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CONTAMINATED AND NATURAL LEAD ENVIRONMENTS OF MAN*

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Introduction

The author of this paper is a geochemist, who, with his colleagues, has studied trace occurrences of lead in the earth and the oceans. In the course of this work it became necessary to consider the quantitative influence of industrial lead contamination and it was discovered that there has been a profound effect by this agency on the lead content of the oceans and of the atmosphere of the northern hemisphere (1) (2) (3) (4) (5).

A prevailing belief is that industrial and natural sources contribute more or less equal amounts of lead to the body burdens of the general population. It is also commonly believed that the significant range of natural lead concentrations in the blood is not much displaced from the interval between an average natural level and the average toxic level. A new approach to this matter suggests that the average resident of the United States is being subjected to severe chronic lead insult.

Natural Lead States

In our country, the concentrations of lead in the blood of people not visibly ill from lead poisoning will range between upper limits that are associated with acute intoxication and lower limits corresponding to levels which existed in man's prehistoric ancestors. The term "normal" has frequently been applied to concentrations of lead falling within this range, usually for cases not involving acute plumbism and not involving obvious occupational exposure to industrial sources of lead. In this report, the term "typical" is used in such instances, and the term "natural" is applied to the lower limits of the ranges of lead concentrations in contaminated environments.

As used here, the term "natural" refers to lead levels in body and environment which are equivalent to those which prevailed during the creation and evolution of our physiological responses to lead. The term "contaminated" refers to lead levels

which have been elevated above natural levels by man in activities which are an outgrowth of his abstract intellect. These definitions should help us to distinguish between "typical" and "natural" lead levels; they attach a needed physiological significance to the term "natural"; and they should help us keep in mind that uses of leaded pigments and cosmetics are acts equivalent in toxicological function to the more sophisticated acts of using leaded gasolines and insecticides.

The industrial use of lead is so massive today that the amount of lead mined and introduced into our relatively small urban environments each year is more than a hundred times greater than the amount of natural lead leached each year from soils by streams and added to the oceans over the entire earth (1). There are indications that about nine-tenths of the lead in the upper mixed zones of the open oceans in the northern hemisphere originates from lead mines (2) (3), and that the atmosphere of the northern hemisphere contains about a thousand times more than natural amounts of lead (5). It is difficult to exclude industrial contaminant lead even from the laboratory during sampling and analysis of materials for trace quantities of lead (6) (7) (8). The possibility of pervasive and high contamination of substances by industrial lead before sampling and analysis has already been recognized (9) (10) (11). The ratio of contaminant lead to natural lead may be very high in typical foodstuffs and biological materials, and lead concentrations in typical people and environments in the United States today may not even approximate natural levels. The process of contamination from mined lead has been operative on a significant scale for thousands of years. A study of tree rings (49) from one tree showed an increase in lead content in recent decades, and a study in the early thirties of supposed natural lead states in some Mexican Indians (48)

showed significantly low concentrations of lead in their blood. The analytical methods used in the latter study have been superceded by better ones, and it was also discovered that the Indians were subjected to contamination from lead glazed dishes, so that further investigations along these lines would be helpful and are expected. Frozen mammoth tissues, dated mammalian teeth and bones preserved in arid cairns, tree rings, fresh mammalian blood and tissues together with plants from remote islands or continents should provide helpful information in the future. In the absence of such information now, it is both necessary and revealing to set aside typical lead levels reported in the literature and to estimate natural lead levels by geochemical means.

The chemical composition of the biosphere is determined to a significant degree by the composition of the environment in which it evolved. To be sure, the bulk of the biosphere is made up of only a few elements which possess decisive chemical properties and which in some instances are only sparingly abundant, but the metallic elements, and in particular those which tend to form simple ionic bonds, show a (continued on page 3)

close correspondence between their abundances in the earth and their abundances in the biosphere. Consider the alkali metals. There are fundamental differences among the physiological activities of sodium, potassium, rubidium, and cesium in a living organism, but these differences neither cause nor obscure the striking similarity shown in table 1 between the abundances of the alkali metals on the earth's surface and in man.

We can use this principle to interpolate a body burden of lead from relations between concentrations of other trace elements in the earth and in the body. The nearest neighbors of lead according to mass are mercury and thallium on the lighter side and bismuth on the heavier side. Lead is also the heaviest of a series of elements of progressively increasing mass, germanium, tin, and lead, which are related by having an identical outermost atomic structure. The industrial production of these six elements in the United States for 1962 is listed in table 2. If contaminations are proportional to industrial production, the ratio of contaminant to natural amounts of these elements in the general population should be very much greater for lead than for the rest, regardless of contamination levels for the latter.

Calcium is abundant both in the earth's crust and in human bodies, and since we are interested only in relative abundances, the concentrations of the above six trace elements can be expressed as parts of the element per million parts of calcium in the earth's crust and as parts of the element per million parts of calcium in the body. The discrimination exerted by the body against these elements in favor of readily accepted calcium will then be reflected by concentration differences. Listed first in table 3 are the abundances (in calcium) of germanium, tin, and lead. As shown in the first column, their crustal abundances increase progressively with increasing atomic mass. According to relationships in table 1, we might therefore expect the natural body-burdens, which are given in the second column, to also increase in progressing from germanium, through tin, to lead. This is not the case, however,

Table 1

Alkali Metals in the Earth and Body

Metal	Atomic weight	Concentration in Earth's Crust (ppm)(12)(13)	Body Burden (gms/70 Kg)(14)
Na	23	26,000	105
K	39	27,000	140
Rb	85	120	1.2
Cs	133	1	<0.00001

Table 2

Production of some Heavy Metals in the United States during 1962 (15)

<u>Metal</u>	<u>Production (thousands of metric tons)</u>
lead	1,000
tin	79
mercury	2.3
bismuth	0.9
germanium	0.02
thallium	0.0 (?)

