

TOXIC METALS IN INDOOR DUST COLLECTED FROM HOUSES INCLUDED IN THE “6X6X6” PROJECT (COVILHÃ, PORTUGAL) DURING THE COOLING SEASON

M. Ramiro Pastorinho¹, João Carlos Lanzinha^{2*}, Luís Taborda-Barata¹, Maria Assunção Vaz-Patto¹, Marisa Monteiro², Miguel C.S. Nepomuceno², Ana C.A. Sousa¹

1: CICS-UBI Health Sciences Research Centre
Department of Medical Sciences
Faculty of Health Sciences
University of Beira Interior
6201-001 Covilhã, Portugal

e-mail: {rpastorinho, tabordabarata, mariavazpato, anasousa}@fcsaude.ubi.pt,
web: <http://www.ubi.pt>

2: LABSED – Health Laboratory in Buildings
C-MADE: Centre of Materials and Building Technologies
Department of Civil Engineering and Architecture
Faculty of Engineering
University of Beira Interior
6201-001 Covilhã, Portugal

e-mail: {jcgl, mcsn}@ubi.pt; m-r-monteiro@hotmail.com, web: <http://www.ubi.pt>

Keywords: Toxic metals, health, indoor environment, housing.

Abstract *We spend more than 90 per cent of our daily lives indoors. Managing the indoor environment so that we feel comfortable and healthy is therefore very important. However, this management is only possible if rigorous data on both construction parameters and indoor environment exists. Moreover, the evolution of construction techniques and materials used modulates both aspects. As a consequence, it is necessary that the information obtained reflects a long period of time. That is the main driver for the project “Six by Sixty by Six” undertaken by the Civil Engineering and Architecture Department and the Faculty of Health Sciences of the University of Beira Interior. Six houses constructed in six different decades (1960’s-2010’s) were surveyed for sixty days regarding a set of parameters (e.g., temperature, humidity, CO₂, CO, VOCs). Additionally, dust was collected by the vacuum cleaner bag and was analyzed for specific contaminants. This work presents the results obtained for the toxic metals mercury, arsenic, cadmium, and lead in dust samples. There is no legislation regulating maximum permissible levels for any of the studied metals in dust. A comparison with “Portaria N° 1450/2007” which regulates the maximum metal contents in dredged sediment that can be safely deposited in soils (here used as an action threshold) reveals that the values for As in all houses were transcended. For Cd and Hg they were only transcended in one house, whereas for lead all the values were below this threshold. Linear regression with age of construction used as independent variable revealed no relationship between this variable and metal concentrations with the exception of As (R^2 adjusted= 0.41). Findings are discussed under the light of potential health outcomes upon the residents.*

1. INTRODUCTION

Nowadays people spend between 85 and 90% of their time indoors [1] being the majority of this time expended at their homes. On average, an adult will remain the equivalent to 15.7h/day at home [2]. Considering the magnitude of this time and that the presence of contaminants in the domestic environment is both frequent and diversified, the need to study this environment becomes self-evident. This need is reinforced by the knowledge that the concentration of several classes of contaminants in the indoor environment is higher than that of the external environment [3] (and references therein). Public perception will allocate the presence of contaminants in the domestic environment as the result of transport from the external environment. However, activities and commodities that we deem indispensable for our everyday comfort may be contributors to the presence of these chemicals. Contaminants can arise from within house sources such as fuel used in heating systems, tobacco smoking, building and decoration materials, cleaning and maintenance products, personal hygiene products and from activities such as gardening, pottery, and soldering [4]. Managing the indoor environment so that we feel both comfortable and healthy is therefore very important. However, this management is only possible if rigorous data on both construction parameters and indoor chemicals exists. Moreover, the evolution of construction techniques and materials used modulates both aspects, thus it is necessary that the information obtained reflects an extended period of time. Those are the main drivers for the project “Six by Sixty by Six” undertaken by the Civil Engineering and Architecture Department and the Faculty of Health Sciences of the University of Beira Interior. The subject of this paper is the environmental evaluation of the houses enrolled in the project.

