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International Analysis of Sources and Human Health Risk Associated with Trace Metal Contaminants in Residential Indoor Dust

Cynthia Faye Isley,* Kara L. Fry, Xiaochi Liu, Gabriel Michael Filippelli, Jane A. Entwistle, Adam P. Martin, Melanie Kah, Diana Meza-Figueroa, John T. Shukle, Khadija Jabeen, Abimbola O. Famuyiwa, Liqin Wu, Neda Sharifi-Soltani, Israel N. Y. Doyi, Ariadne Argyraki, Kin Fai Ho, Chenyin Dong, Peggy Gunkel-Grillon, C. Marjorie Aelion, and Mark Patrick Taylor

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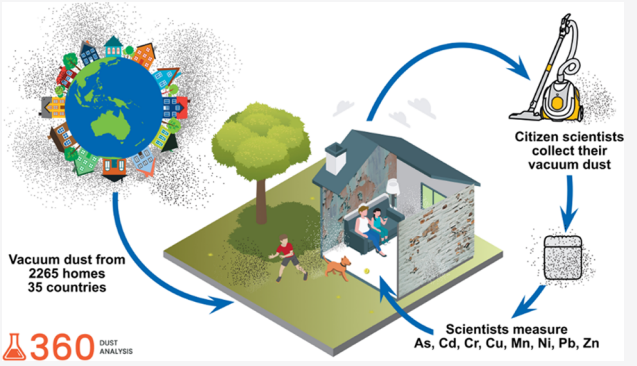
ABSTRACT: People spend increasing amounts of time at home, yet the indoor home environment remains understudied in terms of potential exposure to toxic trace metals. We evaluated trace metal (and metalloid) concentrations (As, Cu, Cr, Mn, Ni, Pb, and Zn) and health risks in indoor dust from homes from 35 countries, along with a suite of potentially contributory residential characteristics. The objective was to determine trace metal source inputs and home environment conditions associated with increasing exposure risk across a range of international communities. For all countries, enrichments compared to global crustal values were Zn > Pb > Cu > As > Cr > Ni; with the greatest health risk from Cr, followed by As > Pb > Mn > Cu > Ni > Zn. Three main indoor dust sources were identified, with a Pb−Zn−As factor related to legacy Pb sources, a Zn−Cu factor reflecting building materials, and a Mn factor indicative of natural soil sources. Increasing home age was associated with greater Pb and As concentrations (5.0 and 0.48 mg/kg per year of home age, respectively), as were peeling paint and garden access. Therefore, these factors form important considerations for the development of evidence-based management strategies to reduce potential risks posed by indoor house dust. Recent findings indicate neurocognitive effects from low concentrations of metal exposures; hence, an understanding of the home exposome is vital.



KEYWORDS: *dust, trace metals, homes, human health risk, lead, sources, enrichment, modeling*

1. INTRODUCTION

Given the amount of time people spend indoors, residential environments are perhaps the most important, yet understudied environments with respect to human exposure to contaminants. Globally, people spend up to 90% of their time indoors.[1] This increased during COVID-19 lockdowns,[2] with children spending on average 17% more time indoors, enhancing potential indoor exposure pathways. Infants, in particular, spend most of their time at home and indoors,[3] and their developing bodies are also more sensitive to trace metal exposures,[3,4] as are fetuses.[5] Due to this evolving dependency on the indoors, identifying, characterizing, and mitigating the risk of household contaminants at an international scale is of high importance. Health risks posed to residents from exposure to potentially contaminant-laden dust[6−11] has been noted, however, the international perspective on the contaminants of common concern to all countries remains relatively unexplored.

Minimizing elevated concentrations of potentially toxic trace metals and metalloids, hereafter referred to as "trace metals", in the home environment remains a persistent issue and should be targeted as a high priority given that exposure can lead to negative lifelong impacts.[12] Children with higher lead (Pb) exposures and higher blood Pb concentrations are unlikely to gain the same cognitive abilities in adult life as those less exposed.[12] Exposure to trace metals is also considered an emerging risk factor for neurodegeneration and neurotoxicity.[13] Increased levels of trace metals including manganese (Mn), copper (Cu),[14] and zinc (Zn)[15] have been associated with amyotrophic lateral sclerosis; Mn with Parkinson's disease[16] and Pb with adult cognitive decline[17] and Alzheimer's disease,[12] even for antenatal Pb exposure.[18] Thus, the identification of elevated trace metals in the

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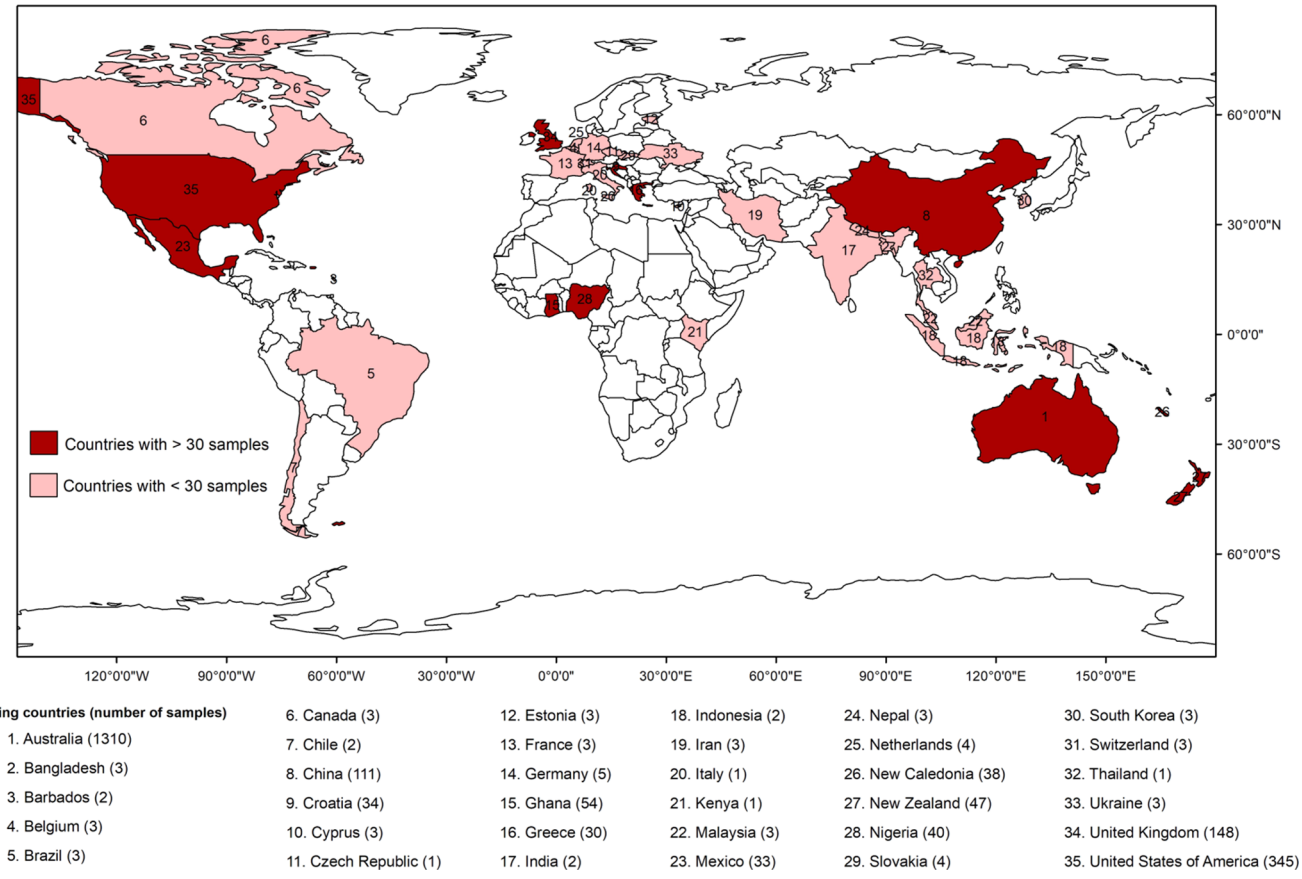

Figure 1. International dust sampling locations and the number of samples from each location. Primary countries (n > 30 samples) and secondary countries (n < 30 samples) are shaded dark red and pale red, respectively. The figure was prepared using ArcGIS 10.8.

predominant environment of occupation (i.e., homes), is a cause of action to control, reduce, and mitigate exposures over a lifetime.