Table 3

Abundances of Some Heavy Metals in the Earth and Body

<u>Metal</u>	<u>Atomic weight</u>	<u>ppm in Ca of earth's crust (a)</u>	<u>ppm in Ca of body (h)</u>	<u>Conc. in earth Ca / Conc. in body Ca</u>
Ge	72	37 (b)	~ 6 (i)	6
Sn	119	150 (c)	6 (j)	25
Pb	207	550 (d)		
Hg	200	18 (e)	0.2 (k)	90
Tl	204	37 (f)	~ 0.6 (l)	60
Pb	207	550 (d)		
Bi	209	74 (g)	~ 0.4 (l)	180

References: (a): (16)
 (b): (17)
 (c): (18)
 (d): (1)
 (e): (20)
 (f): (21)
 (g): (22)
 (h): (14)
 (i): (23)
 (j): (24)
 (k): (25)
 (l): (26)

because another factor becomes important in determining body burdens when the total spread in abundances is small.

In a subgroup of the periodic table, the lightest metal tends to be most nutritive while the heaviest metal tends to be most toxic, as is shown, for example, in the three subgroups: potassium, rubidium, and cesium; calcium, strontium, and barium; and zinc, cadmium, and mercury (19). The body therefore tends to discriminate against the heavier metal of the series in favor of the lighter, and we cannot expect the slight crustal abundance trend in the germanium subgroup to be faithfully recorded in the body. It may, instead, be eliminated or even reversed. It is possible that the discrimination factors, which are given in the third column, increase in going from germanium to lead, so that the extrapolated natural body burden of lead might be less than 10mg Pb / 70 Kg body.

Such a value agrees with that indicated by the second group of metals listed in table 3, mercury, thallium, lead, and bismuth. Here, for an approximately constant but heaviest mass, the discrimination factors are uniformly high, and a similar discrimination value for lead would yield a burden of a few mg Pb / 70 Kg body. The uniformly high discriminations exhibited for each of the heavy metals regardless of their valence suggests that the relative crustal abundances of this second series of heavy metals should be approximately reflected in the body. The abundance relations of these six metals in the earth's crust and in the body suggest, by interpolation, that the natural body burden of lead is some few mg Pb / 70 Kg body.

We have arrived at this estimate through the use of three principles governing relationships between metal abundances in the earth and in the body which may be summarized as follows: (1) very large crustal abundance differences within a metal subgroup are approximated in the body; (2) the body discriminates against the heaviest metal in a subgroup in favor of the lightest, and this modification becomes significant when crustal abundance differences within a subgroup are small; and (3) metals with heaviest atomic masses and lowest crustal abundances are excluded from

the body with a uniformity which is relatively insensitive to group in the periodic table, so that their abundance differences in the crust are approximately reflected in the body.

Interest in the problem of poisoning by Sr^{90} , has recently led to the accumulation of data concerning the amounts of calcium, strontium, and barium in plants, man's diet, and in man's body which are considerably more accurate and extensive than for any other subgroup in the periodic table. These data are presented in table 4, and they illustrate beautifully and clearly the first two principles enumerated above, even to the extent of showing the influence of compounded discrimination by animal products in man's diet. Fortunately, we can apply these valuable data to lead with great confidence not only because of the third principle mentioned above, but in particular because the chemistry of lead and barium are alike in many respects, so that in the body lead exhibits a great similarity in behavior to the alkaline earths with respect to deposition in and mobilization from the skeleton. The average natural level of lead in man which is estimated with useful reliability from the data in table 4 is about 2 mg Pb/70 Kg body.

We can also estimate natural rates of lead ingestion and absorption from these same data. The average normal diet provides about 0.7 gm Ca/day (38), which corresponds to a natural ingestion of 20 μ Pb/day. This amount of lead in 2 Kg of a mixture of vegetable and animal products would yield a natural concentration of about 0.01 ppm Pb in food. The approximate fractions of alkaline earths that are absorbed by the intestine upon ingestion are: calcium 50% (39); strontium 25% (40); and barium 5% (41). If the alimentary absorption factor for lead in food is similar to that for barium, the amount of lead naturally absorbed by man is about 1 μ Pb/day, or about a thousandth of the natural body burden. This latter ratio is maintained for all alkaline earths, regardless of their absolute abundance in the body.

Table 4

Alkaline Earth Metals and Lead in Man and his Environment

(parts of metal per million parts of calcium)

Metal	Atomic Weight	Earth's Crust	Terrestrial Plants	Man's Diet	Mg metal/70 Kg Man
Ca	40	1,000,000(a)	1,000,000(e)	1,000,000(f)	1,000,000 (i)
Sr	88	7,400(b)	3,000(e)	1,500(g)	300 (j)
Ba	137	7,400(c)	1,100(e)	430(h)	22 (k)
Pb	207	550(d)			
(Estimated Pb)			80	30	2

References: (a): (12)

(b): (27)

(c): (28)

(d): (1)

(e): (29)

(f): (30)

(g): (31)(32)(33)(34)

(h): (33)(35)

(i): (14)

(j): (14)(27)(35)(36)(37)

(k): (14)(33)(35)(37)

Although the calcium content by wet weight of animals is about a percent, and the calcium content of plants is only a little less, the calcium content of man's diet is only a few hundredths of a percent. This is a consequence of selecting, for food, those portions of plants and animals that are low in mineral content, so that man's natural intake of lead may be considerably reduced over that which would result from an indiscriminate ingestion of entire plants and animals. This accounts for the probability, according to table 4, that natural lead may be about 0.03 ppm in whole animals, about 0.4 ppm in whole plants, and may still only be 0.01 ppm in uncontaminated human food.

If, in an uncontaminated situation, essentially all of the lead ingested by man originates from food, the amounts of ingested lead originating from water and air would be insignificant. Studies have been made of processes which determine the natural lead contents of water and air, and such studies can be used to check this possibility.

