Title: Limited retention of wildfire-derived PAHs and trace elements in indoor environments

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Key points:
- 14 months after a major wildfire, house dust concentrations of PAH and heavy metals were similar or lower than in background studies
- Ashes from buildings contained high concentrations of As. Traces of these ashes were detected in dust from highly affected neighborhoods
- Ashes from forests contained elevated concentrations of PAH, but not heavy metals. We did not detect evidence of residual forest ash.
Abstract

Wildfires are increasing in prevalence and intensity and emit large quantities of persistent organic and inorganic pollutants. Recent fires have caused elevated concerns that residual pollutants in indoor environments pose a long-term health hazard to residents, however, to date no studies have investigated how long fire-derived pollutants are retained in indoor environments. We quantified polycyclic aromatic hydrocarbons (PAHs) and toxic trace elements in ground ashes from the 2016 wildland-urban interface fires in Fort McMurray (Alberta, Canada) and in house dust from 64 homes. We document residual arsenic pollution from local building fires, but found no evidence that forest fire ash remained in households 14 months after the fire. Overall, house dust pollutant concentrations were equal or lower than in other locations unaffected by wildfires. Given the current and future concerns over wildfire impacts, this study provides importance evidence on the degree of their long-term effects on the residential environment.

1. Introduction

Wildfires release large quantities of toxic trace elements and persistent organic pollutants like polycyclic aromatic hydrocarbons (PAH) [Zhang and Tao, 2009; Shen et al., 2013; Plumlee et al., 2014]. Even more pollutants are emitted by wildland-urban interface fires that burn buildings, adding construction materials and household chemicals to the fire [Plumlee et al., 2012]. While the fate of fire-derived pollutants has been studied extensively in atmosphere [Liu et al., 2016; Reid et al., 2016; Landis et al., 2018; Wentworth et al., 2018], freshwater systems [Emelko et al., 2011; Smith et al., 2011; Verkaik et al., 2013; Bladon et al., 2014; Bodi et al., 2014; Silva et al., 2015; Abraham et al., 2018; Nunes et al., 2018], and soils [García-Falcón et al., 2006; Stankov Jovanovic et al., 2011; Vergnoux et al., 2011; Campos et al., 2015, 2016; Pereira et al., 2017; Alberta Health, 2018], no studies have been conducted to determine if and how long fire-derived pollutants are retained in indoor environments.

Addressing this knowledge gap is critical given the documented health impact of fire-derived pollutants [Plumlee et al., 2014; Reid et al., 2016]. Large quantities of ashes are deposited in indoor environments during typical fire situations, and char particle have been detected in indoor wipe samples 6 months after a fire [Ward, 2014]. In many cases, such ashes from wildland and urban fires were found to contain elevated pollutant concentrations. Long-term pollutant retention has been demonstrated for cigarette smoke, which can contaminate indoor environments for months to years after smoking stops [Sleiman et al., 2010; Acuff et al., 2016; DeCarlo et al., 2018], but it remains unclear if this applies to pollution from a single, large wildfire event. Closing this knowledge gap is important because residents spend most of their time indoors (85% for Canadians) [Leech et al., 2002]. Given that climate change will lead to more frequent wildfires [Westerling et al., 2006; Jolly et al., 2015], particularly in North America [Doerr and Santín, 2016], and expanding settlements will mean that wildfires are more likely to impact urban settlements [Field and Jensen, 2005], it is critical to understand the long-term effects of wildfires on returning residents.

The 2016 Horse River Wildfire in Alberta, Canada, was the costliest natural disaster in Canadian history [Insurance Bureau of Canada, 2016; Snowdon, 2017] and forced the evacuation of all 88 000 residents of Fort McMurray on May 2nd, 2016. Re-entry was delayed for up to five months when initial tests found elevated concentrations of fire-derived pollutants (e.g. arsenic) in ground ashes [Slaughter, 2016]. After re-entry, many residents expressed concern about residual pollution in their homes and
reported deteriorating health conditions, which they often attributed to residual toxins in their homes [The Globe and Mail, 2017]. Understanding if fire-derived pollutants are retained in homes would help local authorities and the scientific community address these types of concerns.

