Environmental Research 126 (2013) 171–178

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

Surface dust wipes are the best predictors of blood leads in young children with elevated blood lead levels

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ARTICLE INFO

Article history:
Received 22 January 2013
Received in revised form
11 June 2013
Accepted 12 June 2013
Available online 26 July 2013

Keywords:
Lead isotopes
Children
Blood
Environmental samples

ABSTRACT

Background: As part of the only national survey of lead in Australian children, which was undertaken in 1996, lead isotopic and lead concentration measurements were obtained from children from 24 dwellings whose blood lead levels were ≥15 μg/dL in an attempt to determine the source(s) of their elevated blood lead. Comparisons were made with data for six children with lower blood lead levels (< 10 μg/dL).

Methods: Thermal ionisation and isotope dilution mass spectrometry were used to determine high precision lead isotopic ratios ($^{208}Pb/^{206}Pb$, $^{207}Pb/^{206}Pb$ and $^{206}Pb/^{204}Pb$) and lead concentrations in blood, dust from floor wipes, soil, drinking water and paint (where available). Evaluation of associations between blood and the environmental samples was based on the analysis of individual cases, and Pearson correlations and multiple regression analyses based on the whole dataset.

Results and discussion: The correlations showed an association for isotopic ratios in blood and wipes ($r=0.52$, 95% CI 0.19–0.74), blood and soil ($r=0.33$, 95% CI −0.05–0.62), and blood and paint ($r=0.56$, 95% CI 0.09–0.83). The regression analyses indicated that the only statistically significant relationship for blood isotopic ratios was with dust wipes ($B=0.65$, 95% CI 0.35–0.95); there were no significant associations for lead concentrations in blood and environmental samples. There is a strong isotopic correlation of soils and house dust ($r=0.53$, 95% CI 0.20–0.75) indicative of a common source(s) for lead in soil and house dust. In contrast, as with the regression analyses, no such association is present for bulk lead concentrations ($r=-0.003$, 95% CI −0.37–0.36), the most common approach employed in source investigations. In evaluation of the isotopic results on a case by case basis, the strongest associations were for dust wipes and blood.

1. Introduction

Studies over more than three decades have indicated that residential dust is an important contributor to blood lead levels in children (Bornschein et al., 1985; Brunekreef et al., 1981; Duggan, 1983; Lanphearand Roghmann, 1997; Lanphear et al., 1998; Laxen et al., 1987; Thornton et al., 1990). Other studies have shown strong associations between house dust and soil and investigations in the US have indicated that soil lead accounted for about two-thirds of house dust (US EPA, 1998).

For the past two decades there has also been considerable investigation into leaded paint and associated lead in dust and soil, although e.g., Mielke et al. (1997), (2010) studying entire metropolitan areas have argued that the legacy of gasoline lead is still relevant in urban environments. The scale and scope of the current study is on lead isotopic case studies of individual children and their environment.

Interest in drinking water as an important contribution to blood lead in adults and children has undergone a resurgence in recent years. This is especially so in the US where elevated levels of lead in drinking water were identified in Washington DC and North Carolina, attributed to changes in chlorination procedures (Edwards et al., 2009; Guidotti et al., 2007; Miranda et al., 2007). A recent critical review by Triantafyllidou and Edwards (2012) concluded that lead in water may be more important as a source of blood lead than previously believed.

The associations between environmental samples and blood lead have been generally based on lead concentrations, rarely have lead isotopes been used to evaluate such associations (reviewed in Gulson, 2008).