The concentration of lead in fresh surface waters during prehistoric times can be estimated from two figures: soluble lead entered the oceans during the last few hundred thousand years at an average rate of about 1.7×10^{10} gms Pb / yr (1); and today the total discharge of rivers to the oceans is 3.7×10^{16} liters / yr (12). In earlier periods this discharge may have been larger or smaller, but a reasonable estimate of the natural lead concentration in fresh surface waters would be about 0.5 μ g Pb / liter, or 0.0005 ppm.

In the order of decreasing importance, six significant sources of natural lead in the atmosphere are silicate dusts from natural soils, volcanic halogen aerosols, volcanic silicate smokes, forest fire smoke, aerosolic sea salts, and meteoritic smokes. Contributions from the first source are obtained from measurements of dust in rural air (42) and from the concentration of lead in silicates. An upper limit to annual contributions from the second source has been estimated (1) and 10% of this

can be assigned to an atmospheric reservoir. Contributions from the third source can be estimated by assuming that 1% of the mass of lavas and pumice extruded by volcanic activity is smoke and by assigning 10% of this smoke extruded annually to the atmospheric reservoir. Less than 1Km^3 is extruded each year and the lead concentration is less than 10ppm. Somewhat less than 0.1% of existing timber acreage is burned each year by lightning (42), and an upper limit to contributions from the fourth source would be obtained by burning 0.1% of the terrestrial biosphere each year, using the mass of the terrestrial biosphere (12) (corrected for water in cell fluids), the natural lead concentration in vegetation suggested by the data in table 4, making no correction for undispersed ash, and assigning 10% to an atmospheric reservoir. 6×10^{14} gms of sea salts are washed out of the atmosphere above land surfaces of the earth each year and are returned, via rivers, to the oceans (12), and this figure can be used to estimate natural contributions to the atmosphere from the fifth source mentioned above. This figure can be used to typify all areas of the globe, 10% of the resulting mass can be assigned to an atmospheric reservoir, and a concentration of 0.007ppm Pb in sea salts (2) can be used to arrive at a contribution. The input of meteoric and meteoritic material to the earth is about 1×10^{12} gms / yr (44), and the concentration of lead in this material is about 1×10^{-7} ppm (45) (46), so that an upper limit for contributions from the sixth source can be made by assigning all of this material to smoke and 10% of the mass to an atmospheric reservoir. The natural amounts of lead in the atmosphere, originating from these sources, are listed in table 5.

In summary, figures in this section indicate that in an uncontaminated environment man ingests about 20 μ Pb / day in food, about 0.5 μ Pb / day in water, and about 0.01 μ Pb / day in air, and possesses a burden of about 2mg Pb / 70Kg body.

(7a)

Table 5

Estimated Amounts of Lead in the Atmosphere from Natural Sources (γ Pb/m³)

<u>Source</u>	<u>Amount</u>
Silicate Dust	5×10^{-4}
Volcanic Halogen Aerosols	3×10^{-5}
Volcanic Silicate Smoke	6×10^{-6}
Forest Fire Smoke	6×10^{-6}
Aerosolic Sea Salts	1×10^{-6}
Meteoritic and Meteoric Smoke	2×10^{-9}

Existing Lead States

Listed in table 6 are the results of three surveys of lead in foods. The mean concentration of lead in the first five items in British foods is about 1.4 ppm. In the United States, the mean in such foods is about 0.2 ppm Pb. The fraction of the lead ingested by humans which is absorbed into systemic blood and then excreted by urine is small. That is, lead concentrations in feces yield good approximations of lead ingestion rates, and, indirectly, concentrations of lead in foods and beverages. A survey of a large number of persons in three cities in one state (48) gave a mean of 232 γ Pb in feces / person / 24 hrs, while another survey of a smaller number of persons in ten other cities in the United States (48) gave a mean of 398 γ Pb in feces / person / 24 hrs. The mean of these two figures corresponds to an ingestion of about 330 γ Pb / person / day, because about 5% of that ingested is excreted in the urine. If this amount of ingested lead is contained in 2 Kg of food, the concentration of lead in food obtained indirectly from these data is about 0.17 ppm, a figure similar to those obtained by direct observation and listed in table 6. In the United States today, the average concentration of lead in foods appears to be about 0.2 ppm.

A conservative estimate of lead concentrations in existing urban atmospheres, which would be applicable for plus or minus a decade, would be about 1.3 γ Pb / m³ (50) (51). That for existing rural atmospheres might be about 0.05 γ Pb / m³ (2) (5). An additional important contribution to respiratory lead exposure originates from tobacco smoke. The average 1½ pack-a-day cigarette smoker is exposed to about 24 γ Pb / day by inhalation.

The mean concentration of lead was found to be 54 ppm in the dissolved solids of ten rivers draining inhabited areas of North America, and 43 ppm for six rivers draining uninhabited areas (52). 25 ppm of lead was found in the dissolved solids

Table 6

Lead in foods from three surveys

<u>Substance</u>	Lead concentration (ppm)		
	<u>British Survey*</u> (ref. 47)	<u>U. S. Survey**</u> (ref. 48)	<u>U. S. Survey*</u> (ref. 49)
Beverages	4.2	0.18	0.05
Cereals	0.5	0.4	0.4
Fruits	0.4	0.5	0.2
Meats	1.5	0.3	0.2
Milk	0.3	0.03	0.2
Vegetables	0.3		0.2
Fish	0.9		0.5
Spices	3.4		0.3

*Average values.

