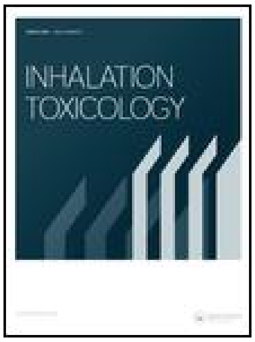

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INDOOR AND OUTDOOR PM10 AND ASSOCIATED METALS AND PESTICIDES IN ARIZONA

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The National Human Exposure Assessment Survey study in Arizona (AZ NHEXAS) sampled trace metals in multimedia in and outside of 176 representative homes in Arizona. PM10 was collected using low-flow impactors indoors and out. Primary metals evaluated from monitoring of indoor and outdoor air were lead, cadmium, chromium, nickel, and arsenic. Secondary metals were also evaluated. They were analyzed using gas chromatography–inductively coupled plasma/atomic emission spectroscopy (GC-ICP/AES). Air concentrations of metals do not contribute appreciable amounts to total concentrations of metals, and none were above levels of concern. Measurements were made of selected pesticides (primarily chlorpyrifos and diazinon) from particulate matter (PM10) filters obtained indoors and outdoors. Indoor air pesticide exposures represent about 25% of the total exposure to these pesticides. The highest 10% of exposures were related to questions about pesticide usage, at home and at work.

PM10 and lead are regulated by National Ambient Air Quality Standards. Indoor PM10 and lead are known to contribute a large amount to total exposure to both (as mentioned in the U.S. EPA Air Quality Criteria Document; U.S. EPA, 1986, 1992, 1996). In addition, other elemental metals can cause significant health effects. However, the population distributions of exposures to

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these metals in different media are unknown (Sexton et al., 1995). Inhalation of inorganic arsenic (As) is associated with respiratory, cardiovascular, neurological, teratogenic, and carcinogenic effects (ATSDR, 1993a). Human exposure to cadmium occurs through ingestion and inhalation (ATSDR, 1997a). Smokers experience double the daily exposure of nonsmokers to cadmium. Acute and chronic inhalation can result in damage to the lung including cancer, and chronic inhalation affects other organ systems. Chromium(III) and nickel are essential nutrients, but chromium(VI) and nickel exposures can cause adverse health effects; it is often assumed that total Cr can thus cause such adverse health effects. Among the sensitized, exposure to chromium or nickel provokes dermal and inhalation allergy, chronic bronchitis, and reduced lung function (ATSDR, 1993b, 1997b). Long-term inhalation of both metals is associated with an elevated risk of lung cancer (ATSDR, 1993b, 1997b). Daily exposure to all of these elemental metals is common.

Chlorpyrifos and diazinon have widespread use (Lewis et al., 1988; Whitmore et al., 1994; Buckley et al., 1997). These organophosphate (OP) pesticides are known to have toxic endpoints and may have carcinogenic endpoints. OP insecticides accounted for one-third of all reported pesticide poisonings in the United States in 1990, and diazinon and chlorpyrifos accounted for 50% of the OP reports (Kamrin, 1997). Acute toxicity from OPs involves acetylcholinesterase inhibition, with multiple organ system effects (Salem & Olajos, 1988). Low-dose chronic exposure can lead to the same effects, and other neurological symptoms (Kamrin, 1997).

The study is primarily concerned with residential exposures, using monitoring, questionnaire and time-activity pattern data, as part of an attempt to obtain population distributions of exposure, by media and total. The specific objectives of the overall study have been elucidated before (Lebowitz et al., 1995). In this paper, the focus is on the efforts to document the occurrence, distribution, and some determinants of exposure to airborne PM10, metals, and pesticides in the general population.

METHODS

The National Human Exposure Assessment Survey (NHEXAS) study in Arizona employed a population-based probability design and contacted 1200 households (Lebowitz et al., 1995). Of these, 176 homes were targeted for intensive multimedia sampling. The study design, questionnaires employed, and field and laboratory analytical techniques were previously presented. PM sampling (for metals and pesticides) was carried out at 4 L/min with a personal sampler pump (model 224-PCXR8, SKC, Inc.). Outdoor air was sampled to give an integrated 24-h sample over a 3-day period using a timer for intermittent sampling. Indoor air was sampled in the same manner for an integrated 12-h period over 3 days, and personal air was sampled similarly to give an integrated 8-h sample over a 1-day period. These schedules were sufficient to measure PM10, and metals in prior studies and in chambers. Personal, fixed indoor, and fixed outdoor air sampling for pesticides was accomplished using the URG-200 sampler unit with 10-μm particle inlet, Teflon-coated glass fiber filter (25 mm diameter, Pallflex TA60A20), and polyurethane foam (PolyUrethane Foam; 25 mm × 76 mm) sorbent (Gordon et al., 1999). Because of the small number of personal and outdoor air samples with detectable levels of pesticides, such sampling and analysis was discontinued during the course of the project. It was determined that the levels of pesticides outdoors had degraded (from ultraviolet and other meterological phenomena) to the point of having mostly nondetects from outdoor air samples, and the personal air samplers were not sensitive enough for pesticides. PM10 air filters were weighed to calculate PM10 concentrations, x-rayed for metals, and shipped to Battelle for analysis. Pb, Cd, Cr, Ni, and secondary metals were evaluated by Battelle using inductively coupled plasma–atomic emission spectroscopy (ICP-AES). Detection limits and quality assurance methods were provided previously (Lebowitz et al., 1995).

