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Exposure and risk estimates

FOR ARIZONA DRINKING WATER

WHEN EXPOSURES TO METALS AND VOLATILE ORGANIC CHEMICALS IN DRINKING WATER WERE COMPARED FOR THE POPULATION OF ARIZONA AND THAT OF BORDER COMMUNITIES, AVERAGE DAILY EXPOSURES AND ASSOCIATED RISKS WERE FOUND TO BE SMALLER IN THE BORDER COMMUNITIES THAN IN THE STATE POPULATION.

This study was conducted as part of the National Human Exposure Assessment Survey (NHEXAS) Arizona, a multiple-media, multiple-route, and multiple-pollutant study (Lebowitz et al, 1995). One of this study's objectives was to assess the Arizona population's exposure to metals, particles, volatile organic compounds (VOCs), and pesticides from inhalation, ingestion, and dermal absorption pathways and compare that exposure with corresponding exposures of the Arizona border population. The border study focused on exposure assessment of communities along the Arizona–Mexico border. There has been concern that exposures of the border communities may be higher than those of other parts of the state because of their proximity to Mexico, where environmental quality is not of equal concern as it is in the United States. To determine the validity of these concerns about elevated exposure, the authors compared oral exposures to metals and VOCs and associated risks from drinking water for the Arizona and border populations.

BACKGROUND

Occurrence of metals and VOCs in drinking water. Arsenic, barium, cadmium, copper, manganese, nickel, lead, zinc, several VOCs, and pesticides may be present in drinking water and may cause health effects to humans (IRIS, 2000). Several types of cancers as well as cardiovascular and neurological diseases are among the reported health effects (Calderon, 2000). Oral exposure via drinking water is reported to account for nearly 100% of the total intake of disinfection by-products, e.g., trihalomethanes (THMs) (van Dijk-Looijaard & van Genderen, 2000). Metals used in domestic distribution systems can be found in drinking water at high levels, especially if the water was stagnant

TABLE 1 Descriptive statistics for drinking water chemical residue concentrations in tap and nontap water

	Arizona Population: Concentration—*μg/L*						Border Population: Concentration—*μg/L*					
	Tap			Nontap			Tap			Nontap		
Chemical Residue	Median	Mean	90th Percentile	Median	Mean	90th Percentile	Median	Mean	90th Percentile*	Median	Mean	90th Percentile
Arsenic	4.76	6.68	16.0	0.14	2.25	4.23	5.09	6.64	10.7	0.23	2.19	4.67
Chromium	1.09	4.89	11.3	0.28	0.95	1.74	1.00	1.40	4.34	0.36	3.49	4.08
Lead†	0.39	0.70	1.59	0.05	0.81	0.94	0.28	0.32	0.60	0.17	0.38	0.95
Nickel†	0.32	2.34	9.62	3.90	4.78	5.89	2.27	3.56	5.62	0.33	2.80	4.27
1,2-dichloroethane	0.29	0.65	1.94	0.30	0.65	2.28	0.29	0.42	0.77	0.08	0.29	0.72
1,3-butadiene							0.048	0.062	0.11	0.031	0.044	0.11
Dichloromethane	0.074	0.21	0.57	0.067	0.26	0.64					NA‡	NA
Chloroform†	0.03	2.60	2.04	0.05	1.30	2.00	0.11	0.39	1.19	0.15	0.74	0.87
Toluene	0.22	2.14	4.51	0.57	2.35	6.78	0.49	1.54	5.71	0.50	1.21	2.67

*Because the sample size was not adequate for distribution fitting, percentile value was calculated from empirical cumulative distribution.
†Median tap water concentrations for Arizona and border populations were significantly different with the Mann Whitney test.
‡NA—not available

before it was ingested (van Dijk-Looijaard & van Genderen, 2000).

Among THMs, chloroform (also known as trichloromethane) is generally reported to have the greatest concentration—as high as 200–300 μg/L (Fawell, 2000). Chloroform concentrations found in tap water of homes in New Jersey (Weisel et al, 1999) and the Lower Rio Grande Valley in Texas (Berry et al, 1997) had medians of 16 and 5.0 μg/L and ranges of 0.04–200 and 1.1–26.1 μg/L, respectively. Chloroform concentrations in New Jersey homes had been measured earlier during a Total Exposure Methodology (TEAM) study (Wallace et al, 1984). Results of the TEAM study showed that chloroform concentrations in tap water measured in the range of 11–225 μg/L with a median of 128 μg/L.

Lead concentrations in tap water and blood samples have fallen substantially since the 1980s, but researchers contend even the much lower levels still constitute a public health problem in relation to bottle-fed infants (Watt et al, 2000). Watt and co-workers estimated that in Glasgow, Scotland, 15.5% of infants may be exposed to tap water lead concentrations exceeding the World Health Organization (WHO) guideline of 10 μg/L. In a study in the United Kingdom, 14–37% of the drinking water samples showed lead concentrations greater than guideline values (Alexander et al, 1993). For all nine homes in the Lower Rio Grande Valley study, lead in drinking water was found at concentrations ≤2.0 μg/L (Berry et al, 1997).

Drinking water arsenic concentration levels <10 μg/L (the WHO provisional guideline for arsenic in drinking water) are found in many public water supplies; higher concentrations are encountered in certain geographic areas, especially in geothermal spring waters (Buchet & Lison, 2000). On the other hand, some metals (e.g., zinc, copper, selenium, iron, and manganese) have beneficial as well as adverse effects. Therefore, dose–response relationships are not available for all metals (van Leeuwen, 2000).

Preliminary results of the NHEXAS Arizona study (O'Rourke et al, 1999) found that when all routes of exposure are considered, the different population subgroups had different levels of exposure. Furthermore, results indicated that non-Hispanics had greater exposures to chromium and nickel, whereas Hispanics had greater exposure to lead. This study also found that nontap drinking water had less metal content than tap water.

Research objectives. The main objective of this research was to estimate and compare risks for both Arizona and border populations from oral exposures to metals, VOCs, and pesticides through drinking water. Pesticide concentrations in almost all samples were below detection limits, however, and are not included in this work.

