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Relationship of Pb in House Dust and Ambient Air

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Abstract

We evaluated the relationship of lead (Pb) using high precision Pb isotopes from ambient air particulates and dust fall accumulation in 59 residences in Sydney New South Wales Australia by the Petri Dish Dust method (PDD) to determine if the dust is a reliable indicator of exposure in cases where air Pb data may not be available. Over the period 1993-2002, Pb values in air samples were higher in winter whereas the Pb loadings for PDD values were slightly higher in spring and summer. These differences are probably the result of differences in sampling times of the air particulates (24-h) and PDD (~3 months).There was no seasonal or suburb effect for the isotopic ratios. Both air and PDD samples showed a strong increase in $^{206}Pb/^{204}Pb$ over time. PDD data were predicted by the air data ($p < 0.001$) and provide a useful adjunct in monitoring exposures.

Keywords: Dust particles; Sampling times; Exposures

Introduction

Exposure to house dust is the most important contributor to blood Pb (PbB) in young children [1-8]. The usual method of estimating exposure to dust is collection by vacuum cleaners [9,10] or surface wipes [9], summarized in US EPA [11]. These sampling methods have various limitations including lack of information about deposition rates, unless resampling over specific time periods is specifically undertaken. An alternative method makes use of collecting trays [12] or dishes [13-16] that monitor exposure for varying lengths of time. Material collected in these vessels is via airborne pathways and may derive from such sources as activities in the house (e.g., renovation, smoking), tracked in dust, and windblown through doors and windows. The exterior sources may be resuspended soil and dust which may contain a legacy of past leaded-gasoline use or leaded paint [17-19], a fact that is continuously misinterpreted by most people in the community (internationally) who think Pb is no longer an issue because Pb has been removed from gasoline and paint in many countries.

Ambient air has been monitored over decades mainly by environmental protection agencies and some researchers [11]. Although air Pb levels have decreased dramatically with cessation of the use of leaded gasoline in many countries (e.g., Thomas et al. [20]) the contribution of Pb in ambient air is still of importance in monitoring environmental exposures to the community and individuals in residences. For example, monitoring of Pb in Total Suspended Particulates (TSP) from High Volume (HV) air filters in Sydney by New South Wales Environment Protection Authority (EPA) showed the annual concentration decreasing from 0.75 μg/m^3 in 1991 to <0.1 μg/m^3 in 2000 [11], levels being so low for their laboratory methods that Pb measurements were discontinued from 2002. Nevertheless Pb has been monitored in Sydney $PM_{2.5}$ particulates over decades by Cohen and colleagues at ANSTO [21,22] using sensitive ion beam methods.

In spite of rapid reductions in air Pb associated with removal of Pb from gasoline and use of Pb in paint, air Pb may still be an important contributor to PbB especially in children. For example, Brink et al. [23] found that Pb measured in the US EPA's National Air Toxics Assessment was a significant predictor of PbB ≥10 μg/dl in children. Using data from the National Health and Nutrition Examination Surveys (NHANES) III and 9908 and air Pb data in TSP from the US EPA, Richmond-Bryant et al. [24] concluded that a larger relative public health benefit among children may be derived from decreases in air Pb at low air Pb exposures. In a US study of 3 urban neighborhoods which measured 23 trace elements from 24-hour $PM_{2.5}$ particulates in outdoor, indoor and personal samples, Adgate et al. [25] found that personal exposure is likely to be underestimated by outdoor central site monitors. Earlier, Tu and Knutson [26] concluded that a compliance with outdoor air quality standards did not ensure a satisfactory indoor situation.

Previous studies have usually evaluated associations between soil-exterior dust- house dust-paint and indoor air with personal monitors [27,28] but generally not exterior ambient air. Some of the studies evaluating the relationship of urban air to house dust include those of Angle and McIntire [29], Manton et al. [30], Laidlaw et al. [19], Rabinowitz et al. [31]. In this study we evaluated the association, using high precision Pb isotopes, between Pb in air collected from high volume air filters in Australia's largest city, Sydney, and house dust collected by the petri dish method to determine the usefulness of data from the petri dish method for other studies where air Pb data are unavailable. This is a follow-up to earlier papers which focused on Pb isotopic relationships on Sydney air [32,33] or elemental associations in $PM_{2.5}$ particulates [33,34].

