Title

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Authors
Matt, Georg E
Quintana, Penelope JE
Hoh, Eunha
et al.

Publication Date
2021

DOI
10.1016/j.jtemb.2020.126656

Peer reviewed

Journal of Trace Elements in Medicine and Biology 63 (2021) 126656

Contents lists available at ScienceDirect

Journal of Trace Elements in Medicine and Biology

journal homepage: www.elsevier.com/locate/jtemb

# Tobacco smoke is a likely source of lead and cadmium in settled house dust

Georg E. Matt [a,*], Penelope J.E. Quintana [b], Eunha Hoh [b], Nathan G. Dodder [c], E. Melinda Mahabee-Gittens [d], Samuel Padilla [c], Laura Markman [b], Kayo Watanabe [b]

[a] San Diego State University Department of Psychology, San Diego, CA, USA
[b] San Diego State University School of Public Health, San Diego, CA, USA
[c] San Diego State University Research Foundation, San Diego, CA, USA
[d] Cincinnati Children's Hospital Medical Center, University of Cincinnati College of Medicine, Cincinnati, OH, USA

ARTICLE INFO

*Keywords:*
Thirdhand smoke
Secondhand smoke
Environmental tobacco smoke
Lead
Cadmium
Multiunit housing
House dust

ABSTRACT

*Introduction:* Environmental exposure to lead (Pb) and cadmium (Cd) are risk factors for adverse health outcomes in children and adults. This study examined whether thirdhand smoke residue contributes to Pb and Cd in settled house dust.
*Methods:* Participants were 60 multiunit housing residents in San Diego, California. All had indoor smoking bans during the study period, and 55 were nonsmokers. Wipe samples from different surfaces and vacuum floor dust samples were analyzed for nicotine, a marker of thirdhand smoke, and for Pb and Cd using liquid chromatography-triple quadrupole mass spectrometry and inductively coupled plasma-mass spectrometry, respectively.
*Results:* Examined in each sample type separately, Pb and Cd loadings were significantly correlated ($r = 0.73$, vacuum floor dust; 0.52, floor wipes; 0.72, window sill/trough wipes; all $p < 0.0025$). Pb and Cd loadings from different sample types were not correlated (all $p > 0.30$). Nicotine loading in dust was significantly correlated with Pb and Cd loading in dust ($r = 0.49$ for Pb; $r = 0.39$ for Cd, all $p < 0.0025$). Pb and Cd loadings on floor or window surfaces, showed no association with nicotine loading in dust, on floors, or on furniture (all $p < 0.30$).
*Conclusions:* Tobacco smoke is a likely source of Pb and Cd that accumulates in settled house dust in multiunit housing, suggesting that Pb and Cd are constituents of thirdhand smoke that lingers long after smoking has ended.

## 1. Introduction

Environmental exposure to lead (Pb) and cadmium (Cd) are risk factors for adverse neurodevelopmental outcomes and disruptions of the endocrine and immune systems in children and cardiovascular disease incidence and mortality in adults. [1,2] Both metals absorb from the soil into plant tissues and have been found in dry tobacco at concentrations ranging from 0.4 to 12.2 μg/g and 0.7–3.6 μg/g for Pb and Cd, respectively [3,4]. As tobacco is smoked, Pb and Cd become constituents of secondhand tobacco smoke (SHS). Pinto et al. estimated that 46–60 % of Pb and 81–90 % of Cd transfers from dry tobacco to the particulate phase of tobacco smoke [5]. Elevated levels of Pb and Cd are present in the blood and lung tissue samples of smokers, and there is a linear association between SHS exposure and blood Pb and Cd levels in youths and adults [6–10].

As SHS mixes with air and moves throughout an indoor space, its gas- and particulate-phase chemical constituents adsorb to surfaces and accumulate as house dust [11]. Also known as third hand smoke (THS), laboratory and field studies demonstrate that numerous tobacco smoke constituents (e.g., nicotine, tobacco specific nitrosamines, and polycyclic aromatic hydrocarbons) can accumulate and persist in indoor environments and may contribute to the exposure to tobacco smoke toxicants long after SHS has dissipated. Little is known, however, about the contribution of tobacco smoke to heavy metals in THS residue in dust and on surfaces.