We conducted a study that addresses three research questions: (1) what are the concentrations of wildfire-derived pollutants in house dust, one year after the fire; (2) do pollutant concentrations in houses from heavily affected neighborhoods where houses were destroyed during the fire differ from less affected neighborhoods; and (3) are pollutant concentrations elevated relative to baseline studies [Rasmussen et al., 2001, 2013; Maertens et al., 2008]. Here we report initial results from our study in order to promptly address these urgent concerns of Fort McMurray residents and local health officials. These results will also provide useful information to inform recovery and remediation efforts in response to future wildfires.

2. Methods

Volunteer recruitment and selection. We collected dust samples from 71 houses of voluntary participants. Recruitment was conducted via social media and other online platforms (email, webpage). Participants were offered no incentives except the results from their houses. Out of 230 initial responses, the 71 participating homes were chosen based on availability during our sampling campaign, equal representation of neighbourhoods, and absence of indoor smokers. The locations of the homes are shown in Fig. 1. 38 houses were in highly affected areas where houses were lost during the fire (Abasand, Beacon Hill, Wood Buffalo, Stoney Creek, and Saprae Creek), whereas the remaining 33 homes were located in less affected areas. Seven houses were under renovation or otherwise unoccupied at the time of our visit and are not included in the results presented here. The total number of houses for which we report data here is 64. Descriptive statistics of the participating houses are provided in Supporting Information 1.

Dust sampling. House visits were conducted July 2017, which is 14 months after the beginning of the wildfire. House dust was sampled following a protocol from a recent child health cohort study [Takaro et al., 2015]. Briefly, samples were collected with two commercial vacuum cleaners (Sanitaire 3683 or 3684) outfitted with a custom-made head (“hognose”) that fits two filter thimbles (Dustream DU-FL-2; 40 micron nylon filters). At each house, dust samples were collected from one often frequented room, typically an adult’s or child’s bedroom. The sampled area was adapted in accordance with the amount of dust collected. We aimed to collect around 5 g of dust per sample, which corresponded to 1-5 m² carpet. In rooms with hard flooring, we vacuumed the entire floor, including corners of the room and below beds. Despite these efforts, samples from hard floors were often limited to around 300 mg of dust. All samples were sieved to 150 µm prior to analysis.

Ash samples. We collected a total of 15 ash samples during the fire. Eleven ash samples were collected the boreal forest outside the urban area of Fort McMurray and four ash samples were collected from three different (burned) Fort McMurray neighborhoods [Abasand (n=2), Beacon Hill (n=1), and Waterways (n=1)]. The samples were processed in the same way as house dust samples, except that these samples were sieved to 125 µm instead of 150 µm.

Analytical methods. Trace elements were quantified by inductively coupled plasma mass spectrometry after total digestion with HNO₃/H₂O₂/HF. PAHs were quantified by thermal desorption gas chromatography mass spectrometry after QuEChERS extraction and stir-bar sorption extraction (adapted
from Pfannkoch et al 2015). A detailed description of our analytical methods is provided in Supporting Information S2.

**Data analysis.** We applied three strategies to identify fire-derived pollution. First, we compared pollutant concentrations to regulatory guidelines and background studies. Currently, no regulatory guidelines for pollutant concentrations in house dust are issued by the Canadian or provincial government. We therefore compare the pollutant concentrations found in this study to guidelines for residential soils in the Alberta Tier 1 Soil and Groundwater Remediation guidelines [Alberta Environment and Parks, 2016]. Extensive data on trace element and PAH concentrations in Canadian houses are available from the Canadian House Dust Study [Rasmussen et al., 2013] as well as two studies of Ottawa house dust [Rasmussen et al., 2001; Maertens et al., 2008]. Concentration distributions in literature data were approximated with log-normal distributions. Typically, these datasets report the geometric mean (GM) and several percentiles (median, 75th percentile, 95th percentile). We used these to estimate the GM and geometric standard deviation (GSD), which formed the basis for the distributions plotted in Fig. 2.