Methods

Air filters

Particulates collected on filters ('air filters') were obtained from an ongoing air quality monitoring program carried out by the NSW EPA.

Filters were analyzed from samples collected in Sydney's Central Business District (CBD) and from a nearby suburb (Rozelle), situated about 5 km west of the CBD. Air filters from Rozelle have been analyzed for Pb isotopic ratios monthly from January 1991 till May 1996, and several were also analyzed from 1987 to 1989 (total n=138). Those from the CBD have been analyzed on a monthly basis only from April 1994 with four additional samples from 1992 and 1993 (n=36). To evaluate differences over a wider area, filters from a number of other locations in Sydney were occasionally analyzed.

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A High Volume Air Sampler (HVAS) following Australian Standard AS 2724.3-1984 was used. Sampling was carried out continuously for 24 hours on a one-day-in-six cycle. Particles in the approximate size range 0.1 µm to 50 µm were collected. Rozelle samples were collected on a filter which was situated within the HVAS 1.13 m above ground level. The HVAS at Rozelle was located in a parkland setting several hundred meters from a major thoroughfare (>70,000 vehicles per day). The CBD HVAS was located 4m above ground on a street awning.

Preliminary measurements for 4 filters collected within 1 week showed small isotopic variations but which were not statistically significantly different (p 0.08- 0.7 in $^{207}Pb/^{206}Pb$ ratios for 4 different time intervals) so that ongoing sampling was restricted to filters that were collected in the first week of each month.

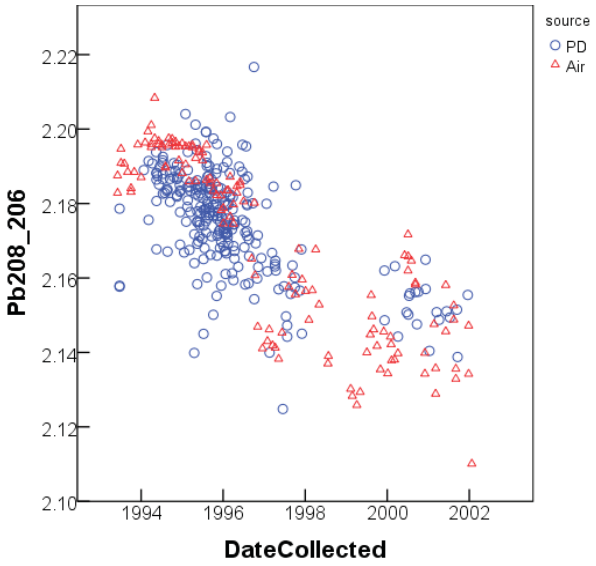

Figure 1: Measured $^{208}Pb/^{206}Pb$ ratios versus Date sampled for air filters and petri dish dust.

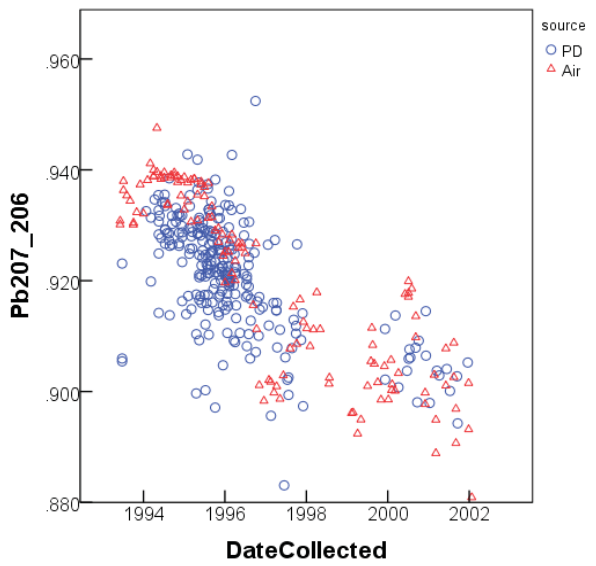

Figure 2: Measured $^{207}Pb/^{206}Pb$ ratios versus Date sampled for air filters and petri dish dust.

