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Exposure Assessment of Allergens and Metals in Settled Dust in French Nursery and Elementary Schools

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Abstract: The aim of this study was to characterise the contamination in settled dust in French classrooms and to provide an overview of the influencing factors of dust contamination. Cat, dog and dust mite allergens and metals were measured in 51 classrooms at 17 schools. The concentrations of pet allergens in settled dust were generally low (mean value of 0.1 $\mu g \cdot g^{-1}$), with carpeted and rug-covered floors presenting higher dust and cat allergen concentrations. The highest metal loadings in dust were observed for manganese (Mn) and copper (Cu), while the lead (Pb) loadings were lower (16 ± 19 $\mu g \cdot m^{-2}$) and fell below the French guideline. Higher metal leachability was found for cadmium (Cd), Cu, Pb and strontium (Sr) at values of approximately 80%, which suggest that, in cases of dust ingestion by children, a large proportion should be assimilated through the gastro-intestinal tract. The intra-classroom and intra-school variabilities of the metal concentrations in settled dust were lower than the variability between schools. Classrooms with tiled floors had higher Pb loadings than classrooms with wood or vinyl floors. In addition, wet cleaning less than once a week resulted in greater loadings of Cu and Pb in the settled dust. Lastly, enrichment factors showed that metals in settled dust of classrooms were not only from the contribution of the natural background concentrations in soils.

Keywords: settled dust; allergens; metals; classrooms; enrichment factors

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1. Introduction

The indoor environment of school buildings has attracted the attention of the scientific community due to scientific evidence associating poor indoor air quality (IAQ) with negative impacts on student health, academic performance and attendance [1–3]. Among the numerous indoor pollutants found in classrooms, settled dust can serve as a source of allergens and metals that could potentially affect student health [4,5].

The main allergens identified in schools are cat and dog allergens, followed by cockroach and mite allergens [6]. Children who are exposed to cat and dog allergens can develop perennial symptoms, allergic inflammation reactions and asthma [2]. Pet allergens have even been detected in educational facilities when animals are not present. Children's clothes are known to act as the main transfer mechanism for delivering allergens to classrooms. Higher concentrations of cat and dog allergens have been found in the clothing worn by students with pets (cats and dogs) than in the clothing worn by students without pets [6].

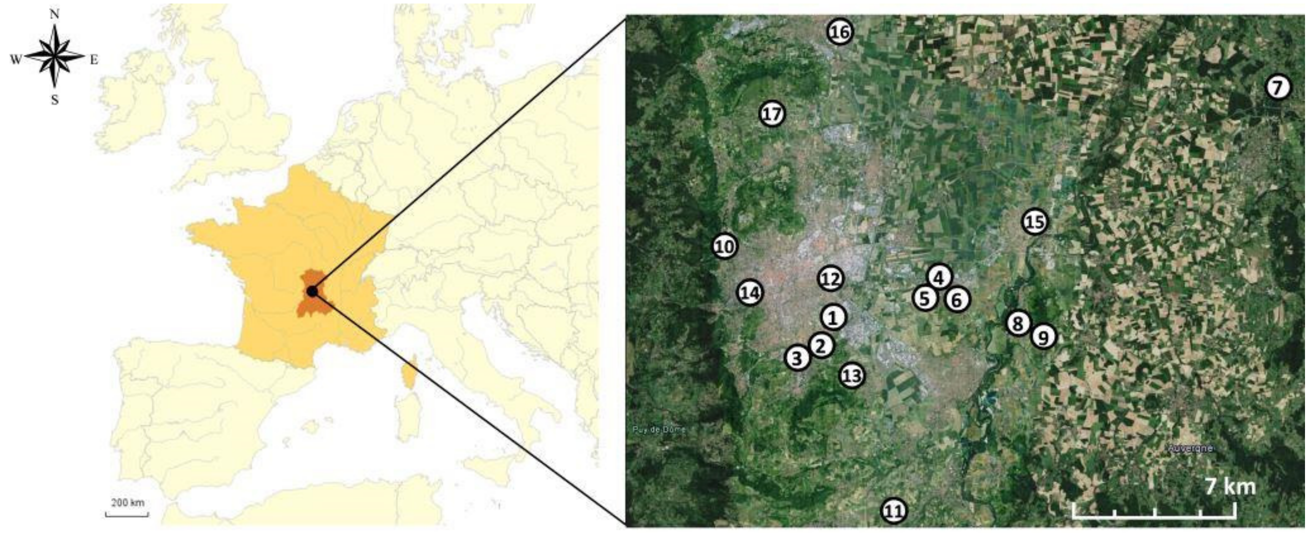
Indoor settled dust contains metals and metalloids, which are natural constituents of the Earth's crust and are generated from anthropogenic sources. Thus, humans can be exposed to these substances through settled dust. The health effects of metals are well known [7]. While environmental lead (Pb) levels have significantly decreased in recent years [8], Pb remains a public health concern due to the absence of a known effect threshold [9]. Because children exhibit more frequent hand-to-mouth contact [10], they are more likely to be exposed to metals in settled dust. To assess the effects of exposure in classrooms, it is necessary to examine the metal concentrations in the settled dust in these environments [11–13]. Few studies have specifically focused on the exposure of students to metals through settled dust in classrooms.

Due to limited knowledge regarding school environment in France, the French Indoor Air Quality Observatory (OQAI) was commissioned to assess the degrees of child exposure to various indoor air pollutants in nursery and elementary schools. A study was conducted in 51 classrooms across 17 French nursery and elementary schools [14]. The objectives of the present study were to characterise allergens and metal concentrations in settled dust and explore the relationships between concentrations and various parameters, such as sampling location, flooring material and the frequency of cleaning. In addition, the leachable fraction (%) of each metal, which is a key parameter for assessing internal exposure dose due to ingestion, was determined. Finally, this study is the first to measure the concentrations of crustal and non-crustal metals in settled dust found on school floors.

2. Methods

2.1. Study Design and Sampling Site Description

The study area includes the town of Clermont-Ferrand and its surrounding area in the Auvergne region of central France. It covers a total area of 43 km^2 and has a population of 139,860 inhabitants. Seventeen schools voluntarily participated in this study, including seven nursery schools and 10 elementary schools. The locations of the studied schools are shown in Figure 1. All of the schools were located in urban areas, except for school 7, which was located in a rural area.



(**a**) (**b**)

Figure 1. Location of the study area in the Auvergne region (dark orange) of central France (**a**), and the locations of the 17 schools in the Clermont-Ferrand area (**b**).

Schools 1 through 10 were sampled during the heating season (from 12 January 2010 to 2 April 2010), during which the outdoor mean temperature and relative humidity reached 6.4 ± 5.7 °C and 59 ± 11%, respectively. Schools 11 through 17 were evaluated during the non-heating season (from 26 April 2010 to 25 June 2010), during which the outdoor mean temperature and relative humidity reached 17.0 ± 3.8 °C and 60% ± 10%, respectively. Three classrooms were sampled at each school over one week. The mean indoor temperatures during the occupied period were 22.5 ± 1.5 and 23.6 ± 1.3 °C for the heating and non-heating seasons, respectively. The mean indoor relative humidity during occupancy in the heating season was 31% ± 5%, while in the non-heating season, the mean value was 47% ± 8%.