**Mid-range values. Ranges only are given in reference.

of hundreds of samples of Maine lake waters (53) (54). The average concentration of lead in dissolved solids of rivers may be 50 ppm and for an average salinity of 150 ppm (12), this converts to 0.008 ppm Pb in existing river waters. Some earlier data yield, for a mixture of 14 well and 23 surface waters drawn from municipal systems, a concentration of 0.011 ppm Pb in United States water supplies (55). One of the earliest reports gave 0.016 ppm Pb in municipal United States water supplies (56). Analyses reported by the United States Water Quality Sampling Network (57), yield quite low concentrations of lead. About 80% of the water ingested by persons in the United States originates from rivers (42), and an average concentration of 0.008 ppm Pb may be a lower limit. Further contamination within municipal water distribution systems may have elevated this figure to about 0.011 ppm Pb, although this may be less than the average lead concentration in water actually consumed, since lead contaminations in the home and kitchen have not been studied.

The foregoing data can be used to summarize the rate at which the average person ingests lead today. Much of this lead is unmetabolized, however, and it is revealing to consider the amounts of lead absorbed by the body from different sources. Respiratory absorption amounts to about 40% of that inhaled (58), while alimentary absorption amounts to about 5% of that ingested (59), and using these figures, estimates of lead ingestion and absorption are tabulated and shown in table 7. The population can be divided into different groups according to variations in respiratory exposure, and their total rates of ingestion and absorption can be compared. This is done in table 8. By considering the distribution of the population according to where they dwell (43), what fraction are adults (43), and what fraction of the adults smoke (61), the fractions of population falling into the four groups listed in table 8 can be estimated. Blood lead concentrations tend to increase with increasing rates of lead absorption (48) (62) (63) (64), and recent studies have shown that urban

Table 7

Ingestion and Absorption of Lead by Contaminated Man

Substance	Intake/day (ref. 38)	Pb Concentration (this report)	Pb Ingested (γ Pb/day)	Fraction Absorbed (this report)	Pb absorbed (γ Pb/day)
Food	2 Kg	0.2 ppm	400	0.05	20
Water	1 Kg	0.01 ppm	10	0.1	1
Urban Air	20 m ³	1.3 γ /m ³	26	0.4	10
Rural Air	20 m ³	0.058 γ /m ³	1	0.4	0.4
Tobacco Smoke (ref. 60)	30 cigarettes	0.8 γ /smoke of one cigarette	24	0.4	10

(9a)

dwellers have higher blood lead levels than rural inhabitants and that smokers have higher blood lead levels than non-smokers. As shown in table 9, these blood data exhibit a proportional relationship which is in remarkable agreement with the relationship of estimated lead absorptions given in table 8. This close correlation shows that large differences in blood lead levels of large segments of a population result from different respiratory exposures to lead.

Not all of the lead which is absorbed into the systemic blood is excreted in the urine, so that, typically, a body-burden of lead is accumulated slowly over a period of years by the retention of a fraction of this metabolized lead. Total body burdens of lead that have been established in people by rates of absorption listed in table 8 have been evaluated by various investigators. Some estimates of the average body burdens of lead are listed in table 10.

Contaminated Lead Environments

Striking differences between natural and existing lead states are:

- (1) Existing average body burdens of lead are about one hundred times larger than natural burdens.
- (2) Existing rates of average lead absorption are about thirty times higher than natural rates.
- (3) Under existing conditions, atmospheric sources of lead make highly significant contributions to absorbed lead, while such sources make insignificant contributions under natural conditions.

Mention has already been made of the evidence for extensive contamination of the oceans and the atmosphere by lead from industrial sources. Now we see that a quantitative evaluation of possible differences between typical and natural lead levels in man suggests that man himself is severely contaminated. Let us

Table 8


Ingestion and Absorption of Lead by Populations
Exposed to Different Atmospheric Contamination

Group	Approximate Number of Persons	Pb Ingested (from Table 7) (γ Pb/day)	Pb Absorbed (from Table 7) (γ Pb/day)
Urban Smoker	3×10^7	461	41
Urban Non-smoker	7×10^7	437	31
Rural Smoker	3×10^7	436	31
Rural Non-smoker	7×10^7	412	21


Table 9

Correlation between Rates of Absorption and Blood Lead Levels for Different Respiratory Exposures

		<u>Rural Dweller</u>		<u>Urban Dweller</u>	
<u>Smoker</u>	[31]	(.17)		[41]	(.21)((.20))
			((.11))		
<u>Non-Smoker</u>	[21]	(.11)		[31]	(.17)((.15))



 Lead alkyl contamination



 Lead arsenate contamination

[] γ Pb/day absorptions estimated from exposures (table 8).

() ppm Pb in blood (ref. 65).

(()) ppm Pb in blood (ref. 66). Rural smokers unidentified.

Table 10

Total Body-Burden of Lead in 70 Kg Adult

<u>Estimate</u>	<u>Burden (mg Pb)</u>
ref. (14)(Tipton)	90*
ref. (67)	220*
ref. (48)	100 - 400
ref. (9)	750
probable average	200

*Includes 1 mg Pb from teeth (ref. 68).

**Includes soft-tissue values from Tipton (14).

consider whether the various possible sources of industrial contaminant lead can reasonably account for existing lead concentrations in food, air, and water.

If the natural concentration of lead in food is about 0.01 ppm, and the existing concentration is about 0.2 ppm, then most of this lead which amounts to some 2×10^7 gms in the total amount of food ingested yearly in the United States, should originate from industrial sources. Since the amount of lead produced industrially in the United States each year amounts to some 1×10^{12} gms (15) and is fifty thousand times larger than the yearly amount of food lead, it must be regarded as a possible major contributant to ingested lead. The significance of industrial lead contamination of food may be appreciated by considering ways in which some manufactured lead products enter food chains, and by evaluating the degree of contamination which results.