Second, we compared pollutant concentrations among highly affected neighborhoods where buildings burned during the fire and other, less affected neighborhoods that were exposed to ash depositions, but not to local fires. Difference between these two groups of houses were verified by Student’s t-tests. Third, we compared the concentrations of pollutants with tracers for biomass burning, namely the alkyl-PAH retene and the element potassium expressed as potassium/sodium ratio (K/Na) [Standley and Simoneit, 1994; Lee et al., 2008; Pio et al., 2008]. We tested for correlations between tracers and pollutants using Pearson’s correlation coefficient. All statistical analysis was conducted using the statistical programming environment R version 3.2.3 [R Development Core Team, 2015]. Pollutant concentrations were log-transformed prior to statistical tests to ensure normal distribution.

3. Results

3.1. Concentrations of organic and inorganic pollutants in house dust

Pollutant concentrations in Fort McMurray house dust samples are shown in Figs. 2a, 2b and Table S3. Most trace elements were present at similar concentrations as those in house dust samples from other Canadian cities (2a), and none of the elements was present in higher concentrations than in the background studies. Some elements were present in significantly lower concentrations than in background studies, with GM concentrations 10-fold lower for Pb, 6-fold lower for Cd, and 4-fold for Co. Slightly (25-50%) lower concentrations were also found for As, Cu, V, Zn, and Ni. House dust concentrations generally remained within regulatory guidelines, except for Zn and Cu. Similarly, the concentrations of all detected PAH were significantly lower than in Ottawa house dust (2b). However, the concentrations of several PAH exceeded regulatory guidelines, including naphthalene, phenanthrene, anthracene, benz[a]anthracene and fluoranthene. The sum concentration of the quantified PAH ranged 0.415 to 31.2 mg kg⁻¹ (median: 0.915 mg kg⁻¹, arithmetic mean: 2.02 mg kg⁻¹, GM: 1.16 mg kg⁻¹), which is ten-fold lower than house dust in Ottawa (median: 9.53 mg kg⁻¹) [Maertens et al., 2008] and at the low end of reported literature values (arithmetic means: 0.13 – 115 mg kg⁻¹) [Ma and Harrad, 2015].

We detected few differences between highly affected and less affected neighborhoods (Fig 2c, 2d). Most importantly, arsenic was present at a 62% higher concentration in neighborhoods where buildings burned during the fire (t=2.08, p=0.041). Samples from houses in these neighborhoods also contained
significantly higher V concentrations (+37%). The concentrations of other trace elements and PAHs did not differ among most and less affected neighborhoods.

### 3.2. Concentrations of organic and inorganic pollutants in wildfire ashes

Pollutant concentrations in ash samples differed strongly between samples collected outside of the urban area of Fort McMurray (FMM) and those collected in neighborhoods with house fires. Samples collected at sites of urban fires (Fig 1a, 2a; black symbols) contained high concentrations of toxic trace elements, exceeding both guidelines and house dust concentrations by 1-2 orders of magnitude. The concentrations of 8 elements in urban ash samples exceeded guidelines, with median concentrations of 880 mg As kg\(^{-1}\), 1630 mg Cr kg\(^{-1}\), 1170 mg Cu kg\(^{-1}\), 57 mg Sb kg\(^{-1}\), 142 mg V kg\(^{-1}\), 3120 mg Zn kg\(^{-1}\), 9.6 mg Mo kg\(^{-1}\), and 55 mg Ni kg\(^{-1}\). The concentrations of Co, Cd, Pb, and U remained below regulatory guidelines. Trace element concentrations in ashes collected outside the FMM urban area (grey symbols), in contrast, largely remained below the guidelines. One exception to this general trend was Zn with a median concentration of 1400 mg kg\(^{-1}\).

