Analysis and reduction of the uncertainty of the assessment of children’s lead exposure around an old mine

Philippe Glorennec*

Ecole Nationale de la Santé Publique, Avenue du Pr Léon Bernard, 35043 Rennes Cedex, France

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Abstract

Exposure to lead is a special problem in children, because they are more highly exposed than adults and because this pollutant, which accumulates in the body, induces neurobehavioral and cognitive effects. The objective of this study was to determine the probability density of the lead exposure dose of a 2-year-old child around an old mine site and to analyze its uncertainties, especially those associated with the bioavailability of lead in soil. Children’s exposure was estimated indirectly from environmental samples (soils, domestic dust, water, air) and parameters (volume inhaled, body weight, soil intake rate, water intake, dietary intake) from the literature. Uncertainty and variability were analyzed separately in a two-dimensional Monte Carlo simulation with Crystal Ball® software. Exposure doses were simulated with different methods for accessing the bioavailability of lead in soil.

The exposure dose per kilogram of body weight varied from 2 μg/kg day at the 5th percentile to 5.5 μg/kg day at the 95th percentile (and from 2 to 10 μg/kg day, respectively, when ignoring bioavailability). The principal factors of variation were dietary intake, soil concentrations, and soil ingestion. The principal uncertainties were associated with the level of soil ingestion and the bioavailability of lead. Reducing uncertainty about the bioavailability of lead in soil by taking into account information about the type of mineral made it possible to increase our degree of confidence (from 25% to more than 95%) that the median exposure dose does not exceed the Tolerable Daily Intake. Knowledge of the mineral very substantially increases the degree of confidence in estimates of children’s lead exposure around an old mining site by reducing the uncertainty associated with lead’s bioavailability.

Keywords: Lead exposure; Health risk assessment; Lead bioavailability; Monte Carlo simulation; Lead mining

1. Introduction

Lead is a cumulative pollutant that induces neurobehavioral and cognitive effects in children (IPCS (International program on Chemical Safety), 1995). Harmful effects, especially on intellectual quotients (Canfield et al., 2003), have been reported for relatively low blood lead levels, below 100 μg/L (10 μg/dL). The studies so far published do not appear to show any threshold blood lead level below which there is no health effect (INSERM, 1999).

Exposure to lead occurs principally through the lungs and the gastrointestinal tract: the cutaneous pathway is considered negligible in humans (IPCS, 1995).

Young children (0–6 years), because of their behavior, especially their hand–mouth habits, are particularly exposed to lead. Such exposure may take place in homes with a history of lead-based paints but also around industrial sites that release or once released lead (Hivert et al., 2002).

In France, an INSERM (National Institute for Health and Medical Research) expert advisory group (INSERM, 1999) recommends screening children in at-risk areas, especially those identified around industrial sites. As screening for lead poisoning requires a blood sample, which is an invasive procedure, especially in young children, the Institute for Health Surveillance...
recommends (Glorennec et al., 2002) a preliminary assessment to examine population exposure around an industrial site before determining the need for a screening program.

This procedure was followed in Trémuson, France, a site where lead, silver, and zinc ore were mined from 1690 through 1931. Although active mining no longer takes place on the site, exposure from soil and dust continues; studies (Bjerre et al., 1993; Gulson et al., 1994; Murgueytio et al., 1998; Sterling et al., 1998; Malcoe et al., 2002) conducted around old mines have observed higher mean blood lead levels in such communities. Many studies (Xintaras, 1992; Mielke and Reagan, 1998; White et al., 1998) stress the importance of soil and dust in population exposure to lead. Soil particles (either directly or indirectly after transformation into house dust (Mielke and Reagan, 1998; White et al., 1998)) can be ingested by adults and children through unintentional hand–mouth contact, geophagia, or dust inhalation. A study (Lanphear et al., 1996) among children in Rochester (NY, USA) indicates that dust lead content explains most of the variance in blood lead levels. A review (Mielke and Reagan, 1998) found a positive correlation between lead levels in soils and those in blood.