Traditionally, the evaluation of interior environmental quality is performed using air as the reference matrix. However, the large spatial and temporal variability of contaminants in the air precludes their quantitative and qualitative integration, particularly over large temporal scales [5]. In face of these limitations, the use of house dust was proposed as an appropriate matrix for the evaluation of indoor environmental quality [5]. House dust is a heterogeneous mixture of soil particles, settled atmospheric particles, hair, fibers (natural and artificial), fungi, pollen, allergens, bacteria, virus, animal fur, human skin, smoke associated chemicals (from heating and food preparation), building and furniture components, to cite only the most common [6]. The composition and physical-chemical characteristics of house dust will determine the type of contaminants that will be associated to it, with the particle size, type, texture, polarity and lipophilicity playing a fundamental role [3]. Thus, house dust reflects and integrates not only air contaminants (that will adsorb to suspended particulate matter and will settle [3]) but also contaminants from other sources (both outdoor and indoor) present in the indoor environment, acting as a concentrator and reservoir. In dust, these chemicals are less exposed to alteration and degradation that naturally occurs when they are freely circulating in the environment (e.g., by the influence of sunlight, temperature fluctuations, microbial action) [3,6,7]. Among these chemicals are metals, which typically occur in dust at higher concentrations than those present in soils of the same area [8]. Generally, metals in dust are found under particle form (at least in inorganic speciation states, the most frequent) [9]. Aside from well-known cases such as that of lead, many other metals are normally present in the indoor environment and can have impact in human health (even those that are essential) [9].

House dust is considered an important source of metal exposure, particularly for children [10]. Since in modern houses the use of a vacuum cleaner is standard, the collection chamber of these appliances (particularly when bags are used) offers itself as the ideal collection spot for representative samples. This work presents the results obtained for the toxic metals mercury, arsenic, cadmium, and lead found in the dust collected in the vacuum cleaner bag from six houses in Covilhã (a small city in the interior center of Portugal) constructed in six different decades (1960's-2010's) after a survey of sixty days during the cooling season (May-July 2015).

2. MATERIAL AND METHODS

2.1. Sampling area

Covilhã is located in the mountainous interior center of Portugal in the Cova da Beira Region at an altitude of 700m and with approximately 50,000 inhabitants. For decades the Municipality had a very strong textile industry, and to this day Covilhã is synonym of fabrics. However, the crisis experienced by the sector in the 1980's, led to a profound reconversion of the local economy and currently 219 small businesses in the transformation sector are distributed between two small industrial parks to the north and south of the city [11]. Approximately 30 km to the southeast a major mining complex ("Minas da Panasqueira") can be found and has been in continuous operation for the last 100 years. During the period of the study the wind regime varied. May was characterized by a dominant wind direction from NW with an average speed of 6.3 Km/h. In June the predominant wind direction was WNW with average speed of 3.9 Km/h, shifting in July to a W dominance and an average speed of 3.3 Km/h (information obtained from the University of Beira Interior weather station at <http://webx.ubi.pt/~goa/>).

2.2. Sample collection

Houses in the 6x60x6 project are located in the Covilhã urban area and were built between 1960 and 2014. Inhabitants were given an informed consent to sign previously to any sample collection. At the beginning of the sampling period vacuum cleaners at each house were fitted with a new vacuum bag (Wonderbag Compact WB 305120) and it was requested for the appliance not to be used in automobiles and garages. After 60 days the bag was collected and transported to the lab inside a sealed polyethylene bag.

2.3. Sample processing

Samples were sieved through a 5 mm mesh stainless steel sieve for larger debris removal, and the sample thus obtained re-sieved through a 500 µm mesh stainless steel sieve. Final samples were stored in polyethylene tubes and maintained at room temperature and protected from the light until analysis.

2.4. Metal quantification

Mercury was quantified through Atomic Absorption Spectroscopy (with thermal decomposition and gold amalgamation in a Nippon Instruments Corporation (NIC) MA-3000) in accordance with USEPA 7473 method [12]. The NIC MA-3000 does not require sample pre-treatment. Total mercury was determined in 50 mg of dust. Calibration was achieved with successive dilutions of a mercury standard (TraceCERT® Sigma-Aldrich, 1000 mg/L Hg metal).

Arsenic, cadmium, and lead were quantified through atomic absorption spectroscopy (PerkinElmer AAnalyst 800 Spectrometer) with graphite furnace and Zeeman correction. Samples (200 mg) were digested with a 5:1 mixture of HNO₃ (Optima grade, Fisher Scientific) and H₂O₂ (Sigma-Aldrich) in a BERGHOF Speedwave Four microwave digestion system. Calibration of the apparatus was achieved using standards (1000 mg/L, SIGMA-ALDRICH) for each of the analyzed elements.

Procedural Blanks and reference materials were run (*NIST Standard Reference Materials 2583- Trace Elements in Indoor Dust*) to ensure precision and accuracy of all analyses.

