Lead Isotopes as a Supplementary Tool in the Routine Evaluation of Household Lead Hazards

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The advent of magnetic sector inductively coupled plasma-mass spectrometry (ICP-MS) allows rapid, accurate, and precise measurement of lead isotopes in environmental and biological samples at a lower cost than traditional methods. This may increase the feasibility of including lead isotope measurements as a routine tool to identify household sources of lead exposure to children. Here, we present three household case studies to illustrate how lead hazard evaluations by an environmental specialist could be supplemented with routine lead isotope analyses of potential lead sources and blood. Sampling for lead isotopes was undertaken following the U.S. Department of Housing and Urban Development regulatory guidelines for the evaluation of lead hazards in housing, and with the consideration of minimizing the additional costs associated with lead isotope measurements. The range of isotopic ratios within a single residence was large enough to allow the characterization of different lead sources, particularly when both major (e.g., $^{207}Pb/^{206}Pb$) and minor (e.g., $^{206}Pb/^{204}Pb$) isotope ratios were considered. These cases illustrate the utility of the lead isotope method to identify main source(s) of lead exposure to the child; discard unlikely sources of exposure to the child; point to sources of lead to dust; and substantiate or refine the environmental assessment based exclusively on lead concentrations and loadings. Thus, a more effective evaluation of household lead hazards would likely benefit from considering *a*) lead concentrations and loadings in and around the household environment; *b*) all isotopic ratios of potential lead sources within that environment; and *c*) information about behavioral habits, as well as an evaluation of viable pathways of exposure to the child. *Key words*: blood lead, ICP-MS, lead exposure, lead hazard assessment, lead isotopes, lead paint.

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The elimination of leaded gasoline and the increasing public awareness about the dangers of lead exposure have resulted in a decrease in the blood lead (PbB) content of children in the United States. However, childhood lead poisoning remains a significant public health problem. Data collected between 1991 and 1994 indicate that approximately 890,000 children, or 4.4% of all 1- to 5-year-old children in the United States, possessed elevated blood lead levels (≥ 10 µg/dL) (*1*). These data are alarming because existing evidence indicates that even moderate elevations of lead during early development produce enduring cognitive impairment (*2,3*).

The reliable identification of the sources of lead exposure to children with blood lead levels above the Centers for Disease Control and Prevention guideline of 10 µg/dL (*4*) is critical to devising effective intervention strategies to reduce their lead intake. At highly elevated blood lead levels, it is likely that the environmental lead source is distinct and readily identifiable through an environmental hazard assessment of the household. For example, in urban areas when the child's blood lead level is > 25 µg/dL, the source of exposure is usually considered to be pica of leaded paint. In contrast, when blood lead levels are between 10 and 25 µg/dL, the sources of exposure are likely more diffuse and could include house dust from deteriorated paint or from soil tracked indoors (*5*). Because > 95% of lead-poisoned children have moderately elevated blood lead levels (10–25 µg/dL) (*6*) and are exposed to a variety of sources, identification and mitigation of the sources of lead intake have become increasingly more difficult.

Epidemiologic studies have identified the major environmental contributors of lead to children's blood and provided the basis for establishing health-based residential standards for maximum limits of lead on floors, windows wells and sills, and soils. A statistical analysis of data pooled from 12 epidemiologic studies in multiple communities indicated that lead-contaminated dust was the major source of lead exposure for children (*7*). However, the utility of this conclusion in the identification of sources of lead exposure in individual cases is limited for several reasons. First, dust lead loadings account for no more than 44% of the variance in blood lead levels (*8–12*). Second, household dust, though an immediate lead source, is nonetheless a pathway of lead exposure. Dust control could be an effective interim intervention strategy, but to make a residence lead safe, the contributors of lead to dust should be identified and mitigated. Third, controlled studies on the efficacy of dust control measurements showed that this type of intervention yielded moderate benefits for children with blood lead levels > 25 µg/dL (*13*), but contradictory results have been obtained in studies where children had average blood lead levels < 25 µg/dL and dust control and educational intervention took place (*14–19*).

Faced with the challenge of identifying the lead sources in the residence of a child with a blood lead level > 10 µg/dL, the lead hazard assessor has to rely on a questionnaire and measurements of lead loadings and concentrations to identify sources of exposure (*20*). The ability of the lead hazard assessor to identify and control the sources of exposure may be limited because the identification of lead sources is speculative and because meeting dust clearance standards is a temporary measure. Consequently, to maximize the effectiveness of often-limited intervention resources and to make the residence permanently lead safe, the ultimate sources of lead to dust should be identified and controlled.

