Shaw, S., Scott, S., Pell, M., Raiche-Gill, J., Tomasallo, C., & Meiman, J. (2023). Lead source attribution by stable isotope analysis in child risk assessment investigations. *Journal of Environmental Health*, *85*(9), 8–19.

Corresponding Author: Summer R. Shaw, CSTE Applied Epidemiology Fellow, Bureau of Environmental and Occupational Health, Wisconsin Division of Public Health, Wisconsin Department of Health Services, 1 West Wilson Street, Room 150, Madison, WI 53703. Email: summer.shaw@dhs.wisconsin.gov.

Note. This supplemental file was submitted by the authors along with the respective peer-reviewed article and has been posted online due to space limitations at https://www.neha.org/jeh-supplementals. The *Journal of Environmental Health* did not copy edit this file, nor was it peer reviewed. The authors have provided this supplemental file as an extra resource should the reader want more information.

Supplemental Text: Additional Methods

Isotope Sample Preparation

All acid reagents used for laboratory procedures were Optima grade purchased from Fisher Scientific. Different sample types required slight modifications to digestion and preparation procedures prior to purification.

Tap Water

Tap water samples were collected in clean two-liter fluorinated ethylene propylene bottles. Two samples were collected: a "first draw" sample representing water that had been sitting in pipes overnight, and a "5-minute flush" sample after running the tap for five minutes. Approximately 1 liter of tap water was weighed into a clean 1liter polytetrafluoroethylene (PFA) jar and dried on a hotplate at 110°C.

Dust Wipes

Dust wipes were weighed into clean 30 mL PFA jars. 10 mL of concentrated (16M) HNO_3 were added to each container and then sat at room temperature overnight. Next, ten mL of 18.2 MΩ•cm H_2O were added to each container, sealed, and then digested overnight on a hotplate at 110°C. The HNO_3 solution was decanted into separate 23 mL PFA jars, and the wipe residue was transferred to a clean low density polyethylene centrifuge tube. Any remaining liquid was decanted into the 23 mL sample jar, and final solutions were dried at 110°C.

Pipe solder, pipes, paint, spices, and cosmetics

Lead service lines were present at three of the five properties and two were able to be sampled. Small portions of pipe solder, pipes, paint, spices, and/or cosmetics were weighed into 15 ml PFA jars. 5 ml of concentrated (16M) HNO_3 were added to each container and then sat overnight at room temperature. Samples were heated on a hotplate overnight at 110°C to digest. Digested samples were then dried at 110°C.

Soils

Soils samples were weighed into 15 ml PFA jars. 5 mL of concentrated (16M) HNO_3 and 2 mL of concentrated (29M) HF were added to each sample and sat overnight at room temperature. Next, samples were heated on a hotplate overnight at 110°C, and solutions were dried at 110°C. When applicable, NIST2709 (San Joaquin Soil) was included for quality control.

Whole Blood

Approximately 1 ml of whole blood aliquots (sometimes less due to small sample size) were weighed into 15 mL PFA jars using a low retention pipette tip. 5 mL of concentrated (16M) HNO_3 were added to each sample and left to react overnight at room temperature. Early samples in the study were then subjected to an additional digestion step involving 2 mL concentrated (16M) HNO_3 and 2 mL of concentrated (12M) HCl. This step was found to be unnecessary and subsequent samples were digested only using the initial HNO_3 step. Samples were then heated on a hotplate overnight at 110°C, and sample solutions were dried at 110°C. Whole blood reference materials (UTAK) were included for quality control.

Lead purification

All dried samples were dissolved in 1M HBr, and Pb was purified using the standard anion-exchange technique (Strelow and Toerien, 1966). BioRad Polyprep columns were loaded with 1.0 ml of previously cleaned AG1-X8 anion exchange, and additional washes of 18.2 MΩ•cm H_2O and 6M HCl were used to remove any remaining Pb. Columns were conditioned with 1M HBr, and the whole 1M HBr sample solution (typically 1 mL) was loaded onto the columns. The sample matrix was eluted using washes of 1 mL, 2 mL, 2 mL, and 8 mL of 1M HBr. Pb was removed and collected using two 1 mL washes of H_2O followed by two 6 ml washes of 1M HNO_3. The purified Pb solutions were evaporated to dryness in preparation for mass spectrometry.

Lead Isotopic Analysis

All isotopic analyses were performed on a Neptune Plus MC-ICP-MS (Thermo Fisher Scientific Inc., Waltham, MA). Purified Pb fractions were re-dissolved in 2% HNO_3, and samples were introduced into the plasma using one of two introduction systems. One setup used the standard quartz cyclonic spray chamber (standard introduction system), the standard nickel sampler cone, and H-series skimmer cone. Sensitivities using this setup range from ~80 - 120 V/ppm total Pb. The second setup used the Aridus3 (Teledyne Cetac) desolvating nebulizer system, the Jet series sampler cone, and X-series skimmer cone. Sensitivities using this setup range from 3000 – 4000 V/ppm total Pb. Tl was added to each sample to a Pb/Tl ratio ~ 3 to correct for mass bias using the exponential fractionation law and assuming $^{205}Tl/^{203}Tl = 2.387$. Pb and Tl isotopes were analyzed in static mode using 6 Faraday collectors; a seventh collector was used to monitor Hg interferences. isotope ratios are measured for 60 cycles of 8 second integrations. International isotopic reference material NBS981 was analyzed at the beginning of analytical sessions and periodically throughout. All samples, reference materials, and NBS981 bracketing standards were blank-corrected for on-peak baselines (2% HNO_3). Pb and Tl concentrations (in 2% HNO_3) in the final analyte solutions were diluted to ~ 100 ng g^{-1} (ppb) Pb + 20 ng g^{-1} Tl using the standard introduction system or ~ 5 ng g^{-1} (ppb) Pb + 1 ng g^{-1} Tl. Isotope ratios are normalized to the days average of NBS981 ratio for each individual analytical day with values reported relative to those NBS981 ratios determined by Thirlwall (2002).

Total Pb recovered from the sample processing and analysis was estimated by comparing the concentration of the diluted sample and the known concentration of NBS981 standard from the isotopic analysis. The concentration of the sample solution was then multiplied by the dilution factor to obtain the total Pb. The total Pb value was then used to calculate an estimated concentration (Tables 1–6, Supplemental Table).