Origin of Lead in the United States Diet

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We report ²⁰⁸Pb/²⁰⁷Pb and ²⁰⁶Pb/²⁰⁷Pb ratios for 1001 duplicate diets collected from mothers and children, 1304 samples of house dust and hand wipes, and 64 samples of aerosols that were taken in Omaha, Nebraska, during the period from 1990 to 1997. A plot of ²⁰⁸Pb/²⁰⁷Pb versus ²⁰⁶Pb/ ²⁰⁷Pb for the dust and hand wipes indicates that they contain lead from ores mined in Idaho, Missouri, and Mexico. The absence of lead from Utah suggests that this mixture is not representative of the whole country. The lead in the aerosols has a narrower range of isotope ratios and resembles aerosols collected elsewhere in the United States. Most dietary collections contain a large component of house dust. Some, especially those from infants, are dominated by uranogenic lead with high ²⁰⁶Pb/²⁰⁷Pb ratios. Its source is taken to be calcium-supplemented food where the calcium is derived from limestone. Another source of lead is thorogenic and is ascribed to lead in tin coatings. Agricultural lead, whether from soil (estimated from recently published analyses of sedimentary materials), fertilizer, or agricultural lime, could not be unambiguously identified in the diets. Lead derived from aerosols, if present at all, is insignificant.

Introduction

In the United States dietary intakes of lead reached a maximum between 1978 and 1980 when a woman's intake was about 60 and a child's intake was between 35 and 45 μ g/day (1). By the end of the 1990s, thanks to the removal of lead from gasoline, the elimination of soldered cans, and awareness of potential sources such as ceramic ware, wine, and shellfish (2), intakes for both groups had fallen by a factor of 10 (*3*, *4*), raising the question whether lead in the diet is still largely anthropogenic or whether natural, agricultural sources of lead may be identifiable or even dominant.

Isotopic techniques could be used to distinguish between anthropogenic and agricultural lead in the U.S. if it were demonstrated that historically used ores have isotope ratios distinct from those of uncontaminated soils. While the ratios of anthropogenic lead are readily obtained by sampling dusts or aerosols, those of pristine soils can only be assumed to fall close to the semiempirical Stacey–Kramers curve (5), which defines the evolution of lead in the continental crust, but their distribution about this curve is unknown. Sediment, whether that currently being transported by rivers or that deposited as deep sea turbidites, may be used as an analogue. Lead isotopic analyses of both types of material have been published recently (6, 7). Although only a few samples in each category are from the United States or its margins they can nonetheless be taken as representative of the scatter that might be exhibited by North American soils which formed on a continental region containing rocks spanning the whole range of geologic time.

In the 1990s we carried out a study in Omaha, Nebraska, directed to determining how young children acquired lead. During its course we analyzed for lead concentration and isotope ratio over 1300 environmental samples of hand wipes and house dust and 1001 duplicate diets from children and their mothers. The results as they pertained to the blood lead profiles of the mothers and children have already been published (*3*, *8*), but only the ²⁰⁶Pb/²⁰⁷Pb ratios were reported. Here we use both their ²⁰⁸Pb/²⁰⁷Pb and ²⁰⁶Pb/²⁰⁷Pb ratios along with those published for sediment in an attempt to understand how lead enters the United States diet.

Experimental Section

Subjects and Samples. The subjects were women who lived in pre-1950 housing in Omaha, Nebraska. They were recruited during pregnancy, and they and their infants were followed for up to 24 months after delivery. Besides blood and urine, a duplicate diet sample and a hand wipe were taken monthly from each. If the mother breast-fed, diets from the child were taken as soon as it began to take solid food. Dust samples were taken from windowsills, from carpeted and noncarpeted play areas, and from beneath doormats. Some subjects voluntarily dropped out of the study before 24 months, and others were continued longer, but 432 hand wipes were taken from 30 children and 491 house dust samples from 26 homes. A total of 461 duplicate diets were collected from the children and 252 from their mothers. In addition 288 diet, 327 hand wipe samples, and 54 house dust samples were obtained from 21 2-6 year old children as part of a pilot study, carried out in 1990 to 1992. At this time 64 samples of suspended aerosols were also collected.