The lead isotope methodology using thermal ionization mass spectrometry (TIMS) has been used to identify sources of exposure to humans (*21–28*). This technique is potentially the most accurate indicator of the source(s) of lead to the child because there are often measurable differences in lead isotope abundances between different sources

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of lead exposure. The isotopic composition of lead in the child's blood can often be traced to the isotopic fingerprint of the environmental source(s) of exposure. However, there are several limitations to the widespread use of this methodology: this approach is useful when the potential sources are few and isotopically distinct (*29*), and sample preparation for TIMS is relatively costly and time consuming. In addition, TIMS instruments are not readily available at certified commercial or governmental laboratories.

The use of magnetic-sector inductively coupled plasma mass spectrometer (ICP-MS) instruments may serve as a viable alternative for the routine application of lead isotopic composition measurements in lead hazard evaluation. Magnetic-sector ICP-MS measurements of environmental samples (soils, dusts, paints) are about four times less precise than those by TIMS (based on external sample measurement reproducibility), but are typically five times more economical. Furthermore, sample processing for ICP-MS does not require column chromatography for sample lead purification, and ICP-MS sample throughput is 5–10 times higher than TIMS throughput. These advantages, and increased availability of magnetic-sector ICP-MS instruments, are important benefits that can offset the poorer external precision of ICP-MS. Use of the more common 'older generation' quadrupole ICP-MS instruments to identify sources of household lead exposure via lead isotopes achieved only mixed success because the long-term external precision (> 1%, 1σ) and accuracy were generally too poor to allow the discrimination of lead sources on the basis of their isotopic fingerprints (*30,31*). Other studies conducted with quadrupole ICP-MS have reported better external precision of 0.35% (1σ) (*32*) and 0.3% (1σ) (*33*). Overall, the utility of the quadrupole instrumentation in the evaluation of household sources of lead exposure will depend on the spread of isotopic compositions observed in each particular case.

Here, the lead isotope method was applied in three household case studies to illustrate the benefits and limitations of this technique, when used within the context of current practices of routine household lead risk assessment. It would have been possible to refine the identification of household lead sources by including sampling of diet over consecutive days, sampling of indoor air with pumps, or dust collection schemes lasting for days. However, the extent and type of sampling that we used were in accordance with the U.S. Housing and Urban Development (HUD) guidelines (*20*) used to identify household sources of lead exposure on the basis of lead concentrations and loadings and routinely followed by a household risk assessor. This approach is justified because in order for this technique to become a supplementary, common, and effective tool in public health investigations, it should be economical and should not considerably increase the workload of environmental lead hazard assessors.

Materials and Methods

Approach

We have developed a new, simple method to measure lead isotopes in environmental and biological samples using a magnetic sector ICP-MS (*34,35*). Lead isotopic ratios of environmental samples measured by this method and by the traditional TIMS are in full agreement. Sample processing effort and time are substantially reduced and measurement time is short (minutes/sample) in comparison to TIMS analyses. The external measurement precision (i.e., sample reproducibility) of this ICP-MS method is < 0.2% (2σ) for the major ratios $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ in environmental samples. For comparison, the external precision of TIMS measurements on the same ratios in environmental samples is about 0.05% (2σ) (*36*). The external precision on the minor ratios (e.g., $^{206}Pb/^{204}Pb$) in environmental samples is 0.25% (2σ) by the ICP-MS method and 0.1% (2σ) by the TIMS method (*36*).

Lead isotope ratios of blood measured by both methods also agree, but less well for some ratios ($^{208}Pb/^{206}Pb$) (*32,34*), which is 0.33% heavier by the ICP-MS method. However, because there is no certified blood lead isotopic standard, it is not possible to assess which method is more accurate. This slight disagreement between both methods has no significant impact on the interpretation of the isotopic data presented here. The external precision of measured blood isotopic ratios using both methods is similar at ~0.2%–0.4% (2σ) (*37,38*).

Overall, the utility of this analytical tool will depend on the magnitude of the sample measurement error compared to the range in isotopic ratios in a single residence. In the cases presented here the lead isotopic composition measurement precision was not a limitation, but this could be different in other dwellings.