Because Cd and Pb can have multiple sources (e.g., leaded paint, contaminated soil, tobacco smoke), their presence in house dust and on surfaces cannot be unequivocally attributed to tobacco smoke. In contrast, nicotine in dust and on surfaces is a specific marker of THS residue. Here we examined the association between nicotine and Pb and Cd in settled house dust and on surfaces collected from vacuum and multiple wipe samples. If tobacco smoke is a significant contributor to

* Corresponding author at: Department of Psychology, San Diego State University, San Diego, CA USA.
*E-mail address:* gmatt@sdsu.edu (G.E. Matt).

https://doi.org/10.1016/j.jtemb.2020.126656
Received 25 July 2020; Received in revised form 15 September 2020; Accepted 23 September 2020
Available online 28 September 2020

Pb and Cd in house dust, comprehensive smoking bans and remediation efforts for THS pollution should become part of integrated, comprehensive Pb and Cd exposure prevention and intervention strategies.

## 2. Methods

### 2.1. Participants

Participants were recruited from a larger observational study of THS pollution in low-income multiunit housing, in which 220 homes were screened for eligibility for a THS cleaning intervention study [12]. Of the 60 homes included in the present study, N = 37 provided environmental samples from the screening phase only, and N = 23 provided samples from the screening and intervention phases. Five participants were smokers, and two reported use inside the apartment during the month prior to screening. To be eligible for the intervention, participants had strict policies against indoor use of tobacco product, and no tobacco or vaping products were used inside the apartment. Participants received gift card incentives for participation. Informed consent was obtained from the San Diego State University Institutional Review Board. Supplemental Table S1 provides additional information about the participants and their apartments.

### 2.2. Environmental measures

#### 2.2.1. Surface wipe samples from floors, window sills or troughs for Pb and Cd

At each home visit, a surface wipe sample (Ghost wipe; SKC-West, Inc., #225-2413) was collected from a 929 $cm^2$ (1 square foot) surface area within a paper template (Ghost wipe; SKC-West, Inc., #225-2416) placed on a carpet-free area of the floor. For homes participating in the cleaning intervention, a second surface wipe sample was collected over a 100 $cm^2$ area within a paper template (5cm × 20cm) (SKC-West, Inc., #225-2415) from a window sill (N = 11) or trough (N = 9).

#### 2.2.2. Surface wipe samples from doors and furniture for nicotine

Two wipe samples, each from a 100 $cm^2$ area within a template, and a field blank were collected at each home visit following the protocol detailed in Quintana et al. [13] Briefly put, prescreened cotton rounds were wetted with 2 mL of 0.1 % ascorbic acid and wiped over vertical (e.g., door panel) and horizontal surfaces (e.g., underneath a table, shelf, or desk). Field blank nicotine levels were subtracted from the corresponding surface sample level (field blank Geo Mean =0.62 ng/wipe), and vertical and horizontal wipe samples were averaged to yield one nicotine surface wipe measure per visit.

#### 2.2.3. Settled floor dust vacuum samples for nicotine, lead, and cadmium

A vacuum house dust sample was collected at each home visit before and after the cleaning intervention from a 1 $m^2$ area (or from a larger area if needed) with a high-volume-small surface-sampler (HVS4, CS3, Venice, Florida, USA) cyclone vacuum into a Teflon bottle.

### 2.3. Analytical methods

#### 2.3.1. Pb and Cd

Surface wipe and house dust samples (100 mg) were digested with trace metal grade concentrated (16 M) nitric acid solution. Levels of Pb and Cd were measured using an Agilent 7900 inductively coupled plasma-mass spectrometer (ICP-MS) (Santa Clara, CA). Calibration solutions were prepared in 2% nitric acid from a multi-element stock solution (Inorganic Ventures, Christiansburg, VA) and an internal standard solution (Spex CertiPrep, Metuchen, NJ). NIST Standard Reference Material 1640a (Trace elements in Natural Water) was used to verify the accuracy of the calibration curves for Pb (91.3 % accuracy) and Cd (102.4 % accuracy). Limits of quantitation (LOQ) for Pb and Cd loadings were $\leq$0.050 μg/$m^2$ and $\leq$0.025 μg/$m^2$, respectively.

#### 2.3.2. Nicotine

The nicotine results described here are a subset of those described in Matt et al. [14], which includes descriptions of the sample preparation for surface wipe nicotine, sample preparation for dust nicotine, and the isotope-dilution liquid chromatography tandem mass spectrometry LC–MS/MS analysis and associated quality control criteria. For surface wipes, the LOQ was 0.30 ng/sample and the estimated method detection limit (MDL) was 0.19 ng/sample, corresponding to an LOQ of 30 ng/$m^2$ and MDL of 19 ng/m [2]. For dust, the LOQ was 0.20 ng/sample and the estimated MDL was 0.13 ng/sample, corresponding to an LOQ of 4 ng/g dust and an MDL of 2.6 ng/g dust.