The 51 studied classrooms had a mean volume of 175 ± 33 m^3 that ranged from 90 to 310 m^3. Regarding the locations of the classrooms, 63% (n = 32) were positioned on the ground floor, 35% (n = 18) were located on the first floor and only 2% (n = 1) were located on the second floor. The mean number of children per classroom was 24 ± 4, and the ages of the children ranged from 3 to 10 years.

Regarding the types of ventilation, 73% of the classrooms (n = 37) were naturally ventilated, while 27% (n = 14) were equipped with a mechanical ventilation system (two classrooms with balanced system and 12 classrooms with exhaust-only system). The studied classrooms presented different types of flooring: vinyl (49% of all classrooms, n = 25), tile (47% of all classrooms, n = 24) and wood (4% of all classrooms, n = 2). Detailed data regarding each classroom are described in Table 1.

A questionnaire was designed and administered to school managers to gather specific information from each school, including the type of flooring and the frequency of classroom cleaning.

Table 1. Detailed information of the 30 classrooms monitored during the heating season (from 11 January to 2 April 2010) and of the 21 classrooms monitored during the non-heating season (from 26 April to 25 June 2010).

Season	School	Classroom	Type	Year[1]	Floor Level	Area (m^2)	Volume (m^3)	Type of Floor	W_o (m^2)	Decor[2] (m^2)	Nr Pupils	Mean Age	Type of Ventilation
Heating Season	S1	C1	E	1967	1	70	195	Tiles	6 (14)	15	28	8	Natural
		C2	E	1967	1	58	162	Tiles	5 (13)	20	23	6	Natural
		C3	E	1967	1	58	162	Tiles	5 (13)	10	22	7	Natural
	S2	C1	E	2000 *	1	54	173	Linoleum	3 (6)	20	26	6	Natural
		C2	E	2000 *	1	59	189	Linoleum	2 (9)	18	25	9	Natural
		C3	E	2000 *	0	44	150	Linoleum	3 (3)	15	12	9	Natural
	S3	C1	N	1980	1	55	153	Linoleum	3 (4)	13	22	4	Natural
		C2	N	1954	1	55	178	Linoleum	5 (10)	10	23	5	Natural
		C3	N	1980	1	64	177	Linoleum	6 (17)	14	23	3	Natural
	S4	C1	N	1977	0	59	186	Linoleum	3 (4)	20	24	4	MV1
		C2	N	1977	0	61	192	Linoleum	3 (4)	25	23	3	MV1
		C3	N	1977	0	59	186	Linoleum	3 (4)	20	25	5	MV1
	S5	C1	E	2005	0	68	177	Linoleum	5 (13)	12	23	6	MV1
		C2	E	2005	1	69	186	Linoleum	5 (20)	15	19	9	MV1
		C3	E	2005	1	64	160	Linoleum	5 (20)	12	21	8	MV1
	S6	C1	E	1957	0	57	177	Tiles	5 (9)	8	26	10	Natural
		C2	E	1957	1	56	180	Tiles	5 (9)	12	27	6	Natural
		C3	E	1957	1	55	179	Tiles	4 (7)	11	22	9	Natural
	S7	C1	N	1887	0	36	119	Linoleum	2 (8)	5	24	4	Natural
		C2	E	1887	0	37	113	Linoleum	2 (5)	7	13	9	Natural
		C3	E	1997	0	38	90	Linoleum	4 (6)	6	12	7	Natural
	S8	C1	E	1983	0	59	156	Tiles	3 (3)	10	22	6	Natural
		C2	E	1983	0	62	166	Tiles	3 (3)	12	26	7	Natural
		C3	E	1983	0	60	159	Tiles	6 (6)	11	25	9	Natural
	S9	C1	N	2002	0	58	157	Linoleum	7 (6)	12	28	5	MV1
		C2	N	2002	0	60	162	Linoleum	8 (7)	12	25	4	MV1
		C3	N	2002	0	58	157	Linoleum	10 (9)	14	25	4	MV1
	S10	C1	E	1971	0	56	167	Tiles	5 (14)	10	20	7	Natural
		C2	E	1971	1	57	171	Tiles	5 (10)	6	22	8	Natural
		C3	E	1971	1	57	171	Tiles	5 (10)	11	22	10	Natural
Non-Heating Season	S11	C1	N	1977	0	75	210	Tiles	4 (6)	16	30	5	Natural
		C2	N	2007	0	62	236	Tiles	2 (1)	13	29	4	MV2
		C3	N	2007	0	61	232	Tiles	2 (1)	22	26	3	MV2
	S12	C1	E	1965	1	56	168	Linoleum	5 (8)	18	22	6	Natural
		C2	E	1965	1	56	168	Linoleum	5 (8)	6	21	9	Natural
		C3	E	1965	1	56	168	Linoleum	5 (8)	11	26	8	Natural
	S13	C1	N	2001	0	69	173	Tiles	2 (2)	6	28	3	Natural
		C2	N	2001	0	74	311	Tiles	4 (2)	10	28	4	Natural
		C3	N	1975	0	80	224	Tiles	7 (13)	8	26	5	Natural

Table 1. *Cont.*

Season	School	Classroom	Type	Year[1]	Floor Level	Area (m^2)	Volume (m^3)	Type of Floor	Wo (m^2)	Decor[2] (m^2)	Nr Pupils	Mean Age	Type of Ventilation
Non-Heating Season	S14	C1	E	1955	2	56	179	Tiles	4 (8)	25	22	7	Natural
		C2	E	1955	0	56	224	Tiles	3 (8)	17	23	10	Natural
		C3	E	1955	0	56	224	Tiles	3 (8)	7	24	8	Natural
	S15	C1	E	1958	1	54	162	Tiles	4 (8)	9	25	7	Natural
		C2	E	1958	0	58	174	Wood	4 (8)	15	23	7	Natural
		C3	E	2000 *	0	58	157	Wood	2 (7)	14	23	8	Natural
	S16	C1	N	1975	0	53	153	Linoleum	4 (7)	7	24	4	Natural
		C2	N	1975	0	53	153	Linoleum	4 (7)	8	27	4	Natural
		C3	N	1975	0	55	153	Linoleum	4 (7)	8	27	3	Natural
	S17	C1	N	1973	0	55	176	Linoleum	2(1)	15	26	3	MV1
		C2	N	1973	0	61	171	Linoleum	3 (2)	5	26	4	MV1
		C3	N	1973	0	59	165	Linoleum	3 (2)	8	25	5	MV1

Notes: E: Elementary; N: Nursery; W$_o$: nr of windows to outdoor; MV1: Mechanical Exhaust System; MV2: Balanced Ventilation System; [1] Construction Year or last refurbishment year (*) [2] Surface of decorative paper in the walls.

