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French children's exposure to metals via ingestion of indoor dust, outdoor playground dust and soil: Contamination data

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ABSTRACT

In addition to dietary exposure, children are exposed to metals via ingestion of soils and indoor dust, contaminated by natural or anthropogenic outdoor and indoor sources. The objective of this nationwide study was to assess metal contamination of soils and dust which young French children are exposed to. A sample of 484 children (6 months to 6 years) was constituted in order to obtain representative results for young French children. In each home indoor settled dust was sampled by a wipe in up to five rooms. Outdoor playgrounds were sampled with a soil sample ring ($n = 315$) or with a wipe in case of hard surfaces ($n = 53$). As, Cd, Cr, Cu, Mn, Pb, Sb, Sr, and V were measured because of their potential health concern due to soil and dust ingestion. The samples were digested with hydrochloric acid, and afterwards aqua regia in order to determine both leachable and total metal concentrations and loadings by mass spectrometry with a quadrupole ICP-MS. In indoor settled dust most (total) loadings were below the Limit of Quantification (LOQ), except for Pb and Sr, whose median loadings were respectively 9 and 10 μg/m². The 95th percentile of loadings were 2 μg/m² for As, <0.8 for Cd, 18 for Cr, 49 for Cu, <64 for Mn, 63 for Pb, 2 for Sb, 56 for Sr, and <8 for V. Median/95th percentile of loadings in settled dust on outdoor playgrounds were 2/16, <0.8/1.3, 17/53, 49/330, 99/424, 32/393, 2/13, 86/661 and 10/37 μg/m² for As, Cd, Cr, Cu, Mn, Pb, Sb, Sr, and V respectively. In outdoor playground soil median/95th percentile of concentrations (μg/g) were 8/26, <0.65/1, 25/52, <26/53,391/956, 27/254, 0.7/4, 54/295, 23/57 for As, Cd, Cr, Cu, Mn, Pb, Sb, Sr, and V respectively. These results are comparable with those observed in other countries. Because of their representative nature, we can assess children's exposures to these metals via soil and dust and the associated risks in urban and rural environments. Ratios of leachable/total concentrations and loadings, calculated on >LOQ measurements, differed among metals. To a lesser extent, they were also affected by type of matrix, with (except for Cd) a greater leachability of dust (especially indoor) compared to soils.

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1. Introduction

Metals (and metalloids) are natural constituents of the Earth's crust and are persistent in outdoor soils and indoor settled dust; these two environmental media can therefore contribute to human exposure to metals (Caussy et al., 2003; Hivert et al., 2002). This is particularly true for children because of their more intense hand to mouth contacts (Calabrese and Stanek, 1998). As a consequence, knowing that most metals have toxic properties, it is necessary to assess metal exposure through soil and dust ingestion, in order to quantify associated health risks (Granero and Domingo, 2002; Stanek et al., 2001). Assessing exposure to metals via soil and dust ingestion requires knowing i) quantity of ingested soil and dust per unit of time, or number of hand to mouth contacts per day and mean quantity ingested per contact and ii) concentration of metals in soil and dust. The first is rarely assessed in specific risk assessments and default values from literature (e.g. 100 mg of ingested soil and dust per day for a child, 50 mg per day for an adult (US EPA (United States Environmental Protection Agency), 1999)) are often used despite their uncertainty and variability among populations. The second is case-specific because metal concentrations depend on location, varying with crustal content, and industrial and built environment (Caussy et al., 2003).

For outdoor soils, most of the studies deal with metal concentration in agricultural soils or in industrial areas. Relatively few studies are exposure-oriented and examine contamination of urban soils where children may have a significant contact with soil, especially in playgrounds. Many studies have also focused only on lead contamination of soil since it is often associated with children's blood lead level (Mielke and Reagan, 1998). Apart from natural occurrence and

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industrial sources, urban soil contamination is also determined by particle deposition due to automobile traffic, waste disposal, and corrosion of building materials (Wong et al., 2006). When assessing direct ingestion of soil, the main sampling issue is to focus on surface (0–1 cm) soils, that are in contact with people, and especially children (U.S.Environmental Protection Agency, 1996).