Most indoor dust studies have been small-scale, focussing on a locality already known to be contaminated,[19,20] a single city,[8,21,22] or small sample numbers (10 preschools[9] and 16−24 homes[7,23]). While broadscale studies have been conducted in single countries such as the United Kingdom (n = 5228 samples analyzed for Pb in predominately metal/mining communities[24]); Canada (n = 1025[25]); Germany (3282 samples over 7 years[26]) and China (n = 122,[27] 381 households[28]), these have been researcher-led as compared to community science. Shi and Wang[11] compared international indoor dust studies, yet their analysis was limited due to different methodologies and analytes between individual reports.

The DustSafe program, also known as "360 Dust Analysis," is the first international study to apply a standardized method to dust collection, analysis, source identification, and health risk calculation across 2235 indoor dust samples from 35 countries. DustSafe is also unique in its collection of a range of metadata on home characteristics, enriching understanding of the human−environment nexus, and allowing detailed investigation of the characteristics that increase health risks from potentially toxic trace metals.

Although the general public may be concerned about trace metal hazards in their home, few know how to quantify and interpret this risk or to access laboratory services. The DustSafe community science program aims to close that gap by providing anyone, internationally, with rapid, low/no-cost testing for trace metals in household dust. Participants follow simple instructions on collecting and sending their dust, along with an online questionnaire, which records demographic and household information (e.g., age of home, home construction material, etc.). On receipt of their data, participants are provided guidance on trace metal concentrations specific to their living environment, associated health risks, and intervention strategies to reduce their potential exposure. The DustSafe project has yielded a valuable data set for researchers, providing insights into the extent and pattern of excess levels of trace metals (above typical background levels) that are anthropogenically derived in residential environments at an international scale. Specifically, this study examines the following research questions: (1) How do household dust trace metals and modeled health risks vary across countries? (2) What is the relevance of home characteristics and their association to increased trace metal concentrations in dust? (3) Can we identify a common international trace metal signature in residential indoor dust?

2. MATERIALS AND METHODS

DustSafe was advertised via social media, radio, open days, and e-mails to invite participants to send vacuum dust samples to participating international universities. A total of 2265 samples from 35 countries (Figure 1, Section 2.1) were collected, since 2016, and analyzed using a standardized method. The majority (75%, n = 1703) of samples were collected across 11 countries (Figure 1), for which a more detailed analysis is provided herein.

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2.1. Sample Collection and Analysis. The DustSafe project protocols were subject to ethical review and approval via Macquarie University, Australia (project #2446); Indiana University, USA (project #1810831960); and Northumbria University, the U.K. (project #2598). Participants completed an online survey (https://www.360dustanalysis.com/dust/get-started) and dispensed the contents of their vacuum cleaner into a clean, sealable polyethylene bag. While an increasing number of vacuum cleaners now have bagless cylinders, we did not test for potential trace metal contamination from vacuum bags or from the cylinders themselves. This is a limitation of the current study, however, given the magnitude of most trace metal concentrations in the dust samples, any metal addition from the vacuum bag or cylinder was considered negligible. In countries where vacuuming is uncommon, collecting material by sweeping was advised. Regardless of how dust samples were collected they will hereafter be referred to as 'vacuum dust'. Samples were posted at the participant's expense to the nearest collection location.

Samples were sieved to 250 μm using either a stainless-steel sieve or single-use polypropylene mesh. Samples were analyzed for eight trace metals: arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), manganese (Mn), nickel (Ni), lead (Pb), and zinc (Zn), with the exception of 59 of 111 samples from China, which were assessed for As, Cd, Cr, Cu, Pb, and Zn. These eight trace metals were selected as they are known to occur in potentially toxic concentrations in residential environments[8,10,29−32] and to allow comparison with other international dust studies.[10,11,24,26−28] Trace metal concentrations were determined primarily using X-ray fluorescence spectrometry (portable (pXRF) and energy-dispersive (ED-XRF)) techniques. A small number of samples (n = 59) from China were analyzed by inductively coupled plasma atomic emission spectrometry (ICP-AES). Analysis of additional trace metals, such as mercury (Hg) and aluminium (Al) would have been beneficial, however was not achievable due to pXRF method and analytical limitations.[33−35] Specific techniques used in each analysis location, instrumental and analytical limits of detection (LoD), and quality control measures are provided in Supporting Information 1.0. Previous research[36] has established that XRF and ICP-AES are considered acceptable for comparison in this context.

At the commencement of the DustSafe program, guidelines for the assessment of relative bioavailability and bioaccessibility[37] required the 250 μm fraction soil/dust size fraction to be used, in alignment with exposure guidelines for young children.[38] More recently, the US EPA[39] modified this advice, recommending the 150 μm fraction be used for assessment of Pb ingestion. Doyi,[40] however, determined no significant difference between Pb bioaccessibility or absolute bioavailability when using the 150−250 μm fraction compared to the 90−150 μm fraction of a subset of indoor DustSafe dust samples. A small number of samples from China (27), representing 1% of the total international sample numbers, and 24% of Chinese samples, were sieved to 150 μm, due to the availability of equipment at the participating university. Although previous studies have shown that some trace metals may be more concentrated in the 150 μm fraction,[40] the inclusion of these samples is unlikely to significantly affect the results.[40] Analytical data from the 27 samples from China that were sieved to 150 μm were combined with the other samples from China, which were sieved to 250 μm, and analyzed by pXRF at Macquarie University, Australia. The combined data set (n = 111 Chinese samples) has been considered for all further calculations.

Following analysis, participants were provided with a report outlining trace metal concentrations in their dust sample alongside comparative average values for their region and/or regionally relevant soil guidelines (Supporting Information Figure 2.1). Soil guideline values were applied as benchmark values because of the absence of the corresponding indoor dust measures. Data were then made publicly available on the international mapping platform "Map My Environment"[41] (www.mapmyenvironment.com), manipulated with a "double-jitter" before display to prevent location identification at a household level.

For concentrations below the LoD (Supporting Information Table 1.2), a value of half the LoD, varying for each instrument, was applied for calculations. For Cd, only 15% of samples reported concentrations above the LoD; hence, Cd was removed from further calculations. Anomalous values were identified (Supporting Information Table 1.2) and removed (n = 30), leaving 2235 international vacuum dust samples.

2.2. Metadata Analyses. Metadata were provided by participants via an online survey (https://www.360dustanalysis.com/dust/get-started). Simplified surveys were used in some locations due to translation or logistical requirements, as detailed in Supporting Information 9.0.

Trace metal data were compared using ANOVA[42] in Minitab 18 and, as appropriate, Tukey−Kramer multiple comparison tests to assess homes with the presence versus absence of pets, peeling paint (interior and exterior), smoking, garden access, recent renovation, and hobbies such as shooting or fishing. Impacts of increasing home age, vacuuming frequency, construction material, flooring, heating fuel, and home type were also considered.

2.3. Enrichment Factor. To decipher the influence of anthropogenic activities on indoor dust concentrations, an enrichment factor (EF) was calculated for As, Cr, Cu, Ni, Pb, and Zn in each country. Enrichment factors were calculated by normalizing vacuum dust trace metal ratios to ratios from global crustal values selected from relevant crustal assessments in the literature[43−46] (Supporting Information 3.0). The EF was calculated using eq 1[47]

$$\mathrm{EF} = \frac{\left(\frac{\mathrm{TE}}{\mathrm{RE}}\right)\mathrm{dust}}{\left(\frac{\mathrm{TE}}{\mathrm{RE}}\right)\mathrm{crust}} \quad (1)$$

where TE is the trace metal concentration determined in dust and RE is the reference element. An example of the EF calculation is provided in Supporting Information 3.2. Due to the limited suite of trace metals available in this international study, Mn was selected as the most appropriate reference element due to its correspondence with natural soil sources (Section 3.3.3) and the lowest variability of the trace metals assessed (standard deviation equal to 95% of the mean, compared to 130% to 340% for other trace metals, Supporting Information Table 4.1). This selection also corresponds to previous approaches.[47,48] Enrichment factors were calculated based on the mean and 95% confidence values for each country to express both the average concentrations on a national scale and also the inherent heterogeneity of indoor dust.[49]