Subjects

Three cases of children who spent most of their waking hours at home and had blood lead levels > 15 µg/dL were identified by the Childhood Lead Poisoning Prevention Program (CLPPP) of Santa Cruz County, California, and referred to us for isotopic determinations of lead in household samples and in blood. These subjects were the first three cases with blood lead levels > 15 µg/dL identified after the collaboration was formalized with the CLPPP. Their residences were visited jointly by the author (R.H.G.) and a California certified lead hazard assessor. During this visit informed written consent was obtained and an interview was conducted according to the CLPPP standard questionnaire. This questionnaire evaluated the habits of the child, the time he/she spent in the different rooms, and the potential lead hazards in the household environment. Information was also gathered on the possibility of exposure via the parents through occupational sources and parental hobbies, as well as potential exposure from other sources such as a previous residence, other environments the child frequented, toys, cooking utensils (including lead-glazed ceramic-ware), folk remedies, idiosyncratic foods, and cosmetics.

Case 1. Case 1 involved a 5-year-old female who lived in Santa Cruz but had lived in Mexico from 1 to 3 years of age and in the United States thereafter. Over a period of 6 months, her blood lead level rose from 14 to 25 µg/dL and then decreased back to 16.2 µg/dL, stabilizing at 14 µg/dL 9 months after the first blood lead determination. Isotopic analyses were done on the two most recent blood samples (16.2 and 14 µg/dL). The family lived in a small two-room dwelling.

Case 2. Case 2 involved a 1.5-year-old male who lived in Santa Cruz and had a blood lead level of 15 µg/dL at the time of blood collection. Three and 6 months earlier the blood lead had been 19 µg/dL. He had lived all his life in the same house, which was estimated to be 65–70 years old. The most noticeable feature of the house was the deteriorated exterior lead-based paint.

Case 3. Case 3 involved a 1-year-old female who lived in the town of Watsonville, California, and had a blood lead level of 22 µg/dL. This child had an elevated blood lead level (16 µg/dL) at 9 months of age. Renovation of the house started when the child was 2 months old and was still ongoing at the time of this study, including scraping of exterior lead-based paint and sanding of leaded lacquer on woodwork inside the house.

Sample Collection

Sampling was done according to HUD guidelines. The number and types of samples collected were based on the responses obtained during the interview and on a visual inspection of the residence. The same sampling was done for lead concentration and for lead isotopes because we hypothesized that this would be the sampling protocol if the lead isotope technique were to be routinely used in household lead hazards evaluation. We collected paint samples from all deteriorated painted surfaces, as well as from intact

paints that appeared to contain lead, based on a visual inspection. Household dust was collected using a cyclone vacuum and from bare soil using a stainless steel trowel. No drinking water samples were taken because water lead concentrations in the Santa Cruz area are well below U.S. Environmental Protection Agency limits (15 µg/L) as reported by the Santa Cruz municipality to the California Department of Health Services. Venous whole blood samples (5–7 mL) were drawn into low-lead heparinized Vacutainer tubes (#367734; Becton-Dickinson, Franklin Lakes, NJ) by a certified phlebotomist at the clinic where the child received medical care. In all cases, samples were collected within 1 month after the home visit.

Analytical Techniques

All sample processing was conducted under trace metal-clean HEPA filtered air (Class 100) laboratory conditions using clean techniques (*39*). Laboratory water was high purity grade (Milli-Q system, 18MW-cm^2; Milliport Corporation, Burlington, MA), and acids were subboiling quartz double distilled. Laboratory-ware was acid cleaned following procedures described by Flegal and Smith (*40*).

All analytical procedures have been described by Gwiazda et al. (*34*). Briefly, environmental samples were transferred into 30 mL polyethylene bottles with 25 mL 0.5 N HCl, and leached at 25°C for 24 hr on a shaker. The leachate was filtered with a syringe through a 0.45 µm Teflon filter into a 15 mL Teflon vial, evaporated to dryness, and redissolved in 1 N HNO_3. Blood aliquots (0.5–1 mL) were transferred to Teflon vials, weighed, and evaporated to dryness. Dried blood was digested overnight with 2 mL hot concentrated HNO_3 in closed vials, evaporated to dryness, and diluted with 1 N HNO_3. All samples were diluted to approximately 10 ng Pb/mL for analyses by ICP-MS. Bismuth-209 (^{209}Bi) was added as an internal standard to the samples before injection into the instrument. Sample lead concentrations and isotopic compositions were measured simultaneously using a Finnigan MAT Element magnetic sector-inductively coupled plasma (ICP) mass spectrometer (ThermoQuest, San Jose, CA), measuring masses of ^{201}Hg (used to correct for ^{204}Hg interference of ^{204}Pb), ^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb, and ^{209}Bi. External standardization for lead isotopes was via the National Institute of Standards and Technology (Gaithersburg, MD) standard reference material 981 lead isotopic standard. Measurement errors are listed in the "Approach" section and are based on replicate measurements of blood and environmental samples.