The main objective of this research was to estimate and compare risks for both Arizona and border populations from oral exposures to metals, volatile organic compounds, and pesticides through drinking water.

Two approaches were applied to estimate exposure. The deterministic approach calculated exposure and risk values of each subject using measured concentrations, drinking water consumption, and body weight specific to each subject. Sample subjects were determined using a probability-based sampling framework to ensure a representative sample of the entire population. The probabilistic approach estimated exposure and risk distributions using Monte Carlo simulation methods. Uncertainties associated with these distributions were estimated using the bootstrap technique. The drinking water oral exposure of several population subgroups and drinking water sources was estimated and compared.

METHODS

This section briefly describes the study design, sampling, and chemical and data analysis; more detailed information can be found in the literature (Moschandreas et al, 2001; Gordon et al, 1999; O'Rourke et al, 1999; Lebowitz et al, 1995). Additionally, this section summarizes the methods used to estimate oral exposure and risk and to compare exposures of population subgroups.

Study design and sampling. A probability sampling design was used by the NHEXAS Arizona study. In phase 1, 48 primary sampling units were selected from 400–600 combined block groups of predetermined regions. Primary sampling units were divided into area segments, each having 20–30 housing units. In phase 2, five area segments, called secondary sampling units, were selected. In phase 3, five housing units were selected from each secondary sampling unit. In phase 4, a primary respondent was selected from each house, but other residents of each house were also asked to participate in the study.

Of the 1,225 households selected, 900 households participated in stage 1 in which baseline and descriptive questionnaires were administered to the primary respondents. In stage 2, 450 households participated in less

Populations from Arizona and the Arizona–Mexico border were studied to assess and compare their exposures and risks associated with drinking water.

TABLE 2 LOC* and percentage of actual and estimated subjects with higher drinking water concentrations

Chemical Residue	LOC *µg/L*	Arizona Population Sample *% > LOC*	Arizona Population Estimated *% > LOC*	Border Population Sample *% > LOC*	Border Population Estimated *% > LOC*
Arsenic	50†	0	0.10	0	0.2
Chromium	100‡	0.8	0.22	0	0.2
Lead	15†	0	0.02	0	0.0005
Nickel	40†	0.8	0.16	0	0.7
1,2-dichloroethane	5†	0	2.3	0	0.02
Dichloromethane	5†	0.8	0.7	NA§	NA
Chloroform	100†**	0	0.4	0	0.002
Toluene	1,000†	0	0.01	0	0.0

*LOC—Level of concern
†US Environmental Protection Agency guideline
‡State of Arizona guideline
§NA—not available
**For total trihalomethanes

intensive environmental sampling and additional questionnaires. Of these 450 households, 179 randomly selected households agreed to participate in stage 3's intensive environmental sampling and monitoring. These samples were used to determine exposures to chemical residues in drinking water and associated risks for 133 subjects. A similarly selected database was used for the border study; because this study included only 34 subjects, however, the border database is not a comprehensive one.

Water sampling and chemical analysis. Tap water samples were taken after a 3-minute flush period. Other drinking water samples were taken as supplied by the respondent. Water sample containers were wrapped individually in bubble wrap, set on ice, and sent by overnight express for analysis to a US Environmental Protection Agency (USEPA) contract laboratory. Samples were analyzed according to USEPA method 200.8, using inductively coupled plasma–mass spectrometry for metals. A USEPA contract laboratory also analyzed VOCs using gas chromatography–mass spectrometry. All field and laboratory procedures were subject to quality assurance/quality control evaluations, including replicate-, split-, and spike-sample collection, precision, recovery rate, and minimum detection level calculations (Gordon et al, 1999; O'Rourke et al, 1999).

Data analysis. Concentrations below detection limit (BDL) denote censored data. This study employed the robust probabilistic technique (Helsel, 1990) to fill in for BDL values. Concentrations other than BDL values were fitted with a distribution using a software program.[1] BDL concentrations were assumed to also follow this distribution, and a random fill-in value between zero and the detection limit was generated for each BDL sample using the software program's random number distribution functions. Then all data (including the fill-ins) were used to estimate the population statistics.

For each chemical residue in drinking water, the Arizona guideline was used as the demarcation level to determine the actual proportion of sample subjects with drinking water residue concentrations higher than the level of concern (LOC). In the absence of state guidelines, the USEPA drinking water limit or guideline values were used for the same purpose. The percentage of the population with drinking water residue values higher than the LOC was also estimated using the cumulative distribution of residue concentrations. When both guidelines were available, the lower of the state or USEPA values was used.

The drinking water consumption of the NHEXAS database was recorded as the number of 8 oz (0.24 L) cups consumed each day for four consecutive days. In this study, the consumption for each person was converted to litres. The analysis of variance test was used to test the null hypothesis of "median drinking water consumption is the same for the four days" versus the alternative hypothesis of "at least one pair of median daily consumption values is different."

Oral exposure. ***Deterministic approach.*** The authors used the indirect approach to estimate oral exposures on a person-by-person basis. For each person, oral exposure was calculated using the following equation (USEPA, 1997a):

$$ADE = \frac{RC^{*}WC}{BW} \qquad (1)$$

in which *ADE* is the average daily exposure (µg/kg/d), *RC* is the residue concentration in water (µg/L), *WC* is the water consumption (L/d), and *BW* is the subject's body weight (kg). Point estimates were used for the deterministic estimation of oral exposure. Drinking water consumption values of each subject were obtained from a four-day-period diet diary questionnaire. Chemical residue concentrations were measured one day after the four-day period and were assumed to be applicable to the four-day period. The body weight and consumption data of each person were obtained from the NHEXAS questionnaires.

