968	0.379
Southern	0.653	0.007	0.098	0.096	0.484	0.118	0.966	0.359
Western	**0.766**	**0.008**	**0.120**	0.106	**0.567**	**0.138**	**1.133**	**0.423**
Family income[c]								
Poverty 0–130%	**0.715**	**0.008**	**0.122**	**0.127**	**0.595**	**0.132**	**1.094**	**0.420**
Poverty 131%+	0.638	0.007	0.098	0.103	0.484	0.121	0.986	0.362

[a]Underlined and bold values indicate the maximum exposure estimate for each of the eight contaminants for each subpopulation group.
[b]The regional classification is as defined by the USDA, and is based upon US Census Bureau regions.
[c]Annual household income as a percentage of the Poverty Index.

dietary intake of chemical residues . The estimated dietary exposure levels of the subpopulation groups are lower than the corresponding RfD except for dietary exposure to arsenic (RfD=0.3 μg/kg BW/day for inorganic arsenic), which is dependent on the specific species constituting total ingested arsenic (inorganic plus organic forms). Concentrations reported in the DEPM and the NHEXAS-AZ databases are for total arsenic. It is generally believed that total arsenic in foods is associated with the less toxic, organic forms, and that the predominant sources are seafoods (Mohri et al., 1990; Reilly, 1991). One should note that at the present time, there are no RfD values for benzene, diazinon, and lead although these chemicals have been associated with adverse health effects (WHO, 1993; WHO, 1998; USDHHS, 1997; Melnyk et al., 2000). When a chemical is a probable human carcinogen, with specified weight of evidence, the relationship between dose and response is estimated by a toxicity value, usually the cancer potency factor. Cancer potency factors were not used in this evaluation.

Forty-three food items appear at least once among the merged ranked food items from the two DEPM ranking methods using the CNRD residue data. These 43 food items contribute from 68% to 91% of the total estimated dietary exposure of the US population to the eight chemical residues. Adding other individual food items contributes minimally to the US population total estimated dietary exposure to target residuals. Table 3 lists 43 ranked food items and their proportional contribution to the estimated total dietary exposure to each of the eight target pollutants. The ranked items consist of 10 Grains/Grain Product items, 9 Seafood items, 9 Beverage items (including drinking water), 5 Fruit items, 4 Vegetable items, 2 Meat/Poultry items, and 1 item from each of the following categories: Dairy/Egg Products, Infant Foods, Legumes/Nuts, and Miscellaneous. The significant effect of residues in drinking water from tap (ECF code B025) is apparent for all chemicals except chlorpyrifos. It contributes 50% of dietary exposure of US population to benzene, 47% to diazinon, 39% to lead, 35% to arsenic, and 30% to nickel. Not only does it account for the largest percentage of total exposure, it is also most likely to result in a few other water-based items with high ranking, e.g., coffee (B012) and tea (B023). For chlorpyrifos, 40% of the exposure to it comes from orange juice (F094). Furthermore, it is evident that Fish and Seafood items, e.g., shellfish (S014), finfish saltwater (S005), and finfish breaded (S001), are among major contributors to exposure to metals, particularly arsenic (probably organic). The nine ranked Fish/Seafood items are responsible for 41% and 27% of total dietary ingestion exposure to chromium and nickel, respectively.

The "drinking water from tap" food item (B025) is the primary residue source of dietary exposure based on the residue data included in the CNRD database. Yet, it is not known if these residue values are typical of treated drinking water or are included in the database because they are unique occurrences. Residues used to estimate exposures are mean values included in the respective databases; however, it is suspected that certain water data may have been included because they represented unusually high values, thus making the mean values higher than typical mean values for water. This is especially relevant for diazinon, a pesticide not commonly found in drinking water, but reported in the Pesticide Residue Information System for one positive sample at a level of 3.2 ppm. Without the inclusion of the water residue, the total exposure would be reduced, but the ranking of the remaining nonwater-based food items would be relatively unchanged.