RESULTS

For the 176 subjects, females were overrepresented, and the study population contains one Hispanic for every two non-Hispanics (consistent for each gender and all age groups and consistent with the decennial census data; Robertson et al., 1999).

The proportion of time spent indoors at home was 77–80% for those younger than 5 yr and older than 65 yr (the remainder being mostly outdoors); it was 63% for the others. Work or school time was 10-12% and transit was 5–8% for those aged 6–65 yr.

The cumulative distributions of PM10 mass from impactor sampling are shown in Figures 1 (for outdoors) and 2 (for indoors); the 90th percentiles were 48.2 μg/m^3, respectively.

Metals

Lead and calcium went undetected in air. Indoor nickel, zinc, and barium were detected in about 1% of the samples (with maxima of 15.5, 1.8, and 0.3 μg/m^3, respectively).

Chromium had no values at or below the 95th percentile, and its maxima were 0.29 μg/m^3 indoors and 0.45 μg/m^3 outdoors. Outdoor manganese did have a 90th percentile (0.05 μg/m^3); its maximum outdoors was 0.21 μg/m^3; indoors the maximum was 0.12 μg/m^3.

Arsenic was detected in about 30% of the indoor and 32% of the outdoor air samples; the 75th percentiles were 0.004 μg/m^3 and 0.006 μg/m^3, respectively; the 90th percentiles were 0.008 μg/m^3 and 0.01 μg/m^3, respectively; and the maxima were 0.022 and 0.026 μg/m^3, respectively. These results indicate that exposure to metals from air is very low for the studied population. For arsenic, residence in mining towns delineated half of those in the upper

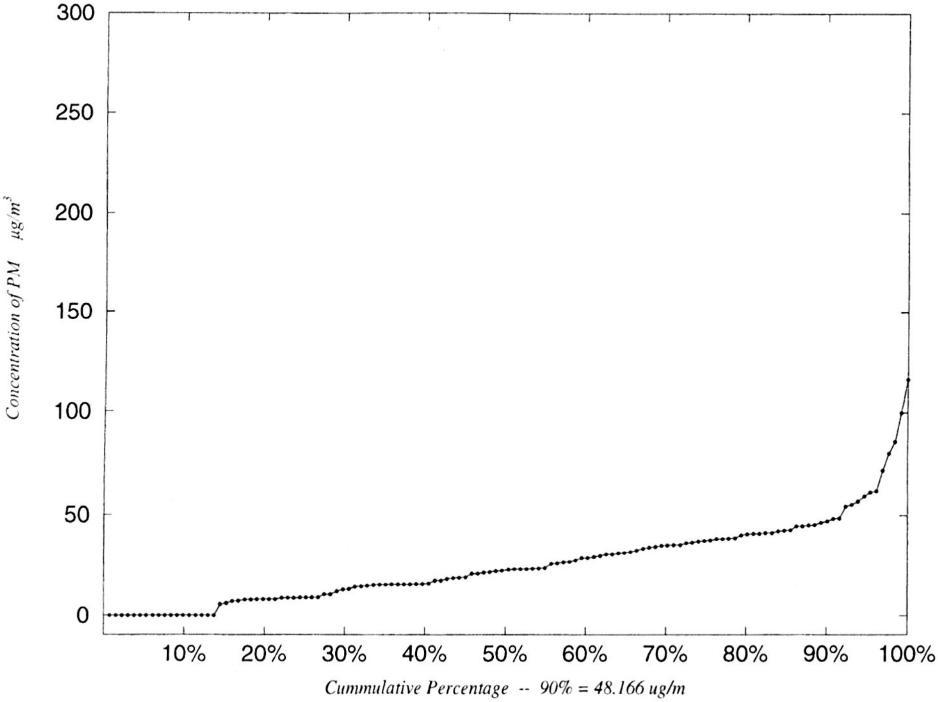

FIGURE 1. Cumulative distribution of outdoor PM10.