Probabilistic approach. In addition to the deterministic estimation of the oral exposure to each chemical studied, oral exposure was also estimated probabilistically using Monte Carlo simulations (USEPA, 1997b). Oral exposure (Eq 1) was calculated repeatedly (10,000 times) to estimate the distribution of oral exposure. Uncertainty associated with oral ex-

TABLE 3 **Descriptive statistics for average daily oral exposure from drinking water**

	Arizona Population: Exposure—*ng/kg/d*						Border Population: Exposure—*ng/kg/d*					
	Deterministic (*n* = 133)*			Probabilistic (*n* = 10,000)			Deterministic (*n* = 34)*			Probabilistic (*n* = 10,000)		
Chemical Residue	Median	Mean	90th Percentile	Median	Mean	90th Percentile	Median	Mean	90th Percentile	Median	Mean	90th Percentile
Arsenic	17	54	151	24	64	180	0.9	7.6	24	20	66	185
Chromium	6	64	84	20	69	147	1.2	136	196	19	64	141
Lead	3	12	20	6	13	26	0.5	0.9	2.3	7	56	100
Nickel	19	51	140	14	56	145	1.7	4.9	14	25	77	176
1,2-dichloroethane	2.4	8.5	25	8	15	36	0.2	0.9	2.3	3.5	6.9	17
1,3-butadiene							0.07	0.1	0.24	0.4	0.6	1.4
Dichloromethane	0.62	2.80	7.30	0.57	4.03	7.76					NA†	
Chloroform	0.42	25.7	25	1.13	35	35	0.4	0.7	2.3	5	14	32
Toluene	3.39	27.3	61	3.40	38	66	1.1	2.7	4.8	7	17	42

*Slightly different for volatile organic chemicals
†NA—not available

posure distribution was estimated using the bootstrap technique (Decisioneering Inc., 1998; Montgomery & Runger, 1998). A two-step process was used. The first step used Monte Carlo sampling to randomly select a set of input to the model variable values and estimate an exposure value. This step was repeated 1,000 times to formulate one exposure distribution. The second step repeated this process 200 times to generate 200 exposure distribution curves. The median and 90th-percentile mean, standard deviation, and range values of the oral ADE distribution are reported here.

Oral exposures for population subgroups. Seven subpopulations (subgroups) were identified for subgroup exposure analysis. These subpopulation categories can be separated into two groups determined by personal and building variables. Personal variables included the exposed individuals' gender, age, ethnicity, education, and income. Building variables included the building type and age. Conclusions reached from hypothesis tests for exposure were also applicable to risk estimates because risk was directly proportional to exposure (explained in detail subsequently).

ADE values were calculated deterministically for each individual. Comparisons of oral exposure values for comparable subgroups were performed using nonparametric statistical tests—Mann Whitney tests with two subgroups and Kruskal Wallis tests with more than two subgroups. In both cases, locations (median) of respective distributions were tested. Two comparisons were considered for ethnicity category: minority/nonminority and Hispanic/non-Hispanic. Because of sample size limitations, this study compared only Hispanic/non-Hispanic groups. All NHEXAS border subjects were in the subgroup.

Oral exposures from tap and nontap drinking water. Oral exposures from tap and nontap (e.g., bottled or filtered) drinking water were calculated for each subject deterministically, and the difference in median values was tested statistically using the Mann Whitney test. Information obtained from daily diaries recording whether subjects drank either tap or nontap water was used to determine whether a subject was a tap or nontap user.

The proportion of oral exposure attributable to tap and nontap water was estimated by comparing distributions of exposure from tap and nontap water. Median, mean, and 90th-percentile ADE values of tap and nontap water were used in a sample-size-weighted proportion calculation (Eqs 2 and 3).

$$\% \text{ Tap} = \frac{n_{tap} \times ADE_{tap}}{(n_{tap} \times ADE_{tap}) + (n_{nontap} \times ADE_{nontap})} \quad (2)$$

$$\% \text{ Nontap} = \frac{n_{nontap} \times ADE_{nontap}}{(n_{tap} \times ADE_{tap}) + (n_{nontap} \times ADE_{nontap})} \quad (3)$$

in which ADE is the average daily exposure and n is the sample size.

Risk estimates. This study used the hazard quotient (HQ)—a ratio of oral exposure and reference dose (RfD) for oral exposure—to estimate risk from oral exposure to chemical residues in drinking water for noncarcinogenic health effects. The HQ was calculated as follows (USEPA, 1999):

$$HQ = \frac{Exposure}{RfD} \quad (4)$$

in which exposure is a daily exposure value and RfD is the reference dose. Both nominator and denominator are expressed in units of μg/kg/d. ADE is used for exposure in the equation. HQ values ≥1 are of concern and indicate potential for health risk. The USEPA Integrated Risk Information

TABLE 4 **Analysis of probabilistic ADE* estimates**

	Arizona Population: ADE—*ng/kg/d*						Border Population: ADE—*ng/kg/d*					
	Median ADE			90th Percentile ADE			Median ADE			90th Percentile ADE		
Chemical Residue	Mean	SD†	Range	Mean	SD	Range	Mean	SD	Range	Mean	SD	Range
Arsenic	22.2	2.0	19.7–24.5	172.3	11.1	159–187	19.65	1.76	17.4–21.6	186	14	168–205
Chromium	18.1	1.1	16.7–19.3	148.6	12.5	121–147	19.41	1.15	17.9–20.8	143	12	129–157
Lead	6.4	0.3	6.1–6.7	26.8	1.6	24.8–29.0	7.49	0.61	6.8–8.4	104	12	89–118
Nickel	15.3	1.3	13.6–17.1	138.5	11.6	123–152	25.22	1.63	23.3–27.3	177	14	159–194
1,2-dichloroethane	7.6	0.4	7.1–8.0	34.8	2.2	31.7–37.5	3.54	0.21	3.3–3.8	17	1	15.9–18.4
1,3-butadiene							0.37	0.02	0.35–0.39	1.4	0.1	1.3–1.5
Dichloromethane	0.6	0.1	0.53–0.65	7.6	0.8	6.5–8.6						
Chloroform	1.2	0.1	1.0–1.3	36.2	5.0	30–43	4.90	0.29	4.6–5.3	32	3	29–35
Toluene	4.0	0.4	3.6–4.5	65.8	7.3	58–77	6.97	0.39	6.5–7.5	39.7	2.7	36–43

*ADE—average daily exposure
†SD—standard deviation

System (IRIS) database was the source for reference doses. The database listed an RfD for select chemicals for chronic oral exposure for noncarcinogenic health effects. The RfD is a benchmark dose derived from the no-observed-adverse-effect level by consistent application of uncertainty factors to reflect various types of data sets used to estimate RfDs. A modifying factor is also used sometimes. The RfD is an estimate of a daily oral exposure to the human population that is likely to be without an appreciable risk of adverse effects during a lifetime (USEPA, 1999).