The ingestion exposure values estimated by the three estimation approaches are summarized in Table 4. First, the results from the NHEXAS-AZ approach were compared with the corresponding results obtained for the western region subpopulation from the DEPM approach. The two approaches agree quite well for arsenic. The NHEXAS ingestion exposure to lead is much smaller than the national exposure values. The reason for the observed difference is not clear, other than it is known that lead levels in food have diminished over the years, and DEPM estimates may be a reflection of previous residue levels. On the other hand, ingestion exposures to cadmium, chromium, and nickel as estimated using the NHEXAS-AZ database are higher than the estimated national values based on CNRD database. A possible explanation is that there are many mining districts in Arizona, which may be the cause of greater values of exposure to these metals. Moreover, the DEPM uses zero for residue values reported below the limits of the detection while the NHEXAS-AZ estimation uses the robust method, which may have led to somewhat higher average exposure estimates. Regardless of the direction, there are two more potential reasons for the noted difference. First, the NHEXAS samples are from Arizona only, while the DEPM western region estimates include several states; and they were based on residue data from all geographical regions. Second, the food sampling method in the NHEXAS relies on the participating subjects to collect the food and fill out the diet diary questionnaire. While it is a more direct measurement, the portion size recorded in the diet diaries is subjective and may lead to errors in our estimates.

The results from the third estimation approach — using the NHEXAS data with the CNRD residue database — were compared with those estimated from the other two approaches. Most metal exposures estimated *via* the third approach have lower values than those estimated using solely the NHEXAS data. Since the two approaches used the same data except the residue values, the likely reason for the difference is that the NHEXAS-measured residue values are higher than the national values. Hence, the mining activities aforementioned may have caused higher

Table 3. Forty-three DEPM ranked food items and corresponding proportions of total exposure (%).

Number	ECF code	Food name	Percent of total exposure							
			Arsenic	Benzene	Cadmium	Chlorpyrifos	Chromium	Diazinon	Lead	Nickel
1	B003	Carbonate beverages noncola	0.15	0.01	0.28	7.92	0.00	0.35	0.70	0.00
2	B008	Citrus juice drinks	0.15	0.32	0.07	0.89	0.06	0.22	0.19	0.12
3	B010	Coffee decaffeinated	1.23	1.75	0.46	0.00	0.50	1.65	1.38	1.05
4	B012	Coffee	9.73	13.82	3.61	0.07	3.95	13.20	10.91	8.33
5	B015	Fruit-flavored drinks from powder	2.29	3.88	0.89	1.99	0.92	3.10	2.62	1.95
6	B018	Noncitrus fruit drinks	0.05	0.06	0.04	2.34	0.02	0.14	0.14	0.04
7	B020	Orange drinks	0.44	0.84	0.17	7.19	0.18	0.63	0.50	0.38
8	B023	Tea	5.32	7.56	1.97	0.01	2.16	7.15	5.97	4.56
9	B025	Drinking water from tap	35.10	49.85	12.97	0.00	14.26	47.05	39.34	30.04
10	D005	Chicken eggs	0.07	0.00	0.98	0.03	2.82	0.04	0.38	1.38
11	F005	Apple juice	0.28	0.00	0.16	1.68	0.00	0.13	0.64	0.00
12	F010	Apples, red, raw	0.01	0.00	0.01	1.71	0.00	0.11	0.12	0.00
13	F057	Fruits, Ns, juice, and nectars	0.16	0.35	0.15	4.53	0.06	0.26	0.24	0.12
14	F093	Orange and tangelo, raw, canned, or frozen	0.00	0.00	0.00	2.85	0.00	0.06	0.00	0.00
15	F094	Orange juice	2.05	2.77	0.84	40.79	0.79	3.03	2.67	1.67
16	G009	Corn mixed dishes	0.71	0.00	1.55	0.02	0.94	0.02	0.16	0.63
17	G026	Mostly oats, ready-to-eat cereals	0.00	0.00	0.00	1.53	0.00	0.05	0.00	0.00
18	G041	Oatmeal, cooked cereals	0.59	0.82	0.23	0.30	0.23	0.80	0.66	0.49
19	G048	Rice mixed dishes	0.58	0.14	1.08	0.04	0.66	0.15	0.22	0.49
20	G052	Rice white, cooked with fat	0.92	0.76	1.04	0.17	0.22	0.88	0.85	0.46
21	G054	Rice with beans dishes	0.06	0.06	0.12	0.02	0.02	0.07	0.11	0.03
22	G066	Wheat soups, meatless	4.28	0.62	9.10	0.19	5.24	0.71	1.36	3.74
23	G082	Wheat with tomato sauce and meat dishes	0.17	0.21	0.90	0.75	0.24	0.89	1.17	0.21
24	G084	Wheat with tomato sauce, cheese, and meat dishes	0.14	0.14	0.74	0.37	0.71	0.56	1.31	0.41
25	G161	Wheatflour yeast breads	0.49	0.33	0.65	1.65	0.42	0.46	0.62	0.36
26	I090	Soy-based infant formula	0.80	1.14	0.29	0.04	0.32	1.09	0.89	0.68
27	L008	Dried bean mixtures	0.30	0.39	0.38	0.13	0.11	0.45	0.44	0.23
28	M007	Beef in gravy sauce or cream	0.07	0.15	0.58	1.29	0.14	0.47	0.61	0.10
29	M010	Beef sandwiches	0.09	0.06	0.52	0.45	0.09	0.53	0.74	0.07
30	S001	Finfish, breaded	2.70	3.31	2.31	0.03	14.59	0.00	1.21	9.18
31	S002	Finfish, freshwater	0.28	0.15	0.13	0.01	1.34	0.00	0.10	0.92
32	S004	Finfish, Ns	0.29	0.00	0.06	0.00	0.60	0.00	0.06	0.45
33	S005	Finfish, saltwater	3.18	0.86	1.00	0.01	10.45	0.03	0.83	7.35
34	S008	Seafood soup	0.76	0.03	0.47	0.04	3.03	0.06	0.30	2.01
35	S009	Seafood with vegetable dishes	1.44	0.06	1.71	0.10	1.76	0.09	0.40	1.23
36	S011	Seafood with wheat or rice dishes	1.19	0.36	1.82	0.02	2.49	0.03	0.34	1.62
37	S012	Shellfish, breaded	2.08	0.00	4.52	0.00	2.80	0.00	0.38	1.86
38	S014	Shellfish	3.14	0.01	6.83	0.02	4.20	0.00	0.57	2.79
39	V107	Potato, baked or boiled without peel	0.00	0.00	1.86	0.10	0.00	0.00	0.02	0.00
40	V110	Potato, fried without peel	0.05	0.06	3.45	0.16	0.14	0.06	0.11	0.09
41	V123	Seaweed and algae cooked	5.10	0.00	0.19	0.00	0.42	0.00	0.33	0.37
42	V148	Tomatoes, raw	0.00	0.00	0.77	0.43	0.00	0.35	2.35	0.00
43	X008	Gravy from meat, poultry, fish base	1.53	0.03	2.79	0.05	7.79	0.05	0.48	1.80
		Total	**87.96**	**90.89**	**67.69**	**79.90**	**84.67**	**84.93**	**82.42**	**87.21**