### 2.4. Statistical analyses

Logarithmic transformations were applied to all Pb, Cd, and nicotine measures to control for positive skew and heterogeneous residual variances. Distributional characteristics are reported in terms of geometric means and their 95 % confidence intervals as well as Tukey's five-number summary statistics [15]. Bivariate associations between Pb, Cd, and nicotine from different sample types using Spearman correlations ($r_S$) showed equivalent results to Pearson correlations ($r_P$). Means from different sample types were compared using paired t-tests. The Type I error rate was set at 5 % with Bonferroni correction. Statistical analyses were conducted using Stata version 16.

## 3. Results

### 3.1. Nicotine in dust and on surfaces

Nicotine was detected in all samples with surface nicotine loadings being on average 3.5 times higher than nicotine dust loadings (Geo Means: 6.3 μg/$m^2$ vs 1.9 μg/$m^2$; see Table 1 for additional information). In homes where surface and dust samples were collected, nicotine loadings were significantly higher on surfaces than in dust ($t(51) = 12.29$, $p < 0.001$).

### 3.2. Lead and cadmium in floor dust and on surfaces

Pb was detected in all vacuum dust and surface wipe samples. In contrast, Cd was detected in 100 % of vacuum dust, 46 % of floor wipes, and 87 % of window wipe samples. Loadings for both metals were highest from window samples (Pb: 82.8 μg/$m^2$, Cd: 5.0 μg/$m^2$), followed by floor vacuum dust (PB: 31.4 μg/$m^2$, Cd: 1.6 μg/$m^2$), and floor wipe samples (Pb: 12.8 μg/$m^2$, Cd: 0.7 μg/$m^2$). Based on homes with floor dust and window surface samples, Pb and Cd loadings were significantly higher on windows compared to vacuumed floor dust (Pb: $t(43) = 6.12$, $p < 0.001$; Cd: $t(43) = 2.50$, $p < 0.022$). Table 1 provides additional information.

### 3.3. Associations between nicotine, lead, and cadmium

#### 3.3.1. Pb and Cd

Pb and Cd loadings were significantly correlated for vacuum dust samples ($r_s = 0.73$; Bonferroni $p < 0.001$), floor surface wipes ($r_s = 0.52$; Bonferroni $p < 0.001$) and window sill/trough wipes ($r_s = 0.72$; Bonferroni $p < 0.001$). In contrast, Pb levels on floors surface and window wipes ($r_s = 0.17$) and Cd levels on floors surface and window wipes ($r_s$ =-.0.02) were not significantly correlated (all Bonferroni $p > 0.30$). Similarly, Pb and Cd levels were not associated when measured on floor and window surface wipe (floor Pb – window Cd: $r_s = 0.12$; window Pb – floor Cd: $r_s$ =-0.02; all Bonferroni $p > 0.30$).

#### 3.3.2. Nicotine with Pb and Cd

Vacuum dust nicotine loading was significantly associated with dust Pb ($r_s = 0.49$; Bonferroni $p = 0.0012$) and dust Cd ($r_s = 0.39$; Bonferroni $p = 0.0382$) loadings. Fig. 1 illustrates the association between dust