Indoor dust is a heterogeneous mixture of particles derived from indoor and outdoor sources (Rasmussen, 2004), transferred either by wind, pets or shoes. Lead has been intensively studied in indoor dust due to its presence in paints, and because of the relationship between indoor dust lead and blood lead levels (Lanphear et al., 1998). But, as recently reviewed by Ibanez et al. (2010), information on other metals in home dust is generally sparse. Their concentrations may be influenced by outdoor environment, because they mainly originate from soil (Mn, V (Fergusson et al., 1986)), from traffic and notably brakes (Cu, Sb (Amato et al., 2009)). But indoor dust concentrations depend also on building (ex: paint (Mielke et al., 2001)) or furnishing materials (carpets: Cr, Cd (Kim and Fergusson, 1993)), and on human behaviours such as smoking (Cr, Cd (Madany et al., 1994)), cooking and cleaning habits. That is the reason why indoor dust concentrations cannot be predicted only by outdoor concentrations; therefore specific measurements are necessary to assess dust exposure to metals (Ibanez et al., 2010).

Settled dust can be sampled by various methods including vacuuming, gentle sweeping, or wiping (Lioy et al., 2002; Sterling et al., 1999). For lead, the wipe method is commonly used to imitate the hand's ability to pick up and retain particles, as described in ASTM E1728-03 (American Society for Testing and Materials (ASTM), 2003a) and is recommended in guidelines for the evaluation and control of lead-based paint hazards in housing (U.S.Department of Housing and Urban Development, 1995). This sampling method can also be used to determine other metal concentrations in micrograms per square metre of wiped surface; in this case they are often referred to as loadings.

The risk posed by a metal depends on its concentration in exposure media, sometimes on its chemical speciation that is more or less toxic, but also on its potential to be mobilized and to enter the human blood (referred to as bioavailability). Bioavailability depends on metal species, dust organic content, and particle sizes (Rasmussen et al., 2008). Therefore, the knowledge of total concentration of a metal may not be sufficient to allow a realistic estimate of risk. For example, in an industrial context, taking into account bioavailability of metals is very important (Luthy et al., 2003). Furthermore, bioavailability can be highly relevant for decision making in health risk management (Casteel et al., 2001; Glorennec and Declercq, 2007; Mahaffey, 1998). The use of humans for bioavailability measurement is unlikely to be feasible, and animal use is also challenging in terms of cost, time and ethical issues (Saikai et al., 2007). So, in vitro tests have been developed to measure bioaccessibility as a surrogate of bioavailability. Bioaccessibility is a measure of the amount of contaminant that may be solubilized in human gastrointestinal fluids, aiming at mimicking the solubilization process in humans (Ruby et al., 1999). It is used in soil exposure studies (Denys et al., 2007; Saikai et al., 2007) as an alternative to oral bioavailability measurements with more or less sophisticated methods. Such tests are very sensitive to the operating conditions: choice of acid, enzyme use, duration and temperature of acidic dissolution (Turner, 2011). Different types of tests have been developed, especially for soil in Europe. For example, Oomen et al. (2002) developed a model that simulates digestion conditions in three compartments including the mouth, the gastric tract and the intestinal tract. Other protocols have used mixtures of pepsin and/or acid at different pH levels (Rasmussen et al., 2008; Rieuwerts et al., 2006; Turner and Simmonds, 2006). In France, the regulatory procedures for lead management require measurement of the leachable lead (Agence française de normalisation (AFNOR), 2008). This regulatory standardized method simulates stomach acid (pH = 1.5) and has the advantage of being simple, inexpensive, reproducible and thus easy to implement for routine investigations. It has been adapted by Le Bot et al. (2010) to measure on a single environmental sample both leachable and total concentrations (and loadings) of lead and other metals. We chose this latter method because of i) its easiness to implement regarding the number of samples ($N = 484$ dwellings with up to 5 dust samples and a soil sample per dwelling) to be measured within this nationwide survey, and ii) to be able to compare lead results to routine measurements in France.

Assessing health risks linked to metal ingestion via dust and soil therefore requires local contamination and bioaccessibility data (in addition to default values for ingested quantities). The objective of this study is to determine the concentrations and/or loadings and leachability for nine metals or metalloids selected for their health interest (As, Cd, Cr, Cu, Mn, Pb, Sb, Sr, and V) in house dust and playground soils frequented by young French children. This study is unique in that it is not focused on an industrial, urban or rural area, but reflects the diversity of residential locations in France. Moreover, it relies on a nationwide survey and has been designed to provide results that are representative of children's homes and playgrounds.

