Within- and Between-Person Variation in Environmental Concentrations of Metals, PAHs, and Pesticides Measured in NHEXAS-MD

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1 Issue

- Environmental exposure is often assessed using a single measurement per individual, but exposure concentrations can vary greatly from day to day and season to season.
- Greater attention to exposure variability can lead to more meaningful characterization of exposure and should be considered in the planning of future exposure studies.
- Current information on variance components is sparse.
- Quantifying the between- and within-person components of variance provides guidance for apportioning resources between numbers of subjects and numbers of repeated measurements and can reduce measurement error.

2 Approach

- We partitioned variance into between- and within-person components to calculate Intraclass Correlation Coefficients (ICC) and examine consistency across compounds within chemical classes and media.
- Data from NHEXAS-MD, a longitudinal study with up to six sampling events per household (N=80) over one year (Table 1), were analyzed.
- Previous analyses of the NHEXAS-MD data have reported significant temporal (within-person) variability in exposure, but analyses were limited to a small subset of the target chemical compounds.
- We extend the analyses to all primary chemicals in each of the three classes measured in multiple media (metals, PAHs, and pesticides).

3 Methods

Table 1. Sample collection and analysis methodology for NHEXAS-MD.

Media	Collection	Sampling Methods	Chemical Analysis
Air	Days 1-8	Intermittent sampling (10 of every 70 min) at 4 L/min. Metals at inhalable fraction (<10 µm) by inertial impactor onto cellulose filters. Pesticides/PAHs with quartz filter/ polyurethane foam for particulates and vapors. Outdoor air Pesticides/PAHs not measured.	Inductively coupled plasma-mass spectrum-etry (ICP-MS) for metals. Soxhlet extraction followed by gas chromatography/mass spec-trometry (GC-MS) for pesticides and PAHs.
Dust	Day 1	House dust >5 µm in diameter collected from carpet (2 m^2) with high-volume small-surface sampler (HVS3). Particles >150 µm removed by sieve in the laboratory.	Extraction and analysis similar to air samples.
Soil	Day 1	Play area, garden, and foundation soil composited. Particles >150 µm removed by sieve in the laboratory. Not collected at all cycles.	Extraction and analysis similar to air samples.
Food and beverages	Days 3-6	"Duplicate plates" of all consumed foods and beverages collected separately in 1-gal polyethylene containers, refrigerated, and shipped on ice.	Homogenized samples analyzed for metals by ICP-MS and for pesticide residues (after extraction and clean-up) by gas-liquid chromatography with electrolytic conductivity detection.
Urine	Day 2 or 8	First morning void (approximately 250 mL) collected, shipped on dry ice to the Centers for Disease Control and Prevention (CDC) for analysis.	Metals analyzed by graphite furnace atomic absorption spectrometry (AAS). Pesticide metabolites analyzed by gas chromatography tandem mass spectrometry (GC/MS/MS).
Blood	Day 2 or 8	56 cc total collected in Vacutainer tubes by venous puncture in the home by licensed phlebotomist. Shipped on dry ice to CDC.	Metals in blood analyzed by graphite furnace atomic absorption spectrometry (AAS). Pesticides as in urine.

Total variability was partitioned into within- and between-person components using the following linear mixed-effects regression model (Equation 1):

$$Y_{ij} = \ln(X_{ij}) = \mu_y + \beta_i + \varepsilon_{ij} \qquad (1)$$

for $i = 1, 2, \ldots, k$ individuals, and
for $j = 1, 2, \ldots, n_i$ measurements of the ith individual,

where X_{ij} represents the measurement for the ith individual on the jth day, and Y_{ij} is the natural log-transformed value of X_{ij}. In this model, μ_y represents the true unknown mean, β_i represents the random effect for the i_{th} individual and ε_{ij} represents the residual error for jth observation on the ith individual. β_i and ε_{ij} are assumed to be independent and normally distributed with means of zero and variances of σ^2_B and σ^2_W, representing the between- and within-person components of variance, respectively.

The variance components were then used to estimate the "intraclass correlation coefficient" (ICC), using the following equation (Equation 2):

$$ICC = \sigma_B^2/(\sigma_B^2 + \sigma_W^2) \qquad (2)$$

The value of ICC ranges from 0 to 1, with higher values indicating that measurements for a given individual are consistent over time across repeated measurements.

4 Results

- We observed some consistency in ICCs among chemicals within compound classes in indoor air, housedust, and urine (Figure 1).
- In indoor air, pesticides had a higher ICC (0.86 ± 0.05, mean ± standard deviation) than PAHs (0.30 ± 0.09) or metals (0.06 ± 0.07). The same pattern was observed in housedust with ICCs of 0.66 ± 0.09, 0.49 ± 0.04, and 0.44 ± 0.11 for pesticides, PAHs, and metals, respectively.
- The relatively high ICCs estimated for pesticides in indoor air and dust and for PAHs in soil suggest that a reliable estimate of exposure can be obtained with relatively few measurements.
- The large range of ICCs for metals and pesticides in soil and blood suggest that variability is compound-specific in those media.

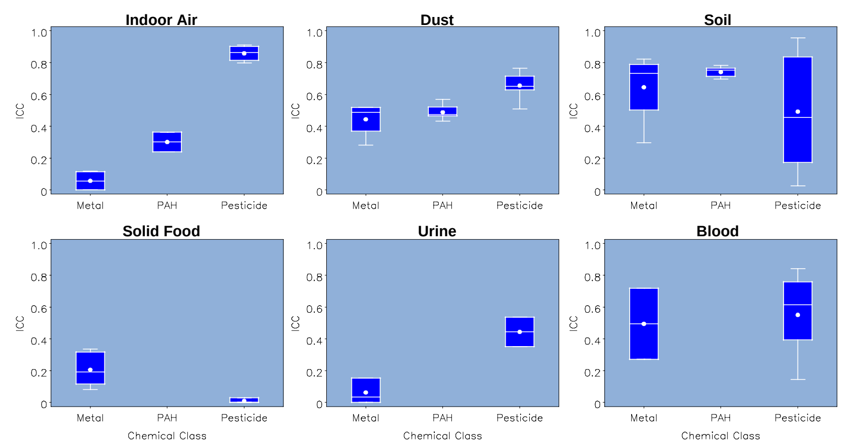
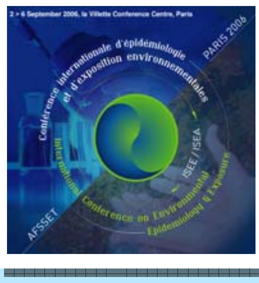


Figure 1. Box-and-whisker plots of ICCs by chemical class and sample matrix.

5 Discussion

- Quantification of variance components allows sampling schemes to be optimized for future human exposure studies. A low ICC (*e.g.*, metals in indoor air) indicates substantial temporal variability, with many repeated samples needed for a reliable estimate of exposure.
- These results suggest that where information on variance components for a specific metal, PAH, or pesticide in indoor air, housedust, or urine is not available, the variance components can be estimated based on compound class and sample medium.
- Variance components may, in turn, provide guidance in selecting sample size and in apportioning resources between numbers of subjects and numbers of repeated measurements.



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