2.4. Principal Component Analysis. Variation between trace metal concentrations of individual samples across the countries was examined using Principal Component Analysis

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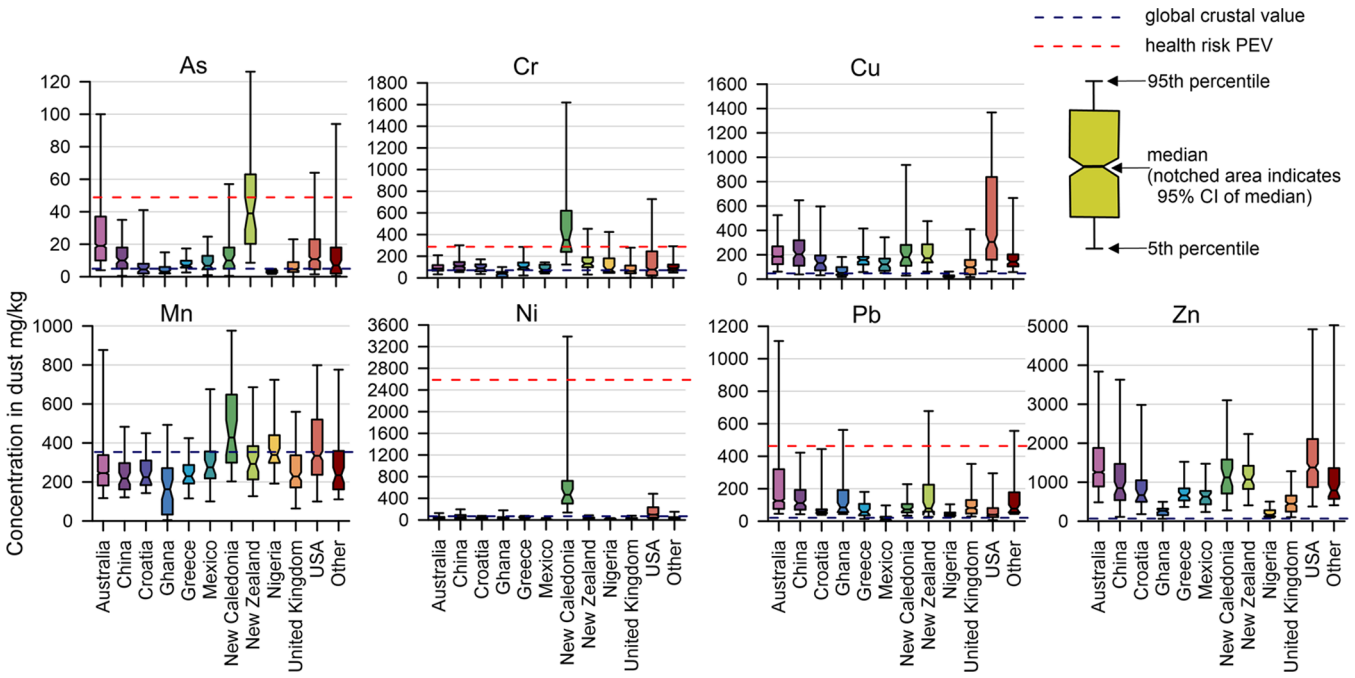

Figure 2. Concentration (mg/kg) of trace metals in dust, global crustal trace metal concentrations (blue dashed line, data in Supporting Information 4.0), and the potential exceedance value (PEV, red dashed line; Supporting Information Table 8.5) at which noncarcinogenic health risk tolerable limits may be exceeded (data in Supporting Information 8.0). Possible exceedance values for Cu (1800 mg/kg), Mn (1600 mg/kg), and Zn (>10 000 mg/kg) are beyond the extent of *y*-axes. Outliers are not shown here but are included in Supporting Information Figure 7.1. Statistics for all countries are shown in Supporting Information Tables 7.1 and 7.2. "Other" countries include Bangladesh, Barbados, Belgium, Brazil, Canada, Chile, Cypress, Czech Republic, Estonia, France, Germany, India, Indonesia, Iran, Italy, Kenya, Malaysia, Nepal, Netherlands, Slovakia, South Korea, Switzerland, Thailand, and Ukraine, detailed in Supporting Information Table 7.2. The figure was prepared using Golden Software Grapher 14.

(PCA). Trace metal concentration data were standardized as part of the PCA method (distribution shown in Supporting Information 4.0). The PCA was undertaken for all 35 countries (Supporting Information 5.0), with the primary 11 countries presented here. The PCA expresses the critical dimensions of the total data set as seven principal components. The first three dimensions explained 67% of data variance. The PCA was completed using FactoMineR[50] and factoextra,[51] detailed in Supporting Information 5.0.

2.5. Positive Matrix Factorization. The data were also analyzed using the United States Environment Protection Agency (US EPA) multivariate factor analysis Positive Matrix Factorization (PMF) 5.0.14 model[52] with the objective of identifying specific indoor dust source factors. The usefulness of PMF lies in its differentiation of sources (factors) that share common elements,[53] which can then be related to emission sources. Methods, uncertainty inputs, and error estimation are detailed in Supporting Information 6.0.

A three-factor model, to align with the PCA, was run initially for the combined international data set and then separately for each country with >30 samples (n = 10). Given that Australian data comprised 57% of the international data set, separate considerations of each country provided a more accurate analysis. Australia, China, Ghana, Greece, Mexico, New Caledonia, New Zealand, the U.K., and the USA returned results within the acceptable error (Supporting Information 6.0); while Croatia and Nigeria did not.

2.6. Human Health Risk Assessment. Modeling was undertaken using standard US EPA[54] methods to quantify the noncarcinogenic and carcinogenic health risks posed to children (<2 years old) from the vacuum dust samples. Data were assessed at the national level, with the upper 95th confidence interval used as the representative value for health risk calculation. This approach was selected to avoid the uncertainty associated with estimating the true average of each trace metal, and the differing sample sizes from each country, as recommended by the US EPA.[55] Noncarcinogenic health risk assessment was undertaken for As, Cr, Cu, Mn, Ni, Pb, and Zn. Given that Cu, Mn, and Zn are not recognized as human carcinogens,[56] carcinogenic (lifetime cancer risk) assessment was evaluated only for As, Cr, Ni, and Pb.

The chronic daily intake (CDI) for ingestion, inhalation, and dermal exposure pathways was estimated using the equations provided in Supporting Information Table 8.1 along with the exposure factors detailed in Supporting Information Table 8.2. Where available, values specific to young children (<2 years old), indoor dust and residential environments were selected. Values provided by the US EPA Regional Screening Level (RSL) calculator[57] and the US EPA Exposure Factors Handbook[58] were prioritized.

Following the calculation of the CDI for each exposure pathway, the exposure risk was determined using the reference dose (RfD; ingestion; noncarcinogenic), reference concentration (RfC; inhalation; noncarcinogenic), dermal reference dose (DRfD; dermal; noncarcinogenic), oral slope factor (OSF; ingestion; carcinogenic), inhalation unit risk (IUR; inhalation; carcinogenic), and dermal slope factor (DSF; dermal; carcinogenic; Supporting Information Table 8.3) using equations provided in Supporting Information Table 8.1. The relevant values were extracted from the US EPA's RSL calculator[57] using the residential exposure scenario. In the absence of appropriate values provided by the RSL, the