A smaller number of samples (n=13 each) were measured for $PM_{2.5}$ Teflon filters over the period 1998 to 2004 for 2 locations in Sydney: one from Mascot, a suburb close to the CBD and a high trafficked area, and the other, a relatively rural (background) setting at Richmond, 20km west of the CBD.

Personal air monitoring for a 24-hour period was undertaken within 12 houses in the inner Sydney suburbs. Although the air samples were collected by different methods and this would affect Pb values, it is not an issue for the Pb isotopic results. The methods for analysis of these filters are detailed in Chiaradia et al. [32], Cohen et al. [34] and Gulson et al. [14].

House dust

The use and advantages of petri dishes to provide ongoing dust fall accumulation was described in earlier publications [14,16]. The PDD collections were usually for a period of about 3 months. The dishes were placed in at least 3 locations: kitchen, main living area, child's bedroom. As early analyses showed only small differences between the different locations in a residence, thereafter the solution from each dish was combined for the appropriate time interval. Dust was collected from 56 houses with the number of collections ranging from 1 to 10 resulting in data for 261 dishes.

Statistical analyses

The analyses were based on 130 air measurements collected between the 5/6/1993 and 23/1/2002 and 261 PDD measurements collected between 23/6/1993 and 18/12/2001. Additional air measurements were available before and after the above periods but these were omitted from analyses so as to maximize the time overlap between the air and PDD samples.

Initial mixed model analyses with each of the Pb isotopic measures as dependent variables included the following independent variables: source (air or PDD), suburb (seven nested under source = PDD and five nested under source = air), season, and time at which the sample was collected, the last coded as fractional number of years since the date the first sample in the analysis subset was collected (5/6/1993). The initial analyses also included a quadratic term for time ($time^2$) and interactions between source and time and $time^2$ respectively. The model also included a random factor referred as location which allowed for the correlation between observations obtained at a common collection point (e.g., the living room of a house).

Results

Isotopic compositions

The data sets used in the statistical analyses are presented in Figures 1 to 3 and predicted isotopic results shown in Figures 4 to 6. Outputs of the results from the statistical analyses are given in the supplementary notes.

The dominant trend is a decrease in $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ ratios and a positive increase in $^{206}Pb/^{204}Pb$ ratios over time which is slightly sharper for air measurements than for PDD measurements. The steady increase in $^{206}Pb/^{204}Pb$ (or decrease in $^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$) over time in both types of samples probably reflects the gradual phasing out of Pb from gasoline and increasing contributions of Pb from natural materials such as soils and industrial materials.

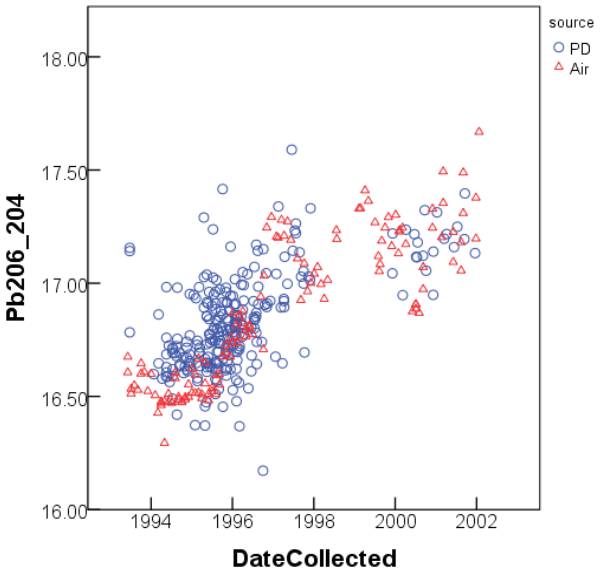

Figure 3: Measured $^{206}Pb/^{204}Pb$ ratios versus Date sampled for air filters and petri dish dust.