At Trémuson, an exposure assessment had already been conducted (Glorennec et al., 2001) and submitted to the local health authorities to determine the usefulness of screening for childhood lead poisoning. The aim of the work presented here is to help identify the types of information that should be collected in priority in this type of situation. A means of describing the sensitivity with respect to different exposure factors is the technique of Monte Carlo simulation. In this probabilistic approach, all variables and parameters used in risk assessment may be regarded as distributions throughout the analysis. A process of repeated simulations is then used, during which the estimated quantity (risk in this case) is calculated many times with randomly chosen values of variables and parameters, covering their range of variability and reproducing the assumed distribution density. The final result is given in the form of a probability distribution of risk (Biesiada, 2001). Beyond ranking the sources of uncertainty and variability, this study presents the probability density of the exposure dose of a 2-year-old child with its degree of certainty, according to the extent of consideration and knowledge of lead bioavailability.

2. Material and methods

2.1. Population studied

The village of Mines, a subdivision of Trémuson, is a rural hamlet of approximately 400 inhabitants in a wooded valley. The homes are old single-family houses, packed densely together. Public premises are limited to a cafe/bar, a sports field, footpaths, and a children’s playground.

The area that may have been contaminated by air pollution from the mining activities is limited to the village of Mines, downhill from the buildings used for mineral extraction and treatment. We initially studied the population of children aged 0–6 years (N = 20) who regularly spent time in the area. The probability density of the exposure dose is presented here for a 2-year-old child, the most exposed in that kind of environment (INSERM, 1999).

2.2. Exposure by ingestion to outdoor soils and indoor dust

Samples were taken from the houses (N = 14) where young children lived; no indoor samples were taken from 1 house (refusal) and no outdoor samples from another (no courtyard or garden). For indoor house dust, three samples were taken for each home, from dust collected from all smooth surfaces in rooms (average 15–20 m²/room) used by the children (according to the parents): living room, kitchen, and bedroom. Samples were taken with a rubber blade, so that all of the dust on the room’s surfaces could be collected without scratching. Outside soil samples were taken with a trowel at a depth of 0–0.01 m. Three soil samples were taken at each house. Each sample was the mixture of one primary sample and four peripheral samples 1 m away from the places most used by the children, according to their parents. The distributions of the concentrations were interpolated with Crystal Ball® software, based on concentrations measured by the following procedure (US EPA, 1997): graphic choice of the shape of the distribution and verification that it is not rejected by the Anderson–Darling test, which is appropriate for considering the values of the distribution tail.

The ingestion rates that we used for soils and dust are those recently proposed by Stanek et al. (2001) for a two-dimensional Monte Carlo simulation. This is a normal distribution N(31, 31) of soil ingestion with its associated uncertainty, in the form of a deviation correlated with soil ingestion (the uncertainty is higher for higher ingestion rates) with normal distribution N(4;0). This is, to our knowledge, the only published study that separates variability and uncertainty and presents an estimate of long-term soil ingestion. This distribution is consistent with the recommendations for point estimates of soil intake rates of the US Environmental Protection Agency (2001) for this age group.

2.3. Dietary exposure

Because of the low transfer rate of lead toward fruits and vegetables (IPCS, 1995) and the absence of
consumption of local food products (Glorennec et al., 2001), exposure from food was presumed equal to the national mean.

Because the distribution of dietary lead in France is not known precisely, we derived it, in part from the most recent French spot estimate for children aged 2 years (INSERM, 1999), i.e., 30 µg/day. Dispersion around this value was estimated from a study (Leblanc et al., 2000) of the lead content in French meals (N = 93): Leblanc et al. thus observed a lognormal distribution of the arithmetic mean, which was 15 µg/meal, and of the standard deviation of 4.5 µg/meal, with a range of 8.5–37 µg/meal. The distribution used here, assuming a similar coefficient of variation for the two distributions, is therefore a lognormal distribution of an arithmetic mean of 30 µg/day and a standard deviation of 9 µg/day (limited to 17–74 µg/day, in accordance with Leblanc et al.’s data).

2.4. Exposure in water

Because the age of the homes meant that their plumbing might use lead pipes, water samples were taken in homes (N = 14) with young children. After letting the water run for 3 min at 5 L/min, the faucet was turned off to keep the water sitting. After 30 min, a 2-L sample was taken, without preliminary draining.