Risk for carcinogenic effects attributable to lifetime oral exposure was calculated for each individual using deterministically calculated ADE values. The following equation was used for carcinogenic risks (LaGrega et al, 1994).

$$R = CDI \times SF \tag{5}$$

in which R is the probability of excess lifetime cancer from oral exposure to a chemical, CDI is the chronic daily intake of the chemical (ng/kg/d), and SF is the slope factor of the chemical $(\text{ng/kg/d})^{-1}$. In this study, the slope factors were also obtained from the IRIS database. The calculated ADE values were equivalent to the potential dose (ng/kg/d) values, which were used as CDI in Eq 5.

Of the chemical residues studied here, only arsenic, chloroform, dichloromethane, and 1,2-dichloroethane are classified as carcinogenic. This study estimated the following statistics: median, mean, and 90th-percentile risk values for carcinogenic and noncarcinogenic risk.

Chemical ranking. Five methods were used to rank residues. Method 1, ranking by residue concentration, was carried out by estimating the proportion of population with residue concentrations greater than the drinking water guideline values of the state of Arizona or USEPA (the smaller the portion, the lower the rank). Methods 2 and 3 ranked the chemicals from low-to-high median and low-to-high 90th-percentile risk values for noncarcinogenic and carcinogenic risks, respectively. Residue chemicals were also ranked by reference dose (method 4) and slope factor values (method 5).

RESULTS AND DISCUSSION

Results and discussions are organized under three main sections: exposure assessment, risk assessment, and synthesis, which includes chemical rankings and comparisons of Arizona and border populations. Because the border database was partial (data from only ~60% of households could be used), results for this population are preliminary.

Exposure assessment. ***Chemical residue concentrations.*** All residue concentrations had a right skewed distribution. Lognormal distribution fit best to concentrations of all four VOCs and two metals (chromium and lead) of NHEXAS Arizona data and to two VOCs (chloroform and toluene) and three metals (chromium, lead, and nickel) of NHEXAS border data. For the other metals, i.e., arsenic and nickel, in the NHEXAS Arizona database, the best-fit distributions were gamma and Weibull, respectively. For the border database, Weibull was the best-fitting distribution to concentrations of 1,2-dichloroethane and 1,3-butadiene, and gamma was the best fit to arsenic concentrations. Both metal and VOC concentrations were higher for the NHEXAS Arizona subjects than for the border populations.

Median metal concentrations in tap water were higher than in nontap water for both Arizona and border populations (Table 1). VOC concentrations were generally at about the same levels for tap and nontap water for both Arizona and border groups. Tap and nontap residue con-

TABLE 5 **Results of hypothesis testing on subgroup ADE***

Category	Subgroups	Sample Sizes‡	Arizona Population—*p-value*†							
			Arsenic	Chromium	Lead	Nickel	1,2-dichloroethane	§	Chloroform	Toluene
Gender	Male/Female	46/87	0.99	0.50	0.62	0.17	0.78	0.28	0.68	0.93
Age	<18/≥18	13/120	0.58	0.66	0.04	0.23	0.15	0.55	0.49	0.90
Ethnicity	Hispanic/ Non-Hispanic	50/83	0.06	0.88	0.07	0.17	0.44	0.001	0.12	0.21
Education	≤ High school/ ≥College	62/71	0.39	0.56	0.24	0.83	0.39	0.35	0.06	0.28
Income	<$20K/≥$20K	32/97	0.34	0.94	0.14	0.02	0.27	0.01	0.82	0.15
Structure type	Mobile home/ House/Apartment	7/121/5	0.58	0.55	0.83	0.77	0.14	0.08	0.06	0.89
Year structure built	≥1980/1960–79/ ≤1959	40/52/34	0.097	0.40	0.54	0.15	0.93	0.61	0.99	0.20

Category	Subgroups	Sample Sizes‡	Border Population—*p-value*†							
			Arsenic	Chromium	Lead	Nickel	1,2-dichloroethane	§	Chloroform	Toluene
Gender	Male/Female	9/25	0.38	0.52	0.06	0.59	0.15	0.38	0.41	0.63
Age	<18/≥18	1/33								
Ethnicity	Hispanic/ Non-Hispanic	26/8	0.90	0.17	0.64	0.31	0.06	0.32	0.57	0.83
Education	≤ High school/ ≥College	16/18	0.18	0.35	0.13	0.09	0.27	0.09	0.96	0.28
Income	<$20K/≥$20K	15/17	0.30	0.49	0.45	0.55	0.02	0.25	0.46	0.27
Structure type	Mobile home/ House/Apartment									
Year structure built	≥1980/1960–79/ ≤1959	8/12/12	0.72	0.21	0.64	0.42	0.40	0.66	0.50	0.19

*ADE—average daily exposure

†*p*-value for Mann Whitney test when two subgroups were tested, for Kruskal Wallis when three subgroups were tested

‡There were slight differences among chemicals.