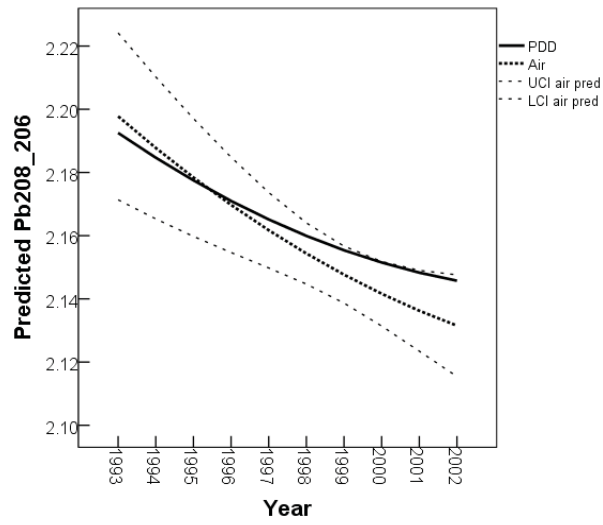

Figure 4: Predicted $^{208}Pb/^{206}Pb$ ratios versus Date sampled for air filters and PDD. The bold solid line is the curve fitted to the PDD measurements, the bold dotted line is the curve fitted to the air measurements and the light dashed lines are the upper and lower 95% confidence intervals for the prediction of the air measurements from the PDD measurements, derived as described above.

Initial analyses showed that while there was a curvilinear change in the measurements over time for each dependent variable, this did not vary with source, so only the main effect of time^2 was retained in the models the results of which are reported here. The interaction between linear time and source was significant, however, and was included in later models. Initial analyses also revealed three air observations the standardized residuals for which were near to 5. These data were excluded from subsequent analyses.

The general pattern of results was the same for each dependent variable (Supplementary material, which gives the ANOVA tables and the parameter estimates for the mixed model analyses). Suburb and season were non-significant (although slightly lower values of $^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$ were observed for autumn), while the main effect of time^2 and the interaction between source and time was significant. The proportion of variance of the dependent variables accounted for by all independent variables (calculated according to the method given by Snijders and Bosker, [35]) ranged from 0.53 to 0.59, while the unique variance accounted for by the source by time interaction ranged from 0.031 to 0.049.

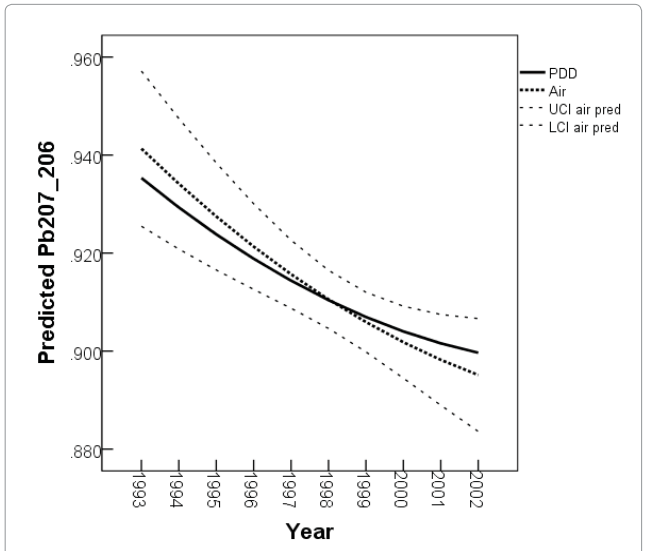

Figure 5: Predicted $^{207}Pb/^{206}Pb$ ratios versus Date sampled for air filters and PDD. The bold solid line is the curve fitted to the PDD measurements, the bold dotted line is the curve fitted to the air measurements and the light dashed lines are the upper and lower 95% confidence intervals for the prediction of the air measurements from the PDD measurements, derived as described above.