We are aware of only one published study about the consumption of water by young children in rural areas of France (Gofti-Laroche et al., 2001). Gofti-Laroche et al. estimated that the distribution of the consumption (N = 22) of those aged 0–4 years (N = 22) in their study area was lognormal, with a mean of 1.19 L/day and a standard deviation of 0.85 L/day.

2.5. Exposure by inhalation

The air samples (N = 2) were taken at a height of 1–1.5 m, with a Gilair 5 pump (pump standard, NFX 43–265; flow 2.5 L/min, collection filter, 37-mm cassette with MCEF 0.8-µm filter for lead analysis). The filters were analyzed at the end of a week. The weather conditions were sunny and dry, favorable to the redispersion of dust.

The distribution of the inhalation rate was considered normal (6.8; 1.275 m³/day), based on American data from the US Environmental Protection Agency (2002).

2.6. Analytic methods

The soil and dust (terrestrial or atmospheric) samples were stored in sealed glass vials. The analyses were performed with inductively coupled plasma-atomic emission spectroscopy (standard EN ISO 11885). Analyses of water complied with the NF T90-119 standard.

2.7. Body weight

We used the distribution of body weight in France that has been reported by the Ministry of Health (Ministère de l’Emploi et de la Solidarité, 1995).

2.8. Bioavailability of lead contained in soils and dust

We tested several levels of information about knowledge of the chemical form of lead, because bioavailability is a useful parameter for exposure models (Technical Review Workgroup for Lead (TRW), 1999):

- ignoring bioavailability, intake equal to 100%,
- considering a default bioavailability of 30%,
- recognition of the existence of the uncertainty associated with bioavailability but no information: bioavailability can vary from 1 to 100% (TRW, 1999),
- knowledge of the lead form in soils, here galena (PbS) (BRGM (Bureau de recherches géologiques et minières), 1995). We used a uniform distribution of 1–22% of relative (compared with food) bioavailability, i.e., the range noted by Ruby et al. (1999) for galena (PbS) in his literature review. This distribution is consistent with the values obtained from in vivo tests (on piglets being weaned) (Henningansen et al., 1998) of mineral soils including PbS; he observed bioavailability of less than 25% from galena-based mineral soil. The range is relatively large because the chemical form alone does not determine bioavailability; the size of the particles also plays an important role (Steele et al., 1990; Mushak, 1998).

Table 1 summarizes the exposure parameters.

2.9. Estimated daily dose (EDD)

The estimated daily dose of lead was calculated according to the formula $EDD = \sum (C_i \times Q_i)$, where $C_i$ and $Q_i$ represent, respectively, the lead concentration in medium $i$ and the mean daily consumption of medium $i$, by kilogram of body weight. Concentration for the dust and soils exposures was weighted by the bioavailability of the lead.

The Tolerable Daily Intake (TDI) of 25 µg/kg of body weight per week recommended by the joint FAO/WHO committee (JECFA, 1999) was used as the reference.

2.10. Simulation parameters

The uncertainty and variability of the distributions were estimated by Monte Carlo simulation (Cullen and Frey, 1999), with Crystal Ball® software, professional academic edition v5.5. The sensitivity analysis used 1000
simulations, and each of the separate analyses of uncertainty and variability used 250.

The following variables were correlated: body weight and dietary lead intake \((r = 0.2334\), AFSSA (French Agency for Food Safety), personal communication) assuming that the latter is proportional to total diet lead levels in soils and dust \((r = 0.34\), observed from the data collected), and soil ingestion and its associated uncertainty \((r = 0.96\), with uncertainty highest for the highest lead levels \((Stanek et al., 2001)\).

The following parameters were considered variable: soil and dust concentrations and extent of contact with the various media (consumption of water, dietary intake, soil intake rate, inhaled volume, and body weight). The following parameters were considered uncertain: bioavailability, ingestion rate, and atmospheric and water concentrations (because the true values, below the detection limit, are unknown).

### 3. Results

#### 3.1. Lead levels observed in the environment

**Soils and dust:** The lead content of soils varied substantially (cf. Table 1) from one place to another and sometimes even within a single dwelling. The dust levels were generally lower and presented less variability (cf. Table 1), both between and within dwellings.