**Table 1**
Descriptive statistics of nicotine, lead (Pb), and cadmium (Cd) loadings (μg/m$^2$).

| | N-Samples | N-Homes | GeoM | 95 % CI | <LOQ | Min | Q1 | Mdn | Q3 | Max |
|---|---|---|---|---|---|---|---|---|---|---|
| **Loading** | | | | | | | | | | |
| Dust Vacuum | | | | | | | | | | |
| Floor | | | | | | | | | | |
| Nicotine | 63 | 24 | 1.86 | [1.27;2.61] | 0 % | 0.04 | 0.32 | 1.27 | 3.91 | 59.41 |
| Pb | 63 | 24 | 31.35 | [24.07;40.73] | 0 % | 2.00 | 18.30 | 24.70 | 61.60 | 716.40 |
| Cd | 63 | 24 | 1.55 | [1.17;2.01] | 0 % | 0.10 | 0.60 | 1.00 | 2.40 | 25.10 |
| Surface Wipe | | | | | | | | | | |
| Door, Furniture | | | | | | | | | | |
| Nicotine | 100 | 60 | 6.30 | [4.41;8.85] | 0 % | 0.04 | 1.27 | 4.68 | 18.15 | 613.88 |
| Floor | | | | | | | | | | |
| Pb | 100 | 60 | 12.77 | [10.20;16.92] | 0 % | 1.08 | 5.38 | 11.30 | 26.91 | 342.29 |
| Cd | 100 | 60 | 0.70 | [0.42;1.03] | 54 % | 0 | 0 | 0 | 1.08 | 916.01 |
| Window | | | | | | | | | | |
| Pb | 60 | 21 | 82.81 | [52.92;129.28] | 0 % | 6.46 | 25.83 | 65.12 | 194.83 | 43294.56 |
| Cd | 60 | 21 | 4.98 | [2.93;8.09] | 13 % | 0 | 1.08 | 3.23 | 1.76 | 1031.18 |
| **Concentration** | | | | | | | | | | |
| Dust Vacuum | | | | | | | | | | |
| Floor | | | | | | | | | | |
| Nicotine | 63 | 24 | 1.63 | [1.21;2.13] | 0 % | 0.03 | 0.45 | 1.20 | 2.88 | 12.69 |
| Pb | 63 | 24 | 29.81 | [24.45;36.34] | 0 % | 7.03 | 20.79 | 27.25 | 34.63 | 1852.68 |
| Cd | 63 | 24 | 1.22 | [1.00;1.47] | 0 % | 0.25 | 0.76 | 1.13 | 1.91 | 7.45 |

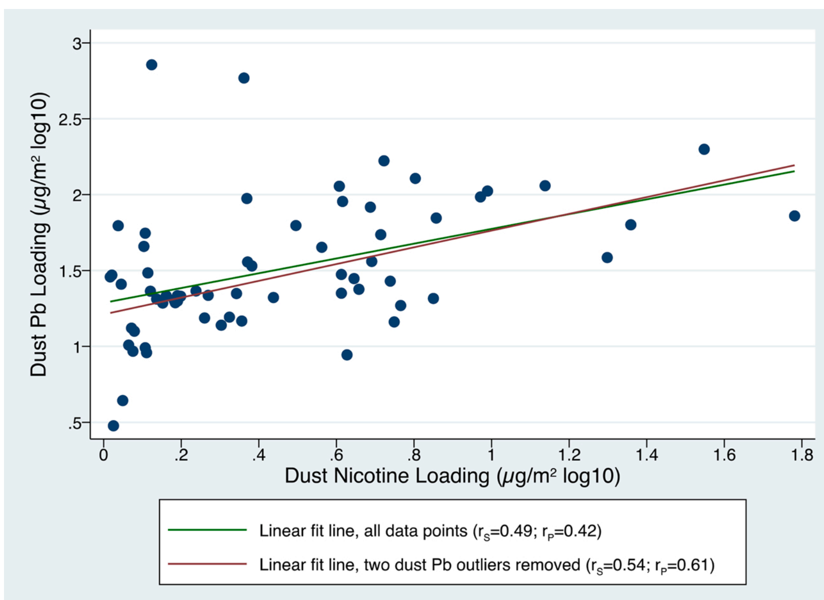


**Fig. 1.** Scatterplot of the association between lead (Pb) and nicotine loadings from settled house dust vacuum samples (N = 63).
Note: $r_S$: Spearman rank-order correlation; $r_P$: Pearson product moment correlation

nicotine and dust Pb loadings, and reveals the presence of two influence points (Cook's distance: 0.27, 0.58). Removing these influence points increased the correlation to $r_s = 0.54$ (Bonferroni $p < 0.0025$). Vacuum dust nicotine was not significantly associated with Pb and Cd levels from surface wipes from floors or windows (Bonferroni $p > 0.30$). Similarly, surface nicotine levels from floor wipes were not associated with Pb and Cd measured in vacuum dust, on floors, and on windows.

## 4. Discussion

This study showed that tobacco smoke is a likely source of Pb and Cd in vacuum house dust found in multiunit housing homes. Together with nicotine, polycyclic aromatic hydrocarbons, and tobacco specific nitrosamines, Pb and Cd should be included as constituents of THS that accumulate and persist in house dust, and considered in future models of health risks associated with THS.