As part of the only national survey of lead in Australian children (Donovan, 1996), high precision lead isotopic measurements were undertaken for a subset of children from 24 households whose

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http://dx.doi.org/10.1016/j.envres.2013.06.002

blood lead was ≥15 μg/dL in an attempt to determine the source(s) of their elevated blood lead. The data for the elevated Blood lead group were compared with six subjects whose blood lead was < 10 μg/dL. These blood lead levels were "action" levels recommended by the US Centers for Disease Control and Prevention (1991). For levels ≥15 μg/dL, US Centers for Disease Control and Prevention recommended that "If the blood lead level is 15–19 μg/dL, the child should be screened every 3–4 months, the family should be given education and nutritional counselling……., and a detailed environmental history should be taken to identify any obvious sources or pathways of lead exposure". The value of 10 μg/dL was widely reported as the "level of concern" and children whose blood lead levels exceeded 10 μg/dL were commonly described in the literature as "lead-poisoned". The recommended action for such children was re-testing within 12 months. In 2012, the US Centers for Disease Control and Prevention (2012a) adopted the US National Toxicology Program (US DHHS 2012) and US Centers for Disease Control and Prevention Scientific Advisory Committee (2012b), recommendations to discontinue the notion of an acceptable blood lead exposure level and use a reference value of 5 μg/dL.

Limited isotopic results and interpretations were reported in Appendix 4 of the Donovan report but this report is not widely available and no statistical evaluation of the isotopic data was undertaken. The objectives of the current paper were to evaluate associations in high precision lead isotopic ratios and lead concentrations of blood and environmental samples of surface dust wipes, soil, drinking water and paint as a guide to the source of lead in blood of the 30 children for whom isotopic data were obtained. Details of the environment for each subject and graphical illustration of the isotopic results ($^{207}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$) are given in the Supplementary notes.

2. Methods

Brief details of the methods have been taken from Donovan (1996).

2.1. The national survey and subjects

The Australian Institute of Health and Welfare conducted a nationwide survey during 1994 of blood lead levels in a representative sample of children aged 1 to 4 years (inclusive) with the following features:

- a nationally representative area-based sample using Australian Bureau of Statistics. Census Collector Districts numbering 27,673 of which 24,593 were classed as 'non-remote', and 3080 were 'remote'. A sample of 4000 children, in which each dwelling across Australia had the same 0.4% probability of selection, was identified in multiple stages;
- public relations and other measures to maximise response;
- questionnaire administration in homes using a national network of interviewers with transmission of questionnaires to the Australian Institute of Health and Welfare;
- blood collection in homes using networks of sample collectors with experience in collecting blood from children;
- notification by the Australian Institute of Health and Welfare of blood lead levels to parents and, if parents so requested, to a family doctor;
- with parental consent, notification by the Australian Institute of Health and Welfare to State and Territory health authorities of individual blood lead concentrations ≥15 μg/dL (0.73 μmol/L), for them to investigate and take remedial public health action as necessary;
- collection of water, house dust, paint flake and bulk soil samples from homes; and
- measurement of the relative abundance of lead isotopes in blood samples where blood lead level was ≥15 μg/dL and in environmental samples from corresponding households.

The 1575 children from whom blood was successfully taken came from 1300 households. For all 1575 specimens, the geometric mean blood lead level was 0.24 μmol/L (5.05 μg/dL). Of the 1575 blood lead readings, 1460 (92.70%) were less than the Australian National Health and Medical Research Council target level of 0.49 μmol/L (10 μg/dL) and 115 (7.30%) were greater than or equal to this level. In the overall survey 24 subjects had blood lead ≥15 μg/dL (the elevated group). Blood and environmental samples were measured for lead isotopic ratios for these 24 subjects and compared with another randomly chosen group of six with lower blood lead (< 10 μg/dL). It is these 30 subjects (24 with elevated blood lead and 6 with lower blood lead) who are the subject of the analyses reported in this paper.

2.2. Sampling

Blood was collected in the home using 4 mL draw Greiner vacuettes with ethylene diamine tetra acetic acid anticoagulant. Collectors had the choice of a butterfly needle and tube set leading directly to the vacuum container, or of a needle and syringe. Tubes were inverted several times immediately after collection of the specimen to prevent clot formation. Collectors also had to adhere to Australian standard AS 2636 'Sampling of venous and capillary blood for the determination of lead or cadmium concentration' as it related to venepuncture. The only variation was that it was recognised as unlikely that children could sit still for 10 min, so the survey requirement was that the children were encouraged to be quiet while their parents were interviewed. At the end of each day's sampling, blood samples were refrigerated and airfreighted in a cool container to a central laboratory for analysis. Storage requirements were those of AS 2636.