90th percentile of exposure. The other metals were also distributed geographically in a way that will contribute to knowledge of sources. None of the air values exceeded Arizona standards.

Comparisons of metals in smokers versus nonsmokers and smoking versus nonsmoking homes (18% homes with smoking) (using nonparametric comparisons, $p < .05$ for significance) showed that smokers had higher values than non-smokers for blood Pb, Cd, and urinary Cd; homes with smokers had higher levels of indoor PM10 than homes without smoking; and there were no significant differences in Cd or Pb in other media between two groups. In regard to tobacco smoke exposure in nonsmokers, there were detectable but not statistically significant differences in blood Pb, Cd, and urinary Cd between nonsmoking residents of smoking homes ($n = 25$) and of nonsmoking homes ($n = 101$). However, for ages less than or equal to 16 yr ($n = 33$), there were detectable and statistically significant greater concentrations of blood Pb, Cd, and urinary Cd for children who reside in homes with smoking ($n = 10$) than those who reside in households without smokers ($n = 23$) (S. Rogan, thesis, unpublished).

Pesticides

Chlorpyrifos and diazinon indoors had a 63–65% detection rate. The distributions were at best lognormal. The 50th percentiles were 10.7 ng/m^3 and

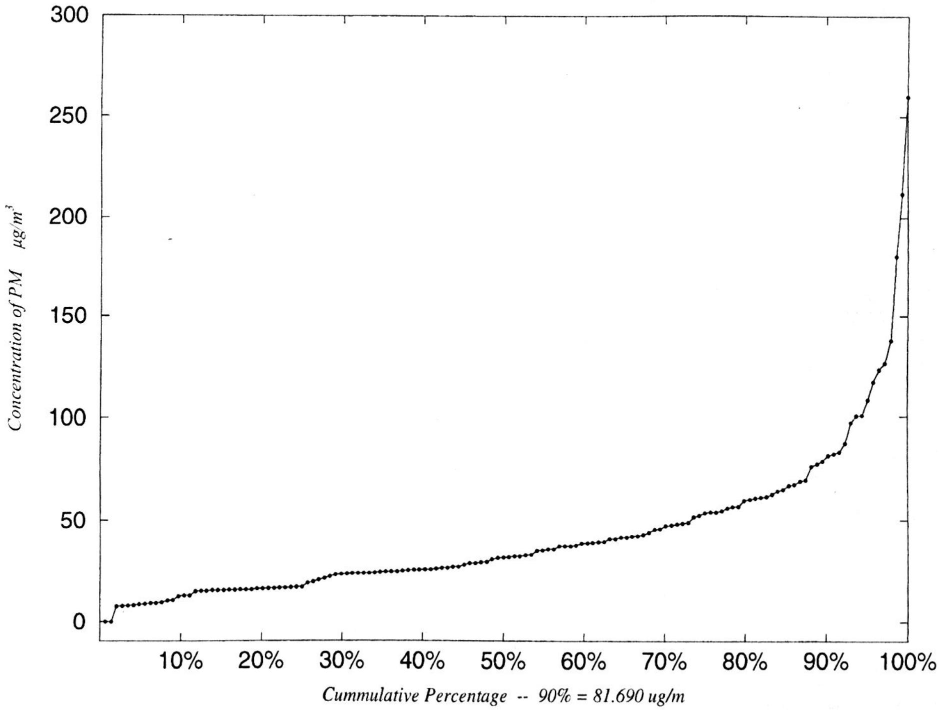

FIGURE 2. Cumulative distribution of indoor PM10.

7.3 ng/m^3, respectively. The 90th percentiles were 78.8 ng/m^3 and 58.3 ng/m^3, respectively. The 99th percentiles were 155.1 ng/m^3 and 124.1 ng/m^3, respectively. Preliminary analysis indicates that the contribution to total exposure from indoor air exposure for chlorpyrifos is at least 25% (for an adult male). Further, the top 10% of those exposed had questionnaire responses indicating pesticide usage either at home and/or at work recently.

DISCUSSION

The relative metal analyte contributions from different media are quite different (O'Rourke et al., 1999). Almost all fixed-site air samples (indoors and out) were below the detection limit or, when detected, values were in the nanograms per cubic meter range. Lacking compelling evidence related to particle deposition site, population susceptibility, association with a specific disease, or pharmacokinetics of a given analyte, the importance of air concentrations to total metal concentrations leading to total exposure may be limited.

For the pesticides, the median levels found in indoor samples agreed well with other studies, although the levels corresponding to the upper 0.1–1% of the population were considerably higher than levels reported elsewhere (Gordon et al., 1999).

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