2. Material and methods

2.1. Targeted metals

Metals were selected on the basis of health risk via dust ingestion assessed using mean contamination in dust and toxicity (both available in literature and toxicological databases). Metals whose potential ingested dose was higher than 5% of the tolerable daily intake were considered: As, Cd, Cr, Cu, Mn, Pb, Sb, Sr, and V (Le Bot et al., 2010). Al was initially selected but discarded because of wipe contamination.

2.2. Population

Children were randomly selected among the children enrolled in a national lead survey (Etchevers et al., 2010). Inclusion and survey procedures are described in Etchevers et al. (2010). Briefly, a two-stage sampling (Lumley, 2010a) was implemented: In the first stage, the primary sampling units were the hospitals; in the second stage, children were sampled from these primary sampling units. The hospitals were stratified by areas suspected to be at high risk of lead poisoning. High-risk hospitals were intentionally oversampled, aiming to increase the probability of sampling children with high BLLs and, indirectly homes with high lead contamination. This oversampling is taken into account in the analysis by sampling weights to correct for imperfections in the sample that might lead to bias.

The sub-population for this environmental investigation included 484 children between 6 months and 6 years old. Components of the sampling design (sampling weights, stratification and stages) were taken into account to estimate the concentrations and loadings of metals for the dwellings occupied by children between 6 months and 6 years old. Post-stratification of sampling weights (inverse of the probability of occurring in the sample) was used to increase the precision of estimates and to make the sample more representative of the population (Lumley, 2010a), based on dwelling characteristics (age, location, individual or collective housing). The statistical adjustment items were the construction period (before or after 1949), the type of dwelling (individual or collective), and its location (22 French administrative regions). Statistical work was performed with the "survey" package of R® 2.9.0 software (Lumley, 2010b).The parents of enrolled children were informed about the purposes of the study and gave their consent. An individual written report on the results was sent to each family. Authorization from the Commission Nationale de l'Informatique et des Libertés (French Data Protection Commission) was previously obtained (Authorization number no. 908326).

Results concerning lead have already been published (Lucas et al., in press), but are also partially reported here to ensure consistency.

2.3. Settled dust and soil sampling

In each home, up to 5 rooms were selected using the US-HUD protocol (U.S.Department of Housing and Urban Development, 1995): child's bedroom, living room, hall, kitchen, play room or bedroom of the immediately younger or older sibling. In addition if the child lived in an apartment, dust sample were collected in the stairwell and the building entrance. In each room a floor area of 0.1 m² was chosen where child routinely played and dust was sampled there, according to the NF X 46–032 standard procedure (Agence française de normalisation (AFNOR), 2008) (similar to the ASTM: E 1728-03 standard (American Society for Testing and Materials (ASTM), 2003a)), with a moist lead-free wipe ("Lead Wipe" by Aramsco) that meets ASTM© standard E1792-03 (American Society for Testing and Materials (ASTM), 2003b). The wipes (one per room) were then placed in 50 ml single-use screw-top polyethylene tubes (digitube, SCP science).

If the child played outdoors in a garden or playground in the close vicinity of the home, the soil was sampled using a ring (2 cm deep) or a wipe (0.1 m²) for hard surfaces, with the same collection and packaging procedure as for indoor dust. For hard surfaces, a composite soil sample was constituted from 10 subsamples in the 0–2 cm layer and was prepared according to the NF ISO 11464 standard (Norme Francaise ISO 11464, 1996). The soil samples were air-dried at a temperature below 40 °C and sieved to less than 250-μm particle size.

2.4. Analysis

Wiped dust and soil samples were treated to extract metals following the method described in detail by Le Bot et al. (2010). This method can determine leachable and total metal concentration (or loading) on the same sample. Briefly, it consists of a two-step digestion stage: firstly, 0.15 N hydrochloric acid is added to the sample at 37 °C to solubilize leachable metal; then aqua regia (3:1 HCl/HNO_3) is added to an aliquot at 95 °C to solubilize residual (i.e., non-leachable) metal. Dissolved metals were analyzed by mass spectrometry with a quadrupole ICP-MS (Agilent Technology 7500ce) according to the ISO 17 294–2 standard (Agence française de normalisation (AFNOR), 2005).

2.5. Quality control

For each batch of samples, reference samples SRM 2583 for dust and CRM SS2 for soil with one blank were included in all digestion and analyzed series. The results made it possible to calculate relative standard deviation (RSD) and measurement uncertainties (+/−2RSD) for each element. For all reference samples, RSDs were lower or near 20%.