2.5. Statistical analysis

Statistical analysis was performed in the Minitab software package (V.16) at a significance level $\alpha=0.05$.

RESULTS AND DISCUSSION

The metals levels registered in each household are depicted in Table 1. There is no legislation regulating maximum permissible levels for any of the studied metals in house dust. Since a threshold was needed in order to characterize risk of exposure, a comparison with "Portaria N° 1450/2007" which regulates the maximum metal contents in dredged sediment that can be safely deposited in soils was used. Despite being originally destined to evaluate risk from a different source, the basic premise is the same: it represents an action threshold that when transcended presents a high probability that biota and humans will suffer toxic effects, since house dust can enter the human body through dermal contact, inhalation, but mostly through ingestion (e.g., dust settled into food items, hand-to-mouth contact) [3, 13]. Estimation of dust daily intakes varies according to authors with some indicating daily ingestions as high as 100 mg for adults and 200 mg for children [13].

Table 1. Maximum, Minimum, geometric average and median values for the metals analyzed ($\mu\text{g/g}$). The values determined as threshold action level by *Portaria N° 1450/2007* are indicated.

	Hg	As	Cd	Pb
Max.	1.08	273.73	1.14	23.26
Min.	0.14	38.72	0.36	0.44

Geometric Mean	0.27	117.81	0.56	3.78
Median	0.23	127.76	0.52	7.54
Portaria N° 1450/2007	0.5	20	1	50

Table 1 presents the Maximum, Minimum, geometric average and median values for the metals analyzed. When compared with "Portaria N° 1450/2007", arsenic values were above the threshold at all houses. Only house C registered any other transgression, specifically for Hg and Cd. No house registered Pb levels above the legal limit. House C differs from the other by being in a "very exposed zone" (meaning a building for which there are no obstacles that attenuate wind, according to the "RCCTE, Decreto-Lei n.º 40/90") in the upper reaches of the city. Since there are frequent forest fires in the area this house can be exposed to higher rates of metal rich particles settlement, thus justifying the higher concentrations measured in the collected dust.

Table 2. Spearman correlation coefficients between construction year and metals analyzed.

	Const. Year	Hg	As	Cd	Pb
Constr. Year	1.00				
Hg	-0.11	1.00			
As	0.64	0.21	1.00		
Cd	-0.24	0.88	-0.14	1.00	
Pb	-0.38	0.90	-0.19	0.88	1.00

The presence of arsenic in the environment can result from natural and anthropogenic activities. Since Covilhã is located in the vicinity of a mining complex (one of the most important anthropogenic activities for this metal), it is quite feasible that wind transport could account for enrichment of soils well above background levels. Even if no favorable winds were registered during the sampling period, the activity of the complex has been maintained for a long period of time (>100 years), which would still justify, if not by direct deposition the integration in house dust of the high environmental levels of arsenic. The concentrations determined in the dust of the surveyed houses can be used to calculate the daily intake of its inhabitants per kilogram of body mass. Calculations indicate exposure levels inferior to the Maximum Tolerable Uptake Intake Level (UL) for arsenic set by JEFCA in 2.1 µg/kg/day [14] (using conservative estimations of adults ingestions of 50 mg of dust/day, children 100 mg of dust/day, and body masses of 65 kg and 20kg, respectively). However it should be noted that IARC considers arsenic a Group I carcinogen, and deems that "exposure should be limited to the lowest possible levels" [15], since even below threshold exposure levels, not conducive to cancer, chronic exposure to low levels has been proven to contribute to diabetes, gastrointestinal disease, encephalopathy and cardiovascular and haematological alterations (e.g., anaemia) [16, 17].

Table 3. Summary of linear regression parameters between year of construction and each analyzed metal.

	Hg	As	Cd	Pb
Adjusted R ²	0.01	0.41	0.06	0.14
F	0.05	2.81	0.26	0.48
p	0.83	0.17	0.64	0.54

Since the number of samples is limited the application of statistical procedures is undermined by power issues, which implies that the obtained results should be analyzed with caution.

Table 2 presents the results for Spearman correlation between the variables and construction year. None of the correlations was significant.