Duplicate Diets. In this method of dietary sampling the mother saves in a container an identical serving of whatever she or her child ate or drank over a 24 h period. The containers were wide mouth 2 L polyethylene jars that had been acid-cleaned. Food was homogenized in a blender, and aliquots were siphoned into 500 mL Teflon PFA bottles.

Chemical Decomposition and Pb Separation. All samples were decomposed in sub-boiling quartz-distilled nitric acid: the food in the Teflon bottles on a hot plate and the hand wipes and house dust in a microwave. Lead was separated from the food samples by anion exchange chromatography, but by Ba(NO₃)₂ coprecipitation and electrodeposition in the case of the hand wipe, dust, and aerosol samples. Processing was done under class 100 conditions, and appropriate blank corrections were made.

Mass Spectrometry. All isotopic analyses were made on a Finnigan MAT 261 thermal ionization mass spectrometer. Addition of a small quantity of ²⁰⁵Pb to each sample during processing allowed both Pb concentration and isotopic composition to be measured simultaneously. As this spike contains 13% ²⁰⁴Pb its use precludes accurate recovery of ²⁰⁴Pb abundance; thus, only ²⁰⁸Pb/²⁰⁷Pb and ²⁰⁶Pb/²⁰⁷Pb ratios are reported. Performance of the mass spectrometer was monitored by analysis of the standard SRM 981.

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FIGURE 1. ²⁰⁸Pb/²⁰⁷Pb vs ²⁰⁶Pb/²⁰⁷Pb plots for environmental samples. Panel a: 1304 hand wipe and house dust samples measured for 51 children and 46 homes in Omaha, Nebraska. A line has been fitted to 999 of the data points. Panel b: 64 aerosol samples from Omaha and a line fitted to them. This plot is drawn to a different scale, but the average of the dust samples of Panel a and the Stacey—Kramers growth curve (5) for the evolution of Pb in the Earth's crust, shown in both plots, allow the two lines to be oriented with respect to each other.

Results

Hand Wipes, House Dust, and Aerosol Samples. The isotopic analyses are shown as ²⁰⁸Pb/²⁰⁷Pb versus ²⁰⁶Pb/²⁰⁷Pb plots in Figure 1. Most of the hand wipes and house dust (hereafter collectively referred to as dust) fall above the Stacey–Kramers curve and form an inverted triangle, whose two lower vertexes are well defined and lie on Stacey–Kramers, the one close to present day lead and the other between 500 and 750 million years (my) (Figure 1a). The uppermost, however, is poorly defined.

The means and standard deviations of the isotope ratios were 2.454 ± 0.014 for ²⁰⁸Pb/²⁰⁷Pb and 1.201 ± 0.022 for ²⁰⁶Pb/²⁰⁷Pb. A straight line was fitted to the data by the method of York (9) using the program of Ludwig (*10*). This program,

however, handles no more than 999 points, so 305 were randomly omitted. Errors were estimated as ± 0.002 in ²⁰⁸Pb/²⁰⁷Pb and ± 0.001 in ²⁰⁶Pb/²⁰⁷Pb, and the correlation coefficient was taken to be 0.999 since ²⁰⁷Pb is in the denominator of both variables. The resulting equation is

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Pb/ 207 Pb = 1.502 × 208 Pb/ 207 Pb - 2.484

and has uncertainties (σ) of ± 0.017 in the slope and ± 0.041 in the *y*-intercept. The measurements, however, do not fit the line within the limits of the estimates of experimental error.

The aerosols form a linear array (Figure 1b), and the line fitted to them passes close to the average of the dust of Figure 1a, but the range is smaller and the majority of samples have ²⁰⁶Pb/²⁰⁷Pb ratios less than 1.22. This and the fact that the line is steeper, intersecting Stacey–Kramers at about 350 my rather than the 500 my intersection of the dust line, suggest that the aerosols and the dust belong to different populations.

Dietary Pb Concentrations. As the lead contents of the 24 h collections tended toward being log–normally distributed their mean contents are expressed as geometric means. These and their geometric standard deviations and ranges are given in Table 1 which also lists the U.S Food and drug Administration's (4) estimates of lead intakes for various age groups over the period from 1991 to 1996.