2.2. Sample Collection and Analytical Methods

2.2.1. Allergens

Dust samples were collected from smooth floors (tile, vinyl or wood/parquet) in each classroom. If a carpet, rug or wall carpet was present, an additional sample was collected (20% of the classrooms). Trained technicians collected dust samples by vacuuming surfaces at a rate of 2 min·m^{-2} with a Roventa Silence Force® 1,500 Watt vacuum cleaner. Smooth floors were vacuumed until enough dust was collected (2 to 5 m^2 areas were sampled). For carpeted floors, areas of 1 m^2 were sampled. Dust samples were collected using a Mitest Collector (Indoor Biotechnologies, Charlottesville, VA, USA).

Overall, 61 dust samples were collected from the 51 studied classrooms. Samples were collected from smooth (51 samples) and carpeted floors, and from rugs and wall carpets (10 samples). The mean area of smooth floor samples was 4.4 ± 0.9 m^2, with an average dust collection of 490 ± 470 mg. The mean surface area of the carpeted floor and rug was 1.3 ± 0.5 m^2, with an average dust collection of 970 ± 590 mg.

The samples were stored at ambient temperature and sent to the laboratory within one week. Assays were conducted by the Paris Laboratory of Hygiene. Dust samples were passed through a 355 µm pore mesh sieve and extracted using a phosphate buffered saline solution with 0.05% Tween. The samples were centrifuged and the supernatants were examined for allergens.

The following allergens were quantified: *Felis domesticus* (cat) (Fel d 1), *Canis familiaris* (dog) (Can f 1), *Dermatophagoides farinae* (dust mite) (Der f 1) and *Dermatophagoides pteronyssinus* (dust mite) (Der p 1). These allergens were analysed using commercially available enzyme-linked immunosorbent assay (ELISA) kits (Quantitative ELISA Kits Biotechnologies, Charlottesville, VA, USA). The limits of Fel d 1, Can f 1, Der f 1 and Der p 1 detection were 0.06, 0.08, 0.31 and 0.16 µg·g^{-1}, respectively.

2.2.2. Metals

The wipe sampling method was used to collect dust from hard classroom surfaces by using the AFNOR NF X46-032 standard procedure [15], which is similar to the ASTM E172803 procedure [16]. Trained technicians sampled settled dust from three 32 cm x 32 cm areas (*i.e.*, 0.1 m^2) of each classroom, as described by Le Bot *et al.* [17]. Three sampling locations were examined for each classroom, (1) CA: Crossing areas, (2) NCA-S: Non-crossing areas exhibiting dust accumulation after short periods (e.g., beneath tables and chairs), and (3) NCA-L: Non-crossing areas exhibiting dust accumulation after long periods (e.g., beneath cupboards). After the sampling procedure was completed, the wipes were stored in polyethylene tubes and sent to the laboratory for further analysis. Bioaccessible (or leachable) and quasi-total dust analyses were performed by the French School of Public Health Laboratory, as previously described [17]. A gastric digestion test using hydrochloric acid was performed to determine metal bioaccessibility levels, and a metal analysis was conducted using an inductively coupled plasma mass spectrometer (ICP-MS) equipped with a quadrupole mass filter and octopole reaction system. The results are expressed in µg of metal per square meter. The metals examined using this method included arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), manganese (Mn), lead (Pb), antimony (Sb), strontium (Sr) and vanadium (V). Detailed description of procedures of ICP-MS analysis is available elsewhere [17].

The analysis validation method presented by Le Bot *et al.* [17] was employed. One field blank consisting of a wipe without dust was used for each school and was analysed following the procedure described above. The limits of quantification (LOQs) for the bioaccessible and total metal concentrations are reported in Table 2. The Sr concentrations in the field blanks always exceeded the LOQ; consequently, Sr concentrations were corrected based on the values of each field blank.

Table 2. Limits of quantification ($\mu g \cdot m^{-2}$) of the total and leachable metals.

Metal	Total Form	Leachable Form
As	0.6	0.2
Cd	0.8	0.4
Cr	10	4
Cu	32	15
Mn	64	30
Pb	2	1
Sb	0.8	0.4
Sr	8	5
V	8	5

2.3. Statistical Analysis

An analysis of variance of the results was performed using non-parametric statistics and a significance level of 0.050. Mann-Whitney tests were used for binary independent groups, and Kruskal-Wallis methods were used for multiple independent groups. When the groups were dependent, the Wilcoxon test was used for binary groups and the Friedman test was employed for multiple groups. The values below the limit of quantification were substituted by the output values given by the laboratory for the statistical tests. All of these analyses were conducted using the XLSTAT 2014.1.09 software program.

3. Results and Discussion

3.1. Allergens

Table 3 shows the classroom pet allergen results. Dog and cat allergens were detected in 58% and 57% of the collected samples, respectively. Higher detection percentages of both allergens were found in the carpet/rug samples relative to the sample obtained from smooth floors. For carpets/rugs, detection percentages reached 80% for cat allergens and 70% for dog allergens. The concentration of pet allergens varied from 0.01 to 0.39 $\mu g \cdot g^{-1}$ in Fel d 1 and from 0.01 to 0.94 $\mu g \cdot g^{-1}$ in Can f 1. These values are lower than the ones described by Cyprowski *et al.* [18] who found pet allergens in Polish kindergartens in concentrations ranging from 0.004 to 3.6 $\mu g \cdot g^{-1}$ for Fel d 1 and from 0.01 to 10 $\mu g \cdot g^{-1}$ for Can f 1.

Table 3. Pet allergen concentrations in the settled dust vacuumed from classrooms.

Floor Type	Parameter	Settled Dust Loading ($g \cdot m^{-2}$)	Allergen Concentration ($\mu g \cdot g^{-1}$)	
			Fel d 1	**Can f 1**
All (n = 61)	% > LOQ	-	58%	57%
	Mean ± SD	0.25 ± 0.40	0.11 ± 0.09	0.13 ± 0.14
	Median	0.10	0.08	0.09
Smooth (n = 51)	% > LOQ	-	54%	54%
	Mean ± SD	0.13 ± 0.16	0.09 ± 0.08	0.13 ± 0.15
	Median	0.07	0.07	0.09
Carpet/rug (n = 10)	% > LOQ	-	80%	70%
	Mean ± SD	0.87 ± 0.63	0.16 ± 0.09	0.14 ± 0.09
	Median	0.73	0.17	0.14

Notes: n: total number of samples; smooth floor: tile, vinyl and wood flooring; LOQ: limit of quantification; SD: standard deviation.

Carpeted floors/rugs contained roughly seven times more dust than smooth floors, which suggested that this type of flooring promotes dust accumulation.

