

Sources of Pb contamination in well water and soil at former orchards in the Mid-Atlantic, USA

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Lead and strontium isotopes, rare earth element, and trace metal compositions were measured to infer the sources of anomalously high concentrations of Pb in residential water wells and soils in former fruit orchards in Pennsylvania. The results were used to characterize potential anthropogenic and geogenic sources of lead, to evaluate specific contributions from various distinct sources, and to assess the geochemical relationship between water, soil, bedrock, and anthropogenic materials.

Groundwater from residential wells, in-line well water filters, surface water, Pb-arsenical pesticides, orchard and garden soils, the Ordovician carbonate-dominated bedrock, and coal deposits in the region display variable $^{206}\text{Pb}/^{207}\text{Pb}$ (1.16862-1.2236), $^{208}\text{Pb}/^{207}\text{Pb}$ (2.43551-2.46570) and $^{87}\text{Sr}/^{86}\text{Sr}$ (0.709030-0.730596) isotope signatures. Waters have variable Pb (up to 200 ppb) and As (up to 11 ppb) contents, and mostly low REE contents (La <0.096 ug/L; Yb ~ 0.027 ug/L). Soils are enriched in Pb (up to 2452 ppm) and As (up to 450 ppm), which might be caused by past use of lead arsenical pesticides applied to orchard crops. Rare earth element contents of soils resemble those of bedrock; however, the abundances of Pb and As in bedrock are much lower than in the soils. Pb isotope signatures of all ground and surface waters closely match, however, the waters do not match the Pb (or Sr) signatures of bedrocks, soils, or lead arsenical pesticides. Residential well waters show a broad linear trend in Pb isotope composition and match Pb isotope signatures of sediments captured in water filters. Surface water matches the Pb isotopic signature of the residential well waters. Depth-constrained water samples obtained from new monitor wells sited in bedrock (Martinsburg Fm., Jacksonburg Limestone, and the Epler Member of the Beekmantown Group) generally overlap the trend of the residential well waters, although some samples extend to lower Pb isotope ratios. Soils and well waters plot in broad fields along contrasting slopes in standard Pb isotope diagrams; the isotope fields do not overlap. Contributions of soil-Pb are apparently limited to wells having shallow water-bearing zones and significant sediment content. As a consequence, neither soils nor bedrock can be the exclusive or dominant source of Pb in the majority of the residential or monitor well waters.

Comparison of the Pb isotope data for the well waters, orchard soils, bedrock, and regional and local anthropogenic sources suggests the lasting impact of industrial lead from past leaded gasoline use, power plants, and various near-by metal manufacturing and processing facilities (e.g., smelters, lead-acid battery production), and waste disposal activities. Thus, the groundwaters are thought to have a regional industrial Pb isotope signature before arriving at the former orchard areas. The Pb in water is thought to be derived by mixing of lead from various industrial origins together with a contribution of labile lead from bedrock, rather than from historical application of pesticide in the region.