Field blanks (B) were collected in each dwelling and then analyzed. The following procedure was applied to measured concentrations (or loadings) (C) when B was >Limit Of Quantification (LOQ). When $B \times 100/C \leq 20\%$, then C was unchanged, because B is in the same range as the measurement uncertainty on C. When $20\% < B \times 100/C < 100\%$, then C was replaced by C − B (if C − B<LOQ, then C was replaced by <LOQ). When $B \times 100/C \geq 100\%$, the measurement was invalidated. It was verified that leachable metal concentration was less than total concentration with regards to uncertainty of measurements (leachable concentration<120% total concentration).

3. Results

Homes of 484 children between 6 months and 6 years old were investigated, representing 3,581,991 French housing units. Indoor dust samples were collected in all of them, playground soils in 315, and playground dust in 53.

Table 1 presents the total concentrations and loadings of metals in children's dwellings for indoor dust (arithmetic mean of measured rooms, excluding the stairwell), outdoor playground dust and soils. Regarding metal loading (μg/m²) in indoor settled dust, the most abundant metals in wiped dust were Sr and Pb, whose median loadings were respectively 10 and 9 μg/m². For other measured metals, median loadings were below the LOQ. The 95th percentile of loadings were 2 μg/m² for As, <0.8 for Cd, 18 for Cr, 49 for Cu, <64 for Mn, 63 for Pb, 2 for Sb, 56 for Sr, and <8 for V. Metal loadings (μg/m²) in dust of outdoor playgrounds (i.e. when playground was a hard surface) were higher than indoor. Mn and Sr were the most abundant metals: their median loadings were 99 and 86 μg/m². They were followed by Cu, Pb, Cr and V (with median loadings of respectively 49, 32, 14, and 10 μg/m²). As and Cd were less abundant with median loadings of 2 and <0.8 μg/m² respectively. For metal concentrations in soils of outdoor playgrounds of children's dwellings, the most abundant metal was Mn, whose median concentration (μg/g) was 394 μg/g, followed by Sr (53), Pb (27), Cr (25), V (23) and As (8). For Sb and Cu, median concentrations were 0.7 and <26, respectively.

The leachability (leachable concentration or loading divided by total concentration or loading) of metals in dwellings (indoor dust) and playgrounds (outdoor soil or dust) of children is shown in Fig. 1. A general feature is the large variability among samples. They also differed among metals. To a lesser extent, they were also affected by type of matrix, with (except for Cd but with a limited number of ratio) a greater leachability of dust (especially indoor) compared to soils. For indoor dust, Sr leachability was the highest (median: 95). Cr, Cu, Mn and Pb also had a median leachability greater than 75%. Median leachability was less than 50% for As, Cr, Sb and V. For outdoor playground soils, median leachability was higher than 75% for Cd and Sr, and higher than 50% for Cu, Mn, Pb. Concerning outdoor hard surface soils, median leachability was higher than 75% for Cd, Cu and Sr, and higher than 50% for Mn and Pb.

4. Discussion

This study has provided, for the first time in France, representative estimations of metal concentrations and loadings in settled dust and soils of French children's homes and environment, along with original information about leachability. The 484 children included in this environmental survey were enrolled in the national lead poisoning survey (3800 children), whose representativeness is discussed by Etchevers et al. (2010). When comparing the household groups who declined participation to those who agreed, on the basis of access to free health insurance in France (CMU) – an indicator of poverty, there was no significant observed difference (p-value = 0.9, chi²-test).

Dust was collected by a wipe method. Indoor settled dust can also be collected by a vacuum cleaner, depending on the objectives (Mercier et al., 2011). Using an HVS3 (High-Volume Small Surface Sampler) vacuum cleaner is a standardized method (American Society for Testing and Materials (ASTM), 2005) of collecting settled dust but, as it is expensive and not easy to use, it seems to be more appropriate for small scale studies. The use of a commercial vacuum cleaner could be an alternative but precautions must be taken to avoid sample contamination. Collecting vacuum cleaner bags from households could be used to obtain a large quantity of dust quickly, but dust can be contaminated by the vacuum cleaner itself and is not necessarily representative of dust in contact with children (if the car or chimney ashes are vacuumed, for example). The wipe method is quick and easy to implement and is specifically designed to collect dust likely to adhere to the hands and then to be ingested. However, the quantity of sampled dust is relatively small (about 30 mg for 0.1 m^2 (Glorennec et al., 2007)), which might alter quantification limits.