Folial contamination of food crops can be caused by fallout of decomposed lead alkyls from the atmosphere. Rapid fallout of such lead products in rural areas of the United States amounts to 2.4×10^{10} gms Pb / yr (69), a quantity which is over a thousand times larger than the amount ingested in food, so that contamination by only 0.1% of alkyl lead fallout on crop lands may be significant. Studies of folial contamination by fallout of Sr^{90} (71) (72) indicate that lead, by analogy, may be retained against rainfall washoff to the extent of about 10% on foliage, about 1% on fruits and pruned leafy vegetables, and 0.1% on edible parts of grains. The contamination process might consist of retention of dry fallout of partially soluble dusts on the waxy cuticle, followed by foliar uptake of soluble salts during subsequent rainfall and mechanical retention of insoluble salts on cuticle surfaces.

The pattern of rapid fallout of decomposed lead alkyls in rural areas consists of a network of lines, coincident with the road network. Using a mean width of 2×10^2 meters, the total area of this pattern (excluding all metropolitan, primitive, unimproved, and unsurfaced roads) is about $6 \times 10^{11} \text{ m}^2$ (70). Crop lands

and roads tend to coexist away from mountain areas and forested areas, so that the lead fallout pattern tends to be superimposed on crop lands, covering about 40% of such areas in the United States (42), and averaging about 4×10^{-5} gm Pb / m² / day for all crop lands. Foliage crops, which have an effective exposure of about 50 days and a yield of about 3×10^2 gms crop / m², are most highly contaminated, and concentrations of alkyl derived lead may average 1 μ Pb / gm crop. Fruits and vegetables, with an effective exposure of about 50 days and a yield of about 5×10^2 gms crop / m², may be contaminated by lead alkyls to the extent of 0.04 μ Pb / gm crop. Grains, with an exposure of 10^2 days and a yield of 3.5×10^2 gms crop / m², may be contaminated to the extent of about 0.01 μ Pb/gm crop.

The total amount of lead solder used in metal food containers is about 7×10^9 gms Pb / yr in the United States (73), a quantity which is three hundred times larger than the amount of food lead. Different foods etch different amounts of lead from solder, but with a per capita exposure of about 100 mg lead as solder/day; only small fractions of etched solder are needed to provide the daily supply of ingested lead. If a person ingests the contents of one #2 $\frac{1}{2}$ can (or the combined equivalent in smaller cans), soldered with 100 mg of lead, and if 0.3% of the solder has dissolved, he would receive his entire daily supply from canned food containing 0.3 ppm of lead. This is within the observed range of lead contamination from food can solder (74) (75) and it is reasonable to expect canned foods to contain some fraction of a ppm of lead originating from this source.

Lead arsenate insecticides are used today in the United States at a rate of 2.3×10^9 gms Pb / yr (15), which exceeds the rate of food lead ingestion by a factor of a hundred. The use of lead arsenate has declined greatly in recent years because of competition from organic insecticides. In 1941 about 85% of the lead arsenate used was applied to fruits and other food crops, while 1% was

applied to tobacco(76). Residues of insecticide are removed from some fruit by washing with dilute hydrochloric acid (77) so as to lower the lead content below a maximum permissible level of 7 ppm set by the United States Department of Agriculture. This high MPL suggests that it is difficult to use the insecticide and keep lead residues below a few ppm, and it is therefore reasonable to expect food crops treated with lead arsenate to contain about a ppm of insecticide lead.

The sources of food contamination discussed above are expended in a season and to a great extent are removed from further human contact by being absorbed in the clays of the land and ocean floors. There is another type of food contamination source which acts as a fixed reservoir of available lead which continuously bleeds into food chains, and examples of this are leaded ceramics and glazes in food processing equipment and kitchenware, lead alloys in food manufacturing equipment, and sheet lead used to manufacture chemicals that are in turn used to prepare and process foods and food acids and salts.

The amount of lead used in ceramics and the production of tableware glass, china glazes, and porcelain enamels for food processing equipment and kitchenware is significant but difficult to estimate because a considerable fraction of such glasses, glazes, and enamels are used for items unrelated to foods. If one fifth of the 2.5×10^{10} gms Pb / yr ceramics production (72)(73) is related to food equipment and the stored reservoir is equivalent to five years production, it would then equal 2.5×10^{10} gms Pb. A considerable quantity of lead exists as a reservoir in food processing and manufacturing equipment in the form of soldered joints, farings, and brass fittings. If the size of this reservoir is 1% of the total solder and brass production for the last two decades in the United States (15), the mass is 1.4×10^{10} gms Pb. The sheet lead reservoir in the United States, accumulated during the last two decades, is about 5×10^{11} gms Pb (15). These

reservoirs of lead are each about a thousand times larger than the amount of lead ingested yearly in food and may therefore contribute substantially to the latter by minute abrasion or dissolution.

In summary, even in the absence of data which would show the exact contribution of industrial lead to food, it is clear from the above examples of the amounts of lead and types of exposure that industrial sources could reasonably account for the difference between the existing lead concentration of 0.2 ppm in food and the estimated natural concentration of 0.01 ppm.

Man has disturbed natural leaching conditions in soils through agricultural stimulation of soluble denudation, thereby increasing the salinity and lead content of surface fresh waters above natural levels. Rapid fallout and washout of lead alkyl decomposition aerosols, dispersed lead ammunition, weathering products of lead paints, and seepage from lead metals and compounds in scrap and waste heaps all contribute lead to surface water supplies. These contributions should not affect ground waters, but surface waters supply 75% of public water utilities, which in turn, provide for about 80% of total household services and most potable industrial waters (42). Lead piping, paints, luting compounds and solder, contribute to water during its transport in distribution systems regardless of its initial purity. Lead in paints, glazes, enamels, solders, brasses, plastics, and glasses in kitchenware, tableware, and potable liquid dispensing machines contribute lead to the water after it has left the distribution system.

It is estimated (2) that the salinity of fresh waters has increased from about 40 ppm to 150 ppm as a result of agricultural stimulation of soluble denudation, and that although the natural concentration of lead in soluble salts may have remained about 12 ppm, the total amount of lead in fresh surface waters

may have been raised by this activity from 0.5 γ Pb/liter to about 2 γ Pb/liter.