Cat allergen concentrations were significantly higher on carpeted floors/rugs than on smooth floors (Kruskal-Wallis test with a p-value of 0.014). Dog allergen concentrations did not significantly differ across the floor types (p-value of 0.433). Regarding the ventilation (mechanical *versus* natural) and classroom types (nursery *versus* elementary school), no significant differences were found for either allergen. However, dog allergen concentrations were roughly three times higher (p-value of 0.003) during the heating season (0.18 ± 0.18 $\mu g \cdot g^{-1}$) than during the non-heating season (0.07 ± 0.04 $\mu g \cdot g^{-1}$). A similar trend was found by Cyprowski *et al.* [18] who observed a significant difference between the mean levels of dog allergen in different seasons at Polish kindergartens, with heating season showing almost twice higher levels (1.3 ± 2.1 $\mu g \cdot g^{-1}$) than non-heating season (0.80 ± 7.9 $\mu g \cdot g^{-1}$).

Urban classrooms (n = 48) presented a median value of 0.07 $\mu g \cdot g^{-1}$ for cat allergen (ranging from 0.01 to 0.39 $\mu g \cdot g^{-1}$), while rural classrooms (n=3) presented a median value of 0.05 $\mu g \cdot g^{-1}$ (ranging from 0.04 to 0.11 $\mu g \cdot g^{-1}$). A similar trend was observed for dog allergen with urban classrooms presenting a median value 0.09 $\mu g\ g^{-1}$ (ranging from 0.01 to 0.94 $\mu g \cdot g^{-1}$), while rural classrooms presented a median value of 0.05 $\mu g \cdot g^{-1}$ (ranging from 0.03 to 0.11 $\mu g \cdot g^{-1}$).

Salo *et al.* [6] compiled a review of studies on allergens in schools and day care environments that were published during the last two decades and gathered information from 28 and 19 studies on cat and dog allergens, respectively. The concentrations of allergens measured in the present study are similar to those observed in previous studies, especially in studies conducted in Scandinavian and Asian countries. However, the present results are lower than those observed in studies in the United States. The only French study included in the review is the one conducted by Andrade *et al.* [19], who studied 30 day care centers. These authors observed similar values for cat allergens (within a range of <0.1 - 4.5 $\mu g \cdot g^{-1}$) and higher values for dog allergens (within a range of <0.2–4.5 $\mu g \cdot g^{-1}$). Recently, Chatzidiakou *et al.* [20] studied five primary schools and one nursery school in London (18 classrooms) and found a median cat allergen concentration of 0.27 $\mu g \cdot g^{-1}$ (range: 0–1.76 $\mu g \cdot g^{-1}$) and a median dog allergen concentration of 0.05 $\mu g \cdot g^{-1}$ (range: below the limit of detection −0.68 $\mu g \cdot g^{-1}$).

Mite allergens were only assessed in dust samples from carpeted floors/rugs. No Der p 1 mite allergens were found in any of the 10 samples, and Der f 1 was only found in one sample with a concentration of 1.7 $\mu g \cdot g^{-1}$, which was less than the allergic sensitisation limit of 2 $\mu g \cdot g^{-1}$ proposed by Platts-Mills *et al.* [21]. These results agree with those of previous studies conducted on Swedish and Asian classrooms, but are less significant than those observed for classrooms in the United States [6]. In addition, Chatzidiakou *et al.* [20] found low concentrations of Der f 1 in one school, with a median concentration of 0.17 $\mu g \cdot g^{-1}$.

The low mite allergen concentrations observed in this study could be attributed to the lower values of relative humidity in the studied classrooms (31% ± 5% during the heating season and 47% ± 8% during the non-heating season [4]) relative to those examined in other studies. Dust mites only survive when the relative humidity is at least 55% over a sufficient period because water vapor serves as their only water source [6,22].

3.2. Metals

3.2.1. Loadings in Settled Dust

Table 4 presents the metal loadings in the settled dust samples. The total and leachable forms of As, Pb and Sr were quantified in more than 90% of the analysed samples. The metal loadings in the dust samples decreased as follows: Mn > Cu > Sr > Pb > Cr > Sb > As > Cd ≈ V. Thus, Mn and Cu were the metals found with the highest mean loadings, namely 65 and 43 $\mu g \cdot m^{-2}$, respectively. The median loadings are similar to those reported by Glorennec *et al.* [7] and Rasmussen *et al.* [23] for floor dust in homes across France and Canada, respectively, with the exception of As loadings. In the present study, median As loadings were around four times higher (2.7 $\mu g \cdot m^{-2}$) than the As loadings found in homes, both in France (<LOQ) and Canada (0.7 $\mu g \cdot m^{-2}$). Similar studies of classrooms examining metals loadings were not found in the literature as a means for comparison. However, some studies that focused on metal concentration in indoor dust in schools can be found. Darus *et al.* [24] studied the indoor floor dust in three nursery schools in Malaysia and found that the heavy metal concentrations followed the order Fe > Al > Zn > Pb > Ba > Cu > Cr > Ni, with Fe reaching 7919 $mg \cdot kg^{-1}$. A similar trend was found by Latif *et al.* [25] in ten Malaysian pre-schools, where the order of heavy metal concentrations was Fe > Pb > Zn > Cr > Cd, with Fe reaching 10,739 $mg \cdot kg^{-1}$. Lu *et al.* [26] studied the indoor dust of 46 nursery schools in China and showed that its heavy metal composition was following the order Ba > Mn > Zn > Pb > Cr > Cu > Co > Ni > As, with Ba reaching 2141 $mg \cdot kg^{-1}$.

Only 2% of the settled dust samples (n = 3) contained Pb loadings above the French limit of 70 $\mu g \cdot m^{-2}$ defined by the French Committee of Public Health (HCSP) that motivates childhood Pb poisoning screening [27]. This recently established guideline is consistent with the threshold of 64–128 $\mu g \cdot m^{-2}$ defined by Dixon *et al.* [28] for protecting children from high blood Pb levels (PbB ≥ 10 µg/dL). Additional data regarding the Pb levels in settled dust in classrooms and in wall paints are presented in Derbez *et al.* [29]. Pb remains a health concern due to the absence of a known effect threshold [9].

Table 4. Metal loadings ($\mu g \cdot m^{-2}$) in settled dust from classrooms.