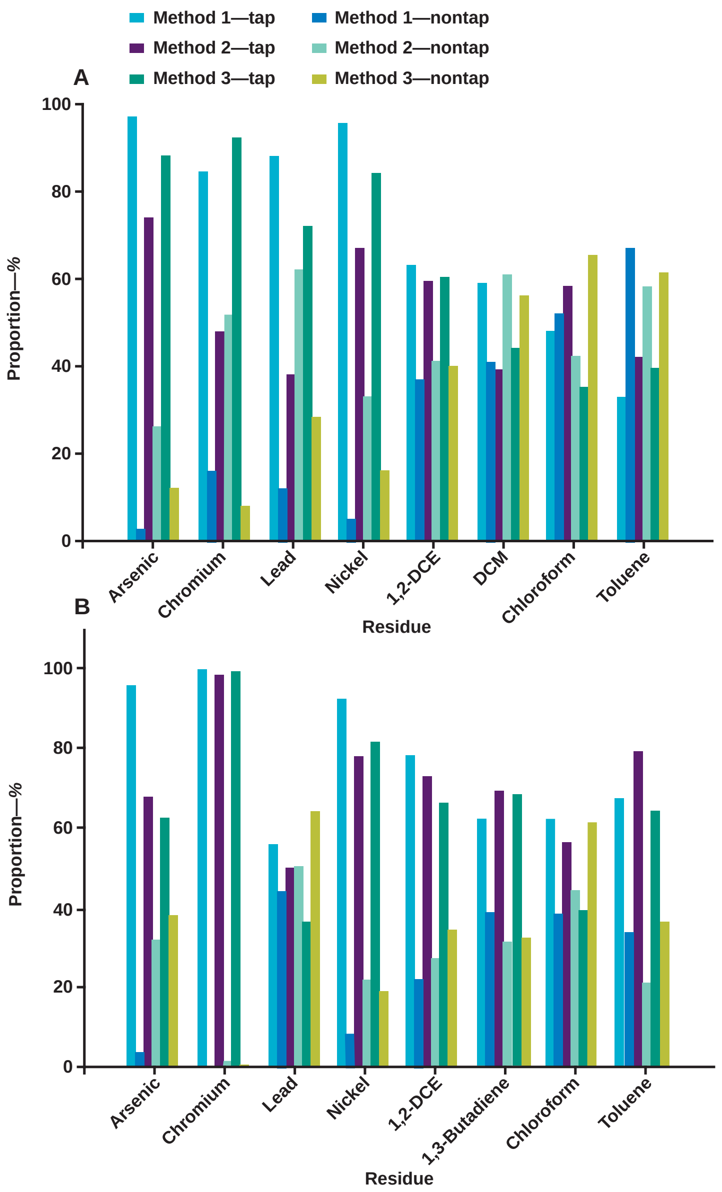
§Dichloromethane for Arizona, 1,3-butadiene for border

centrations of NHEXAS Arizona subjects differed significantly for all four metals and toluene with the Mann Whitney test. Differences between median tap and nontap water concentrations were significant only for arsenic, chromium, and nickel in the border study.

Chemical residue concentrations were compared with Arizona state guideline concentrations and USEPA drinking water limits or recommended levels. The number of sample subjects who exhibited drinking water concentrations greater than the lower of these two values (LOC) was not higher than one per chemical in the NHEXAS Arizona study. No border sample subjects showed residue concentrations higher than LOC values. Table 2 summarizes the guideline values and percentage of subjects who had concentrations greater than these values. Estimated proportions of Arizona and border populations that have residue concentrations greater than LOC values were <1% except for 1,2-dichloroethane; NHEXAS Arizona subjects had residue concentrations of 2.3% for this chemical. In general, these proportions were higher for the Arizona general population than for border communities. Arizona had smaller proportions only for arsenic and 1,2-dichloroethane.

The TEAM study (Wallace et al, 1984) reported that chloroform concentrations in tap water of nine New Jersey homes ranged from 11 to 225 μg/L with a median of 128 μg/L. These concentrations were high compared with concentrations measured in this study; however, the TEAM study was performed in the early 1980s. In the TEAM study, 1,2-dichloroethane concentrations (range = 0.005–0.05 μg/L and median = 0.02 μg/L) were lower than those of this study. Concentrations of chloroform and 1,2-dichloroethane for three North Carolina homes reported by Wallace and colleagues were similar to those found in the New Jersey homes. The National Resources Defense Council estimated that 34 million Americans drink water supplied by systems containing arsenic at levels that pose unacceptable cancer risk (Fuhs, 2000). Arsenic con-

FIGURE 1 Proportions of ADE attributable to tap and nontap drinking water for Arizona (A) and border populations (B)



ADE—average daily exposure, DCE—dichloroethane, DCM—dichloromethane

centrations in New Hampshire drinking water ranged from 0.002 to 66.6 mg/L with a geometric mean of 0.29 mg/L (Karagas et al, 2000). In a study of Maryland drinking water (Ryan et al, 2000), arsenic and lead levels ranged from <0.2 to 13.8 mg/L and from <0.1 to 13.4 mg/L, respectively.

During a two-year period in 1976–77, researchers investigated first morning tap water that had been standing overnight in residential plumbing and measured lead concentrations in the drinking water at 50 mg/L. Occupants of such households with medium or high levels of tap water ingestion had progressively higher patella lead levels than did those with low levels of ingestion. The authors of the study concluded that ingestion of lead-contaminated water was an important predictor of elevated bone lead levels later in life (Potula et al, 1999).

Lead concentrations of both NHEXAS Arizona and border households were comparable to those measured in United States and Europe. Watt and co-workers (2000) reported that most of the households in Glasgow were supplied by drinking water with lead concentrations <10 µg/L. However, 17% of the Glasgow homes had drinking water concentrations >10 µg/L, and 2.1% had concentrations ≥50 µg/L. In comparison, 90% of the NHEXAS Arizona subjects had drinking water lead concentrations ≤1.76 µg/L.

Drinking water consumption. Results of the dietary questionnaire indicated that drinking water consumption by sample subjects was normally distributed for each of the four days surveyed. The sample size responding to the questionnaire was 133 subjects for the Arizona study and 81 for the border sample. The difference in mean drinking water consumption over the four days was not statistically significant ($\alpha = 0.10$) for both Arizona and border subjects. NHEXAS Arizona drinking water consumption had mean and standard deviation values of 0.92 and 0.42 L/d (0.24 and 0.11 gpd), respectively. Mean and standard deviation values for NHEXAS border drinking water consumption were 0.95 and 0.47 L/d (0.25 and 0.12 gpd), respectively. Body weights of subjects were normally distributed, with a mean of 69.7 kg (153.7 lb) and standard deviation of 18.2 kg (40.1 lb) for NHEXAS Arizona subjects and 71.6 kg (157.9 lb) and standard deviation of 15.4 kg (34.0 lb) for border subjects. The null hypothesis of equal means for drinking water consumption and body weight of Arizona and border subjects could not be rejected ($\alpha = 0.10$). The fact that the two populations were similar in terms of body weight and drinking water consumption suggested that any difference in exposure and risk estimates between the two regions would be attributable mostly to chemical residue concentrations.

