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Changes in the lead isotopic composition of blood, diet and air in Australia over a decade: Globalization and implications for future isotopic studies ☆

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Abstract

Source apportionment in biological or environmental samples using the lead isotope method, where there are diverse sources of lead, relies on a significant difference between the isotopic composition in the target media and the sources. Because of the unique isotopic composition of Australian lead, source apportionment has been relatively successful in the past. Over the period of a decade, the $^{206}Pb/^{204}Pb$ ratio for Australian (mainly female) adults has shown an increase from a geometric mean of 16.8–17.3. Associated with this increase, there has been a decrease in mean blood lead concentration from 4.7 to 2.3 μg/dL, or about 5% per year, similar to that observed in other countries. Lead in air, which up until 2000 was derived largely from the continued use of leaded gasoline, showed an overall increase in the $^{206}Pb/^{204}Pb$ ratio during 1993–2000 from 16.5 to 17.2. Since 1998 the levels of lead in air were less than 0.2 μg/m^3 and would contribute negligibly to blood lead. Over the 10-year period, the $^{206}Pb/^{204}Pb$ ratio in diet, based mainly on quarterly 6-day duplicate diets, increased from 16.9 to 18.3. The lead concentration in diet showed a small decrease from 8.7 to 6.4 μg Pb/kg although the daily intake increased markedly from 7.4 to 13.9 μg Pb/day during the latter part of the decade probably reflecting differences in demographics. The changes in blood lead from sources such as lead in bone or soil or dust is not dominant because of the low $^{206}Pb/^{204}Pb$ ratios in these media. Unless there are other sources not identified and analysed for these adults, it would appear that in spite of our earlier conclusions to the contrary, diet does make an overall contribution to blood lead, and this is certainly the case for specific individuals. Certain population groups from south Asia, south-east Asia, the Middle East and Europe (e.g. UK) are unsuitable for some studies as their isotopic ratios in blood are converging towards the increasing Australian values. The increases in blood $^{206}Pb/^{204}Pb$ ratio combined with globalization, which has resulted in the increases in $^{206}Pb/^{204}Pb$ ratio for diet, means that isotopic studies undertaken with a high degree of certainty of outcomes over a decade ago, are now considerably more difficult, not only in Australia but also in other countries where the isotopic differences are even less than in Australia.

Keywords: Lead; Isotopes; Blood; Diet; Air; Sources; Changes; Time

1. Introduction

In spite of the decrease in exposure to lead and attendant decreases in blood lead concentration, lead is still of concern, especially for its potential impact on young children at low blood lead levels (Canfield et al., 2003).

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In earlier decades with widespread use of lead in gasoline, in solder for canned foods and in paint, these materials provided the overwhelming contribution of lead in blood and could be fairly easily identified. Diet is still considered a significant contributing factor to blood lead levels (EPA, 1994; Bolger et al., 1996; Carrington et al., 1993). In many cases, the source of lead is not always obvious, especially at low blood lead levels. In urban environments where low blood lead levels may be present, source apportionment usually relies on 'field' observations, and statistical manipulations of lead concentration data, including structural equation modelling (Clark et al., 2004). Less commonly, lead isotopes have been employed to evaluate sources and pathways of lead in blood and other biological measures but the success of this approach is contingent on there being a sufficient difference in the isotopic composition of the sources of the lead and that in the target medium. For example, Manton et al. (2003) suggest that the lead isotope approach is only successful in 1 out of 5 cases in the US, although it is higher in Australia because of the prevalence of unusual lead isotopic signatures, described later.

In this paper, we present the changes that have occurred for blood, diet and air in Australia over a period of about 10 years. We will then point out the difficulties that could arise in future studies, especially in those countries where the differences in isotopic composition may be small and/or the technology is lacking to undertake the quality of analyses that are required to achieve any realistic outcomes.

2. Methods

2.1. Samples

Blood: We have selected, from past research projects, the venous blood results from 79 subjects, consisting of mostly female adults, 8 males, and some children from diverse communities. The communities include those with mining and smelting activities but in these cases the males or occupationally exposed subjects were excluded. Likewise, the >2000 blood results for migrants to Australia and their existing and/or newborn children were excluded because of the markedly different isotopic composition of the lead in their bodies compared with those in the Australian environment and long-term residents (see Fig. 1, this paper; Gulson et al., 1995a, 1997a, b, 1998).

Apart from the longitudinal sampling of the pregnant Australian subjects for up to 3 years and those in the study involving a lead in calcium supplements study (Gulson et al., 2001), most of the other subjects were sampled only once.

Diet: This data set comprised the results for all 6-day duplicate diets undertaken over the decade, as well as those from quarterly market basket surveys from 1990/1991 (Gulson et al., 1996). Results for the migrant subjects were included, as their food was purchased in Australia and reflected a more complete spectrum of the dietary intakes prevailing over the decade. The data set can be subdivided further into a main cohort, including mainly migrants and Australians covering the period 1993/1998, and another cohort of mainly migrants with somewhat different demography covering the period 1999/2002. The earlier cohort mainly comprised subjects from eastern block countries whereas those from the later cohort were mainly from the Middle East (Fig. 1).

Air: This data set comprised results of total suspended particulates from high volume air filters from two sites in Sydney, one being the central business district and another approximately 3 km west of the central business district in a less trafficked area although not downwind from the central business district. From comparisons of the isotopic composition of gasoline and high volume air filters, Chiaradia et al. (1997) showed that more than 90% of the lead in air for the period 1993–1996 was derived from gasoline. Lead was phased out from most gasoline in Australia by January 2002.

2.2. Analyses

Details of analytical methods have been described in several papers, including Gulson et al. (1996). High precision lead isotopic ratios and lead concentrations by isotope dilution were measured using thermal ionization mass spectrometry (TIMS).

2.3. Statistical analyses

Statistical analyses were performed using SPSS versions 11.5 and 12.0. The data used for the analyses were aggregated over subject, so that there was only ever one observation per subject. However, in some cases there were multiple observations from a given site, and these data were analysed with a mixed model with site as a random factor, in order to take account of the dependence amongst the observations from the same site. When there was a single observation for each site, ordinary least squares regression was used to fit regression lines. A squared time term was included in each model to test for curvilinear relationships.

3. Results

With the use of TIMS and isotope dilution, three isotopic ratios and the lead concentration can be obtained. In our laboratory, the ratios $^{208}Pb/^{206}Pb$, $^{207}Pb/^{206}Pb$ and $^{206}Pb/^{204}Pb$ are measured on the sample