**Water:** Lead concentrations were all below the detection limit of 3 \(\mu g/L\).

**Air:** Lead levels measured were less than the detection limit of 0.5 \(\mu g/m^3\).

**EDD:** Fig. 1 summarizes the EDD distribution according to the degree of uncertainty. The contributions of the different parameters to the variability of the result (uncertainty + variability) are reported in Fig. 2.

### 3.2. Effect of consideration of bioavailability

Fig. 3 reports the results of the simulation with the specific bioavailability of galena taken into account. The effect on the EDD of considering bioavailability is shown in Table 2.

### 4. Discussion

The air was sampled for a 1-week period. Because of this brevity, the meteorologic conditions, and the use of the site, these measurements cannot be considered representative of an annual period. Nonetheless, taking the measurements in the summer, during the dry period, is propitious to redispersion of soil particles and should tend to result in overestimating their concentrations in the air. Even overestimated, inhalation represents only 5% of the exposure dose: the non-representativeness of the air samples therefore has no notable consequences on the estimated exposure.

Dietary exposure was not specifically evaluated but was presumed to be equal to the national mean. The underlying hypothesis is that no local dietary...

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### Table 1

<table>
<thead>
<tr>
<th>Exposure/parameter</th>
<th>Distribution</th>
<th>(P10; P50; P90)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Air</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Inhalation rate ((m^3/day))</td>
<td>Normal distribution (N(6.8; 1.275)) (from the US EPA child specific exposure factors handbook) (US EPA, 2001)</td>
<td>(5.2; 6.8; 8.4)</td>
</tr>
<tr>
<td>Lead concentration in air ((\mu g/m^3))</td>
<td>Uniform ((0; 0.5)) distribution, fitted to measured data ((N = 2))</td>
<td>(0.05; 0.25; 0.45)</td>
</tr>
<tr>
<td><strong>Soil and dust</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ingestion rate ((mg/day))</td>
<td>Normal distribution (N(31,31)) with normal uncertainty (N(0.4)) (from Stanek et al., 2001; Ruby et al., 1999; Leblanc et al., 2000)</td>
<td>(6; 38; 77) (at P50)</td>
</tr>
<tr>
<td>Lead concentration in dust ((\mu g/g))</td>
<td>Lognormal, fitted to measured data ((N = 35))</td>
<td>(65; 168; 439)</td>
</tr>
<tr>
<td>Lead concentration in soil ((\mu g/g))</td>
<td>Lognormal, fitted to measured data ((N = 39))</td>
<td>(43; 323; 2442)</td>
</tr>
<tr>
<td>Relative bioavailability (%))</td>
<td>Uniform ((0.01–0.22)), from Ruby et al. (1999). Other distribution also used for test: Uniform ((0.01–100)) or fixed values</td>
<td>(3; 12; 22)</td>
</tr>
<tr>
<td><strong>Dietary intake</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Central estimate 30 (\mu g/day) (Inserm 99) with dispersion parameters from the distribution of Pb intake observed by Leblanc et al. (2000) in French lunches</td>
<td>(26; 30; 34)</td>
<td></td>
</tr>
<tr>
<td><strong>Water intake</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Water consumption ((L/day))</td>
<td>Lognormal distribution ((1.19; 0.85)) observed in a French rural area (Gofft-Laroche et al., 2001)</td>
<td>(0.4; 1.0; 2.2)</td>
</tr>
<tr>
<td>Lead concentration in tap water ((\mu g/L))</td>
<td>Uniform ((0; 0.3)), fitted to measured data ((N = 8))</td>
<td>(0.3; 1.5; 2.7)</td>
</tr>
<tr>
<td><strong>Body weight</strong></td>
<td>Fitted to observed values (French Ministry of Health)</td>
<td>(10; 12; 14)</td>
</tr>
</tbody>
</table>
specificities or peculiarities should have led to excess exposure, which would require both substantial consumption of local products and their contamination. The population survey showed that this consumption was marginal, and the analyses revealed no particular contamination (Glorennec et al., 2001). Dietary intake was estimated on the basis of INSERM’s general estimate (INSERM, 1999) for children 2 years of age. It is probably overestimated because it relies on data from several years ago, although lead levels in the environment and in food have been decreasing since lead was banned in gasoline. Using foreign data is nonetheless questionable since leaded gasoline was banned in France later than elsewhere. Because food represents more than half the exposure dose for most of the children, the error induced is not negligible. Nonetheless, INSERM’s central estimate (INSERM, 1999) for adults—50 µg/day—remains current: the most recent assessment (Scientific cooperation (SCOOP) on Questions relating to food, 2004) estimated adult intake at 57 µg/day.