A study (Sterling et al., 1999) on the relative efficiency of four sampling methods (wipes, HVS3, and two commercial vacuum cleaners) concluded that the HVS3 vacuum cleaner provided the best correlation

Table 1
Metal concentrations and loadings in children's dwellings. France 2008–2009.

Element	Sample	*n*	*N*	Units	LOQ	min	P10	P25	P50	P75	P90	P95	Max
As	Indoor dust	473	3,461,008	µg/m^2	0.6	<LOQ	<LOQ	<LOQ	<LOQ	0,8	1,3	1.9	7.2
	Playground dust	53	325,647	µg/m^2	0.6	<LOQ	0.8	1.2	2.2	3.7	11	16	50
	Playground soil	315	2,518,808	µg/g	0.3	0.3	3.6	5.7	8.1	12	18	26	114
Cd	Indoor dust	473	346,1008	µg/m^2	0.8	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	11
	Playground dust	53	325,647	µg/m^2	0.8	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	1	3
	Playground soil	315	2,518,808	µg/g	0.7	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	1	3
Cr	Indoor dust	473	3,461,008	µg/m^2	10	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	14	18	121
	Playground dust	53	325,647	µg/m^2	10	<LOQ	<LOQ	<LOQ	14	24	33	53	913
	Playground soil	315	2,518,808	µg/g	7	<LOQ	13	19	25	33	47	52	100
Cu	Indoor dust	473	3,461,008	µg/m^2	32	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	32	49	2433
	Playground dust	53	325,647	µg/m^2	32	<LOQ	<LOQ	<LOQ	49	106	207	331	609
	Playground soil	315	2,518,808	µg/g	26	<LOQ	<LOQ	<LOQ	<LOQ	33	47	53	213
Mn	Indoor dust	473	3,461,008	µg/m^2	64	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	1855
	Playground dust	53	325,647	µg/m^2	64	<LOQ	<LOQ	<LOQ	99	192	386	424	2932
	Playground soil	315	2,518,808	µg/g	13	<LOQ	157	257	391	546	731	956	3913
Pb	Indoor dust	471	3,453,789	µg/m^2	2.0	<LOQ	2	4	9	17	39	63	1412
	Playground dust	53	325,647	µg/m^2	2.0	6	11	17	32	99	188	393	3225
	Playground soil	315	2,518,808	µg/g	1.3	2	12	17	27	60	117	254	3408
Sb	Indoor dust	473	3,461,008	µg/m^2	0.8	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	1.4	2.4	8.6
	Playground dust	53	325,647	µg/m^2	0.8	<LOQ	<LOQ	1.4	2.0	5.1	12	13	31
	Playground soil	315	2,518,808	µg/g	0.7	<LOQ	<LOQ	<LOQ	0.7	1.3	2.3	4.0	41
Sr	Indoor dust	456	3,371,973	µg/m^2	8	8	8	8	10	18	35	57	305
	Playground dust	53	325,647	µg/m^2	8	30	34	53	86	1242	2058	436	1492
	Playground soil	315	2,518,808	µg/g	7	<LOQ	10	18	54	84	205	296	701
V	Indoor dust	473	3,461,008	µg/m^2	8	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	30
	Playground dust	53	325,647	µg/m^2	8	<LOQ	<LOQ	<LOQ	10	14	26	37	301
	Playground soil	315	2,518,808	µg/g	7	<LOQ	11	18	23	34	45	57	169

n: number of dwellings with valid samples – *N*: number of represented dwellings – LOQ: Limit of Quantification.