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Table 1. Summary of Metadata Statistics between Home Characteristics. Statistically Significant Outcomes Are Indicated by Bold Text. Boxplots and, Where Applicable, Tukey−Kramer Multiple Comparison Test Results, and Regression Plots Are Provided for Each Characteristic in Supporting Information 9.0

	p value for presence versus absence of characteristic or for increase, as specified						
characteristic	As	Cr	Cu	Mn	Ni	Pb	Zn
pets	0.23	0.16	0.11	**0.011**	**0.015**	0.38	**<0.001**
peeling interior paint	**0.001**	0.12	0.62	0.07	0.18	**<0.001**	0.97
peeling exterior paint	**<0.001**	**0.011**	0.23	**<0.001**	**0.005**	**<0.001**	0.71
smoking present	0.64	**0.004**	0.21	**0.003**	0.78	0.67	0.45
garden present	**<0.001**	0.37	0.056	0.098	0.50	**<0.001**	0.074
recently renovated	**0.017**	0.081	0.57	0.40	0.34	0.071	0.963
hobbies involving metal exposure (by type)	0.119	0.66	0.93	0.13	0.61	**0.001**	0.16
increased vacuuming frequency	0.18	0.78	0.35	**0.010**	0.071	0.49	0.44
increasing home age	**<0.0001**	0.19	**0.019**	**<0.0001**	**0.96**	**<0.0001**	**<0.0001**
home construction material	**<0.001**	0.18	0.59	**<0.001**	**0.042**	**<0.001**	**0.005**
heating fuel type	0.084	0.053	**<0.001**	**0.012**	0.29	0.26	**0.016**
floor covering	**<0.001**	**0.016**	0.231	**0.001**	**<0.001**	**<0.001**	0.153
type of home (e.g., detached)	**<0.001**	0.29	**0.001**	**<0.001**	**0.001**	**<0.001**	0.27

following tiered approach was used to identify other suitable exposure risk values: California OEHHA,[59] US EPA[54] Superfund Guidance, published values in Ferreira-Baptista and De Miguel[60] and Doyi et al.,[6] and Wignall et al.'s[61] Conditional Toxicity Value (CTV) predictor (Supporting Information Table 8.3). Given that the CTV is not optimized for assessment of inorganic (e.g., trace metal) contaminants,[61] values from this source were only applied where other suitable literature-based estimates were not available.

The sum of risk estimates for each exposure factor provided the noncarcinogenic hazard quotient (HQ) and carcinogenic target risk (TR). Where HQ or TR values were greater than 1.0 and 1×10^{-6}, the tolerable risk is exceeded and intervention actions should be considered.[54,58]

Determining the CrVI content of environmental samples is complicated as Cr can transition between its trivalent (CrIII) and hexavalent state (CrVI), including during sample collection, storage, and analysis.[62] The data reported in this study are the total measured concentrations and exclude speciation analysis. Subsequently, assessment of Cr in its toxic hexavalent form (CrVI) had to be estimated because suitable factors for RfC, DRfD, OSF, and DSF were only available for CrVI. To avoid over-representation of the total Cr health risk, a conservative ratio based on speciation assessments[63−65] of 0.25:1 CrVI:total Cr was adopted[66] and imputed for RfC, DRfD, OSF, and DSF values. Importantly, the modeling approach used here provides conservative estimates of the health risk from trace metals in vacuum dust, as other exposure mediums, external environmental factors, and sample trace metal bioavailability have not been assessed. Further investigation into these factors should be undertaken in countries where the health risk exceeds tolerable limits.

3. RESULTS AND DISCUSSION

Dust trace metal concentrations and associated metadata were interrogated using a variety of spatial and temporal analyses to establish the factors influencing their concentration, enrichment levels, sources, and risk to human health.

3.1. Analysis of Temporal and Spatial Factors in Household Dust. Concentrations of trace metals in dust varied between locations internationally (Figure 2, Supporting Information Tables 7.1 and 7.2). Statistical comparison (Supporting Information Table 7.3) showed that trace metals were different (ANOVA $p < 0.0001$) between countries for each trace metal. Tukey−Kramer multiple comparison tests show that Australia, the USA, and New Zealand are more significantly different than all other countries in their individual trace metal concentrations in dust (Supporting Information Table 7.3). New Caledonia is distinctive in terms of having higher Cr, Mn, and Ni concentrations; Australia had high Pb, New Zealand had high As, and the USA had high Cu and Zn, relative to other countries in the study.

3.2. Metadata Analyses. Metadata provided by international participants revealed insights into the relevance of home characteristics and their association with increased trace metal concentrations in dust (Table 1).

Our data support those of Taylor et al.[30] that internationally, home age is a significant predictor of As, Cu, Mn, Ni, Pb, and Zn indoor dust concentrations.[30] DustSafe data show that every year since home construction was associated with an increase of 0.48 mg/kg As, 3.0 mg/kg Cu, 4.2 mg/kg Mn, 0.63 mg/kg Ni, 5.0 mg/kg Pb, and 22 mg/kg Zn in dust (Supporting Information Figure 9.10). The presence of a garden and interior and exterior peeling paint were associated with the largest percentage differences in As and Pb concentrations (Table 1). Homes with peeling exterior paint had slightly lower Ni levels (14%) than other homes. This is surprising as Ni is also a common ingredient in paint.[67] In the DustSafe data, these Ni concentration differences are small (14%), compared to As (40%), Mn (33%), and Pb (74%) concentration increases under the same conditions (Supporting Information Figure 9.2). Potential sources of Ni that may confound this result include vehicular emissions[68] or even laboratory use of stainless-steel sieves;[69] however, the sieves used in this study were not found to contribute to Ni above the instrument detection limit.[6] Arsenic concentrations were also 19% higher in recently renovated homes. No further associations with trace metal concentrations were found when considering only recently renovated homes more than 55, 60, or 70 years of age (indicative of the Pb paint era).

Concentrations of As and Pb were higher in detached and semidetached homes (Supporting Information Figures 9.17 and 9.18), likely due to the ingress of outdoor legacy trace metals, which commonly include inputs not only from past leaded gasoline usage and exterior Pb paints[30] but also from treated timbers and pesticides.[30] Lead concentration in

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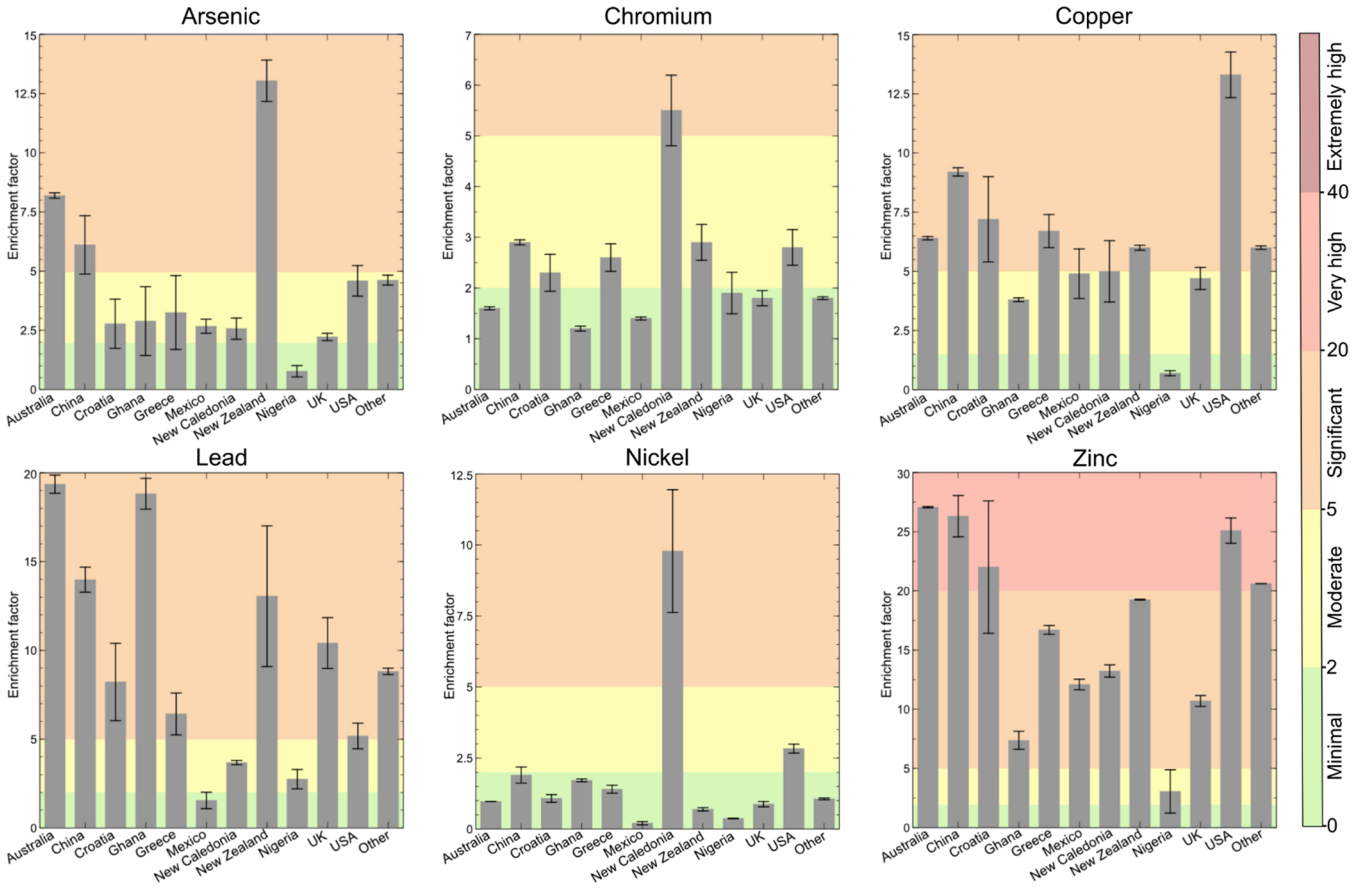