Normalization of pollutant concentrations in ash to those in house dust (geometric means) further demonstrates these differences between locations and pollutant classes (Fig 2e, 2f). Trace element concentrations in ashes collected outside FMM were generally not elevated relative to those in house dust, indicating that these ashes would not be an important contributor to indoor contamination. Trace element concentrations in urban ash samples, in contrast, contained up to 100-fold higher trace element concentrations than house dust. The highest concentrations relative to house dust were found for As and Cr, followed by Cu, Sb, V, Z, Mo, and Co (Fig 2e). Both urban and forest ashes exhibited higher PAH concentrations than house dust. Ash concentrations of 2- and 3-ring PAHs (naphthalene, fluorene and phenanthrene), in particular, exceeded those in house dust by 2-3 orders of magnitude (Fig. 2f).

### 3.3. Biomass burning tracers

As expected, ashes contained high concentrations of retene (GM 23.0 mg kg\(^{-1}\); GSD 1.85), 28 times higher than in house dust samples. We did not detect differences between ashes collected from location within and outside of Fort McMurray (t=1.02, p=0.366). Potassium to sodium ratios (K/Na) were elevated in forest ash samples (GM 18.1, GSD 3.1), whereas concentrations in ashes from burned buildings (GM 0.75, GSD 1.43) were similar to those found in house dust (GM 0.67, GSD 1.41). We did not detect differences in retene concentrations or K/Na ratios between highly and less affected neighborhoods (Figs. 2c, 2d). Furthermore, we detected only weak correlations between either tracer and any of the quantified pollutants in house dust samples: V concentrations was correlated with retene concentrations (R=0.32), while V (R=0.42) and Co (R=-0.25) concentrations were correlated with K/Na (Supporting Information S5).

### 4. Discussion

#### 4.1. Limited retention of wildfire-derived pollution in house dust.

Overall, the house dust concentrations of PAH and trace elements detected in this study were low compared to regulatory guidelines and background studies. Even after the wildfire, the exposure of Fort McMurray residents to house dust pollutants was equal or lower to residents of other Canadian cities. These low pollutant concentrations have likely resulted two main causes. First, due to its remote location, Fort McMurray is exposed to less urban pollution, including vehicular and heating emissions. Industry sites
are located at least 50km north of the city (downwind under typical spring/summer weather conditions).

Second, Fort McMurray houses are younger than in other Canadian cities. Fort McMurray has experienced a rapid growth over the last 3 decades due to the expansion of the oil sands industry [Regional Municipality of Wood Buffalo, 2015]. More than 50% of the sampled homes were built since 2000, and none were older than 1960 (Supporting Information S2). House age has been associated with lower Pb, Cd, and Zn concentrations in some past studies [Kim and Fergusson, 1993; Rasmussen et al., 2013]. Furthermore, the primary heat source in Fort McMurray is natural gas, which is associated with lower pollutant emissions than other methods of heating [Rasmussen et al., 2001].

4.2. Local and regional pollution

Regional pollution. The pollutant concentrations in ash samples indicate that at least two types of toxic ashes were produced during the 2016 wildfire. First, forest fires produced ashes that contained high concentrations of PAHs, whereas the concentrations of toxic trace elements in these ashes were low (typically lower than in house dust samples). These ashes were characterized by high concentrations of the two tracers of biomass burning studied here, i.e., retene and potassium (resulting in high K/Na values). Our results indicate that these regional forest ashes did not significantly increase the pollutant load of Fort McMurray house dust 14 months post fire. While the tracer retene was detected in all studied house dust samples, PAH concentrations remained an order of magnitude lower than in other studies and were not correlated with retene concentrations. The ratio of K/Na in house dust (GM 0.66, 0.70 in highly and less affected areas) was only slightly elevated compared to background studies (0.49) [Rasmussen et al., 2001]. All together, these results indicate that forest fire ashes did not significantly increase the pollutant load in Fort McMurray house dust at the time of our field campaign.

Our study provides some indirect evidence for the initial contamination of house dust with PAH. Elevated PAH concentrations in ash samples from different locations in- and outside of Fort McMurray (Fig 2f), however, suggest that PAH concentrations in these ashes were 1-2 orders of magnitude higher than in house dust. Large quantities of these ashes were found inside Fort McMurray homes after the fire, suggesting that elevated PAH concentrations were present house dust immediately after the fire.