Atmospheric washout of decomposition products of lead alkyls onto rural areas should for the most part be immediately held in the clay fraction of soil, and probably makes little contribution to surface fresh waters. The major contribution from lead alkyls to rivers probably originates from discharge of storm sewers draining urban areas. If metropolitan streets, roofs, and other surfaced drainages intercept one half of the rapid urban fallout of decomposed lead alkyls, and two thirds of this are soluble salts that are washed into sewers, such lead contributions at present amount to about 8×10^9 gms Pb/yr. River discharge in the United States is about 1.8×10^{15} liters/yr, yielding an average concentration of lead in rivers from lead alkyls sources of about 4 γ Pb/liter. As experiments with radioactive Pb^{210} show, concentrations of added lead in river waters decrease as a result of absorption and exchange with solid matter (78), and downstream concentrations of lead derived from lead alkyl sources are decreased by this process. On the other hand the value of the lead alkyl contamination effect given above is low as a result of using too large a volume of water, because river waters are extensively reused along short reaches of stream bed in urbanized areas.

Lead piping may be a significant source of contamination in water distribution systems. If the average 1" I. D. service pipe, which contains 1.5×10^5 gms of lead, admits 6×10^5 liters of water/yr and corrodes on the inside at a rate of about 30 gms Pb/yr (79), the lead content of the water will be about 50 γ Pb/liter. It is claimed that today many of America's greatest cities use lead pipe exclusively for water service connections (80). The lead pipe reservoir accumulated during the last four decades in the United States may be about 3.5×10^{11} gms (15), and may account for approximately 10% of public water

utility services (42). The average per capita drinking water contribution from lead water service pipes would then be about $5 \frac{1}{2}$ Pb/liter.

It is evident that the difference between the concentration of 0.01 ppm of lead found in municipal surface water supplies and the estimated natural concentration of 0.0005 ppm of lead in fresh surface waters can be accounted for as originating from rapid fallout of lead alkyl decomposition products in cities, from lead piping, paint and alloys in water distribution systems, and from agricultural stimulation of soluble denudation. There are no nation-wide data concerning the concentrations of lead in waters and beverages at the stage of consumption, but there are ample reasons for believing that they may be higher, as a result of additional contamination in the kitchen and factory, than at the stage of municipal supply.

Estimates of average contributions made to urban atmospheres by the principal sources of industrial lead, which are lead alkyls, lead paint dust and coal fly-ash, can be made by considering the volume and rate of renewal of urban atmosphere together with the rate of introduction of contaminants into this chamber on a nation-wide basis.

The urban area of the United States is about $6 \times 10^{10} \text{ m}^2$ (43). Below an inversion altitude of about 1 KM, lateral flushing and renewal by fresh air is rapid, compared to vertical mixing above this altitude and to removal of pollutants by rain washout, so that the volume of the urban atmospheric chamber is about $6 \times 10^{13} \text{ m}^3$. Pollutant aerosols may have half-lives of about three hours within this atmospheric chamber for the average large city, and their introduction into the air at ground level establishes steep concentration gradients such that concentrations near the ground may be about twice average concentrations for the total chamber.

Aerosols of decomposed lead alkyls, smaller than 5 microns in diameter, are introduced into the urban chamber described above at a rate of 4×10^{10} gms Pb/yr

on a 12 hr/day cycle because half of the passenger motor vehicle miles are driven in cities (70) and, of the total burned lead alkyls (69), half are converted to stable aerosols (69). The concentration of lead from this source should average about $0.6 \text{ } \mu\text{g Pb/m}^3$ in urban atmospheres near ground level.

Urban flyash production in the United States originates mainly from the burning of coal by power utilities, steel mills, cement mills and manufacturing industries (81). About 20% of this coal is ash, of which about one third leaves the boilers (82). Stable flyash aerosols, less than a few microns in diameter, form about 5% of the mass of ash leaving the boilers and are essentially uncontrolled by flue gas purification devices. This material amounts to about 3×10^{11} gms/yr. The concentration of lead in this silicate material is about 100 ppm (19). Assigning all production to urban areas, the concentration of lead originating from flyash aerosols should average $0.0001 \text{ } \mu\text{g Pb/m}^3$ in urban atmosphere near ground level. Coarse flyash will not be inhaled and is of little significance to problems of lead ingestion, but it will affect analyses of samples obtained by mechanically filtering the atmosphere. This material will have a short half-life in urban atmospheres as a result of rapid fallout, and in addition, about half of the coarse ash is kept out of the air by purification devices so that an upper limit to the average concentration of lead originating from coarse flyash in urban atmospheres near ground level should be about $0.004 \text{ } \mu\text{g Pb/m}^3$.

During the last forty years, 3.4×10^{12} gms of lead have been applied as paint on homes and buildings (largely on the exteriors) in the United States (15). Approximately 50% of the paint is removed by weathering, ablation, and peeling during an approximate seven year period before the paint is protected by new paint (83). The rate of application of leaded paints has diminished

today to one fifth of the rate forty years ago, so that changes in rates of application and ablation can be ignored in consideration of the total effects today from urban soils and burning buildings. Approximately one half the total paint has been applied in urban areas, and one fourth the total has been transferred to urban soils. Accounting for streets and buildings (70), the area of urban soils is about $2 \times 10^{10} \text{ m}^2$. Paint residues alone should have elevated the concentration of lead in the upper 5 cm of urban soils from 15 ppm to 600 ppm, and if half the 0.1 mg/m^3 of urban dust (42) originates from local soil, this source should contribute about $0.02 \text{ } \mu\text{g Pb/m}^3$ to urban atmospheres. About 5% of this material, or $0.001 \text{ } \mu\text{g Pb/m}^3$ is of a size that is significant in problems of lead absorption.