Dietary Pb Isotope Ratios. The isotope ratios of the children's diets are plotted in Figure 2. Those of the youngest (Figure 2a) are characterized by lead enriched in ²⁰⁶Pb, though some samples fall in the field of the hand wipe and environmental samples of Figure 1a. The 13–24 month old children's diets (Figure 2b) for the most part overlap the field of the wipe and house dust lead, but some are enriched in ²⁰⁶Pb and others lie to the right of present day lead as defined by the Stacey–Kramers model (5). The 2–6 year olds' diets lie in the field of the wipes and house dust (Figure 2c). There is, however, a tendency for the points to be shifted to the right of the dust average.

The mothers' diets are plotted in Figure 3. Although some lie in the field of hand wipes and house dust, many fall outside it, a few lying below Stacey–Kramers, some to the right of present day lead, and those with high ²⁰⁶Pb/²⁰⁷Pb ratios above the dust line. Not shown are two diets collected three months apart from the same woman that had ²⁰⁸Pb/²⁰⁷Pb ratios of 3.119 and 3.179 and ²⁰⁶Pb/²⁰⁷Pb ratios of 1.324 and 1.326. The lead contents were 8.4 and 11 μ g and as such were not remarkably elevated with respect to those of the other mothers, which had a geometric mean content of 7.5 μ g of Pb per collection (Table 1).

Discussion

Stacey-Kramers Model. The model, which is based on the lead isotope ratios in galenas (natural lead sulfide) of various ages, purports to show how the isotopic composition of lead in the Earth's continental crust has changed over time (5). While its basic assumptions-that the Earth's continental crust was isotopically homogeneous 3.7 billion years (by) ago and remained closed to uranium, thorium, and lead ever since-are no longer considered valid (11), the Stacey-Kramers curve nonetheless provides a useful datum against which natural leads can be compared. Generally speaking, young upper crustal rocks tend to plot along the extension of the curve to the right of present day lead, while lower crustal rocks, such as are exposed in the geologically oldest terrains, tend to plot below the curve because they are characteristically depleted in uranium. An additional feature of this plot is to shift geologically old rocks that contain excess amounts of ²⁰⁷Pb (produced when ²³⁵U was far more abundant than it is today) below the growth curve and to the left of present day lead.

TABLE 1. Daily Lead Intakes Measured for Mothers and Children in This Study and the U.S. Food and Drug Administration's (4) Estimates of Lead Intakes for Various Age Groups

this work						U.S. Food and Drug Administration	
		μgPb/day					μgPb/day
group	Ν	min	max	geometric mean	$\pm \sigma$	group	est range
0–12 mo	250	0.2	24.5	1.8	+2.3/-1.0	infants	0.8-5.7
13–24 mo	211	0.5	39.0	3.3	+3.0/-1.6	children, 2 yr	2.4-10.1
2-6 yr	288	1.1	41.9	4.1	+3.7/-1.9	children, 6 yr	3.5-13.2
mothers	252	0.8	92.4	7.5	+6.9/-3.6	women, 25–30 yr	3.5-15.6
						women, 40–45 yr	3.3-15.3
1.35 (a) 1.30 - 1.25 - 1.25 - -			1 1 qd ₂₀₀ /	35 (b) 30 - 25 -	, , , , , , , , , , , , , , , , , , ,	1.35 (c) 1.30 - 4 4 02 4	



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FIGURE 2. Plot of 200Pb/207Pb vs 200Pb/207Pb for children's diets. In each panel are also shown the Stacey-Kramers curve (S-K) (5) from 750 my ago to the present, the line fitted to the hand wipe and house dust samples (dust), and the average of the dust data. Panel a: children from birth to 12 months. Panel b: children from 13 to 24 months. Panel c: 2 to 6 year olds.



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FIGURE 3. Plot of ²⁰⁸Pb/²⁰⁷Pb vs ²⁰⁶Pb/²⁰⁷Pb for mothers' diets. The Stacey–Kramers curve (5) for 750 my to the present, the line fitted to the hand wipe and environmental samples (Omaha dust), and the average of the dust data are shown.