Local pollution. Second, ash samples from burned buildings contained high concentrations of multiple toxic trace elements, including As, Cr, Cu, and Sb. Our results suggest that some heavy metals resulted from these localized sources remain in Fort McMurray house dust. Most importantly, they explain the 62% higher arsenic concentrations in neighborhoods with burned buildings. Arsenic concentrations were most elevated in urban ash samples relative to house dust (150 times higher). This high As concentration, along with Cr and Cu, in urban ashes likely resulted from combustion of chromated copper arsenate (CCA) treated wood, which was used to construct of most Fort McMurray houses [Alberta Health, 2018]. Cr and Cu commonly occur at higher concentrations in house dust, such that any increase in their concentration due to fire emissions was likely masked by the Cr and Cu already present in house dust. Background concentrations of As, in contrast, are relatively low, allowing us to observe As at slightly elevated levels if traces of urban ashes were retained in house dust. Nevertheless, it is important to emphasise that that even in highly fire affected neighborhoods, As concentrations were lower than in other cities, and remained below the regulatory guideline.

5. Conclusions
This is the first study to investigate the potential for indoor retention of pollutants after wildfires. Given the increased prevalence of disastrous fires in recent years as well as the high level of concern about residual communities, we communicate our initial finding of the limited persistence of fire-derived pollution in house dust here to inform the recovery efforts of other fire effected communities and ongoing studies in those localities. Based on our observations, we find limited evidence of elevated fire-derived pollutant levels in indoor house dust 14 months after the Horse River Fire. While fire ash in areas with burned buildings showed signatures of residual As, Cr and Cu, the levels of these pollutants in house dust remain lower than or comparable to those in other Canadian cities.

It should, however, be noted that there are several limitations to our study. First, we acknowledge that our toxicological assessment of ash samples remains incomplete – we were not able to test for every potential contaminant. We specifically acknowledge that we did not quantify hexavalent chromium and mercury, two pollutants that can be found in fresh wildfire ashes [Friedli et al., 2001; Wolf et al., 2011; Campos et al., 2015]. Second, we acknowledge two important limitations in our sampling scheme. We collected samples during the summer months, when ventilation is likely higher than in winter, and when a warm and dry climate increases the production of soil-borne dust. These seasonal differences might lead to a dilution of fire-derived house dust with outdoor dust, thus leading to lower pollutant concentrations than can be found during the winter months. We also note that our sampling strategy focussed on often frequented and cleaned areas of house (bedrooms and similar areas). The fact that we found little remaining ashes there does not exclude the persistence of ash reservoirs in less-often cleaned areas of the houses. In the future, we intend to expand our study to address these remaining questions.

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Ethics approval
This work has been approved by the University of Toronto Research Ethics Board under the protocol #34221.

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Figure 1. Location of sampled homes (circles) and ash samples (stars) analyzed.
Figure 2. Concentrations of trace elements (a,c,e) and polycyclic aromatic hydrocarbons (PAH; b,d,e) in wildfire ashes and house dust samples. The plotted data show pollutant concentrations in Fort McMurray house dust compared to house dust in other Canadian cities (a,b). Furthermore, a comparison between house dust pollutant concentrations in the most affected neighborhoods relative to those in other neighborhoods (c,d). Asterisks indicate significant differences between neighborhood types. Finally, pollutant concentrations in wildfire ashes from urban and non-urban locations, normalized to house dust concentrations (e,f). K/Na, Potassium/sodium ratio; Nap, naphthalene; Flu, fluorene; Phe, phenanthrene; Ant, anthracene; Fla, fluoranthene; Pyr, pyrene; Chr, chrysene; BaA, Benz[a]anthracene; BbkF, sum (Benz[b]fluoranthene + Benz[k]fluoranthene); BaP, Benzo[a]pyrene; DBA, Dibenz[ah]anthracene; BeP, Benzo[ghi]perylene, InP, Indo[1,2,3cd]pyrene; Ret, retene.