exposure to metals because they cause higher metal concentrations in food. Similarly, all exposures estimated *via* the third approach have lower values than those estimated using the DEPM. Since the two approaches used the same residue values, the difference was caused by the difference in consumption data of the two populations. Clearly, the consumption data are influenced by demographic characteristics, e.g., age, ethnicity, and income level. The small number of children in the NHEXAS samples may be the reason for lower mean exposure estimates obtained from the third approach, since children generally have higher exposure than adults. Nevertheless, individuals of the same demographic characteristics may still consume different types and amounts of food due to preferences or other factors.

Out of the 43 ranked food items, two items (soy-based infant formula, and seaweed and algae cooked) could not be matched with any of the NHEXAS food items. Therefore,

Table 4. Comparison of the ingestion exposure estimates obtained from the three approaches.

Chemical residue	NHEXAS sample size	Mean exposure (ng/kg BW/day)		
		DEPM[a]	NHEXAS	NHEXAS using CNRD[b]
Arsenic	147	766	700	514
Cadmium	147	120	331	84
Chromium	147	567	1411	312
Lead	147	1133	218	1075
Nickel	147	423	3058	286

[a]Mean exposure of the western region subpopulation of the US, obtained from DEPM.

[b]Exposure estimated using the NHEXAS consumption data with the CNRD residue database.

subsequent analyses did not include these items in the ranked food list. Since there is no one-to-one correspondence among food items in the NHEXAS questionnaire and the DEPM databases, identification of ranked food items in NHEXAS resulted in 57 NHEXAS food items that corresponds to the 41 food items ranked by the DEPM. Ten nonlisted items, one per each food category, were conservatively included into the ranked items group. Each of these items was assigned the median residue value of the all other food items in the category. Therefore, there are totally 67 ranked food items in NHEXAS. The use of Eq. (3) to calculate the exposure from only ranked food items and the exposure from all food items yields the percent contribution of ranked food items for each subject. The average percent contribution of ranked food items, from all subjects and all 4 days, was compared to the corresponding values of the western region subpopulation, which was obtained from the DEPM (see Table 5). Results show that out of 289 food items included in the NHEXAS-AZ dietary questionnaire, Arizona residents consume about 15 items daily. Six of these 15 food items consumed contribute highly to the dietary ingestion exposure. For the ingestion exposure values, the average percent contribution of ranked food items in the NHEXAS samples, over the 4-day period, ranges from 56% to 70%. The corresponding values for the contribution of the 41 ranked food items using the DEPM are higher for all chemical residues, ranging from 78% to 90%. Given that the same residue database is used, the difference of the ranked food contributions is likely to come from three factors: type of food items consumed, amount of food items consumed, and demographic characteristics of samples in the two groups. Assuming comparable demographic characteristics, the result suggests that the NHEXAS-AZ respondents either consumed fewer ranked items than the western US population, or consumed fewer amounts of the ranked items, or both.