ADE. Like residue concentrations, oral ADE to all investigated chemi-

TABLE 6 Descriptive statistics of risk from oral ADE* via drinking water

	Risk for Arizona Population						Risk for Border Population					
	Deterministic			Probabilistic			Deterministic			Probabilistic		
Chemical Residue†	Median	Mean	90th Percentile	Median	Mean	90th Percentile	Median	Mean	90th Percentile	Median	Mean	90th Percentile
Arsenic (C)	2.6E-5	8.3E-5	2.3E-4	1.5E-5	4.9E-5	1.1E-4	1.3E-6	1.1E-5	1.2E-4	1.3E-5	4.4E-5	1.2E-4
Arsenic (NC)	6.0E-2	1.8E-1	4.8E-1	1.1E-3	3.2E-3	8.7E-3	2.8E-3	2.5E-2	8.0E-2	9.8E-4	3.3E-3	9.2E-3
Chromium (NC)	2.0E-3	2.1E-2	3.0E-2	1.0E-2	2.0E-2	5.0E-2	4.0E-4	4.5E-2	7.0E-2	6.5E-3	2.1E-2	4.7E-2
Nickel (NC)	9.5E-4	2.5E-3	6.1E-3	1.0E-3	2.6E-3	6.7E-3	8.5E-5	2.5E-4	7.0E-4	1.2E-3	3.9E-3	8.8E-3
1,2-dichloroethane (C)	2.2E-7	7.7E-7	2.5E-6	8.6E-5	1.7E-4	4.0E-4	1.9E-8	8.1E-8	2.1E-7	3.2E-7	6.3E-7	1.6E-6
1,2-dichloroethane (NC)	7.6E-7	1.7E-7	5.5E-6	1.6E-6	2.8E-6	6.1E-6	2.3E-7	9.4E-7	2.6E-6	7.7E-7	1.3E-6	3.0E-6
Dichloromethane (C)	4.6E-9	2.1E-8	5.6E-8	4.3E-9	3.0E-8	5.8E-8						
Dichloromethane (NC)	1.0E-5	4.7E-5	1.3E-4	9.5E-6	6.7E-5	1.3E-4						
Chloroform (C)	2.6E-9	1.5E-7	1.5E-7	6.9E-9	2.1E-7	2.1E-7	2.2E-9	4.5E-9	1.4E-8	3.0E-8	8.6E-8	1.9E-7
Chloroform (NC)	4.2E-5	2.5E-3	2.5E-3	1.1E-4	3.5E-3	3.5E-3	3.6E-5	7.3E-5	2.3E-4	4.8E-4	1.4E-3	3.2E-3
Toluene (NC)	1.7E-5	1.3E-4	3.3E-4	2.0E-5	1.9E-4	3.3E-4	5.5E-6	1.4E-5	2.4E-5	3.5E-5	8.7E-5	2.1E-4

*ADE—average daily exposure
†C—carcinogenic risk, NC—noncarcinogenic risk

cals had right skewed distributions for both NHEXAS Arizona and border populations. Lognormal was the best-fitting distribution for ADE of six chemicals in the NHEXAS Arizona sample and of all eight chemicals in the NHEXAS border sample. Only arsenic and nickel ADE distributions had gamma distribution.

Table 3 shows descriptive statistics of deterministic and probabilistic oral ADE estimates. For NHEXAS border populations, distributions of deterministic and probabilistic estimates agreed on five of the eight chemicals; the two approaches resulted in different best-fitting distributions for arsenic, 1,3-butadiene, and 1,2-dichloroethane. When deterministic and probabilistic approaches were used for NHEXAS Arizona data, all chemicals had the same type of ADE distribution.

Median oral exposures from tap and nontap water were significantly different for all four metals.

Table 4 shows relevant statistics of probabilistically estimated median and 90th-percentile oral ADE values. This information was used to compare probabilistic estimates with deterministically calculated ADE values. The probabilistic approach generally overestimated exposures relative to deterministic estimates for the NHEXAS border data. However, the two approaches were in general agreement for exposure estimates for the NHEXAS Arizona sample (Tables 3 and 4).

Exposures from tap and nontap water. Median oral exposures from tap and nontap water were significantly different for all four metals. Median oral exposures from tap and nontap water did not significantly differ for all four VOCs. Comparisons of median ADE values showed that exposures from tap water were as much as 44 times higher than exposures from nontap water for the NHEXAS Arizona sample and as much as 614 times higher for the NHEXAS border sample. The difference in border median metal ADE values from tap and nontap water was not significant only for lead. The only significant difference in the border median ADE to VOCs was for 1,3-butadiene.

Figure 1 shows the estimated proportions of the two populations' ADE values attributable to tap and nontap water. In general, the proportion of ADE values attributable to tap water was larger than those attributable to nontap water. The difference between the two proportions for VOCs was not as large as it was for metals.