Data Analyses

Data analyses and interpretation were performed based on graphical presentation and assessment. No formal statistical analysis was applied.

Results

Case 1

Lead hazards evaluation based on lead concentrations and loadings. Two paint samples (exterior window frame, #3, and child's bedroom window sill, #8; Figure 1A) had high lead content and were potential contributors to the child's elevated blood lead level. In particular, #8 was more accessible to the child and covered a friction surface (window jamb). The dust loading of the bedroom carpet (#2) next to this window, at 110 µg/ft^2, almost met HUD clearance standards of 100 µg/ft^2. Nevertheless the recommendation by the environmental health personnel was to control both paints and to thoroughly clean and vacuum horizontal surfaces.

Lead isotopes results. The lead isotopes results indicate that one of the main suspected sources of exposure (paint sample #8) was an unlikely contributor to the child's elevated blood lead level because its isotopic composition is distant from the blood isotopic compositions of either time, t_0 (PbB = 16 µg/dL) or 3 months later at time t_1 (PbB = 14 µg/dL) (Figure 1C). Major ratios of paint sample #3 are close to the blood ratio (Figure 1C); however, when the minor ratios (^{206}Pb/^{204}Pb vs. ^{207}Pb/^{204}Pb) are considered, neither paint sample (#3 or #8) matches the blood ratios (Figure 1C). Other than direct paint ingestion, the likely pathway of exposure from these sources to the child would be through household dust (#1 and #2). However, the isotopic composition of either dust sample does not match the blood isotopic composition in either isotope plot (Figure 1B,C). Isotopically, the closest samples to blood are soil (#4) and dust from the exterior doormat (#1), both isotopically indistinguishable. Thus, no single sample or combination of samples could account for the extreme isotopic composition of the blood. Alternatively, the blood isotopic value could be a result of lead intake from an even more extreme non-sampled source (i.e., one with lower ^{207}Pb/^{206}Pb and higher ^{206}Pb/^{204}Pb ratios) combined with any of the samples on the opposite side of the blood (e.g., soil or bedroom dust, with their higher ^{207}Pb/^{206}Pb ratios) in Figure 1B,C.

Conclusions. The preferred interpretation based on lead isotopes is that a source with a ^{206}Pb/^{204}Pb ratio higher than blood from either outside the home or from endogenous (skeletal) origin supported the high blood lead level of the child, although minor intake from any of the other measured sources in the house is possible and could not be discarded.

Case 2

Lead hazards evaluation based on lead concentrations and loadings. The environmental health personnel identified five paints with high lead content as potential contributors to the child's elevated blood lead level (Figure 2A). The lead hazards evaluation stated that the deteriorated exterior paints (#1, #2, #3) were the most likely source of lead to the child. Other paints (#5, #6, #9) were in good condition, although #10 was not. Household dust loadings (#7 = 450, #8= 2200, and #11= 1150 µg/ft^2) were above the HUD clearance standard of 100 µg/ft^2. The environmental health personnel recommendation was to

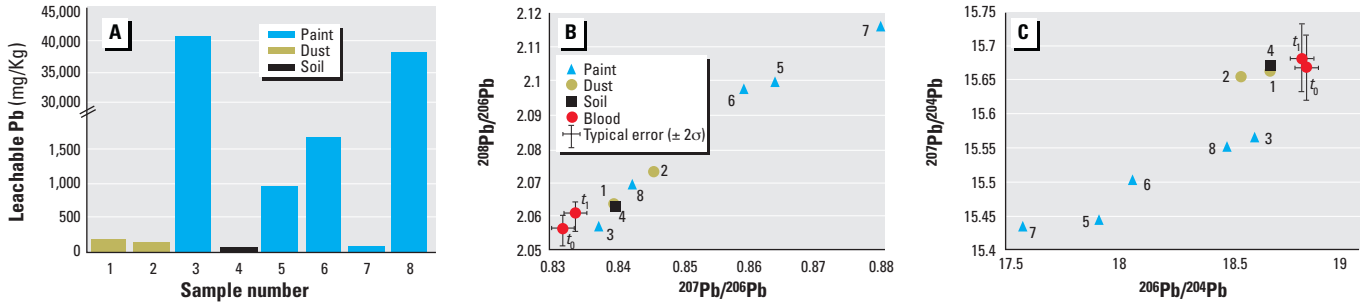

Figure 1. Lead concentration (*A*) and isotopic ratios (*B,C*) of environmental and blood samples from Case 1 [$PbB_{(t_0)}$ = 16.2 µg/dL; $PbB_{(t_1)}$ = 14 µg/dL]. Samples: #1, outside door mat; #2, child's bedroom carpet; #3, exterior window frame; #4, soil; #5, kitchen; #6, kitchen door; #7, dresser; #8, child's bedroom window. Sample numbers in (*A*) identify the samples in (*B*) and (*C*). Leachable lead in (*A*) refers to milligrams of lead leached per kilogram of sample.

control the exterior paint (#1) and to clean floors to meet clearance standards.