Pb Isotope Ratios in Sediments. Hemming and McLennan's measurements of the isotope ratios of lead in turbidites (6) and Millot et al.'s (7) in suspended sediment are presented in Figure 4 as ²⁰⁸Pb/²⁰⁷Pb versus ²⁰⁶Pb/²⁰⁷Pb plots. The ages of the turbidites are stated as modern, which in geological terms usually means less than 1 my old, but, even if some were deposited since antiquity, all with one exception lie in regions far from the influence of early European smelting (6). The sediments analyzed by Millot et al. (7) were collected worldwide from major rivers and their tributaries. The ratios of Al/Pb and Nd/Pb were used to gauge contamination by anthropogenic lead, which resulted in some from heavily industrialized countries being excluded. Millot et al.'s (7) remaining analyses (Figure 4b) are similar to those of the turbidites in Figure 4a and thus appear consistent with them. As the sedimentary materials are derived from source rocks whose ages are the same as those of rocks exposed in North America, their isotope ratios may be taken as an index of the range of ratios to be expected in United States soils.

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Hand Wipe and Dust Pb. Mines in Idaho, Missouri, and Utah and imports from Mexico have been the principal sources of lead historically used in the United States. Each has unique isotopic characteristics that reflect the manner in which the ore formed. Rabinowitz has attempted to define the isotope ratios of each source of domestic lead by analyzing ingots from museums (12). His average values form a triangular area that encloses the triangular area displayed by the hand wipe and dust samples. The mean isotope ratios of the Omaha dust falls midway between the means of the Missouri and Idaho ingots, suggesting that the average Omaha lead is an equal mixture of the two. There is, however, no clear evidence of any lead from Utah, which plots below Stacey-Kramers, and the third component appears to be lead from Mexico, the ratios of which plot close to modern lead (13). The absence of Utah lead suggests that the distribution in Omaha dust is peculiar to that city and not representative of the United States as a whole.

U.S. Aerosols. The aerosols collected over the United States by Bollhöfer and Rosman (*14*, *15*) resemble the Omaha aerosols (Figure 1b) in that (1) most of the measurements have ²⁰⁶Pb/²⁰⁷Pb ratios less than 1.22; (2) the line passes through the dust average; and (3) the mean ²⁰⁸Pb/²⁰⁷Pb and ²⁰⁶Pb/²⁰⁷Pb ratios of 2.444 and 1.184 are almost identical to the Omaha aerosol means of 2.446 and 1.187. The Omaha aerosols therefore appear typical of those encountered elsewhere in the United States and represent a source of contamination distinct from the local household dust.

Soil Pb versus Environmental Pb. It is seen from Figures 1 and 4 that predicted soil lead isotope ratios and environmental leads overlap in the region above the Stacey–Kramers



FIGURE 4. Plot of ²⁰⁸Pb/²⁰⁷Pb vs ²⁰⁶Pb/²⁰⁷Pb for modern sedimentary materials. Panel a: deep sea turbidites recalculated from Hemming and McLennan (*b*). Panel b: suspended fluvial sediment recalculated from Millot et al. (*7*). In both panels the Stacey–Kramers curve (*5*) for 750 my to the present and the line fitted to the hand wipe and house dust samples (Figure 1a) are shown.

curve, but that the region below Stacey–Kramers is uniquely soil lead, as is the region to the right of present day lead along the extension of the curve. Thus in many cases anthropogenic lead and primary soil lead should be distinguishable.

Dietary Pb Concentrations. The U.S. Food and Drug Administration (FDA) periodically measures the levels of contaminants in individual food items consumed in the United States and uses them to estimate dietary intakes for various population groups. The most recent survey covers the years from 1991 to 1996 (4). As lead was not detected in 74% of the items measured, the intakes are given as a range its bottom, the nondetects set to zero; its top, the nondetects set to the detection limit of the method (graphite furnace atomic absorption spectrometry). Table 1 reproduces the relevant FDA figures and shows the geometric means obtained in this study. Although the age groupings are not exactly comparable, our total diet study gave intakes that lie comfortably within the FDA estimates.