Conclusions

This paper identifies subpopulation groups most highly exposed to each of the eight target chemicals, and food items contributing high portions of the dietary ingestion exposure to these chemical residues in food. It also compares the ingestion exposure estimates and the contributions of ranked food items based on different analysis approaches. Analytes examined in this study are the primary target contaminants of the NHEXAS-AZ exposure study. The dietary exposure to each chemical is estimated using either the DEPM approach or the NHEXAS-AZ approach.

Based on the CNRD databases, subpopulation groups with the greatest ingestion exposure to at least one of the target residues were: Nonnursing Infants, Children 1–6, Hispanic, Non-Hispanic Others, Western, Northeast, and Poverty 0–130%. Infants and young children have small

Table 5. Comparison of percent contribution of ranked food items to total ingestion exposure.

	Chemical residue	NHEXAS[a]					DEPM[b]
		Day 1	Day 2	Day 3	Day 4	4-Day average	
Number of subjects		296	265	259	250		
Ranked items/total items[c,d]		6/16	5/15	5/14	6/14	6/15	
Percent[e] based on CNRD	Arsenic	70	69	71	68	70	81
	Benzene	68	67	68	66	67	90
	Cadmium	55	55	57	57	56	68
	Chlorpyrifos	63	64	63	64	64	78
	Chromium	64	63	67	61	64	84
	Diazinon	66	66	66	65	66	84
	Lead	63	63	64	63	63	81
	Nickel	68	67	70	66	68	86

[a]Percent contribution of 67 ranked items to total exposure of the NHEXAS study participants.

[b]Percent contribution of 43 ranked items to total exposure of the western region subpopulation, obtained from DEPM.

[c]Average number of ranked items per average number of total items consumed.

[d]"Nonlisted" food items were considered as ranked items.

[e]Average percent contribution of ranked items to total exposure.

body weights and, therefore, have high daily food consumption per unit body weight (μg/kg BW/day), which contributes to such high values of ingestion exposure. Except for the Age/Gender subpopulation, the difference between the subpopulation with the maximal estimated dietary exposure and other subpopulations within a category is not very large. This indicates that the consumption characteristics of a subpopulation have minimal influence on dietary exposure.

Two methods were developed to rank food items as a function of their contribution to the estimated total dietary exposure. Using the combined chemical residue database, CNRD, we conclude that over 68% of the total estimated dietary exposure of the US population to all eight chemical residues is contributed by 43 food items appearing at least once among the merged list of ranked items. The majority of these items belong to the Grains/Grain Products, Seafood, and Beverage categories. Water from tap, tea, and coffee is estimated to be the primary contributor to dietary exposure; however, it is expected that residue values for water contained in the CNRD database may not be typical of all drinking water.

Different estimation approaches yield different ingestion exposure results. For lead, the value estimated from the NHEXAS study is smaller than that of the western region using the DEPM. The opposite is true for exposure to cadmium, chromium, and nickel. Probable reasons are differences in methodology of estimating exposure, region considered (western region *versus* only a part of the region), and treatment of below detection values. Most metal exposures estimated using the NHEXAS data combined with the CNRD have lower values than those estimated using solely the NHEXAS data, suggesting that the NHEXAS-measured residue values are higher than the national values. Similarly, the lower values of estimates from the third approach than those from the DEPM approach indicate the consumption difference of the two populations caused by demographic characteristics and factors that affect types and amounts of food consumed.

Due to the lack of one-to-one correspondence, the list of 43 ranked food items from the CNRD databases resulted in the identification of 57 corresponding NHEXAS ranked food items. Ten more items that represent the nonlisted food items of each food category were added to the NHEXAS ranked food item list; this increased the number of ranked items to 67 items. On average, the NHEXAS subjects consume a total of about 15 food items per day; six of them are ranked food items that contribute between 56% and 70% of dietary exposure to the eight chemical residues. The percent contributions of ranked food items obtained from the NHEXAS samples are smaller than those obtained from the western US population. This may indicate the difference in consumption characteristics of the two groups with respect to the ranked food items.