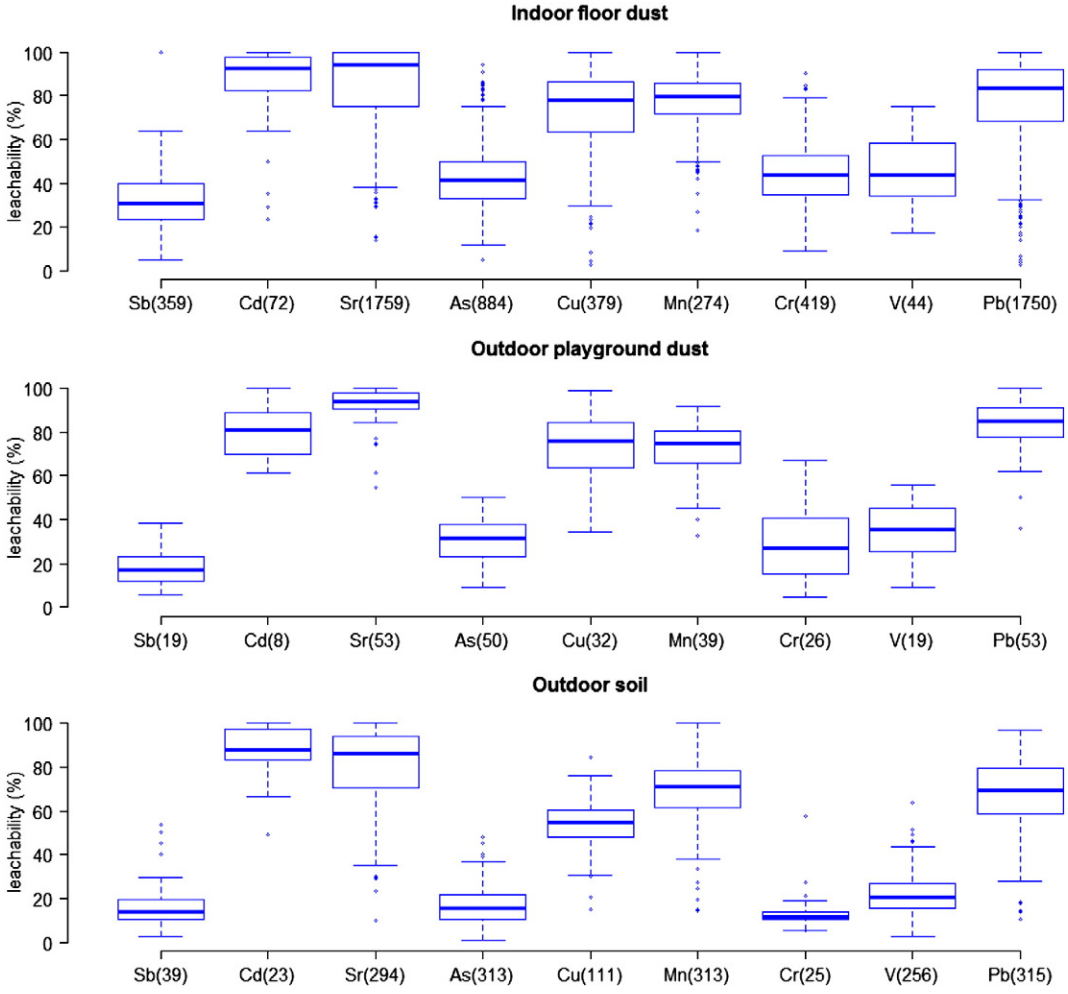


Fig. 1. Metal leachability (unweighted by sampling weights) in dwellings (indoor dust) and playgrounds (outdoor soil or dust) of children. France 2008–2009.

with blood lead levels ($R^2 = 0.33$), followed by wipes ($R^2 = 0.25$); wipes were recommended by the authors for exposure assessment in public health actions, even on carpets and rugs, as they were easier to use and inexpensive. A consequence of the choice of the sampling method is the unit in which concentrations are expressed. The wipe method provides a loading (or surface concentration), expressed in μg/m² whereas the vacuum method provides mass concentration (μg/g). Loading is especially useful for lead, because the relationship between the lead in dust and blood lead levels uses this exposure metric (example (Lanphear et al., 1998)). On the other hand, mass concentration is suitable for measuring daily dust and soil ingestion rates, which are expressed in mg (ingested dust and soil)/day. Otherwise a dedicated assessment of dust loading or a default quantity of dust per m² is required. As dust loading is highly variable, it is not appropriate to use a default value in a given home to assess exposure for an individual. However, it may be acceptable for an estimation of a median dust concentration, or a population-averaged dust exposure, as in a health risk assessment. In a large study (Giovannangelo et al., 2007) in Germany, Sweden & Netherlands ($n = 57$ homes; 117 samples), the median dust mass was around 0.35 g/m²; we observed virtually the same levels (0.3 g/m²) in a pilot study in France (Glorennec et al., 2005). Then simply multiplying metal loading (in μg/m²) by 3 to obtain metal mass concentration (μg/g) is equivalent to assuming an average dust mass of 0.33 g/m².