Metals ($\mu g \cdot m^{-2}$)	n	LOQ	% > LOQ	Mean ± SD	Min	P5	P25	Median	P75	P95	Max
As_{total}	151	0.6	96	3.4 ± 2.6	<LOQ	0.7	1.7	2.7	4.3	8.9	15.4
$\text{As}_{\text{leachable}}$	152	0.2	99	1.3 ± 1.9	<LOQ	0.3	0.6	0.9	1.3	4.2	17.3
Cd_{total}	151	0.8	8	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	1.8	12.4
$\text{Cd}_{\text{leachable}}$	151	0.4	26	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	1.5	3.8
Cr_{total}	151	10	46	12 ± 11	<LOQ	<LOQ	<LOQ	<LOQ	17	32	83
$\text{Cr}_{\text{leachable}}$	152	4	24	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	4	7	16
Cu_{total}	151	32	26	43 ± 195	<LOQ	<LOQ	<LOQ	<LOQ	33	73	2399
$\text{Cu}_{\text{leachable}}$	152	15	59	23 ± 26	<LOQ	<LOQ	<LOQ	17	26	62	193
Mn_{total}	151	64	36	65 ± 72	<LOQ	<LOQ	<LOQ	<LOQ	78	164	643
$\text{Mn}_{\text{leachable}}$	152	30	57	36 ± 25	<LOQ	<LOQ	<LOQ	34	45	74	169
Pb_{total}	151	2	99	16 ± 19	<LOQ	4	7	9	17	46	128
$\text{Pb}_{\text{leachable}}$	151	1	100	12 ± 13	2	3	5	7	14	39	83
Sb_{total}	151	0.8	68	4.2 ± 14.2	<LOQ	<LOQ	<LOQ	1.2	2.3	14.5	117.9
$\text{Sb}_{\text{leachable}}$	152	0.4	37	0.9 ± 2.4	<LOQ	<LOQ	<LOQ	<LOQ	0.6	4.3	18.5
Sr_{total}	151	8	100	35 ± 21	3	11	19	31	47	74	122
$\text{Sr}_{\text{leachable}}$	152	5	100	27 ± 16	4	8	15	22	37	56	89
V_{total}	151	8	23	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	18	86
$\text{V}_{\text{leachable}}$	152	5	0	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ

n: number of samples; LOQ: limit of quantification; SD: standard deviation; P: percentiles.

3.2.2. Leachability

Metal leachability results are shown in Figure 2. Only values exceeding the LOQ were used to evaluate the metal leachability levels. Thus, V was excluded from this evaluation.

The mean metal leachability values were 36 ± 15, 76 ± 30, 29 ± 9, 77 ± 18, 55 ± 17, 80 ± 13, 27 ± 8 and 77% ± 15% for As, Cd, Cr, Cu, Mn, Pb, Sb and Sr, respectively. These values are consistent with those observed by Glorennec *et al.* [7] for floor dust from French homes, except for Mn. In this study, a lower mean Mn leachability value of 55% was observed, which differed from the 80% value reported for French homes. As observed for homes by Glorennec *et al.* [7], considerable levels of leachability variability were found among the settled dust samples, mainly for As, Cd, Cu, Mn and Pb. Overall, these metal leachability values are also consistent with the mean values reported by Ibanez *et al.* [30] in their international review of household dust metal content and bioaccessibility. The high leachability variability observed in the present study may be explained by the specific bioaccessibility of an element in the dust sampled from a given classroom, which depends largely on its speciation that is driven by which indoor and outdoor sources are present and by the subsequent transformations and accumulation processes in the dust matrix [31].

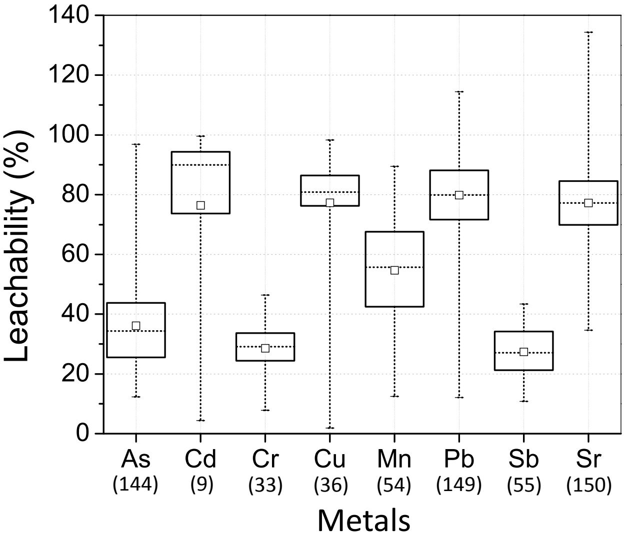


Figure 2. Leachability of metals in settled dust in classrooms. The number of samples, *n*, is shown in brackets. A percentile box of 25%–75% with minimum and maximum values is shown with the mean value.

3.2.3. Inter-and Intra-School Variability

The spatial variability of the metal loadings was assessed at the following three levels: Between schools (inter-school), between classrooms of a given school (intra-school or inter-classroom) and between different sampling locations of a given classroom (intra-classroom). This variability is expressed as the relative standard deviation (RSD, %). Figure 3 shows the RSDs for the total and leachable metals for these levels.

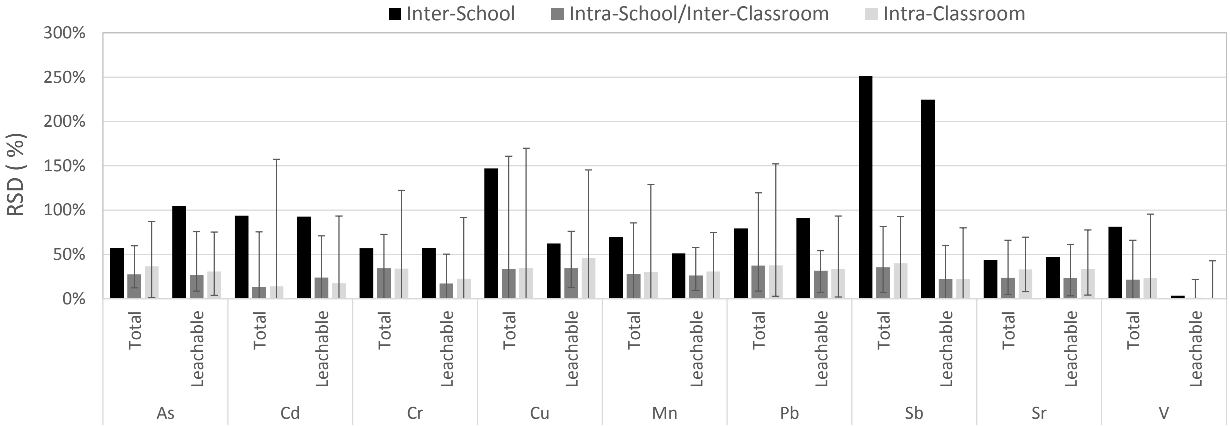


Figure 3. Relative Standard Deviation—RSD (%) values from Inter-School (mean value), Intra- School/Inter-Classroom and Intra-Classroom analyses (mean value, with lower and higher limits representing minimum and maximum RSD values obtained).

The intra-classroom variability varied from 0 to 170% and was in the same order of magnitude as the intra-school/inter-classroom variability, which ranged from 0% to 161%. However, the RSD means for the intra-classroom and intra-school/inter-classroom scales were always lower than the RSD mean for inter-school variability. This result suggests that greater variability was seen between schools rather than between classrooms, indicating that school location (*i.e.*, proximity to nearby local sources) is an important determinant for intra-classroom levels of metals.

A comparison of ground level data (63% of classrooms) with first floor data (35% of classrooms) showed no statistically significant difference (Mann-Whitney, alpha = 0.05), allowing all schools and classrooms to be compared for spatial variability. However, Pb levels on settled dust from the first floor were found to be significantly higher than in the ground level. When comparing inter-school and intra-school variability, a bias could appear due to differences between floor levels. No significant differences in average intra-school variability of Pb concentration were observed when selecting schools with all classrooms at the same level and schools with classrooms at a different level.