Lead isotopes results. Isotopes results support the environmental health personnel hypothesis that the main source of exposure was exterior paint because the blood isotopic composition plots close to those sources (#1, #2, #3) on both major and minor isotope plots (Figure 2B,C). An additional contribution of lead from a source possessing a higher $^{206}Pb/^{204}Pb$ ratio was also possible. The best candidate was dust #11, which exceeded clearance standards. Based on the isotopic ratios, the lead in dust #11 sampled in the immediate vicinity to the washer room (#10) appears to be a mixture of lead from exterior paint (#2, #3) and paint from the washer room (#10). Despite its relatively low lead content (0.1%), the washer room paint (#10) clearly contributed lead to the environment because of its very deteriorated condition. Other paints with high lead content were unlikely candidates because of their very good condition and because of the absence of friction points that might generate lead dust.

The soil lead isotopic value (#4) is relatively close to the isotopic composition of the high lead exterior paint (#2), but its lead content is very low (36 µg/g) compared to the HUD action limit of 400 µg/g for high-contact soil play areas. In contrast, the isotopic values of household dusts (#7, #8, #11) plot within the field delimited by the ratios of all suspected paints, and the dust lead loadings exceeded the clearance standard.

Conclusions. These data indicate that the source of exposure to the child was household dust that contained lead mostly from exterior paint tracked inside and from the washer room paint (#10). Isotope results support the lead hazard assessor's conclusions, but bring attention to an additional source of exposure (#10), and provide further insight into the pathway of exposure.

Case 3

Lead hazards evaluation based on lead concentrations and loadings. Environmental evaluation was conducted when the child was 1 year old, 3 months after the first elevated blood lead level was detected. Six paints with high lead content were identified (Figure 3A). The main suspected sources of exposure were the original exterior paint (#2) and the lacquer on woodwork inside the house (#7; 10% leachable lead content), which had been scraped and sanded during some minor renovation work. Most floors were recarpeted after this renovation. The rest of the lead-containing paints in the house were in fair or good condition, with the exception of washer room area paint sample #9. This paint covered walls that were in the process of being removed, and no precautions had been taken to minimize the release of dust into the room during renovation. Floor dust lead loadings in the washer room area where the paint was being removed and in the child's bedroom (#8) were 710 and 235 µg/ft^2, respectively, both exceeding HUD clearance standards. Dust in the main living area (#6) had lead loadings below clearance standards (70 µg/ft^2). The evaluation from the lead hazard assessor was that past lead exposure resulted from the sanding of exterior paint (#2) and woodwork lacquer inside the house (#7), and could be ongoing through intake of dust derived from paint #9 generated during the current renovation. The recommendation was to follow lead-safe practices in the ongoing remodeling work to prevent intake of household lead by the child.

Lead isotopes results. The lead isotopes results partially support the lead hazard assessor's conclusions about the past sources of exposure. Considering all isotopic ratios (Figure 3B,C), the sample with the closest ratios to blood is the woodwork lacquer (#7), whereas exterior paint (#2, a suspected source of past exposure) does not match the blood isotopic value. Other possible sources that match the blood isotopic composition in $^{208}Pb/^{206}Pb$ versus $^{207}Pb/^{206}Pb$ ratios do not have high lead contents (household dusts #6, #8) or are distant from the blood isotopic composition when the minor ratios are considered (#5, #6, #8) (Figure 3C). Exterior paint from the neighbor's house (# 4) is isotopically similar to the blood isotopic ratio;