Dietary Pb Isotope Ratios. The most striking feature of the isotope ratios of the dietary leads (Figures 2 and 3) is the number of samples having high ²⁰⁶Pb/²⁰⁷Pb ratios that can neither be explained by contamination nor predicted by analyses of sediment. These samples, especially those in Figure 2, parts a and b, appear to be bounded by a line with a negative slope, which could only be generated by a source containing uranium but essentially no thorium. Candidates are phosphate fertilizer derived from the Permian (248-290 my) phosphorite deposits of Idaho, agricultural lime, or calcium salts derived from limestone used to supplement food. In Figure 5a a line has been drawn approximating the left-hand boundary of the diet samples. It is the same line along which the lead isotopes in essentially thorium-free minerals would evolve if they precipitated at the beginning of the Jurassic (206 my) from ocean water having the same isotope ratios as the average continental crust. This age, however, cannot be used to eliminate fertilizer because seawater lead varies widely in isotope ratio on account of its residence time being far less than the oceans' mixing time. To decide the source of the lead we turn to the diets of the children (Figure 2) and note that the diets of the 0-12 month olds, which contained large amounts of calcium-fortified baby formula, are the most influenced by this 206Pb-enriched lead (Figure 2a) and that the number of collections with high ²⁰⁶Pb/²⁰⁷Pb ratios decreases with the age of each group (Figure 2a-c). It is thus more likely that the lead with high $^{206}Pb/$ ²⁰⁷Pb ratios comes from food supplements because, if agricultural lime and phosphate fertilizer contributed significant amounts of lead, more diets would have their isotopic signatures. A further argument favoring calcium supplements is that in heavily contaminated soils the bioavailability of lead can be reduced by the addition of phosphate rock (16) and phosphate-containing biosolids (17) so that little lead from fertilizer may be available to crops. That some of the mothers' diets are also enriched in ²⁰⁶Pb (Figure 3) can be explained by the consumption of calcium-fortified drinks.

Also plotted in Figure 5a are the average ratios of aerosol lead as measured by us in Omaha (Figure 1b) and by Bollhöfer and Rosman (14, 15) in other U.S. cities and the average ratios of sedimentary materials (Figure 4) as measured by Hemming and McLennan (6) and Millot et al. (7). These form the base of a triangular area that has at its apex the isotope ratios of the supposed calcium additives, which for the sake of argument we have taken to be the point marked $\mu' = 80$. This area (Figure 5b) encloses the majority of the diets, which could be taken to mean that the lead they contain is a mixture of these three sources. While this may be true for some samples, such an interpretation ignores the fact that most of the diets fall close to the Omaha dust line whose average falls within the triangle, leading us to conclude that apart from calcium additives much of the lead in the diets of these subjects derives from contamination by household dust. In this regard, we noted in an earlier paper that the amount of lead in a group of children's diets appeared to correlate with the amount of lead wiped from their hands (18). Modern aerosol lead, in fact, seems to be a minor component as few samples cluster around its average value (Figure 5a).

A third, undefined source of lead is required to account for the samples that lie to the right of the triangle as defined above and also to explain the shift of the densest distribution of the diets to the right of the dust average. A possibility is tin which in the 1990s was permitted to contain up to 0.05% lead (19). The U.S. Food and Drug Adminstration does not monitor tin in food, but a British study (20) reports average tin concentrations of 79 ± 18 (σ) ppm for 11 samples of canned fruits such as are commonly eaten in the United States. The associated lead concentrations were 0.06 ± 0.02 (σ) ppm. For the United States, the lead contents of similar canned fruits are lower, averaging 0.016 ppm, but are nonetheless



FIGURE 5. Isotope ratios of dietary leads and proposed mixing components. Panel a: plot of ²⁰⁸Pb/²⁰⁷Pb vs ²⁰⁶Pb/²⁰⁷Pb for 999 duplicate diets obtained from mothers and children. In addition to the Omaha dust line and the Stacey-Kramers growth curve (5) for the evolution of Pb in the Earth's crust for the period from 750 my ago, the plot shows the evolution (limestone evolution) of the lead isotope ratios of thorium-free minerals that precipitated 206 my ago (the Triassic-Jurassic boundary) and contained lead with the ratios prevailing in the crust at that time. Where a sample falls on the line is controlled by its U/Pb ratio and is calculated from its present day ²³⁸U/²⁰⁴Pb ratio, conventionally designated by the symbol μ' . A μ' of 80 approximately equals a U/Pb ratio of 1.2. The plot also shows the average ratios of aerosols and sedimentary materials. Panel b: end member components potentially contributing to dietary lead. The locations of the points for calcium additives and tin coatings are speculative and were chosen as the minimum values to satisfy the data.