Contributions from lead alkyls and lead paints by routes different than those outlined above may be significant, and these include evaporative losses of lead alkyls, secondary transport of alkyl derived lead on asphalt and soil dusts and on smoke from incinerated trash, and secondary transport of paint lead on smoke from burning buildings and incinerated trash. There are no other contributions of lead to urban atmospheres from industrial sources which are comparable to those from lead alkyls, lead paints, and coal ash. Primary lead smelting operations are carried out in rural areas in the United States (84) with the exception of a large plant located in El Paso, Texas. Secondary lead refining operations contribute considerably less lead to the atmosphere than primary smelting.

The difference between existing lead concentrations in urban atmospheres of $1 \text{ } \mu\text{g Pb/m}^3$ and natural concentrations of $0.0005 \text{ } \mu\text{g Pb/m}^3$ can be accounted for by contributions from industrial sources. Essentially all lead that is significant with respect to respiratory absorption originates from lead alkyls.

This brief survey shows that industrial sources can provide the lead we absorb. Whether this is the actual case depends upon the natural levels of lead in different materials. It is virtually certain that all toxicologically significant amounts of lead in air originate from industry because the difference between existing concentrations and inferred natural concentrations of lead in the air is extreme and because contributions from natural sources are known reasonably well. The same is probably true for lead in surface waters. Figures given here will be improved, but the difference between existing concentrations and inferred natural concentrations of lead is large compared to the uncertainties of the values so that the sign and magnitude of the difference is probably real. It is more than just possible that the same is also true for the lead in our food. Although the abundance relationships of the metals in the biosphere, their positions in the periodic table, and their abundances in the earth's crust may be unfamiliar, this does not make it improbable that they can compel us to believe that existing lead levels in our food and bodies greatly exceed natural levels.

It should be possible to measure natural lead levels directly in some materials believed to be free of lead contamination. Because of the pervasive extent of contamination, the collection of such material is a difficult problem. The effects of contamination cannot be easily avoided by going back in time. If we go back to Pleistocene fossils the problem of chemical alteration by the action of percolating ground water becomes very complex. If we estimate natural body burdens of lead from bones or teeth of long dead persons preserved in burial vaults, this presents the problem of environmental control in its most difficult form. Significant fractions of human populations have been exposed to lead contamination for thousands of years, and by choosing this, instead of contemporary material, one simply abandons verifiable controls for virtually nonexistent historical records.

This problem is not easily solved by going to different continents. There is definite evidence that the Indians prized and used galena several thousand years ago in the Mississippi Valley (85).

The problem is not readily solved by the study of extant primitives. Mexican Indians near Toluca were studied in 1931 and the amounts of lead they ingested were reported to be less than one-third of those ingested in the United States (3), but these Indians ate food prepared and served in dishes glazed with lead, and the contributions from this and other natural sources are uncertain. Analyses of contemporary mammalian and plant tissues taken from remote regions of South America or the South Pole might be helpful, but careful consideration should be given to all possible sources of lead contamination before material is collected and analyzed. Foods and their environment might be analyzed at various stages of progress through contemporary production and processing chains.

Some misconceptions have arisen concerning the relative contributions of lead to urban atmospheres from different industrial sources. It has been suggested that lead concentrations have been reduced as a consequence of smoke abatement (86). Data offered in support of this view are questionable because the locations of sampling stations, sources of lead pollution, and filtering efficiencies were not controlled as a function of time. It is possible that groups of associated elements can be investigated and used to trace the different origins of atmospheric lead. The association of chlorine with sea salts or vanadium with fly ash are examples. Consider the atmospheric vanadium to lead ratio at times of maximum smoke pollution in those cities which supposedly experienced a subsequent reduction in atmospheric lead. It was about 0.04 (50) (87). Since the vanadium to lead ratio in coal ash is about 100 (19), the discrepancy of a factor of over a thousand makes it clear that flyash was inconsequential as a lead contributor to the atmosphere of those cities.

Natural Blood Lead Levels

We want to know where typical body burdens of lead lie with respect to natural and toxic levels. Toxic thresholds for total body burdens of lead are less readily defined than toxic thresholds of lead concentrations in the blood, so that it is useful to identify the natural level of lead in the blood. The average natural body burden of about 2 mg Pb/70 Kg body, estimated here, can be converted to an average natural blood lead level by distributing lead in the body in natural states in the same proportions as in contaminated states. In the United States, the significant range of lead in blood is about 0.05 ppm to 0.4 ppm with a mean value of about 0.25 ppm (48) (62) (63) (65) (66), while the mean body burden is about 200 mg Pb/70 Kg body (Table 10). The average natural blood lead levels would then be about 0.0025 ppm. We will consider briefly a few factors which might affect this projection.

If most of the lead is stored in the skeleton in contaminated states, this reservoir might be diminished in natural states, while blood lead levels might remain relatively constant. If this were the case, lead in the blood would comprise 70% of the body burden of lead in the natural state, and would be equivalent to the total amount of lead absorbed in a four year period. This is unlikely, first, because formed elements of the blood which contain most of the blood lead in contaminated states (63), are not sufficiently long-lived to acquire such large concentrations at such small rates, and second, the blood usually contains amounts of metals equivalent to those absorbed during a period of a few days instead of thousands of days.

The concentration of lead in the blood might be under homeostatic control, and would therefore remain relatively constant. The fraction of stored skeletal

lead contained in the active pool of bone-crystal-surface (88) which involves about 1% of total body calcium and which is utilized by the body to maintain a strictly constant serum-calcium level (89), serves as a reservoir of readily available lead whose mobilization is sensitive to those factors which mobilize or inactivate calcium such as vitamin levels, parathyroid activity, acidity of the blood, and nutrition (90) (91) (92) (93), but such responses are inadvertent, as are the responses for any similar trace element, such as strontium (40), for such control is exerted only in response to variations in concentrations of the grossly abundant element, whether it be calcium in the natural state, or strontium in the experimental state. The inadvertent mobilization or inactivation of lead by calcium homeostasis is a secondary, and not a primary controlling factor in determining blood lead concentrations.