Exposures of subpopulations. Table 5 shows the subgroups, sample sizes, and results of hypothesis tests. Among the subgroup categories tested, ethnicity was the subgroup that showed statistically significant differences in median exposures for the largest number of chemicals. Hispanics and non-Hispanics had statis-

TABLE 7 Chemicals ranked from best to worst

Arizona Population

Method 1	Method 2		Method 3		Method 4	Method 5
By %>LOC*	By Median HQ†	By 90th Percentile HQ	By Median CR‡	By 90th Percentile CR	By RfD§	By SF**
1,2-dichloroethane	Dichloromethane	Dichloromethane	Chloroform	Dichloromethane	Toluene	Chloroform
Dichloromethane	Toluene	Toluene	Dichloromethane	Chloroform	Dichloromethane	Dichloromethane
Chloroform	Chloroform	Chloroform	1,2-dichloroethane	1,2-dichloroethane	Nickel	1,2-dichloroethane
Chromium	Nickel	Nickel	Arsenic	Arsenic	Chloroform	Arsenic
Nickel	Chromium	Chromium			Chromium	
Arsenic	Arsenic	Arsenic			Arsenic	
Lead						
Toluene						

Border Population

Method 1	Method 2		Method 3		Method 4	Method 5
By %>LOC*	By Median HQ†	By 90th Percentile HQ	By Median CR‡	By 90th Percentile CR	By RfD§	By SF**
Toluene	Toluene	Toluene	Chloroform	Chloroform	Toluene	Chloroform
Lead	Chloroform	Chloroform	1,2-dichloroethane	1,2-dichloroethane	Nickel	1,2-dichloroethane
Chloroform	Nickel	Nickel	Arsenic	Arsenic	Chloroform	Arsenic
1,2-dichloroethane	Chromium	Chromium			Chromium	
Arsenic	Arsenic	Arsenic			Arsenic	
Chromium						
Nickel						

*%>LOC—proportion of subjects with residue concentrations higher than the level of concern
†HQ—hazard quotient
‡CR—carcinogenic risk
§RfD—reference dose
**SF—slope factor

tically significant differences for exposures to arsenic, lead, and dichloromethane for the Arizona population. Median exposures to nickel and dichloromethane were significantly different for income and structure-type subgroups. The difference in median dichloromethane and chloroform oral ADE values among structure-type subgroups was statistically significant. Differences in median ADE of age subgroups were significant only for lead. Table 5 shows results for structure-type, age, and income categories, even though there were large differences in sample size.

Subgroup comparisons were made only for subgroups that had a sample size sufficiently large enough for statistical comparisons. In general, the differences in median ADE of subgroups were not statistically significant. Median 1,3-butadiene and nickel ADE values were significantly different for education category subgroups. Median chromium ADE values were significantly different for ethnicity and family income category subgroups. Median lead ADE values were significantly different for gender category subgroups. Pair-wise comparisons of the median values for the building age (i.e., year building was built) category indicate that 1960–79 and 1959 or earlier subgroups had statistically different median ADE values for chromium and toluene ($\alpha = 0.10$). Differences in median NHEXAS Arizona ADE values for 1960–79 and 1959 or earlier subgroups were also significant for toluene at $\alpha = 0.10$.

Risk assessment. Carcinogenic and noncarcinogenic risks were calculated using ADE values for each chemical depending on availability of RfD for noncarcinogenic risk and slope factor for carcinogenic risk. Table 6 shows deterministic and probabilistic estimates of risk values.

Among the median values of carcinogenic risk calculated for NHEXAS Arizona subjects, only that for arsenic was >1.0E-6, the de minimis or acceptable risk level. The median and 90th-percentile carcinogenic risk values for arsenic were on the order of 10^{-5} and 10^{-4}, respectively. At the time of this analysis, the arsenic standard for arsenic in drinking water was 50 µg/L. Debate is taking place over whether this standard should be revised to 10 µg/L.

This potential change would not affect the risk estimates of this analysis. Probabilistic estimates of carcinogenic risk for arsenic were also on the order of 10^{-5} for the median value and on the order of 10^{-4} for the 90th-percentile value.

Although the median value of carcinogenic risk for 1,2-dichloroethane was below the acceptable risk, the 90th-percentile value was higher than this level. When estimated probabilistically, both the median and 90th-percentile values of carcinogenic risk were higher than the acceptable risk for 1,2-dichloroethane. Both median and 90th-percentile values for dichloromethane and chloroform were below the acceptable risk. Because a slope factor was not available in IRIS (2000), no risk assessment was performed for lead.

Estimated noncarcinogenic risks had HQ values <1, even in the 90th percentiles for all the chemicals. The HQ value closest to 1 was the 90th-percentile value for arsenic, with an HQ of 0.48. The next closest chemical was chromium with a 90th-percentile HQ value of 0.05. All other exposure levels were much less than the corresponding RfDs, producing noncarcinogenic risk levels of <1.0E-2. Risks attributable to oral exposures from tap and nontap water were significantly different for all four metals. Risks attributable to oral exposures from tap and nontap water did not significantly differ for all four VOCs.

For the NHEXAS border data, the deterministic median carcinogenic risk values were less than the acceptable risk of 1.0E-6 for 1,2-dichloroethane and chloroform but were higher than the acceptable risk for arsenic. Chloroform 90th-percentile deterministic risk values were less than the acceptable risk value. The 90th-percentile risk value of 1,2-dichloroethane was greater than the acceptable risk of 1.0E-6. Probabilistically estimated 90th-percentile carcinogenic risks were greater than the acceptable risk for arsenic and 1,2-dichloroethane and less than the acceptable risk for chloroform.

TABLE 8 **Comparison summary of Arizona and border populations**

	Arizona/Border Ratio			Mann Whitney
	Median	Mean	90th Percentile	*p*-value
Residue concentration				
Arsenic	2.00	1.11	0.69	0.09
Chromium	2.50	1.03	2.39	0.00
Nickel	17.62	1.71	1.08	0.00
Lead	3.62	2.71	2.08	0.00
1,2-dichloroethane	7.76	2.90	2.32	0.00
Chloroform	3.73	9.02	9.86	0.00
Toluene	2.13	2.45	3.03	0.22
Average daily exposure				
Arsenic	39.55	42.48	29.19	0.00
Chromium	7.44	0.87	15.83	0.00
Nickel	27.01	27.01	34.28	0.00
Lead	7.70	14.41	15.89	0.00
1,2-dichloroethane	8.08	7.50	7.60	0.00
Chloroform	1.19	35.47	42.09	0.11
Toluene	3.00	8.28	5.42	0.02
Carcinogenic risk				
Arsenic	39.39	25.57	28.90	0.00
1,2-dichloroethane	8.42	7.77	7.91	0.00
Chloroform	1.16	33.71	41.92	0.13
Noncarcinogenic risk				
Arsenic	48.67	45.45	30.00	0.00
Chromium	8.62	0.87	16.00	0.00
Nickel	28.21	34.18	33.50	0.00
Chloroform	1.17	34.25	40.45	0.12
Toluene	3.09	8.13	5.40	0.02