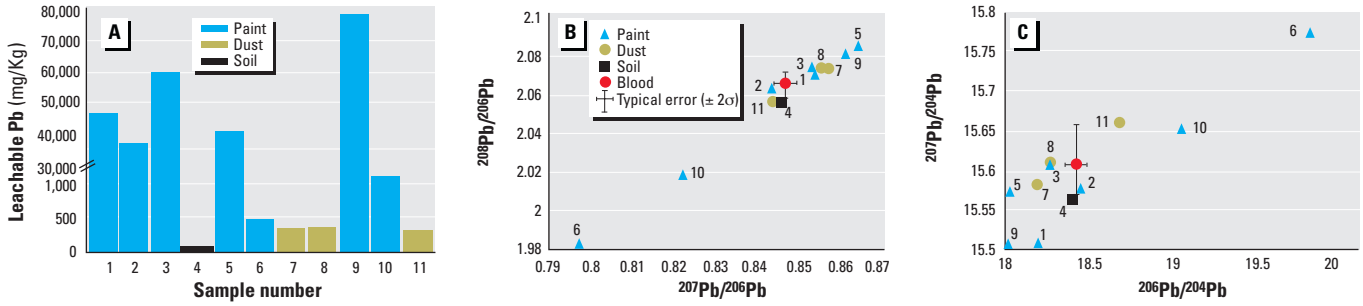

Figure 2. Lead concentration (*A*) and isotopic ratios (*B,C*) of environmental and blood samples from Case 2 (PbB = 14.9 µg/dL). Samples: #1, exterior; #2, exterior window frame (blue); #3, exterior window frame (green); #4, soil; #5, bathroom; #6, child's bedroom; #7, door mat (inside); #8, carpet; #9, eating area; #10, washer room; #11, kitchen mat. Sample identification numbers in (*A*) identify the samples in (*B*) and (*C*). Leachable lead in (*A*) refers to milligrams of lead leached per kilogram of sample.

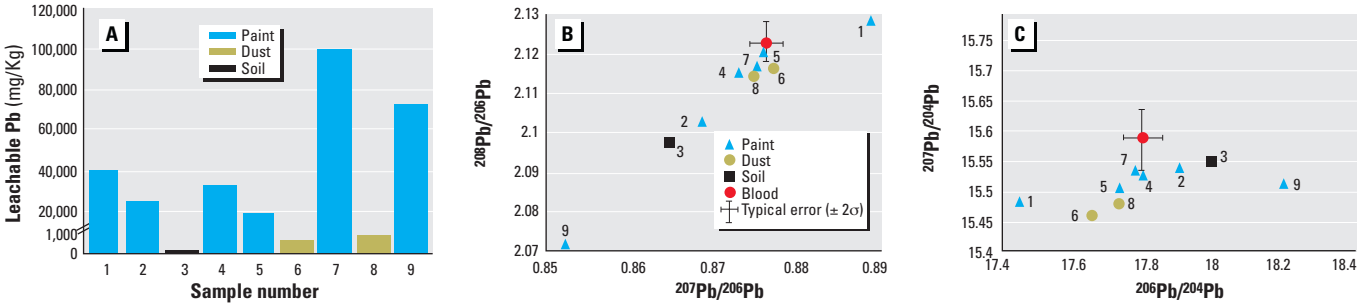

Figure 3. Lead concentration (*A*) and isotopic ratios (*B,C*) of environmental and blood samples from Case 3 (PbB = 22.4 µg/dL). Samples: #1, porch floor; #2, exterior; #3, soil; #4, exterior of house next door; #5, kitchen; #6, living room; #7, woodwork lacquer; #8, bedroom; #9, washer area. Sample identification numbers in (*A*) identify the samples in (*B*) and (*C*). Leachable lead in (*A*) refers to milligrams of lead leached per kilogram of sample.

however, the pathway of exposure to this source would have been indirectly through the soil (#3), which is isotopically different from both the paint and the blood. In contrast to the lead hazard assessor's evaluation, ongoing exposure to washer area paint #9 was unlikely because its isotopic values are very different from the blood values.

Conclusion. The lead isotope results confirm that the elevated blood lead level was due to past exposures. In addition, ongoing exposure to high lead sources within the house is not supported by the isotope data. Therefore, intervention aimed at controlling household lead sources would have no direct effect in reducing the child's blood lead level.

Discussion

With the emergence of the more economical and faster ICP-MS technology, the routine application of lead isotopes to investigate lead sources within household environments could become feasible and affordable. In this study, we illustrated the issues associated with this approach with three household case studies. This approach is aimed at providing an overview of the benefits and limitations of applying lead isotopes in the context of current practices in the evaluation of household lead hazards.

It has long been known that lead isotopes possess great potential as a diagnostic tool for the identification of environmental lead hazards. Traditionally, the use of lead isotopes in environmental investigations was limited to the identification of polluting lead sources on a geographical scale much larger than single household units (*41–45*). However, in some instances lead isotopes measured by TIMS were also used in individual cases of exposure. Most of these studies were successful in characterizing the source(s) of exposure for two reasons: they investigated residences of children with high blood lead levels where the source was distinct (*23*), or they conducted a very comprehensive investigation in which numerous samples were collected for each case. These included continuous collection of diet over a week and airborne house dust for a day or longer, in addition to detailed analyses of mineralogy and chemical speciation (*22,27,28*). This comprehensive approach may in general increase the success of the intervention, but it is not practical if it is used routinely and under the budget constraints normally encountered in lead exposure evaluation by public health personnel.