Figure 3. Enrichment factor (EF) calculations for the assessed trace metals in each country (mean, ±95% confidence interval), normalized against the reference element Mn. Shaded categories indicate the level of enrichment, as stipulated in Barbieri,[47] where an EF less than 2 suggests natural conditions and an EF greater than 2 indicates anthropogenic enrichment. The figure was prepared using Golden Software Grapher 14 and Affinity Designer 1.10.4.

detached homes, as with all homes (Table 1), is also significantly ($p < 0.001$) influenced by the age of the home, with Pb concentration in detached homes increasing by 5 mg/kg with every year of age (Supporting Information Figure 9.19). Detached homes also had higher Mn concentrations, presumably due to soil track-in,[70] yet surprisingly Mn concentration differences were not statistically significant ($p > 0.05$) for homes with versus without garden access. Detached homes did not have significantly higher concentrations of Cr, Cu, Ni, and Zn, suggesting that indoor sources (discussed below and in Section 3.3.3) may be more significant than the type of home for these trace metals. The use of oil for heating was associated with higher Cu concentrations in indoor dust, and wood burning with increased Mn concentrations. Higher Mn concentrations (37%) were also associated with indoor smoking, as was Cr (9%), a known component in cigarette smoke.[71]

Homes constructed with metal had the highest Mn concentrations (Supporting Information Figure 9.11), likely since Mn is an additive to steel.[72] Metal, asbestos and timber homes also had higher Pb concentrations, likely due to paints[73] or wood treatments.[74] Timber floors were also associated with higher As and Pb concentrations, although the reasons for this are unclear. Higher Cr and Ni for tile floors may be due to use of metal-rich slags in tile production[75] and similarly for Mn in concrete floors.[76]

Crafts (lead lighting, jewelry making, woodwork, modeling, and mineral collecting) were associated with higher Pb concentrations (Supporting Information Figures 9.7 and 9.8). Previous studies have documented high levels of Pb exposure in shooting-range participants;[77] however, the sample size (n = 13 shooting hobbies) here was insufficient to identify any significant relationship.

Homes with pets had 14% higher Mn concentrations in dust, which is not unexpected as pets track-in natural soil materials and plant matter; however, significant track-in of other trace metals was not observed. Higher Mn concentrations were associated with greater vacuuming frequency, as noted elsewhere for Pb.[78] Pet owners (who have higher Mn levels, Table 1) vacuum more frequently, with 9% more pet owners vacuuming weekly or more compared to those without pets (Supporting Information Figure 9.9). Hence it is probable that pet ownership is confounding this result.

3.3. Source Modeling. Source modeling was completed using EF, PCA, and PMF. The PCA shows that while some source factors are similar across countries, there are also some clear differences identifiable in the PMF modeling.

3.3.1. Enrichment Factor. An enrichment factor (EF) less than 2 indicates natural conditions, while EFs greater than 2 suggest anthropogenic influence.[47] Zinc returned an EF > 2 in all locations, and As, Pb and Cu in 11 of 12 countries (Figure 3, Supporting Information 10.0), demonstrating anthropogenic enrichment of trace metals in dust internationally. Overall,

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enrichment of Zn was the greatest, followed by Pb > Cu > As > Cr > Ni. Regional variations were present, with the greatest enrichment of As (EF = 13.0) in New Zealand, Cr (EF = 5.5) and Ni (EF = 9.8) in New Caledonia, Cu (EF = 13.3) in the USA, and Pb (EF = 19.4 and 18.8) in Australia and Ghana, respectively. These unique differences reflect local anthropogenic activities adjacent to the sampled residential environments, including the production, use, and burning of Cu−Cr−As treated timber in New Zealand[79] and geology and industry in New Caledonia.[80,81]

Cumulative exposure to trace metals has been significantly associated with health impacts such as obesity, hypertension, and type-2 diabetes.[82] Cao[83] reports that cumulative risks for children's trace metal exposures must be considered, rather than simply considering each trace metal individually. Similarly, interactions between mixed agents (including trace metals) may be significant in the development of neurodegenerative disorders such as Parkinson's disease.[84] To develop a comparative enrichment measure for each country, a cumulative enrichment factor (cEF) was calculated using the sum EFs for As, Cr, Cu, Ni, Pb, and Zn. Cumulative enrichment was the greatest in Australia (cEF = 64.7), followed by China (cEF = 61.4), New Zealand (cEF = 56.0), the USA (cEF = 54.8), New Caledonia (cEF = 40.8), Greece (cEF = 38.1), Ghana (cEF = 36.8), the U.K. (cEF = 31.7), Mexico (cEF = 23.9), and Nigeria (cEF = 10.5).

3.3.2. Principal Component Analysis. Correlations are evident in the data (Figure 4) between distinctive Pb and As,

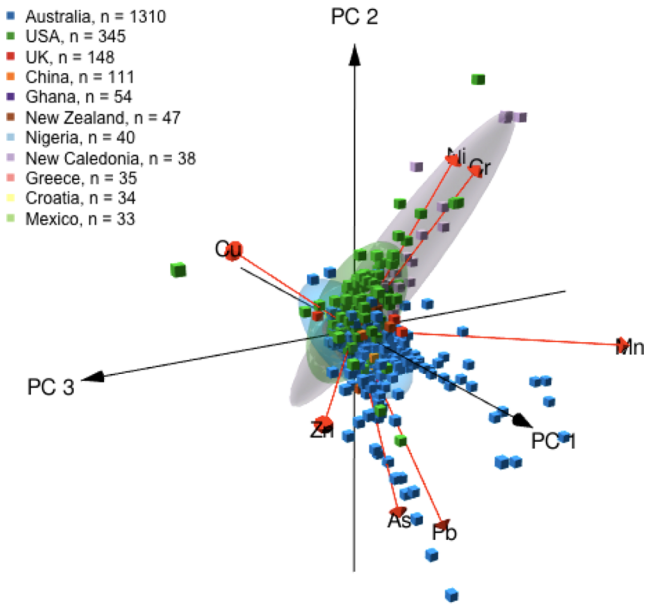

Figure 4. Principal component analysis of international DustSafe samples. Three-dimensional model. The figure was prepared using the pca3d R package.

Ni and Cr, and Mn and Cu components. Zinc is less well-fitted in-between the Pb−As and Cu components (Figure 4). The distinctive clustering evident in data from Australia, the USA, and New Caledonia indicates different underlying dust sources in these locations, as further explored in the PMF below.

3.3.3. Positive Matrix Factorization. Three similar factors: the "Pb−Zn−As" factor, "Zn−Cu" factor, and "Mn" factor (Figure 5) were found consistently in independent PMF model runs across the combined international data set (Supporting

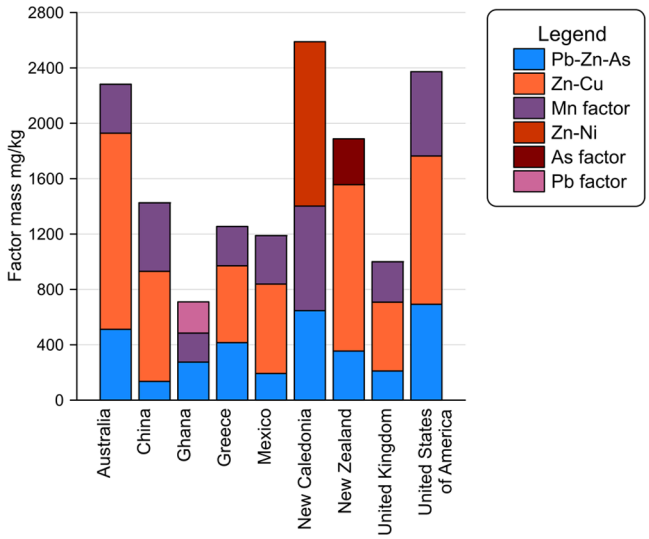

Figure 5. Cumulative mass of factors identified in PMF modeling for each country's household dust. The figure was prepared using Golden Software Grapher 14.