In the performance of this study, only the CNRD database was used. Clearly, use of other databases (or any of the combined CNRD databases used independently) may lead to different results. Conclusions reached by this study are constrained by the effects of the following: (i) demographics are related to consumption data only; (ii) use of either zero for residues below the limit of detection value or the robust method for nondetects; and (iii) the effect of water residue data in the CNRD database.

The ranking method, combination of Methods I and II, developed for this work may be thought of as a source apportionment of dietary exposure because it identifies food items that contribute substantively to dietary ingestion exposure. The ranking method illustrates that a small number of identified food items, 43 of 800 ECFs considered in the DEPM, contribute a large portion of the total estimated dietary exposure. A relatively larger proportion, 67 of 289, is found for the NHEXAZ study. This finding may lead to efficient control strategies for reducing dietary exposure to chemical residues by focusing on the relatively small number of food items that have similar ingredients and contribute high portions to the total dietary ingestion exposure.

References

Berry M.R. Strategy for a dietary exposure research program. *J Expos Anal Environ Epidemiol* 1992: Suppl. 1: 97–110.

Berry M.R., and Tomerlin J.R. A Dietary Exposure Potential Model using extant databases to estimate exposure to chemical residues. In: *Proceedings of the EPA/AWMA International Specialty Conference, "Measurement of Toxic and Related, Air Pollutants (VIP-64)",* Air and Waste Management Association, Pittsburgh, PA, 1996, pp. 819–828.

Dietary Exposure Potential Model (DEPM) Version 3.3.2A software available for evaluation on the internet at http://www.epa.gov/nerlcwww/depm.htm. 2000.

Helsel D.R. Less than obvious: statistical treatment of data below the detection limit. *Environ Sci Technol* 1990: 24(12): 1766–1774.

Lebowitz M.D., O'Rourke M.K., Gordon S., Moschandreas D.J., Buckley T., and Nishioka M. Population-based exposure measurements in Arizona: a phase I field study in support of the national human exposure assessment survey. *J Expos Anal Environ Epidemiol* 1995: 5(3): 297–325.

Melnyk L.J., Berry M.R., Sheldon L.S., Freeman N.C.G., Pellizzari E.D., and Kinman R.N. Dietary exposure of children living in lead-laden environments. *J Expos Anal Environ Epidemiol* 2000: 10(6; Part 2): 723–731.

Mohri T., Hisanaga A., and Ishinishi N. Arsenic intake and excretion by Japanese adults: a 7-day duplicate diet study. *Food Chem Toxicol* 1990: 28(7): 521–529.

Pennington J.A.T., and Gunderson E.L. History of the food and drug administration's Total Diet Study — 1961 to 1987. *J Assoc Off Anal Chem Int* 1987: 70: 772–782.

Reilly C. *Metal Contamination of Food*, 2nd ed. Elsevier, New York, NY, USA, 1991.

Tomerlin J.R., Berry M.R., Tran N.L., Chew S.-B., Petersen B.J., Tucker K.D., and Fleming K.H. Development of a Dietary Exposure Potential

Model for evaluating dietary exposure to chemical residue in food. *J Expos Anal Environ Epidemiol* 1993: 7(No. 1): 82–102.

US Department of Health and Human Service. *Public Health Service, Agency for Toxic Substances and Disease Registry, Toxicological Profile for Arsenic,* 1993a.

US Department of Health and Human Service. *Public Health Service, Agency for Toxic Substances and Disease Registry, Toxicological Profile for Cadmium,* 1993b.

US Department of Health and Human Service. *Public Health Service, Agency for Toxic Substances and Disease Registry, Toxicological Profile for Chromium,* 1993c.

US Department of Health and Human Service. *Public Health Service, Agency for Toxic Substances and Disease Registry, Toxicological Profile for Lead,* 1993d.

US Department of Health and Human Service. *Public Health Service, Agency for Toxic Substances and Disease Registry, Toxicological Profile for Nickel,* 1995.

US Department of Health and Human Service. *Public Health Service, Agency for Toxic Substances and Disease Registry, Toxicological Profile for Benzene,* 1997.

World Health Organization (WHO). Environmental Health Criteria 150, Benzene, 1993.

World Health Organization (WHO). Environmental Health Criteria 198, Diazinon, 1998.