Except for a tendency of the body to reject a larger fraction of an ingested trace element with an increase in the amount ingested (46),

gross tendency of the body is simply to reject with relatively constant efficiency, the heaviest toxic metals. One might therefore expect both the body burden and blood lead levels to fluctuate with the rate of lead ingestion. Lead concentrations in the urine tend to reflect short term fluctuations in lead absorption better than lead concentrations in the blood, but over long periods blood lead levels increase with increased rates of absorption (48) (62) (63) and in a natural state should vary according to differences in natural rates of ingestion. The range of the latter should be small. The significant range of lead in source rocks of soil is about 5 ppm to 25 ppm, with a mean of about 15 ppm (1), while the significant range in genetically effective primeval soils might have been less due to homogenization by weathering and transport and the mean might have been less due to prolonged leaching. Extreme variations which have been mentioned for

soils (48) may not be significant because they represent both contaminated and unnatural materials. It has been pointed out that large populations may today be living off soils that are deleterious because such soils are chemically unnatural with respect to our genetic heritage (94).

There are factors which, by themselves, suggest that typical blood lead levels are unnaturally high. As shown in table 9, persons who did not inhale large amounts of industrially derived lead exhibited greatly reduced blood leads. One may ask whether similar large reductions in blood leads would be exhibited by rural non-smokers if they were to ingest food uncontaminated by industrial lead. If such would be the case, and as we have seen, this is not unlikely, the remaining amount of blood lead would be very small.

As mentioned earlier, in the United States the average concentration of lead in blood is 0.25 ppm, yet the threshold for classical lead poisoning has been said to be 0.8 ppm (48) (62) (63), and most recently has been proposed to be 0.5 ppm (64). In view of the ready response of blood lead levels to changes in rates of lead ingestion and absorption, one may ask why is there so narrow a margin of safety and the body so poorly defended if natural blood levels are about 0.2 ppm.

Experimental checks could be made by determining, by ingestion over relatively long periods, the toxic thresholds of mercury, thallium, and bismuth in the blood of animals, together with natural levels, since contamination by industrial sources of these metals is a problem that can be handled with some certainty. The peculiar affinity of lead for the membranes of red blood cells in the contaminated state might also be utilized as a check. Although investigators have studied the distribution of lead between serum and the formed elements of the blood as a function of increasing concentration of lead, starting with the contaminated state, no one has studied what happens as a function of decreasing amounts of ingested lead over long periods. This could be done in humans by simply searching for and studying cases of

low blood lead levels. Controlled animal studies of lead would be difficult because of contamination problems, but it would be interesting to follow the distribution of mercury, thallium and bismuth between serum and formed elements of the blood in animals as a function of increasing metal concentration, starting at natural levels and ending at toxic thresholds and seeing whether the distribution changes with concentration.

Chronic Lead Insult

0.25 ppm of lead in the blood has been regarded as more or less natural for a long time, but such a level seems actually to lie between an average natural level of about 0.0025 ppm and an acute toxic threshold of 0.5 to 0.8 ppm. This suggests that the average resident of the United States is being subjected to severe chronic lead insult.

Our knowledge to date about classical lead poisoning is largely clinical and morphological, and it usually applies to combinations of rates and periods of lead absorption which yield blood lead concentrations exceeding 0.5 to 0.8 ppm. Resulting changes in cell morphology are widespread throughout the body, but many are not yet recognized as being specific for lead and little is known of the mechanisms which cause them. The evidence is therefore permissive for some lesions or altered cell metabolisms, which either have not been identified or have not been assigned to exclusive origins, to result from lead absorptions which correlate with blood lead concentrations considerably above a natural level, but less than 0.5 ppm.

The economic pressure for expedient exposure to lead can be balanced by different kinds of knowledge. One kind is a basic understanding of mechanisms of lead metabolism. This is acquired slowly, however, and it would be tragic

if, many decades from now, it were recognized from accumulated evidence that large segments of our population had suffered needless disability because another kind of knowledge — early warning signs — went unheeded. Intellectual irritability and disfunction are associated with classical lead poisoning, and it is not impossible that similar impairments on a lesser but still significant scale might occur in persons subjected to severe chronic lead insult. It is interesting and not at all unworthy to consider how the course of human events may have been altered by lead contamination.

Summary

There are definite indications that residents of the United States today are undergoing severe chronic lead insult. The average American ingests some 400 micrograms of lead per day in food, air, and water, a process which has been viewed with complacency for decades. Geochemical relationships and material balance considerations show that this ingestion of about twenty tons of lead per year on a national basis is grossly excessive compared to natural conditions. It probably originates from the million tons of lead dispersed yearly in such forms as lead alkyls, lead arsenates, and food can solder, and from the many millions of tons of lead accumulated throughout past decades and stored as paints, alloys, piping, glazes and spent ammunition. Existing rates of lead absorption are about thirty times higher than inferred natural rates, yielding body burdens of about 200 mg Pb/70 Kg body, and blood concentrations of 0.25 ppm Pb, which values are about a hundred times above inferred natural levels of 2 mg Pb/70 Kg body and 0.0025 ppm Pb in blood. Existing blood lead concentrations have for decades been regarded as natural, although it is well known that the average value lies only slightly below

threshold levels for classical lead poisoning which are 0.5 to 0.8 ppm Pb. It appears that the following activities deserve serious consideration and support: defining natural and toxic lead levels with greater care than has been shown in the past; investigating deleterious effects of severe chronic lead insult; investigating the dispersion of industrial lead into food chains; elimination of some of the most serious sources of lead pollution, such as lead alkyls, insecticides, food can solder, water service pipes, kitchenware glazes, and paints; and a re-evaluation by persons in positions of responsibility in the field of public health of their role in this matter.

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