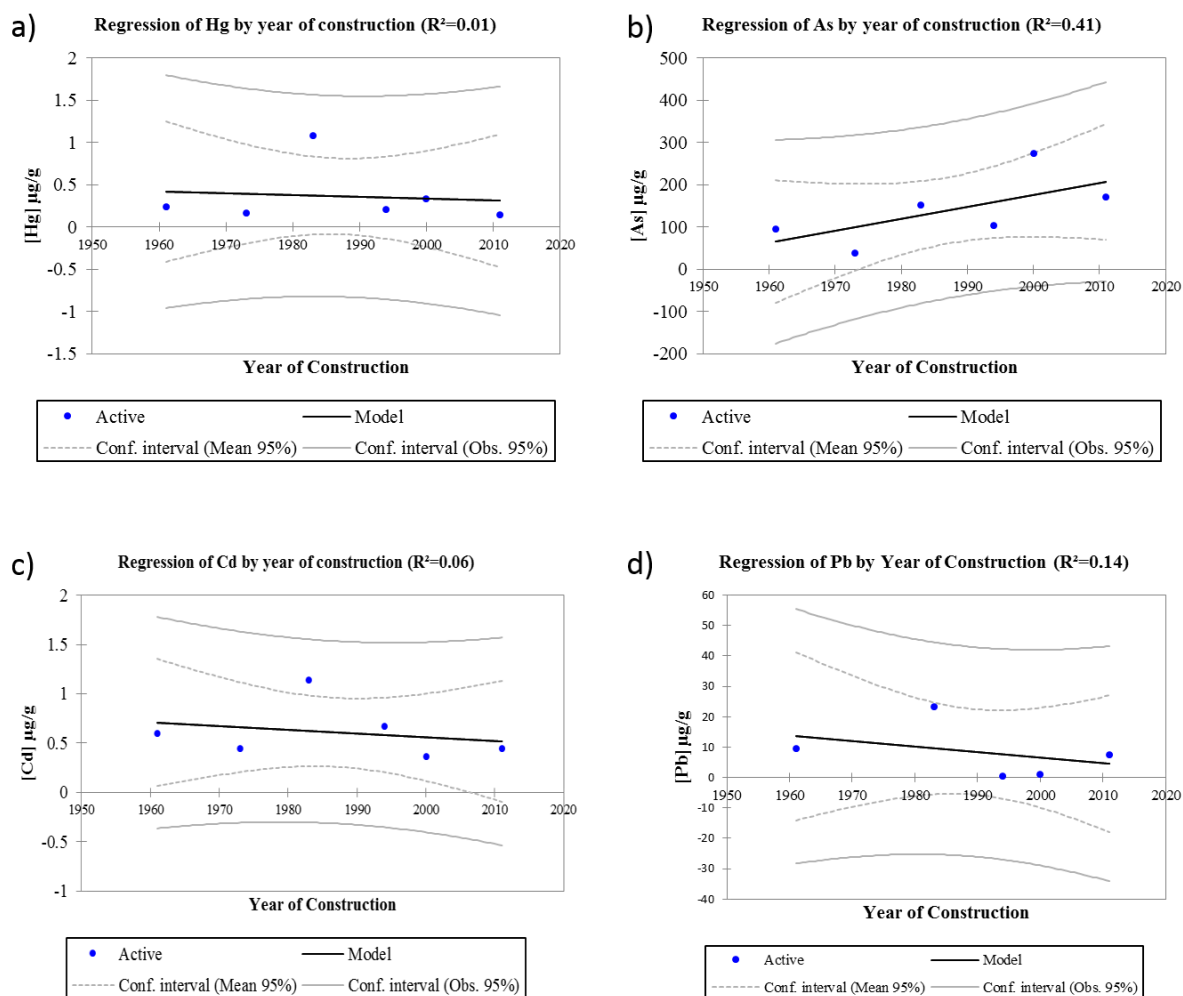


Figure 1. Linear regression between construction year and a) mercury, b) arsenic, c) cadmium, and d) lead.

Observations, regression model and respective 95% confidence intervals are presented.

The application of linear regression (Table 3, and figures 1 to 4) returned models with low goodness of fit after R^2 adjustment to the sample size and values of the F test (Table 3) were all non-significant, allowing us to conclude with a certain amount of caution, that the age of construction of the house is not a good predictor of metal burdens found in that house dust.

CONCLUSIONS

Of all surveyed metals, only arsenic registered above threshold levels in all house dust samples from Covilhã. The calculation of the Maximum Tolerable Uptake Intake Level for the household inhabitants returned arsenic exposure below the value set by JEFCA (2.1 $\mu\text{g}/\text{kg}/\text{day}$). However, since arsenic is considered as a potential promoter of disease in humans, even at non-carcinogenic levels (e.g., cardiovascular disease, diabetes), preventive measures aimed to decrease exposure should be put in place.