Information Figure 11.1) and separately for six countries: Australia, China, Greece, Mexico, the U.K., and the USA (Supporting Information 11). An additional three countries: Ghana, New Caledonia, and New Zealand, returned two out of three of these factors. The exact ratio of elemental combinations in these factors varied by country (Supporting Information Table 11.1). This approach is not measuring fixed, defined emission sources, but a mixture of several different emission sources[53,85] captured and stored in household dust. The Mn factor, which also contained a Cr component in some countries, most likely represents natural inputs, while the other factors suggest anthropogenic influence. Relevantly, the Mn factor was also distinct from other factors in the PCA (Figure 4).

Like the PCA, PMF groups the anthropogenic As and Pb contaminants. While the PCA (Figure 4) shows Zn to be somewhat poorly fitted in-between the As−Pb and Cu groups, PMF separates out the association of Zn with the Pb−Zn−As factor, as well as with the Zn−Cu factor.

3.3.4. Pb−Zn−As Factor. Over the larger DustSafe data set, Pb, Zn, and As concentrations are enriched significantly compared to crustal values (Figure 3), indicating anthropogenic inputs. Indoor dust Pb, Zn, and As have been associated with residential soils[6] tracked indoors[10] with solder, plastic, metals,[69] industry, and tobacco smoke,[86] although no association with indoor smoking was found here (Table 1). We found increased Pb and As concentrations with garden access (As 76%, Pb 94%), peeling paint (40% As, 60% Pb for interior paint; 40%, 33%, and 74% As, Mn, and Pb for exterior paint, respectively), increasing home age (also for Zn, Supporting Information 9.0), and detached home types, as well as renovation (for As) (Section 3.2). Older housing, potentially containing Pb-based paints, remains a risk factor for increased trace metals in indoor dust, with Pb-based paints still sold in some countries[73] and still present in older homes. In Australia, where Pb contamination is notably enriched in DustSafe samples, widespread Pb contamination has also been found across residential yards and gardens,[30] and indoors,[6] particularly older, inner-city areas affected by legacy Pb from leaded petrol and paint usage. Ethics requirements for

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international data sharing in this study did not allow the transfer of location data for most countries; however, these data were available for Australia and New Caledonia, comprising 38% of the total international sample numbers. In New Caledonia, the distance from the city center of Noumea, the nation's capital, (and from the industrial smelting facility) was significantly correlated with decreased concentrations of Cr, Fe, Mn, Ni, and Zn in dust.[87] This is also evident for dust samples from Australia's largest cities, Sydney, Melbourne, and Brisbane, with concentrations of As, Cr, Cu, Mn, Ni, Pb, and Zn all decreasing with increased distances from the city center (Supporting Information Section 12.0). This is an important consideration for the choice of housing by families with children under 5 years old, as Pb in household dust presents significant potential health risk[88] (Section 3.4).

Mining and industrial activity also impact household Pb exposures;[6,89,90] particularly in low- to middle-income countries.[91] Contamination from past leaded gasoline usage pervades cities internationally;[92,93] elevated concentrations occur across a broad range of environmental and biological media, including soils,[30] dust,[6,10,24,94] lichens,[95] bees,[96] and vegetables.[97]

3.3.5. Zn−Cu Factor. In urban environments, Cu and Zn have been attributed to building materials;[98] including galvanized surfaces, wood preservatives, paints, and metal coatings.[99] Enrichment of Cu and Zn compared to crustal values (Figure 3) indicates an anthropogenic source. Likewise, Zn is often more concentrated in dripline soil samples[30,100,101], beneath house guttering. DustSafe data showed an increased Zn concentration with increased property age (Supporting Information Figure 9.10), also indicating contribution from degradation of building materials; still, no relationship was found between home construction materials and Cu, with Zn only significantly lower in cement versus asbestos homes (Supporting Information Figure 9.12). Copper and Zn are also characteristic of atmospheric traffic emissions, including brake and tire wear,[98,99] which accumulate in homes over time. In the absence of detailed location data for all countries except Australia and New Caledonia, we were unable to determine international impacts from agricultural chemicals, which are a potential source of Cd, Cu, Mn, and Zn[102] on dust trace metal concentrations in homes near farmlands. For New Caledonia, all of the samples were from nearby the capital city, Noumea, and Mn and Zn concentrations in dust decreased with distance from the city,[87] not indicating contamination from farming lands. Further sampling in more rural areas of New Caledonia is needed to determine agricultural impacts. Likewise, in Australia, Cu and Zn decreased significantly with distance from the city center for the three main cities (Sydney, Melbourne, and Brisbane, Supporting Information Section 12.0); Mn also decreased with distance from the city center and only significantly for Melbourne. Additional influences to trace metal concentrations include industrial emissions, forest fires, fireworks; track-in of soils,[103] including those that contain fertilizers, pesticides, and herbicides;[104] batteries and personal care products such as sunscreens.[99]

3.3.6. Mn Factor. Given the lower Mn concentrations in vacuum dust samples compared to those measured outdoors[30] (Figure 3), we considered this Mn factor to largely represent a natural soil source,[105] tracked or blown inside residences, where it accumulates, in agreement with Reis[106] (Portugal) and Yadav[23] (Nepal). This is supported by higher Mn concentrations in detached homes, where the potential for track-in is higher (Supporting Information Figure 9.18) due to outdoor soil areas and shorter track-in distances, compared, for example, to apartments. Pets (Table 1) were also associated with increased Mn concentrations, presumably indicating track-in. Manganese is the eighth most abundant crustal metal[107] and is naturally enriched[108] in soils,[109] particularly from dead plant biomass,[110] and is internationally associated with wildfire activity,[111] sea spray, volcanic activity, and animal wastes.[112] The use of wood fuels for heating and indoor smoking were related to a 37% increase in Mn concentrations in vacuum dust samples (Supporting Information 9.0). While these are anthropogenic inputs, they are consistent with Mn contributions from natural wildfire activity.[111] Previously, industrial sources,[113−115] steel mills, ore crushing and welding,[116] and possibly fuel aditives[117,118] have been implicated in contributing Mn to the local environment. Australian soil Mn loadings,[30] however, show no clear correlation between the Mn concentration and distance from the city center, meaning that industrial and higher vehicle traffic areas were not comparatively enriched.

The Cr−Mn source in New Caledonia likely reflects the natural input from ultramafic soils[81,119] or the widely used Cr-rich slag.[120] Likewise, Greece has natural rock and soil sources of Cr,[121] with industrial sources also contributing. The presence of Cr in the USA samples varied widely across and between cities and potential sources include timber treatments[122] and carpet dyes.[123]

3.4. Additional Factors. Modeling New Zealand data by PMF returned a unique "As factor" not seen elsewhere, mirroring New Zealand's high As enrichment (Figure 3).[124] Rare soils adjacent to geothermal activity and geothermal power exploitation in New Zealand may have very high concentrations of As, with a mean of 895 mg/kg (SD range 135−1470).[125] The production, use, and burning of Cu−Cr−As treated timber in New Zealand is a significant As source,[79,126] with As concentrations being higher (internationally) in homes burning wood than oil for heating (Supporting Information Figure 9.13). Anthropogenic As inputs in New Zealand also include pesticides remaining from urbanization of agricultural lands.[125,127]

New Caledonia's Zn−Ni factor is most likely of industrial origin. Pasquet[128] considered Zn in New Caledonian lichen samples to be of industrial origin due to smelting of Ni, although also present in the ultramafic geology, and hence was surprised not to find a Zn−Ni correlation. Here, with samples mainly sourced from Noumea, situated in close proximity to a Ni smelter, we clearly see this Zn−Ni correlation. Further sampling across New Caledonia is needed to characterize sources away from industrial activity and mining.