3.2.4. Influence of Sampling Location within the Classroom

The variations within the classrooms were studied in detail and are presented in Figure 4, which shows the loadings of each metal (total and leachable) for each of the three sampling locations.

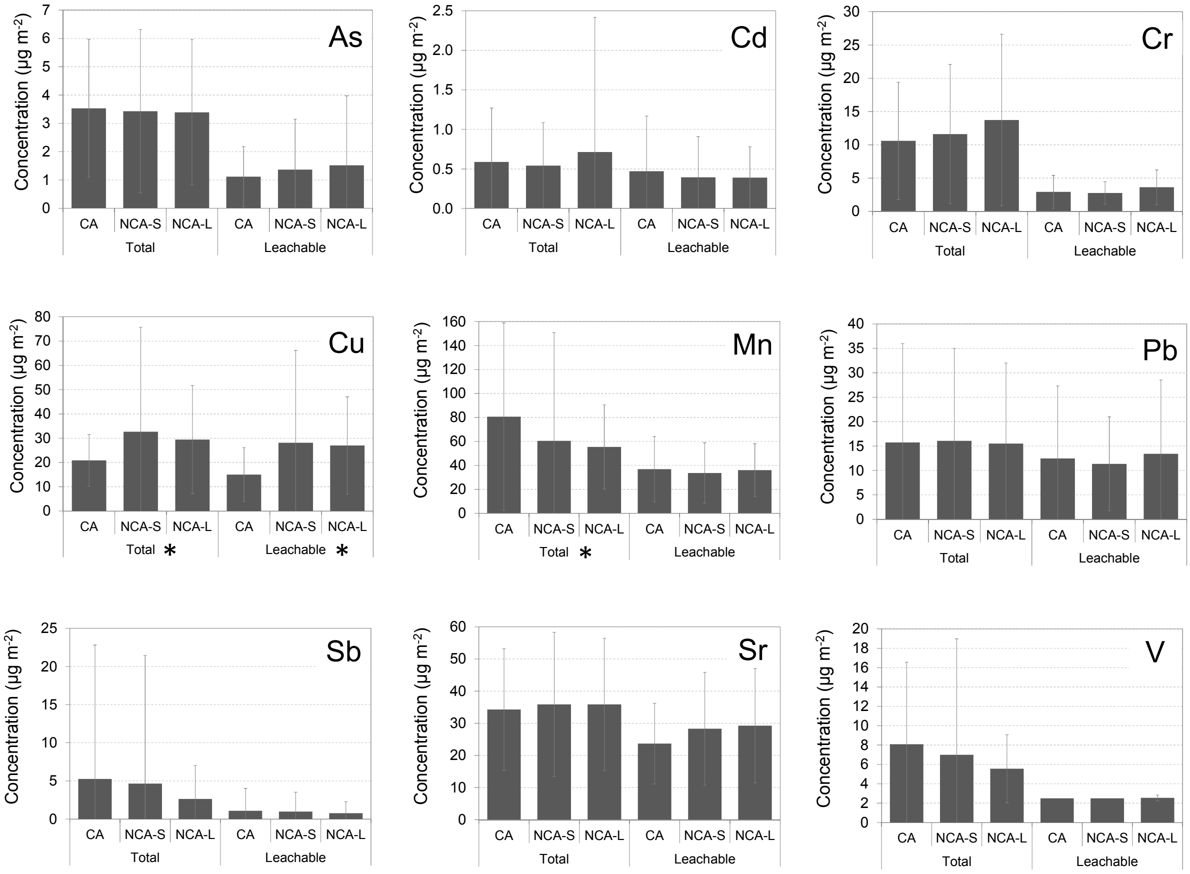


Figure 4. Mean metal loadings (total and leachable; µg·m^{-2}) per sampling location. CA: Crossing area; NCA-S: Non-crossing area with dust accumulated over short periods; and NCA-L: Non-crossing area with dust accumulated over long periods. *: Denotes a significant difference within a group according to Friedman test results (*p*-value < 0.050).

The leachable and total Cu and total Mn loadings varied significantly depending on the type of sampling location (Friedman test with *p*-values of < 0.001, 0.019 and 0.015, respectively). The lowest mean leachable Cu content was detected in the settled dust that was collected from the crossing area and was much lower than the mean values measured for the two other sampling locations (non-crossing areas with dust accumulated over short and long periods) (Wilcoxon test, *p*-value < 0.050). The total Cu loadings were significantly lower in the settled dust from the crossing area than from the other locations (Wilcoxon test, *p*-value < 0.050). In addition, the total Mn loadings were significantly higher in the settled dust that was collected from the crossing area than from the other sampling locations (Wilcoxon test, *p*-value < 0.050). The loadings of the other metals (total and leachable) did not vary significantly across the sampling locations. This finding shows that no significant variations in settled dust loadings occurred between the sampling locations for a set of classrooms. Considering additionally that the variability of loadings comes mainly from the differences between schools, this finding allows for performing the sampling in a unique location in the classroom. Thus, combining settled dust samples from various locations of a classroom, as described by Lu *et al.* [26], is not required.

3.2.5. Influence of Flooring Material

Three different types of flooring were observed in the studied classrooms: Vinyl, tile and wood. Figure 5 shows the metal loadings for each floor type.