Lead isotopes are the most accurate tool available for the lead hazard assessor to identify sources of lead exposure because the isotopic fingerprint of a biological sample (e.g., blood) could be traced to the household source(s) of the lead exposure. For this approach to work, different household sources of lead must possess distinct isotopic compositions that are distinguishable within the error of the measurement. In other words, the difference in isotopic composition between the potential sources should be at least twice the measurement error for each source. In the magnetic-sector ICP-MS method used in this study two sources are considered isotopically distinct when their major and minor ratios are at least 0.4% and 0.5% apart, respectively. As shown in Figures 1–3, there are wide ranges of lead isotopic compositions within a single household. The range was as small as 4.5% (Case 3) and as large as 8.4% and 9.4% for the $^{207}Pb/^{206}Pb$ and $^{206}Pb/^{204}Pb$ ratios, respectively (Case 2). This provided a relatively high level of diagnostic "resolution" to distinguish samples and assess childhood exposures. Similar ranges in a single household were found by Yaffe et al. (*26*) (5.7%, $^{208}Pb/^{206}Pb$) and Rabinowitz (*23*) (4.6%, $^{207}Pb/^{206}Pb$; 4.1%, $^{206}Pb/^{204}Pb$), substantiating that these relatively large ranges measured here are likely to occur in other households in the United States.

However, the fact that potential lead sources (e.g., paint, soil, dusts) plot in linear arrangements with the biological (e.g., blood) samples does not necessarily allow the distinct identification of the actual lead sources to the child. This is because there is no certainty that all actual sources have been sampled and because there are multiple hypothetical combinations of sources that could yield the isotopic value of the biological sample. Even if the isotopic compositions of a single potential source and a biological sample agree, the isotopic composition of this biological sample could still be explained as the result of mixing of lead from two or more sources along the continuum of the linear arrangement.

There are two main reasons for the covariation of major isotopic ratios of industrially processed lead found in households (*29,46*). First, the isotopic compositions of all major ores, the ultimate source from which environmental lead was derived, fall very close to an average line in $^{208}Pb/^{206}Pb$ versus $^{207}Pb/^{206}Pb$ coordinates. Second, the increased use of recycled lead metal in the manufacturing of paint pigments and other industrial leads throughout the latter 1900s has gradually blurred the isotopic differences of lead across different batches of paint or other materials.

Lead recycling notwithstanding, there are still measurable deviations from co-linearity in the lead isotopic distribution of the minor ratios (vs. ^{204}Pb) observed in industrially processed lead found in households (e.g., all cases in this study). Thus the use of all lead isotopes in lead hazard assessment, and not just the major ratios, maximizes the information that can be gained with the application of this technique.

In these cases the minor ratios were helpful in refining the lead source identification, but this may not always be true. Minor isotope ratios of household samples could be co-linear, or despite a large isotopic range, most environmental samples within a single residence could cluster within a narrow interval. At the very least in those instances, isotopic analysis would allow for the exclusion of sources that were not major contributors to blood lead because their isotopic composition could be very different from the blood lead ratios. Therefore, a thorough evaluation of household lead hazards that incorporates the use of lead isotopes would not rely on this tool exclusively, but would consist of three elements: *a*) lead concentrations and loadings in the household environment; *b*) all isotopic ratios of those samples; and *c*) information about behavioral habits, as well as an evaluation of viable pathways of exposure to the child.

Even with a full characterization of the relative contribution of all external lead sources to the child, the isotopic composition of the integrated external input and the isotopic composition of lead in blood may not agree due to the presence of endogenous lead sources. In other words, lead in the bloodstream may also be composed of lead that has been remobilized from endogenous sources, such as the skeleton, that have accumulated lead over medium to long periods of exposure (e.g., > 6 months). The importance of mobilized skeletal lead as a contributor to blood lead levels has been shown in adults (*21,47–50*), and in children (*51*). As a result of the mixing of lead from these different exogenous and endogenous sources, there may be a shift in the blood isotopic ratios away from the isotopic ratios of the main exogenous source(s) of exposure toward the isotopic values of lead released from the endogenous (skeletal) source(s). Further, the isotopic composition of lead released from the skeleton as a whole may not necessarily match the isotopic composition of lead in bulk cortical or trabecular bone because the skeleton is a complex organ that possesses different lead subcompartments with different toxicokinetic behaviors (*52–54*).