A sample of house dust was taken from a 300 mm square of floor where the parents indicated the child spent most of its time. This method gives a measure of lead available to the child (Lanphear et al., 1995). Briefly, a 300 mm square template was dropped onto the floor, and taped around its edges. The template was removed. Wearing a disposable glove, the interviewer wiped the marked square in two directions with a 'baby wipe'. This was then placed into a clean container.

Garden soil samples were taken from as close as possible to the wall at the rear of houses, under a window if there was one. If this area was grassed, a small section of lawn was removed. A sample was also collected from any area indicated by parents as to where the child played. An apple corer was used to obtain the specimens, all of which were placed into the one clean container. If the area was fully paved, dirt and dust scrapings were collected instead with a plastic knife. No sample was taken from multistorey flats where there was no garden area.

A tap water sample was collected if the residents had not collected an early morning sample as requested at the previous visit.

If badly peeling paint was noted, a paint flake was scraped into a clean container using a plastic knife. A window sill was suggested as the most suitable place to sample, because it was a site very likely to show peeling, and because the paint used on window sills was considered more likely than other paint to contain lead (Donovan 1996).

2.3. Analyses

Lead isotopic ratios and lead concentrations were measured in environmental samples of water, soil, paint and household dust wipes and were compared with lead isotopic data for blood samples. Red blood cell samples and whole blood samples in ethylene diamine tetra acetic acid and environmental samples were supplied on request by the Royal Prince Alfred Hospital in Sydney for the isotopic study.

The samples were digested in concentrated nitric acid (ultra-clean) and the lead was purified by anion exchange chromatography. All the corresponding environmental samples provided by Royal Prince Alfred Hospital, except for the waters, were partly digested in nitric acid. An enriched tracer of ^{202}Pb was added to the solution to obtain the lead concentration on the same sample as for the isotopic ratios (the isotope dilution method). The Royal Prince Alfred Hospital digests were analysed because: (1) no additional wipe samples were available and, (2) the potential variability of soils may have added complexity to the results. These samples were further digested with concentrated nitric acid and then converted to bromides. Lead separation using anion exchange chromatography was carried out, and in the case of paint and soil samples the "coarse" lead separation was followed by anodic electrodeposition to achieve a "pure" separation.

If whole blood samples were not available, red blood cells were measured and the results adjusted by a factor derived from the average of nine matched whole blood/red blood cell pairs. The blood lead data are thus expressed as μg/100 g rather than as μg/dL although in the figures in the Supplementary notes the blood lead values from the Donovan report are used and expressed as μg/dL. Blood lead levels measured by the more precise isotope dilution method are on average 22% lower than the values that were measured by inductively coupled plasma mass spectrometry and shown in the Appendix 4 figures in the Donovan (1996) report.

Solubility of lead from paint flakes was measured in specimens from homes where any child had a blood lead level of ≥10 μg/dL and where the lead concentration in those flakes exceeded 10,000 mg/kg (1%). The samples were crushed in an agate mortar and pestle to pass through a nylon sieve of 100 μm mesh size. Bioaccessibility of the lead in the paint samples was measured by leaching the crushed and sieved particles in 0.1 M hydrochloric acid for 2 h at 38 °C, and the dissolved lead measured by inductively coupled plasma emission spectroscopy.

High precision isotopic ratios ($^{208}Pb/^{206}Pb$, $^{207}Pb/^{206}Pb$ and $^{206}Pb/^{204}Pb$) for all the above mentioned samples were measured on a VG Isomass 54E thermal ionisation mass spectrometer. The precision of the $^{206}Pb/^{204}Pb$ ratio was ± 0.1%