Median and 90th-percentile values for noncarcinogenic risks estimated using the deterministic approach were less than the LOC of HQ = 1 for all eight chemicals. The 90th-percentile noncarcinogenic risk values estimated using the probabilistic approach were also under the LOC for all eight chemicals. Because no RfD and slope factor value were available (IRIS, 2000), risk assessments could not be performed for 1,3-butadiene and lead in the NHEXAS border sample. The difference in median risk levels attributable to oral exposures from tap and nontap water was not significant for lead only. The only significant difference in median risk levels attributable to oral exposures from tap and nontap water to VOCs was for 1,3-butadiene.

Results (discussed in the exposure analysis section and shown in Table 5) for differences in exposure levels of population subgroups through hypothesis tests were valid for both carcinogenic and noncarcinogenic risk levels. This was because risk was a product obtained by multiplying oral exposure levels by a constant, i.e., the slope factor for carcinogenic risk and $(\mathrm{RfD})^{-1}$ for noncarcinogenic risk.

Synthesis. *Chemical rankings.* Among the chemicals of the Arizona study, arsenic was ranked the highest (worst) by all ranking methods except method 1, i.e., ranking by medians (see Table 7). Noncarcinogenic risks associated with metals ranked higher than those associated with VOCs.

For the Arizona data, ranking by methods 2–5 provided consistent results, but method 1 results differed from the other four ranking methods. Toluene, for example, was ranked the second best by method 2 but the second worst by method 1. Another example was 1,2-dichloroethane, which was ranked worse than dichloromethane and chloroform by method 3, whereas method 1 ranked dichloroethane better than dichloromethane and chloroform. Arsenic had the worst ranking by four of the five ranking methods for both the border study and the Arizona study. Chemical rankings by exposure and risk were the same for both the border and the Arizona study.

Only ranking by method 1 produced differences between the two population studies. Toluene and lead were ranked as the two best chemicals for the border population, whereas these two chemicals had the two worst rankings in the Arizona study. Chemical rankings were different for the two populations when method 1 was used; chemicals were ranked consistently with ranking methods 2–5.

Comparisons of Arizona and border results. Table 8 shows Arizona-to-border ratio values for median, mean, and 90th-percentile values and *p*-values for difference tests of median values of the two databases. As this table indicates, almost all residue concentrations in drinking water measured in the NHEXAS Arizona sample were higher than those measured in the NHEXAS border sample. Only the ratio for the 90th-percentile value of arsenic concentration was <1. Differences in median residue concentrations were statistically significant for all chemicals except toluene. Median chloroform ADE was only 1.2 times higher for the NHEXAS Arizona study than for the border study, and the difference was not significant. However, chloroform mean and 90th-percentile values for the two studies differed substantially.

Exposures to all other chemicals were larger for Arizona populations than for border communities. The only ratio with a value <1 was the ratio of the means for chromium noncarcinogenic risk for the two populations. The null hypothesis of equal median values for the two populations cannot be rejected for chloroform carcinogenic risks estimated by the slope factor method. Median arsenic and 1,2-dichloroethane carcinogenic risks were statistically different. Differences in median noncarcinogenic risk estimates for arsenic, chromium, nickel, and toluene were statistically significant. The null hypothesis of equal medians for noncarcinogenic risk for Arizona and border communities could be rejected for all chemicals but chloroform.

CONCLUSIONS

Exposure and risk assessments for Arizona and border populations were performed using water samples of 133 and 34 households, respectively. For the border study, all chemical residue concentrations were below the lower guideline concentration values of both the state and USEPA. For the Arizona database, three residences (one residence per agent) had residue concentrations greater than the LOC for chromium, nickel, and dichloromethane. The estimated portion of population with concentrations higher than the lower guideline value was >1% only for 1,2-dichloroethane in the NHEXAS Arizona sample; the proportions for all other residues ranged from 0% to 0.7%. All oral ADE values and risks of chemical residues were lognormally distributed except for Arizona arsenic and nickel, which exhibited gamma distributions.

Results using the probabilistic approach were in general agreement with those from the deterministic approach for the Arizona study region. For the border study, however, the probabilistically estimated exposure and risk values were higher than those established by deterministic values. Estimation of individual exposures and risks on a person-by-person basis (i.e., the deterministic approach) should be preferred whenever data are available. Carcinogenic risks for all substances except arsenic were below the de minimis or acceptable risk level of 1.0E-6 for both NHEXAS Arizona and border populations. Although median carcinogenic risks were below the acceptable risk, 90th-percentile risk values of 1,2-dichloroethane for both Arizona and border populations were greater than the 1.0E-6 value. All noncarcinogenic risks were below the LOC (HQ = 1) for both populations.

Among population subgroups, ethnicity seemed to be the category with the greatest effect on oral exposure. ADE values for arsenic, lead, and dichloromethane in the NHEXAS Arizona study were significantly different for Hispanics and non-Hispanics. In the Arizona study, all categories other than gender had significantly different ADE values for at least one chemical. The only category with significant difference in median ADE values for more than one chemical was the education category in the NHEXAS border sample. Other categories had at least one chemical with significant difference except the category of building age (i.e., year structure was built).

Residue concentrations in tap and nontap drinking water and oral exposures and risks from tap and nontap water were significantly different. Arsenic was ranked highest (worst) and was the chemical associated with the highest risks for both Arizona and border populations. Metal concentrations, exposure, and risks were higher for tap water than for nontap water. Nickel, lead, and chloroform concentrations of tap water were statistically different in the two study groups. The null hypothesis of no difference between the two groups in chemical residues of nontap water could not be rejected. In general, average daily oral exposures to water residues and carcinogenic and noncarcinogenic risks were higher for Arizona subjects than for border subjects.

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FOOTNOTES

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