For intake from water, we considered consumption from a particular area, that of the study that we used, but the geographic disparities in use of tap water (Beaudeau et al., 2003) produce an uncertainty that cannot be quantified. Nonetheless, ignoring it should not have an important effect in this case, given the marginal contribution of water to the EDD. Not taking
The consumption of heated water into account represents an error of 5% (according to the data of Beaudeau et al., 2003) on the mean volume consumed and should therefore not have a notable influence on the result.

For soils and dust, the assessment did not consider all of the premises of the village used by young children; in particular it did not consider a playground in the village center. Three samples were taken from there and analyzed: the lead levels were low, ranging from 14 to 37 ppm. The results presented do not take into account the possibility of lead from sources other than the mine, which would have different bioavailability. Nonetheless, a simulation with a contribution of 50 ppm (equivalent to the regional background level) of lead 60% bioavailable had no notable influence on the results (deviations of 4% and 10% for the 50th and 90th percentile's, respectively, of the EDD).

The rates used here for soil ingestion are less than the consensus parameter chosen by many authors—100 mg/day (Simon, 1998). Two factors support this choice. First, in our case, we are estimating chronic behavior. Second, Stanek et al.’s (2001) study, which we use here, is more recent; it was also used by the EPA in a subsequent document (US EPA, 1997).

For bioavailability, we used in vivo data for piglets being weaned, which Mushak (1998) considers to be the

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**Table 2**

<table>
<thead>
<tr>
<th>Extent of consideration of bioavailability</th>
<th>Mean contribution of the dose (soil and dust to total dose) (%)</th>
<th>Contribution of the uncertainty bioavailability to the variance of the result (%)</th>
<th>Degree of certainty that the P50 of EDD&lt; TDIa (%)</th>
<th>Degree of certainty that the P90 of EDD&lt; TDIa (%)</th>
<th>Level of P50 with a certainty of 95%</th>
<th>Level of P90 with a certainty of 95%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ignoring (100%)</td>
<td>37</td>
<td>—</td>
<td>5%</td>
<td>50</td>
<td>4.1</td>
<td>8.8</td>
</tr>
<tr>
<td>IEUBK default value (30%), i.e., 0.6 relative to food bioavailability</td>
<td>26</td>
<td>—</td>
<td>50</td>
<td>95</td>
<td>3.8</td>
<td>6.5</td>
</tr>
<tr>
<td>1–100% (recognition but no information)</td>
<td>10</td>
<td>6.6</td>
<td>7.5</td>
<td>90</td>
<td>3.8</td>
<td>7.3</td>
</tr>
<tr>
<td>1–22% (knowledge of specific mineral: galena)</td>
<td>3</td>
<td>1.8</td>
<td>&gt;95</td>
<td>&gt;95</td>
<td>3.0</td>
<td>4.5</td>
</tr>
</tbody>
</table>

aTDI: tolerable daily intake (25 μg/kg/week: 3.5 μg/kg/day).
best animal source of information for determining the bioavailability of lead in children. The only study in humans (Maddaloni et al., 1998) concerns adults fasting or immediately after a meal, and its results, unsurprisingly, are extremely variable (absolute bioavailability 1–34%). They do not in any case contradict the values used here. The epidemiologic observations by Rieuwerts et al. (2000) and Berglund et al. (2000) also argue in favor of low bioavailability of the lead in mining soils. We did not consider a difference in bioavailability in dust and soils because soil is the main source for house dust; however, lower particle size in dust could lead to higher values. Soils and dust are mainly silty clay; lead is mainly in the <0.25 mm size fraction (Baubron, 2003).