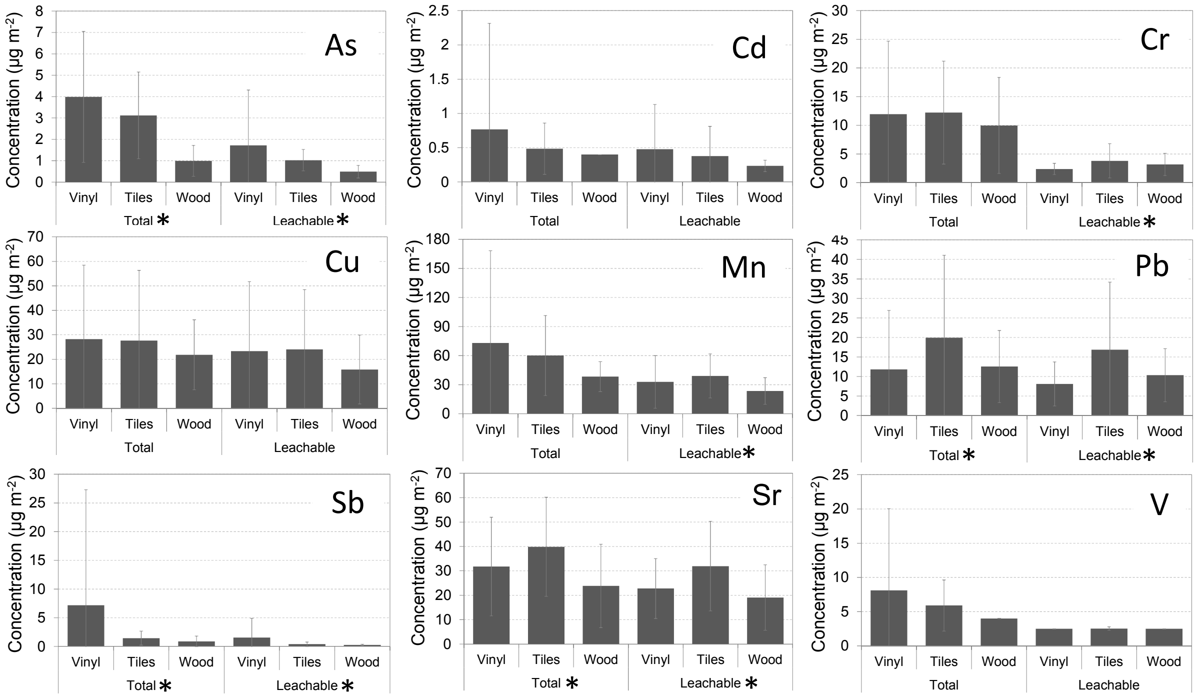


Figure 5. Mean metal loadings (total and leachable; $\mu g \cdot m^{-2}$) per classroom floor type. *: Denotes a significant difference within a group according to Kruskal-Wallis test results (*p*-value < 0.050).

Significant differences between the different floor types were found for loadings of leachable and total As, Pb, Sb and Sr and for loadings of leachable Cr and Mn (Kruskal-Wallis test, *p*-value < 0.050).

Tiled floors showed higher leachable Cr, Pb, Mn and Sr loadings than vinyl floors, and vinyl floors presented higher leachable Sb loadings than tiled floors (Mann-Whitney test, p-value < 0.050). Tiled floors presented significantly higher total Pb loadings than vinyl floors and significantly higher total Sr loadings than wood and vinyl floors. These high loadings of Pb and Sr in tiles, when compared with other flooring materials, may be explained by the production process of tiles, which may include both metals in the ceramic glaze of tiles [32]. In addition, vinyl floors presented significantly higher total Sb loadings than tile and wood floors, which indicate that this type of floor has Sb in its composition. In fact, Sb is known to be used in the production of vinyl floors in the form of antimony trioxide (SbO_3), used as a chemical synergist in chlorinated and brominated flame retardants to increase the retardants' effectiveness [33]. Finally, wood floors presented significantly lower leachable and total As loadings than vinyl and tile floors (Mann-Whitney test, p-value < 0.050).

3.2.6. Influence of Cleaning Frequency

Using data gathered from questionnaires completed by the school managers, it was possible to assess floor cleaning routines, namely, the frequency of dry cleaning (e.g., broom, vacuum cleaner) and wet cleaning (e.g., wet mop). The effects of the frequency of dry cleaning could not be evaluated because it was performed daily in each classroom. Three frequencies of wet cleaning were reported, daily (63% of the classrooms), one or two times each week (25% of the classrooms) and less than once a week (only 12% of the classrooms).

Significant differences between wet cleaning frequencies were found for the loadings of total Cu and Pb in settled dust sampled in the crossing area (Kruskal-Wallis test, p-value < 0.050). Classrooms that were cleaned via wet cleaning less than once a week showed significantly higher total Cu and Pb loadings than those wet-cleaned daily or once or twice each week (Mann-Whitney test, p-value < 0.050). This result shows that wet cleaning procedures should be conducted in classrooms at least once each week to reduce the loadings of total Cu and Pb in floor dust.

3.2.7. Influence of Other Parameters

The relationships with other parameters (e.g., the sampling season, school type and ventilation type) on metal loadings are presented in Table 5.

The total Cu loadings were significantly higher during the heating season, while significantly higher leachable Cu, Mn and Pb loadings and total Mn, Pb, Sr and V loadings were found during the non-heating season. This result potentially occurred because the primary outdoor sources of Cu, Mn, Pb and Sr are transported indoors during the summer as windows are opened with a higher frequency during this season.

Elementary schools showed significantly higher leachable Cr, total and leachable Pb and leachable Sr loadings than nursery schools, and nursery schools only presented significantly higher total V loadings. This observation may be due to the fact that 90% of the elementary classrooms had natural ventilation. In fact, naturally ventilated classrooms showed significantly higher leachable Cr and total and leachable Pb and Sr loadings. However, the total As levels were significantly higher in mechanically ventilated classrooms. No explanation of this phenomenon has been proposed, and no similar studies regarding this variable were found in the literature.

Urban classrooms (n = 48) presented a median value of 102 μg·m^{-2} for the sum of leachable form of all analysed metals (ranging from 55 to 317 μg·m^{-2}), while rural classrooms (n = 3) presented a slightly higher median value of 136 μg·m^{-2} (ranging from 111 to 193 μg·m^{-2}). A similar trend was observed for the sum of all analysed metals (total form) with urban classrooms presenting a median value 138 μg·m^{-2} (ranging from 88 to 2794 μg·m^{-2}), while rural classrooms presented a median value of 222 μg·m^{-2} (ranging from 107 to 291 μg·m^{-2}).

Table 5. The total and leachable mean (± standard deviation) metal loadings (μg·m^{-2}) in the settled dust samples from the 51 classrooms by sampling season, school type and ventilation type. Bold values denote significant *p*-values (Mann-Whitney test).