Regarding the analytical aspects, ICP/MS made it possible to lower limits of quantification (LOQ) in the wiped samples. The leachable element LOQs were comparable with those from the study of McDonald et al. (2011). However, in our case, only a few elements were quantified. One possibility is the low contamination of metals in French home dust; another possible explanation could be the small quantity of collected dust (relatively clean homes).

Indoor dust loadings (total metals) were compared to those of McDonald et al. (2011), who also used the wipe sampling method, and a modified (addition of hydrofluoric acid) version of the ASTM 1644 standard for Pb. Comparison was focused on the 95th percentile because of many metals with median loadings\ <LOQ. Our results are 3–5 times lower than those measured in urban homes (Canada, 2008) for As, Cd, Cr and Sr. Our results are in the same order of magnitude for Pb, and 2.5 higher for Cu. When, as mentioned above, multiplying observed loading by 3 to obtain an estimate of concentration in μg/g, our results fall within the range of previously published values (Ibanez et al., 2010). Considering leachability in indoor settled dust, the values observed here are well in line with the published values of bioaccessibility (Ibanez et al., 2010; Turner and Simmonds, 2006) for As, Cd, Cr, Cu, Pb, and higher for Mn (80% vs. 40–60%). These appear to be the first reported values of leachability in dust for Sb, Sr and V.

Regarding outdoor playground dust loadings, these appear to be the first published values using this sampling method and this measurement unit (per m²), except for lead, but especially in polluted context (for example Mielke et al., 2007).

For playgrounds soils, our results were compared to those from Rasmussen et al. (2001) in residence gardens ($n = 50$, Ottawa, Canada,1993). Rasmussen used ICP-MS, after digestion with nitric acid and hydrofluoric acids. As, Cd, Cr, Cu, Mn, Pb, Sb, Sr, and V median/95th percentile concentrations compared as follows in our and Rasmussen's study: 8/26 μg/g in the present study vs. 3/4 in Ottawa for As, <0.65/1.1 vs. 0.3/0.6 for Cd, 25/52 vs. 43/59 for Cr, <26/53 vs. 12/19 for Cu, 391/956 vs. 532/718 for Mn, 27/254 vs. 33/205 for Pb, 0.7/4 vs. 0.2/1 for Sb, 54/295 vs. 356/418 for Sr, and finally 23/57 vs. 46/70 for V. In the present study concentrations were a little lower for As, Mn and especially Sr, and a little higher for Cu and Sb. Higher concentrations in Cu and Sb may be related to recent use of these metals in brake pads (Hjortenkrans et al., 2006). Sr differences may be due to crustal composition differences. Overall, observed concentrations were in the same order of magnitude. When comparing this study to measurements by Baize (2000), (France, 1990's), one can observe that concentrations in our study were higher for Cu, Pb (for high percentiles), quite comparable for Cr, and lower for Mn. However, comparison is limited because the soils sampled by Baize were in rural areas and were not top soils. Regarding Strontium, our results are close to those observed in natural Mediterranean soils (Roca-Perez et al., 2010). Our results of leachability of soils are difficult to compare to published values because published studies on soil bioaccessibility were performed on contaminated sites (mining wastes or smelters for example) and bioaccessibility is highly variable depending on the site. For example Ruby et al. (1999) displayed values ranging from almost 0 to 0.9.

Concentrations and loadings (data available in supplemental material) were also compared between rural and urban areas, with a cut-off number of 2000 inhabitants in the city (in 1999, http://www.statistiques-locales.insee.fr). For indoor dust, distributions are similar until the 95th percentile, higher in urban areas. Outdoor playground dust loadings are higher in urban areas for Cd, Cr, Cu, Sb, Sr and Pb (for values above the median), whereas As and Mn loadings are higher in rural areas. The same difference is observed for outdoor playground soils, except that there is no difference for Cr, and that higher concentrations for As in rural areas are observed only above the 90th percentile.

5. Conclusion

These results of metal contamination of dust and soils are the first reported in metropolitan France for a representative sample of dwellings. They can be useful to assess total exposure to metals for French children. They also provide a set of default values to be used for local exposure assessments (for example for lead poisoning screening (Glorennec et al., 2006)) and in a regulatory context because of the usefulness of taking into account background exposures (Ronga-Pezeret et al., 2010).

Conflict of interest

The authors declare they have no conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.envint.2012.04.010.

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