The Pb-factor seen in Ghana is likely due to the e-waste recycling that occurs near Accra, the city from which samples were provided. This activity has caused widespread Pb contamination to air and soil.[129]

3.5. Human Health Risk Assessment. For the assessed countries, the noncarcinogenic hazard index (HI) for children (<2 years old) was within tolerable limits (<1.0) for all trace metals and locations, except for As in New Zealand (HI = 1.20) and Cr in New Caledonia (HI = 2.26; Supporting Information 8.4a). Although the noncarcinogenic health risk from Pb was within tolerable limits for all countries assessed, the risk was the greatest in Australia (HI = 0.73) and New Zealand (HI = 0.62). Internationally, Cr presented the highest noncarcinogenic risk, followed by As > Pb > Mn > Cu > Ni >

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Zn (Supporting Information 8.4a). Overall, noncarcinogenic health risk was greatest via the ingestion exposure pathway, returning a median cumulative risk estimate from all trace metals assessed of 0.89 across all locations assessed, followed by dermal and inhalation exposure pathways, with median international cumulative risk values of 0.49 and 0.09, respectively (Supporting Information 8.4b).

Similarly, ingestion posed the greatest carcinogenic risk, followed by dermal and inhalation pathways, returning a median cumulative risk of 6.9×10^{-5}, 1.4×10^{-7}, and 1.1×10^{-6}, respectively (Supporting Information 8.4b). Carcinogenic target risk (TR) can be presented as an acceptable range from 10^{-6}, the upper bound of negligible risk (1×10^{-6}; 1 case per every 1 000 000 people), to the level of unacceptable risk (1×10^{-4}; 1 case per every 10 000 people).[130,131] The upper bound of negligible risk (1×10^{-6}) was exceeded for As, Cr, and Ni in all countries, and in 10 countries for Pb, where only Mexico presented risk estimates within tolerable limits based on calculations at the upper 95th percentile of each data set (Supporting Information 8.4a). The level of unacceptable risk (1×10^{-4}) indicated possible exceedance for As in New Zealand (TR = 1.2×10^{-4}), and Cr and Ni in New Caledonia (Cr TR = 1.7×10^{-4}; Ni TR = 2.3×10^{-4}; Supporting Information Table 8.4a). In locations with documented Cr enrichment, both naturally and via anthropogenic activities like smelting, such as in New Caledonia,[87] precautions should be considered to reduce children's exposure to Cr in indoor dust. Similarly, sources of As exposure in New Zealand may require further investigation and mitigation.

The impact of cumulative trace metal exposure and hazards posed by other indoor contaminants (e.g., microbial exposures, molds, pesticides, and fine particulate matter) have not been accounted for in this model. Additionally, the health risk calculations are approximations, as we cannot account for the complexity of individual living situations, such as household characteristics like the presence of gardens, pets, or renovations (Table 1) at an international scale, nor the individual behaviors that can affect exposure.[132] The upper 95th confidence interval was used for health risk assessment as a conservative measure of risk to the most vulnerable children in each country.

To better quantify risk levels and understand the vacuum dust concentrations at which health risks may be posed,[141] a possible exceedance value (PEV) was calculated based on the parameters applied in the health risk assessment. The PEV is defined as the concentration at which tolerable risk levels for noncarcinogenic risk (>1.0) would be exceeded for a child (<2 years) exposed to trace metals in vacuum dust, which can then be used to estimate probable exceedance.[139,140] The PEVs were most exceeded for As, where 12.3% of international samples, 36.2% of New Zealand samples, and 16.8% of Australian samples were above the PEV of 50 mg/kg (Supporting Information Table 8.5). The vacuum dust Pb PEV was exceeded in 17.3% of Australian samples and 11.6% of samples internationally (Supporting Information Table 8.5). No PEVs were exceeded in vacuum dust samples from Mexico. The PEV calculations rely on generic input parameters that may not capture likely regional trace metal and lifestyle variations.

Data benchmarking is limited by the absence of guidelines or intervention approaches for trace metals in indoor vacuum dust, even though indoor environments and their associated contaminants are becoming an increasing focus of concern.[133−135] The most equivalent comparisons for trace metal contaminants are soil guideline values[136−138] that can vary by orders of magnitude and lack international consensus regarding what is "safe." Subsequently, there is a unique opportunity for future investigations to respond to these assumptions by exploring country-specific housing and behavioral outcomes that may impact trace metal exposure in young children, the age group most at risk. It is paramount that future efforts address this research gap further by exploring a wider range of contaminants and spatial and temporal gradients to better inform decision making, advise preventative measures, and establish data-informed regulatory guidelines for indoor trace metal concentrations. Indeed there has been international interest in setting public health guidelines for chemicals in indoor dust.[141]

Answering the research question, "how do dust trace metals and health risks vary across the world?", trace metal concentrations in indoor dust varied internationally. While Zn was most enriched, it carried the lowest health risk. The noncarcinogenic risk from Cr was above accepted thresholds in New Caledonia and USA samples and As in New Zealand samples presented the greatest carcinogenic risk. While the international average Pb risk was within accepted thresholds, there is potential for the dust hazard to have an unacceptable health risk, particularly from Pb in Australian homes.

While all exposures to Pb in household dust are of concern, bioavailability and bioaccessibility of trace metal constituents should also be considered in additional and more detailed risk assessments.[40] Peeling paint is indeed a factor of concern (Section 3.2) because Pb pigments in older paints are highly bioaccessible,[25] increasing Pb bioaccessibility[142] in dust. In an Australian Pb mining community, Pb bioaccessibility in dust was found to be 68% (mean).[144] Across 38 DustSafe samples from Sydney, Australia, the mean Pb bioaccessibility ranged from 42% to 62% depending on the size fraction.[40] Pb sourced from outside soil sources may have a comparatively lower relative bioavailability.[143] Hong et al.[145] demonstrated the bioaccessible portion of Cr, Cd, Mn, and Ni in soils to be highly variable, finding, respectively, maximum values of 15, 81, 51, and 41% in the stomach and 16, 56, 51, and 17% in the intestine. However, studies in Portugal (urban Lisbon[146]) found relatively high (65% mean) Pb bioaccessibility in soil. Entwistle[147] found Al oxides in the soil to be the main contributors to the bioaccessible Pb fraction. The largest percentage increase in trace metal concentrations, particularly Pb and As, were increasing home age, peeling paint, and garden access (As). These are important considerations for those with children aged 5 and under, for whom Pb exposures present the greatest risk.

We identified two anthropogenic source factors; a Pb−Zn−As factor, reflecting the use of leaded paint in older homes, and a Cu−Zn factor, indicating degradation of building materials and accumulation of traffic pollutants; and the largely geogenic (soil-related) Mn factor. Each of these factors commonly occurred across 7−9 countries (dependant on factors). Ghana, New Caledonia, and New Zealand exhibited distinct factors not seen elsewhere, related to distinct geogenic and industrial sources.

3.6. Environmental Implications. While DustSafe participants have each been provided with a detailed report for their home and measures they may take to reduce trace metal concentrations, the study has broad implications for understanding exposures in a wider international context. For example, participatory community science programs such as