Although experimental data on the amount of lead in children's bones are limited, those data suggest that between the ages of 1 and 5 years the amount of lead in the skeleton increases from 10 to 25 times the amount of lead in the blood compartment (*55,56*). During this age interval, bone mineral turns over every 90–120 days, whereas lead in blood turns over every 15–20 days. Therefore, over the course of one blood lead turnover

period, the skeleton could potentially contribute between 1.5 and 4 times the amount of lead in blood (i.e., 15 days/90 days × 10; and 20 days/120 days × 25). In reality, most remobilized bone lead is quickly resequestered back into bone with bone formation, which is proceeding at a more rapid rate than bone resorption in young children. Although the net contribution of bone lead to blood lead may be much less than the full amount of bone lead released via bone resorption, isotopic equilibration between the bone lead and blood lead occurs as lead is released to the plasma, mixes with plasma lead, and is incorporated back into bone. This would result in a modification of the isotopic ratio of lead in blood, from the initial isotopic ratio of the exogenous source(s) toward the isotopic ratio of skeletal lead. When lead exposure is chronically elevated at the same residence, it is likely that the accumulated skeletal lead fingerprint will resemble that of the chronic exposure and the contribution of skeletal lead to the blood will not obscure the interpretation of lead isotopes and the identification of the exogenous sources.

The cases presented here highlight the importance of the endogenous (skeletal) source of lead and its effect on the isotopic composition of blood. In Case 1, the blood lead isotopic composition did not match any of the suspected in-house sources. This is consistent with the child's history. From 1 to 3 years of age, she lived in Mexico where environmental exposures to lead were higher than in the United States, and she moved to her present residence 2 years before detection of the high blood lead level. Thus, the blood isotopic composition could be a reflection of high past lead exposures through ongoing releases of skeletal lead, as well as current exogenous exposures from her household environment. In Case 2, the child had lived all his life in the same residence, and the blood intake and skeletal isotopic compositions were probably similar or identical.

The most remarkable case is Case 3. At the time of the visit, this child appeared not to be exposed to any major lead source within the household, yet her blood lead level was 22 µg/dL. The proximity in isotopic ratios between the blood and the lacquer (Figure 3B, C; #7) suggests that the lead body burden was acquired several months before the blood test, when the child was not yet able to crawl and when the lacquered surfaces throughout the house had been sanded. The body lead burden acquired over that period, including lead accumulated into the skeleton, possibly supported the elevated blood lead level measured here via the release of skeletal stores of lead.

As the lead isotopic technique becomes more available and is used more routinely, it should be possible to allocate intervention resources more efficaciously. The lead isotope method has limitations, and there will be cases in which little information will be added to that gained through the currently used lead hazard assessment procedures. However, in those cases in which isotopes provide additional information, intervention strategies may be developed and implemented in a more cost-effective manner. For example, in Case 1 the main source of exposure did not appear to be in the house. The main suspected sources identified through the lead hazard assessment (#3 and #8) did not appear to be supported by the isotope results. In Case 2, there were at least five sources with high lead levels (Figure 2A); however, only three of them (#1, #2, #3, the initial main suspected sources of exposure), and an apparently minor one (#10, not identified in the lead hazard assessment), seemed to be major contributors to the elevated blood lead level. Isotope results pointed to dust as the main pathway of exposure. In Case 3, high blood lead levels were due to a source that was no longer a lead hazard, and it is unlikely that intervention efforts would yield a decrease in blood lead level because this elevated level was apparently due to past exposure.

Conclusions

Lead isotopes used in the context of current practices of household lead hazards evaluation can be used in refining the identification of sources of lead exposure. In this approach aimed at maximizing information while keeping the investigation economical, lead isotopes are measured on splits of samples collected for lead concentrations according to regulatory guidelines. The fact that lead isotopes are to be measured does not affect sampling procedures or the number of samples taken. Based on the cases presented here, it is possible to recommend that a thorough evaluation of household lead hazards would benefit by incorporating *a*) lead concentrations and loadings in the household environment; *b*) all isotopic ratios of potential lead sources; and *c*) information about behavioral habits, as well as an evaluation of viable pathways of exposure to the child. In the best scenario, lead isotopes can provide additional information to single out the source of lead that is actually being ingested by the child. In the worst case, where most potential sources have isotopically similar fingerprints, this approach could at least be used to exclude isotopically extreme sources from which the child is not receiving lead. Expected increases in the availability of magnetic sector ICP-MS at commercial laboratories should make this approach more prevalent in lead hazard identification and control.

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