Two previous studies have examined the associations between Pb, Cd, and nicotine in dust [7,8]. Unlike our study, these studies included homes of both indoor smokers and nonsmokers and used dust samples collected from home vacuum cleaners. Our findings are based solely on nonsmokers' homes and dust collected via a cyclone-equipped HVS-4 sampler, collected over a defined area. Willer et al.'s 1993 study reported pollutant concentrations in the subgroup of nonsmokers' homes 6–15 times higher than those in our study (e.g., nicotine: 18 vs 1.2 μg/g; Pb 190 and 160 vs 27 μg/g) and reported no significant association between heavy metals and nicotine concentrations. Willer et al.'s 2005 study reported pollutant levels for Pb and Cd in their combined smoker and nonsmoker sample similar to ours (Pb and Cd: 35 and 1.1 μg/g in fine dust; 23 and 0.8 μg/g in coarse dust) and nicotine levels that were 60 times higher than in our study. They report an association between nicotine and Pb in fine dust, but no association in coarse dust or for Cd. This pattern is consistent with other research on the efficiency of vacuum surface samplers and reinforces that future research on Pb and Cd in dust related to THS should rely on cyclone-equipped dust samplers to efficiently collect fine dust [16,17].

In our data, nicotine vacuum dust and surface wipe loadings were not associated with Pb and Cd levels in surface wipes from floors and windows. This could be due to a greater spatial variability of pollutants measured on different surfaces compared to more homogeneously distributed settled house dust. Because Pb and Cd loadings collected from different sample types were not associated with each other, it is important to rely on multiple sample types when evaluating homes for Pb and Cd pollution.

The observed levels were lower than the current HUD and proposed EPA dust-lead hazard standard value of 10 μg/ft$^2$ for settled dust on floors (108 μg/m$^2$), yet still represents a potentially important source of lead, especially for young children in the home [18]. Considering an average one-bedroom apartment of 700 sq ft (65m$^2$), the total amount of Pb in settled house dust in such an apartment would be approximately 2000 μg. That is, house dust represents a significant potential reservoir of tobacco-related Pb pollution that persists long after smoking bans have been implemented.

### 4.1. Limitations

Our recruitment strategy relied on volunteers and is likely to have created a restricted range with respect to THS pollution because homes with low levels of surface nicotine and homes with active indoor smoking were ineligible for the cleaning intervention phase. Surface wipe samples for Pb and Cd (floor, window sill, trough) were not taken from areas side-by-side with the nicotine wipes (doors, furniture). Restricted range and sampling from different surfaces could have attenuated the observed correlations.

### 4.2. Implications

These results have concerning implications for residents of low-income multiunit housing who are most susceptible to the deleterious effects of heavy metals in dust [19]. THS residue in dust presents another source of Pb and Cd exposure in young children, contributing to the development of adverse neurobehavioral conditions [1]. Bio-accumulation and reduced elimination of heavy metals has been observed in older adults, and exposure to Pb and Cd from THS may contribute to cardiovascular disease, renal disease and osteoporosis [2]. Finally, this study suggests that comprehensive healthy homes programs should include indoor smoking bans to prevent the accumulation of THS and remediation efforts for reduce exposure to toxicants in heavily THS-polluted homes.

## CRediT authorship contribution statement

**Georg E. Matt:** Conceptualization, Methodology, Validation, Formal analysis, Writing - original draft, Writing - review & editing, Visualization, Supervision, Funding acquisition. **Penelope J.E. Quintana:** Conceptualization, Methodology, Validation, Writing - review & editing, Supervision, Funding acquisition. **Eunha Hoh:** Conceptualization, Methodology, Investigation, Resources, Data curation, Writing - review & editing, Supervision. **Nathan G. Dodder:** Methodology, Validation, Investigation, Resources, Data curation, Writing - review & editing, Supervision. **E. Melinda Mahabee-Gittens:** Conceptualization, Writing - original draft, Writing - review & editing. **Samuel Padilla:** Investigation, Data curation, Writing - review & editing. **Laura Markman:** Investigation, Writing - review & editing. **Kayo Watanabe:** Investigation, Writing - review & editing.

## Declaration of Competing Interest

The authors declare no competing interests.

## Acknowledgements

The authors express their gratitude to United Women of East Africa including Sahra Abdi and Bethlehem Degu for their collaboration, to Miriam Adam, Korryn Brackin, Brittney Carroll, Elizabeth Davies, Mayra Duran, Nicolette Dux, Irma Hernandez, Lupita Lombardo, Olga Vergara Lozano, Laura Markman, Maria Martinez, Viridiana Mendoza, Kafisa Mohamed, Dr. Ana M. Navarro, Taylor Shrum, and Faiza Warsame for assisting with participant recruitment and data collection, and to Christine Batikian, Linda Chu, Dana Datuin, Laura Markman, and Mansi Vyas for samples preparation. The authors also thank Joy M. Zakarian for her invaluable contributions as project coordinator for this study and to Peter Ashley and Eugene Pinzer for their comments on an earlier version of this manuscript.

## Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.jtemb.2020.126656.

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