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Table 2. Suggested Best Practice Measures to Limit Risks Posed by Trace Metals in Household Dust

characteristic	measure to reduce trace metals in dust
renovation[150−152]	avoid exposure to and release of metal-rich particles into the home or outdoor environment by:
	•where possible, employing a certified professional to renovate in homes where Pb-based paint is suspected or likely (i.e., older than 1970s)
	•if possible, residing elsewhere during renovations if extensive, especially pregnant women and children. Advise neighbors of plans so that they can also reduce any exposure by closing windows or leaving the house. Keep children away from the work area
	•removing or covering carpets and furniture. Cover entryways (e.g., with plastic sheeting) to minimize dust spread
	•if outdoors, lay plastic sheets around the work area and close windows to prevent dust from traveling indoors
	•vacuuming ceilings to minimize the spread of dust prior to any ceiling work
	•using wet sanding or scraping to minimize dust
	•if working with contaminated paints or materials, wear a disposable Tyvek suit and use a mask with a P1 or P2 filter
	•cleaning up with HEPA vacuum or with wet wipes
	•sealing any potentially contaminated materials (including paint dust, disposable personal protective equipment) in bags and dispose of them responsibly. Ask your local council for disposal instructions
	•after work, remove work clothes and place them in a plastic bag
	•showering when finished and washing clothes separately
	•washing hands and face before eating or drinking
aged home[150]	ensuring paintwork is in good repair and that renovations are conducted in such a way as not to broadly release metal-rich particles from paint and aged building materials into the home environment (see "Renovation")
peeling paint[153]	if the old paint is in good condition, do not remove it (simply paint over it); do not paint over damaged or peeling paint
	paint can be easily tested for Pb content using commercially available test kits.
	Containing dust and preventing personal exposures as per "Renovation"
hobbies involving metal exposure[154]	practicing metal-exposing hobbies outside of main living areas. Showering and changing clothes after finishing work and washing clothes separately
pets[155]	washing or toweling down dirty or muddy animals before allowing them to enter the home
	reducing the access of pets to some areas of the home (eg. bedrooms) to reduce the extent of soil track-in
smoking[156]	smoking outside of the home rather than indoors and away from open windows and doors, wearing a jacket when smoking that can be removed at the door and washed regularly to prevent contaminants entering the home. Regularly cleaning any hard or soft surfaces exposed to smoking
vacuuming[6]	vacuuming frequently (once a week or more) to reduce dust accumulation and using a vacuum with a HEPA filter to prevent dust re-entering the home environment
home with garden/yard, general measures[6]	for any home environment, entry and accumulation of contaminated dust may be minimized by:
	•closing doors and windows on windy days or during dust storms
	•removing shoes at the door
	•establishing an entry mat system (outdoor coarse mat/s and indoor washable fine mats)
	•using a wet rag rather than a dry cloth to wipe surfaces
	•using washable rugs
	•wet mopping instead of sweeping
	•washing dusty and potentially contaminated clothes separately
heating fuel[157] refer to Supporting Information Figure 9.13.	use of electricity is preferred as it does not generate metal dust in the home
	coal burning is most likely to create metal-contaminated dust in the home and should be avoided by switching to a different form of heating
	for wood burning, do not burn treated timbers. Use an enclosed fireplace, seasoned versus greenwood, and regularly maintain a chimney
	for oil burning (potential Ni exposure), ensure the heater has a good flue

DustSafe educate participants about environmental health risks and empower them to take mitigating action (such as mats at the home entry, wet mopping and dusting, sealing areas of leaded paint), to reduce exposure risk, an engagement mode that has been proven effective.[148] A post participation survey of 246 DustSafe participants[149] showed that 39% of participants had taken some action to reduce contaminants in their homes. Recommended actions are summarized in Table 2. Surveyed participants (94%) found the information they received useful and 74% felt safer in their home environment as a result of participation. But perhaps more importantly, this type of work advances our understanding of exposure types and differences in the geochemical "texture" of exposure sources depending on the housing type and age, proximity of housing to urban centers, and country location of housing. Clearly, most of the interior dust samples were from one country (Australia), but as this program continues, one goal is to understand whether country-specific differences exist in interior dust chemistry, and if so, what the role of geogenic versus anthropogenic geochemical inputs is.

The DustSafe program provided a unique opportunity for international science collaboration, resulting in international data, accessible to all, that gives context to trace metal concentrations in individual households. Participants are able to better understand trace metal hazards in their homes and take action to reduce exposures, potentially improving the health of their families.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.1c04494.

1.0 Analysis techniques and quality control: detailed method and tabulated quality control data for each country; 2.0. DustSafe surveys and reports sent to program participants; 3.0 international crustal values

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used in enrichment factor calculations; 4.0 distribution of trace metal concentrations for each element; 5.0 principal component analysis method; 6.0 positive matrix factorization method; 7.0 international trace metal concentrations for each participating country; 8.0. health risk modeling method, worked example, and data tables; 9.0 metadata analyses, detailed graphs, and statistical results for each variable; 10.0 tables of metal enrichment factors' data for each country; 11.0 positive matrix factorization results graphed for each country; and 12.0 association between trace metal concentrations and distance from the city center (PDF)

(PDF)

AUTHOR INFORMATION

Corresponding Author

Cynthia Faye Isley − *Earth and Environmental Sciences, Faculty of Science and Engineering, Macquarie University, Sydney, New South Wales 2109, Australia;* orcid.org/0000-0001-7063-295X; Email: cynthia.isley@mq.edu.au

Authors

Kara L. Fry − *Earth and Environmental Sciences, Faculty of Science and Engineering, Macquarie University, Sydney, New South Wales 2109, Australia*

Xiaochi Liu − *Earth and Environmental Sciences, Faculty of Science and Engineering, Macquarie University, Sydney, New South Wales 2109, Australia*

Gabriel Michael Filippelli − *Department of Earth Sciences and Center for Urban Health, Indiana University-Purdue University Indianapolis (IUPUI), Indianapolis, Indiana 46202, United States;* orcid.org/0000-0003-3434-5982

Jane A. Entwistle − *Department of Geography and Environmental Sciences, Northumbria University, Newcastle-upon-Tyne NE1 8ST, U.K.*

Adam P. Martin − *GNS Science, Dunedin 9054, New Zealand*

Melanie Kah − *School of Environment, University of Auckland, Auckland 1010, New Zealand;* orcid.org/0000-0002-8705-9229

Diana Meza-Figueroa − *Universidad de Sonora, 83067 Hermosillo, Mexico;* orcid.org/0000-0002-8934-0321

John T. Shukle − *Department of Earth Sciences and Center for Urban Health, Indiana University-Purdue University Indianapolis (IUPUI), Indianapolis, Indiana 46202, United States*

Khadija Jabeen − *Department of Geography and Environmental Sciences, Northumbria University, Newcastle-upon-Tyne NE1 8ST, U.K.*

Abimbola O. Famuyiwa − *Department of Science Laboratory Technology, Moshood Abiola Polytechnic, Abeokuta, Ogun State P.M.B 2210, Nigeria*

Liqin Wu − *School of Environmental Science and Engineering, Guangzhou University, Guangzhou 510006 Guangdong, China*

Neda Sharifi-Soltani − *Earth and Environmental Sciences, Faculty of Science and Engineering, Macquarie University, Sydney, New South Wales 2109, Australia*

Israel N. Y. Doyi − *Earth and Environmental Sciences, Faculty of Science and Engineering, Macquarie University, Sydney, New South Wales 2109, Australia*

Ariadne Argyraki − *Department of Geology and Geoenvironment National & Kapodistrian, University of Athens, 15784 Athens, Greece*

Kin Fai Ho − *Institute of Environment, Energy, and Sustainability, The Chinese University of Hong Kong, Shatin, N.T., Hong Kong, China;* orcid.org/0000-0001-7464-3437

Chenyin Dong − *State Environmental Protection Key Laboratory of Environmental Pollution Health Risk Assessment, South China Institute of Environmental Sciences, Ministry of Ecology and Environment, Guangzhou 510655, China*

Peggy Gunkel-Grillon − *Institute of Exact and Applied Sciences (ISEA), University of New Caledonia, 98851 Nouméa, New Caledonia, France*

C. Marjorie Aelion − *Department of Environmental Health Sciences, University of Massachusetts Amherst, Amherst, Massachusetts 01003, United States*

Mark Patrick Taylor − *Earth and Environmental Sciences, Faculty of Science and Engineering, Macquarie University, Sydney, New South Wales 2109, Australia; Environment Protection Authority, Centre for Applied Sciences, Melbourne, Victoria 3085, Australia;* orcid.org/0000-0001-7598-9982

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.est.1c04494

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Notes

The authors declare the following competing financial interest(s): MP Taylor is affiliated with Broken Hill Lead Reference Group (NSW, Australia), LEAD Group (NSW, Australia), and NSW Environment Protection Authoritys Broken Hill Environmental Lead Program, and reports undertaking paid and non-paid work for the NSW Environment Protection Authoritys Broken Hill Environmental Lead Program in relation to the assessment and management of environmental lead contamination in Broken Hill, NSW, Australia. MP Taylor has also provided advice in relation to lead exposure matters to various law firms, relating to mining and smelting lead contamination and human exposures in Australia and Africa, including accepting personal fees from Leigh Day for an investigation of lead contamination in Zambia. MP Taylor also reports managing two community-orientated programmes in Australia that provide advice about lead contamination from garden soils and household dusts with support from Macquarie University. MP Taylor also reports

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