Given the small amount of houses surveyed, definite conclusions regarding metal burdens in Covilhã's households, especially when trying to ascertain the influence of construction year, are difficult to set. However, the "6x60x6" pilot project has provided a set of preliminary results, laid good foundations and established sound practices for a much wider study encompassing the entire Beira Interior region.

REFERENCES

- [1] EEA (European Environment Agency), "Environment and human health", Joint EEA-JRC report. Luxembourg: Publications Office of the European Union, pp. 1–112, 2013.
- [2] S. Brasche and W. Bischof, "Daily time spent indoors in German homes - Baseline data for the assessment of indoor exposure of German occupants". *International Journal of Hygiene and Environmental Health*, vol.208, n°4, pp. 247–253, 2005.
- [3] R. M. Maertens, J. Bailey and P.A. White, "The mutagenic hazards of settled house dust: A review". *Mutation Research - Reviews in Mutation Research*, vol. 567, pp. 401–425, 2004.
- [4] USEPA (United States Environmental Protection Agency), "Care for Your Air: A Guide to Indoor Air Quality", 2008. Retrieved from <http://www.epa.gov/iaq/pdfs/careforyourair.pdf> in May, 15th, 2016.
- [5] T. Whitehead, C. Metayer, P. Buffler, and S. M. Rappaport, "Estimating exposures to indoor contaminants using residential dust". *Journal of Exposure Science & Environmental Epidemiology*, vol. 21, n°6, pp. 549–64, 2011.

- [6] A.G. Oomen, P. J.C.M. Janssen, A. Dusseldorp and C.W. Noorlander, "Exposure to chemicals via house dust". *RIVM Report 609021064*, 2008. Downloaded from http://www.rivm.nl/en/Documents_and_publications/Scientific/Reports/2008/april/Exposure_to_chemicals_via_house_dust?sp=cml2bXE9ZmFsc2U7c2VhcmNoYmFzZT02MzM2MDtyaXZtcT1mYWxzZTs=&pagenr=6337 in May, 16th, 2016.
- [7] E. Bernalte, C. Marín Sánchez and E. Pinilla Gil, "Determination of mercury in indoor dust samples by ultrasonic probe microextraction and stripping voltammetry on gold nanoparticles-modified screen-printed electrodes". *Talanta*, vol. 97, pp. 187–92, 2012.
- [8] Y. Ibanez, B. Le Bot and P. Glorennec, "House-dust metal content and bioaccessibility: a Review". *European Journal of Mineralogy*, vol. 22, n° 5, pp. 629–637, 2010.
- [9] P. Le Cann, N. Bonvallot, P. Glorennec, S. Deguen, C. Goeury and B. Le Bot, "Indoor environment and children's health: recent developments in chemical, biological, physical and social aspects". *International Journal of Hygiene and Environmental Health*, vol. 215, n° 1, pp. 1–18, 2011.
- [10] P. E. Rasmussen, C. Levesque, M. Chénier, H. D. Gardner, H. Jones-Otazo and S. Petrovic, "Canadian House Dust Study: population-based concentrations, loads and loading rates of arsenic, cadmium, chromium, copper, nickel, lead, and zinc inside urban homes". *The Science of the Total Environment*, vol. 443, pp. 520–9, 2013.
- [11] INE (Instituto Nacional de Estatística), "Anuário Estatístico da Região Norte 2013", 2013.
- [12] USEPA (United States Environmental Protection Agency), "Method 7473 (SW-846): Mercury in Solids and Solutions by Thermal Decomposition, Amalgamation, and Atomic Absorption Spectrophotometry" Revision 0, 1998.
- [13] S. D. Coelho, A. C. A. Sousa, T. Isobe, S. Tanabe, and A. J. A. Nogueira, "Flame Retardants in Indoor Dust - A Review on the Levels of Polybrominated Diphenyl Ethers and Hexabromocyclododecanes", *Current Organic Chemistry*, vol. 18, n° 17, pp. 2218–2230, 2014.
- [14] JECFA Joint FAO/WHO Expert Committee on Food Additives, "World Health Organization Technical Report 776", 1989.
- [15] IARC (International Agency for Research on Cancer), "IARC Monographs on the evaluation of carcinogenic risks to humans- Arsenic, Metals, Fibres And Dusts", Volume 100C- A review of Human Carcinogens, 2012.
- [16] ATSDR (Agency for Toxic Substances & Disease Registry), "Toxicological Profile for Arsenic", 2007.
- [17] WHO (World Health Organization), Health Research on Arsenic Poisoning, "Gaps in health Research on Arsenic Poisoning", SEA/ACHR/27/10, 2002.