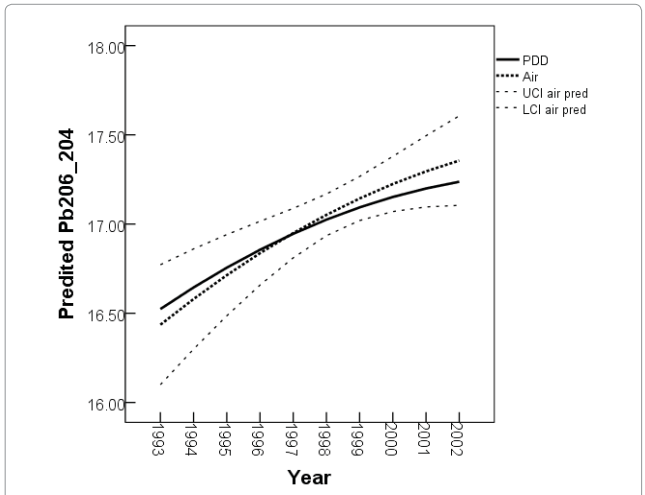

Figure 6: Predicted $^{206}Pb/^{204}Pb$ ratios versus Date sampled for air filters and PDD. The bold solid line is the curve fitted to the PDD measurements, the bold dotted line is the curve fitted to the air measurements and the light dashed lines are the upper and lower 95% confidence intervals for the prediction of the air measurements from the PDD measurements, derived as described above.

Dependent variable	B_{source}	$B_{(source \times time)}$
$^{208}Pb/^{206}Pb$	-0.005237	0.002161
$^{207}Pb/^{206}Pb$	-0.005998	0.001171
$^{206}Pb/^{204}Pb$	0.087706	-0.022941

Table 1: Regression coefficients used in predicting air measurements from PDD measurements.

The predicted values of the three different measurements over time for each source are shown in Figures 4 to 6. The bold solid line in each graph is the curve fitted to the PDD measurements, and the bold dotted line is the curve fitted to the air measurements.

The regression equation derived from the mixed model for each dependent variable provided a basis for making predictions from one source to the other, in particular from PDD measurements to air measurements. A prediction equation for each dependent variable was derived by equating the separate equations for the two sources with respect to time then rearranging and simplifying to give rise to an expression of the following form: for a given time, t,

$$air_t = PDD_t - B_{source} - B_{(source \times time)} \times t \quad (1)$$

Where B_{source} is the regression coefficient for the effect of source and $B_{(source \times time)}$ is the coefficient for the interaction between source and time. The coefficients for each dependent variable are shown in Table 1.

In order to estimate the variability of predictions derived from the present data, bootstrap sampling was used. Estimates of the two coefficients used in the above equation (the observed values of which are shown in Table 1) were obtained from analyses carried out on approximately 1000 bootstrap samples, using the mixed procedure and bootstrap procedures in Stata 13. Predictions of air measurements from the PDD measurements were obtained for each pair of coefficients, and the standard deviation of the predicted values was used as the standard error to construct 95% confidence intervals for the predictions.

The sampling with replacement was performed at the cluster level rather than at the level of individual measurements; that is, all the measurements for a given location, meaning a suburb or house, were either included in the bootstrap (possibly more than once), or not included at all. This method can be regarded as fairly stringent and gave rise to the conservative 95% confidence intervals shown in Figures 4 to 6.

Pb values

There were no significant differences in Pb loadings for PDD between suburbs but there were higher values in spring and summer compared with winter (Supplementary materials).

In contrast to the PDD, during the 4 years of detailed investigation of TSP in the Sydney suburb of Rozelle, Pb concentrations displayed a maximum in late autumn-start of winter (Figure 4) [32]. Similar trends have been observed in other Sydney suburbs (Figure 4) [34]. The higher Pb values in the autumn-winter months are probably related to thermal inversions typical of the cold season or to drier conditions during this period compared with summer. On the other hand the higher Pb contents in summer for the PDD may arise from the propensity of residents for open doors and windows in summer and the higher temperatures giving rise to dust which may then enter the residence. The decreasing Pb concentrations over time (p 0.008) reflect the decreasing use of Pb from gasoline, terminated in 2002.