10 times higher than in their fresh counterparts (21). Why tin contains lead is not understood as the ore, the natural oxide or cassiterite, incorporates less than 5 ppm in its lattice (22). One possibility is that lead is contained in the fluxes used during smelting. Another is that it comes from the galena that is associated with cassiterite in certain types of mineralization. If, however, the ore body is alluvial, as many large deposits are, cassiterite is typically associated with a suite of resistant minerals of high specific gravity, including

the rare earth phosphates monazite and allanite. Both contain thorium so that, if the source rock was geologically old, they will contain lead enriched in the isotope ²⁰⁸Pb. Noting that in the early 1990s Brazil was the principal supplier of tin to the United States (23), much of which came from alluvial deposits associated with the 1.8 by Pitinga granites (24), we propose that the lead in tin was at least moderately thorogenic. In support of this we cite the case of the mother mentioned above who had dietary ²⁰⁸Pb/²⁰⁷Pb ratios greater than 3.1, because we can see no other way in which an American diet could contain geologically old lead derived from a source rich in thorium. An unusual combination of events, storage of the canned food at elevated temperature and leaving the contents in the opened can before transferring them to the collection jar, both of which accelerate the dissolution of tin (25), may have made her case unique. In Figure 5b a point labeled "tin coating" is shown. The ratios were arbitrarily chosen so as to be able to enclose the analyses lying to the right of the join between "average sediment" and 'calcium additives".

Imported food could also influence isotope ratios, and in the 1990s that imported by weight was 11% and included 100% of coffee, tea, cocoa, coconut oil, Brazil nuts, and cashews; 50% of fish and shellfish; and 17% of vegetables (26). The last of these contributes the most lead to the diet, 55% for infants, 30% for 2-6 yr old children, and 20% for 25-30 yr old women (4). In the case of fruit and vegetable imports from Mexico and Canada the uncertainty does not lie so much in the primary soil lead-the ages of the rocks in the main agricultural regions of those countries being similar to those in the U.S.-as in the contribution of any contaminating lead. Sweets contribute 10% of lead to a 2-6 yr old child's diet and 9% to a 25-30 yr old woman's (4). When these contain imports of chocolate products from Côte D'Ivoire and Ghana where the geologic basement is 2 by old (27), primary soil lead may tend to shift the U.S. dietary leads in the direction of the region below the Stacey-Kramers curve. The contribution of fish, which Egan et al. (4) include with meat and poultry, is probably small as the whole category contributes 10% of the lead consumed by children and adults. Without specific isotopically identifiable sources (with the possible exception of cocoa products), it is difficult to say how imported food has influenced the data of Figure 5a except that it has undoubtedly increased the scatter.

At the beginning of this paper we raised the question whether the lead content of food had fallen to the point where primary agricultural sources were identifiable. There is no clear answer. The overlap of the field of dust leads with those of sediment leads in the region between the dust line and the Stacey-Kramers curve renders any data points falling there ambiguous. The field to the right of modern day lead initially appeared to be exclusively that of sediment until evidence was presented that it could also be occupied by lead derived from tin coatings. Only the region beneath Stacey-Kramers is uniquely sedimentary lead. That few diets fall there does not necessarily mean that there is no soil lead in the diet because much of the country's richest farmland is developed on wind-deposited loess which, being derived from the detritus of continental ice sheets, is a mixture in which the isotope ratios of geologically old materials would be masked by the contribution of lead from younger rocks.

A total diet study measures all the lead a person consumes, including that incorporated during the preparation and serving of meals, and is more than just a shortcut to obtaining the combined contributions of lead from individual foodstuffs. With this in mind, it is instructive to return to Table 1 which showed the ranges most recently reported for women and children in the U.S. Food and Drug Administration's Total Diet study (4). Since our mean concentrations fell below the middle of their estimates and since our isotopic data suggest that for all groups, except the 0 to 12 month old children, house dust is a large component of dietary lead, it is likely that the concentrations of lead in the food items where the FDA detected no lead is much less than the graphite furnace detection limit of $0.008 \ \mu g/g$ that they reported (4). If individual foods were isotopically analyzed it may then turn out that very small amounts of lead in them are indeed of agricultural origin.

Acknowledgments

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