		Season			**School**			**Ventilation**		
Metal	**Form**	**Heating**	**Non-Heating**	**p-value**	**Nursery**	**Elementary**	**p-value**	**Natural**	**Mechanical**	**p-value**
As	Total	3.8 ± 2.8	3.0 ± 2.3	0.064	3.5 ± 2.7	3.4 ± 2.6	0.789	3.3 ± 2.8	3.9 ± 2.1	**0.010**
	Leachable	1.6 ± 2.3	0.9 ± 0.5	0.323	1.2 ± 1.2	1.4 ± 2.2	0.466	1.3 ± 2.0	1.5 ± 1.4	0.179
Cd	Total	0.53 ± 0.51	0.73 ± 1.60	0.952	0.80 ± 1.61	0.47 ± 0.35	0.086	0.65 ± 1.25	0.54 ± 0.57	0.855
	Leachable	0.42 ± 0.57	0.40 ± 0.51	0.834	0.49 ± 0.68	0.36 ± 0.41	0.721	0.41 ± 0.51	0.42 ± 0.64	0.208
Cr	Total	11 ± 8	14 ± 13	0.082	12 ± 13	12 ± 9	0.510	12 ± 10	11 ± 14	0.080
	Leachable	3.2 ± 2.6	2.9 ± 1.7	0.939	2.4 ± 1.0	3.6 ± 2.8	**0.002**	3.4 ± 2.6	2.3 ± 1.0	**0.009**
Cu	Total	52 ± 250	31 ± 31	**0.048**	29 ± 38	55 ± 260	0.355	49 ± 230	27 ± 34	0.220
	Leachable	21 ± 26	27 ± 26	**0.006**	24 ± 33	23 ± 20	0.821	24 ± 24	22 ± 31	0.270
Mn	Total	51 ± 43	85 ± 96	**0.001**	79 ± 100	55 ± 37	0.371	67 ± 78	62 ± 54	0.921
	Leachable	31 ± 20	42 ± 29	**0.005**	34 ± 28	37 ± 22	0.107	37 ± 26	31 ± 20	0.169
Pb	Total	14 ± 20	18 ± 16	**0.018**	12 ± 16	19 ± 20	**<0.001**	18 ± 18	10 ± 18	**<0.001**
	Leachable	11 ± 13	14 ± 14	**0.020**	7 ± 5	16 ± 16	**<0.001**	15 ± 15	6 ± 4	**<0.001**
Sb	Total	1.9 ± 2.0	7.3 ± 21.5	0.301	7.5 ± 21.0	1.6 ± 1.2	0.775	5.0 ± 16.5	2.0 ± 2.6	0.861
	Leachable	0.5 ± 0.6	1.5 ± 3.6	0.729	1.6 ± 3.5	0.4 ± 0.4	0.765	1.1 ± 2.8	0.5 ± 0.7	0.515
Sr	Total	32 ± 19	41 ± 21	**0.003**	34 ± 21	36 ± 20	0.301	38 ± 22	29 ± 15	**0.048**
	Leachable	26 ± 17	29 ± 15	0.074	23 ± 13	30 ± 18	**0.013**	30 ± 17	20 ± 10	**0.004**
V	Total	5.1 ± 3.0	9.4 ± 12.6	**0.001**	9.0 ± 12.3	5.2 ± 3.3	**0.006**	7.0 ± 10.0	6.5 ± 3.8	0.125
	Leachable	2.5 ± 0.2	2.5	0.408	2.5	2.5 ± 0.2	0.388	2.5 ± 0.2	2.5	0.553

3.2.8. Enrichment Factors

To examine the crustal and non-crustal origins of the studied metals, the crustal enrichment factor method was employed using Equation (1). In Equation (1), the enrichment factors (EF_X) were calculated using Sr as a crustal reference trace element [34] and the soil compositions described by Mason and Moore [35].

$$EF_X = \left(\frac{[X]}{[Sr]}\right)_{\text{settled dust}} / \left(\frac{[X]}{[Sr]}\right)_{\text{crust}} \quad (1)$$

where EF_X is the enrichment factor of element X, $([x]/[Sr])_{settled\ dust}$, is the ratio between X and Sr in the settled dust in the classrooms and $([x]/[Sr])_{crust}$ is the ratio of X to Sr in the soil given by Mason and Moore [35] or obtained from the available median values for outdoor playground soils in France [7].

Elements with EF values that approach unity are generally considered as predominantly crustal in origin. If the evaluated element shows EF values of more than 10, its provenance is mainly attributed to local, regional and/or long-term transportation phenomena caused by other natural and/or anthropogenic processes [36]. Figure 6 shows EF values for settled dust in the studied classrooms.

Based on soil composition values presented by Mason and Moore [35], the only heavily enriched elements (EFs > 10) were Cd and Sb, which are of anthropogenic origin (e.g., traffic emissions mainly from tire and brake wear [34,37]). Mean EF values for Cu, As and Pb approached 10 and were greater than 10 in some classrooms, which indicates the contribution of specific non-crustal sources for these metal loadings in many classrooms.

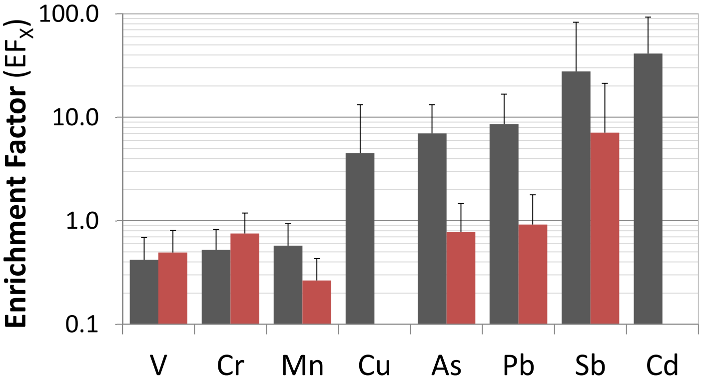


Figure 6. Enrichment factors using Sr as a reference element and the soil compositions presented by Mason and Moore [35] (grey bars) and Glorennec *et al.* [7] (red bars).

Using data for French soils provided by Glorennec *et al.* [7], only the enrichment factors of six elements were assessed. Among these elements, Sb was the only element that showed enrichment characteristics, with 11% of the samples showing strong enrichment characteristics (EFs > 10) (one sample for school 3, seven samples for school 4 and nine samples for school 16).

4. Conclusions

This study is the first to analyse allergens and metals in schools along with building characteristics in France. The concentrations of allergens in settled dust were generally low and fell below the established sensitisation limit regarding dust mite allergens. Floors with carpets or rug presented roughly seven times more dust and significantly higher concentrations of cat allergens than smooth floors. The highest metal loadings in dust were observed for Mn and Cu, and the Pb loadings were lower and fell below the French guidelines. Higher degrees of metal leachability were found for Cd, Cu, Pb and Sr, with values of approximately 80%. In cases of dust ingestion by children, a large proportion should be assimilated through the gastro-intestinal tract.

Regarding metal loadings in settled dust, intra-classroom and intra-school variabilities were lower than the inter-school variability. The sampling location in the classroom did not affect the mean loadings of most metals in both forms, except for Cu and Mn. This finding can help guide future sampling strategy. However, the type of floor had a greater effect on metal loadings than the sampling location. For example, tiled floors showed higher Pb loadings than vinyl or wood floors. Thus, the type of school floor should be carefully selected to limit the exposure of children to harmful substances. In addition, wet classroom cleaning procedures should be used weekly to reduce the metal loadings. Lastly, the enrichment factors indicated the contribution of specific non-crustal sources for some metals, in particular Cd and Sb. Different sources can be attributed to the metals in the settled dust in classrooms: indoor sources such as the materials used in the classroom flooring, outdoor sources such as traffic and soil sources.

Overall, this study optimised sampling and analytical procedures and presented information on the exposure of children to allergens and metals in classroom settled dust. The procedures described in this study are currently being employed in a nationwide survey of 300 randomly selected French schools (600 classrooms). Thus, the results of this study will be generalized using a larger sample of classrooms.

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Author Contributions

Nuno Canha conducted statistical analyses and interpretation and drafted the manuscript. Corinne Mandin and Olivier Ramalho helped in data interpretation and in writing the manuscript. Guillaume Wyart contributed to data cleaning and data analysis. Jacques Ribéron revised the manuscript. Claire Dassonville supervised the local team and the laboratories, helped in the school recruitment and supervised data acquisition. Mickael Derbez made the study design, selected and trained the local team and supervised the whole study. All authors read and approved the final manuscript.

Conflicts of Interest

The authors declare that they have no conflict of interest.

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