Discussion

Comparison with other studies

Earlier studies of house dust did not include exterior air Pb, only reported Pb values (e.g., Succop et al. [28]) and rarely employed Pb isotopes. Furthermore, where Pb isotopes were measured, the dust sampling was by wiping material that had accumulated on surfaces and/or from door mats (e.g., Manton et al. [30]). In their investigation of aerosols, hand wipes, house dust, and a 24-hour duplicate diet Manton et al. [30] concluded that the Omaha aerosols represented a source distinct from the local household dust. In Shanghai, China, Liang et al. [36] found that children's blood lead levels were strongly correlated with air lead and mainly caused by coal consumption and fly ash after phasing out of leaded gasoline.

There are very few published isotopic data from Eastern Australia which would be relevant to compare with our analyses. Using a low volume sampler system powered by a diaphragm pump with a collection duration of from 1 to 3 months, Bollhöfer and Rosman [37,38] published data 2 samples in Sydney in March to May 1994 (2000) and 6 samples in 1994-95 and 6 in 1998 from Melbourne (2002). The sets of data from Bollhöfer and Rosman are encouragingly almost identical to our results for the same time periods even though the collection methods and laboratories were completely different [37]. The 1994 Sydney samples had $^{206}Pb/^{204}Pb$ ratios of 16.49 and 16.45 whereas those for Melbourne were slightly higher varying from 16.53 to 16.69 and in 1998 ranged from 16.8 to 17.3. Bollhöfer and Rosman [38] suggest that Pb isotopic ratios in Melbourne were slightly more radiogenic (e.g., higher $^{206}Pb/^{204}Pb$ ratio) during the summer compared with winter as we observed for summer-autumn but this is not clear from the data presented in their Table 1. Only one sample of gasoline from Western Australia was analyzed to evaluate the relationship between tetra-alkyl Pb and air in contrast to the numerous analyses for Sydney reported by Chiaradia et al. [32]. In the Sydney study, Chiaradia et al. [32] estimated that more than 90% of the Pb in HV air filters was derived from gasoline and gasoline Pb was also the primary source of surface soil and pavement dust.

Monthly sampling over a 15-month period (November 2010-January 2012) of dust deposition gauges placed in the rear yards of 5 houses in Sydney showed higher atmospheric Pb loadings in the summer/autumn with the lowest values in winter [15].

Dust from petri dishes in the houses described here over a 9-year time interval (1993-2002) showed higher Pb loadings in spring and summer compared with winter (Supplementary material). No difference between seasons for Pb loadings was observed by Laidlaw et al. [15], measured using the same petri dish method. In a longitudinal study in Sydney from 2001 to 2006 of 108 houses (1163 analyses) using PDD sampling over 6 monthly intervals, no seasonal effect for Pb loadings was observed [39]. The differences between our results and those of Laidlaw et al. [15] may arise from the different years of sampling and the limited number of 5 houses studied by Laidlaw et al. [15] compared with the 59 houses in our study.

Using vacuum cleaner dust from 82 houses in different Sydney suburbs, Chattopadhyay et al. [40] suggested that while air Pb levels decreased dramatically over decades, Pb concentrations in household dust remain unchanged due to accumulation of Pb from old paints and almost 80 years of leaded gasoline use [41,42].

Conclusions

The tracking of isotopic data for the air filters and house dust indicates that house dust provides an alternative indicator of exposure especially where air monitoring is not possible. The absence of seasonal or suburban effects on the isotopic data indicates applicability of house dust measurements over a wide area; the Sydney Basin area sampled

in our study encompasses some 40 by 40 km. PDD is a simple, non-invasive sampling method compared with personal air monitoring and provides an integrated picture of air-dust relationships over say 3-monthly periods. PDD provide isotopic information that can be used in other environmental studies such as monitoring with mosses and lichens and lake sediments.

In trying to evaluate sources of metals in the environment now that Pb has been removed from gasoline in most countries and the important role of leaded paint has been more strongly publicized, it is worth reiterating a comment by Professor Roy Harrison "instead of having a few dominant sources we are now subject to a large number of relatively weak sources and therefore the source signatures become very obscure in the atmospheric signal" [33].

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