The uncertainty associated with the relative distribution of indoor and outdoor dust was not taken into account in the results presented, which were calculated for an interior/exterior distribution of 55/45. Calculation (data not shown) with alternative values (40/60 and 75/25) did not notably affect the results.

In the case studied here, the principal source of local lead exposure was previous mining activity, and lead levels were much higher than standard background levels of lead in soils (50–100 ppm). Lead levels in water were below the detection limit (3 µg/L) and even further below the WHO recommendation (10 µg/L). Air concentrations were also low, below the detection limit (which equals the WHO recommendation (WHO, 2000)); they are typical of rural environments (INSERM, 1999) and contribute only slightly to exposure. The other major source of exposure—the principal source when we take into account the reduced bioavailability of mining lead—is dietary intake, which is similar to that of the general French population, in view of the lack of consumption of locally grown products. The EDD varies (cf. Fig. 3) from 2 µg/kg/day at P5 to 5.5 µg/kg/day at P95 (2 and 10 µg/kg/day when ignoring bioavailability; cf. Fig. 1). The principal factors of variation are (cf. Fig. 2) dietary intake, soil concentrations, and soil ingestion. The principal uncertainties are associated with the level of soil ingestion and the bioavailability of lead.

As shown in Table 2, quantification of bioavailability by taking the type of mineral into account substantially raised our confidence that P50 of the EDD does not exceed the TDI (from 75% to more than 95%) and P90 of the EDD does not exceed twice the TDI (from 50% to more than 95%).

At the same time, the P50 and P90 for which there was a 95% degree of confidence (represented by the top curves in Figs. 1 and 3) increased from 4.1 and 8.8 µg/kg day to 3.0 and 4.5 µg/kg day, respectively. This increase in confidence is thus especially noteworthy for the highest exposures, because the reduction in the relative contribution of soils and dust is greatest for the most highly exposed.

The reduction of the uncertainty associated with bioavailability makes it possible to cut its contribution to the variance of the EDD by more than two-thirds (from 6.6% to 1.8%).

Institutional recommendations for exposure assessment (TRW, 1999; Glorennec et al., 2002) do not advise that bioavailability be determined locally, because of the difficulty (and especially cost) of obtaining reliable site-specific information; default values are therefore used. We show here that knowledge of the specific mineral, although it does not totally eliminate uncertainty about bioavailability, does reduce the uncertainty of the EDD considerably and sometimes sufficiently, especially for the most highly exposed. Accordingly, in this case, it is not necessary to conduct expensive studies to determine bioavailability more precisely: simple knowledge of the mineral suffices to allow us to conclude with a certainty of 95% that the P50 of the EDD is less than the TDI. Taking the uncertainty associated with bioavailability into account, as in a two-dimensional Monte Carlo simulation, and therefore reducing it when the mineral is known appears to us to be a useful method for optimizing risk management around old mine sites, especially since galena is the form of lead most often encountered (Steele et al., 1990). When the degree of confidence in the estimation, by knowledge of the mineral alone, does not provide sufficient certainty, then a decision to screen for lead poisoning for safety’s sake or to determine bioavailability might be appropriate.

The low air levels observed here are probably typical of an old mine site no longer in activity; on the other hand, water levels may vary more from one site to the next (INSERM, 1999). These were measured in our study. In the absence of information, when we consider that concentrations in water can vary in France for a network with no lead in its public portions from 0 to 50 µg/L (75% from 0 to 10 and 25% from 10 to 50) (Association des hygienistes et techniciens municipaux (AGHTM), 1994), exposure from water is substantial, and the concentration in water can become the major source of uncertainty for calculating the EDD. Obtaining local data about lead concentrations in water is therefore essential.

Improving estimates of lead exposure in France requires the availability of current distributions of dietary lead intake to improve the reliability of the estimates, as dietary intake has been shown to contribute strongly to dose and variance. In Europe, and especially in France, studies of soil ingestion would also help to reduce the uncertainty associated with the use of US data. If such studies are conducted, it would be useful for their protocol to allow separate distributions of uncertainty and variability to be derived.
Acknowledgments

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