

## Polycyclic Aromatic Hydrocarbons (PAHs) in Wildfire Smoke Accumulate on Indoor Materials and Create Postsmoke Event Exposure Pathways

Aurélie Laguerre and Elliott T. Gall\*



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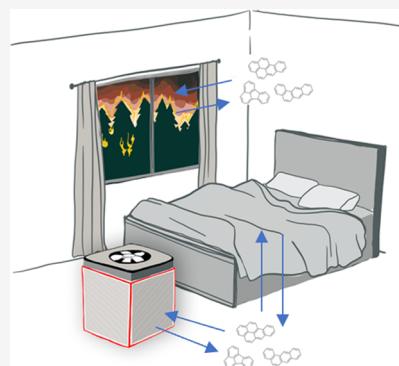
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**ABSTRACT:** Wildfire smoke contains PAHs that, after infiltrating indoors, accumulate on indoor materials through particle deposition and partitioning from air. We report the magnitude and persistence of select surface associated PAHs on three common indoor materials: glass, cotton, and mechanical air filter media. Materials were loaded with PAHs through both spiking with standards and exposure to a wildfire smoke proxy. Loaded materials were aged indoors over  $\sim 4$  months to determine PAH persistence. For materials spiked with standards, total PAH decay rates were  $0.010 \pm 0.002$ ,  $0.025 \pm 0.005$ , and  $0.051 \pm 0.009 \text{ day}^{-1}$ , for mechanical air filter media, glass, and cotton, respectively. PAH decay on smoke-exposed samples is consistent with that predicated by decay constants from spiked materials. Decay curves of smoke loaded samples show that PAH surface concentrations are elevated above background for  $\sim 40$  days after the smoke clears. Cleaning processes efficiently remove PAHs, with reductions of 71% and 62% after cleaning smoke-exposed glass with ethanol and a commercial cleaner, respectively. Laundering smoke-exposed cotton in a washing machine and heated drying removed 48% of PAHs. An exposure assessment indicates that both inhalation and dermal PAH exposure pathways may be relevant following wildfire smoke events.

**KEYWORDS:** *indoor air quality, persistent organic pollutants, partitioning, biomass burning, surface chemistry*



### 1. INTRODUCTION

The frequency and severity of wildfire events are increasing all over the world.<sup>1,2</sup> Wildfires are a substantial source of air pollutants, including polycyclic aromatic hydrocarbons (PAHs).<sup>3</sup> Understanding the pathways and magnitudes of exposure to PAHs is essential because of their known mutagenicity and carcinogenicity.<sup>4</sup> Respiratory effects, developmental issues, infertility, and carcinogenicity are among the health concerns linked to exposure to PAHs.<sup>5,6</sup>

PAHs consist of two or more fused aromatic rings and are produced during the incomplete combustion of organic matter. The U.S. Environmental Protection Agency (EPA) classifies seven PAHs as probable human carcinogens among 16 placed on a priority list because of their human toxicity.<sup>7,8</sup> Recent studies show that other PAHs might be added to this list.<sup>9</sup> PAHs are present in substantial quantities in outdoor air during wildfire events,<sup>10–12</sup> and are also observed in indoor air.<sup>3,12–17</sup> PAHs are semivolatile compounds and will deposit with particulate matter and/or partition to the extensive material surface area that exists indoors. Yet, only few studies to date focus on PAH accumulation due to wildfire smoke on indoor surfaces.<sup>12,18</sup>

Several studies indicate that PAHs may accumulate on surfaces due to exposure to smoke. For example, while differing in many ways from indoor wildfire smoke, PAH accumulation is observed on firefighters' skin and equipment during

firefighting activities.<sup>19–23</sup> Ghetu et al.<sup>12</sup> showed higher accumulation of vapor-phase PAHs indoors than outdoors, measured using passive samplers deployed during wildfire events. These studies imply that PAHs will also accumulate on indoor materials during wildfire events, although there are limited studies investigating authentic indoor materials. Kohl et al.<sup>18</sup> collected dust samples in the bedrooms of 64 homes in the region of a major fire in 2016 in Fort McMurray (Alberta, Canada). This study showed no difference in PAHs in dust 14 months after the wildfire event when comparing homes impacted by the fire to those unaffected by the fire. This study implies the indoor PAH reservoir created by wildfire smoke may not persist above background for time scales of years. However, additional studies are warranted, including the need to complement studies of dust with sampling of materials directly and measuring PAHs in the weeks and months immediately following a smoke event when PAH surface concentrations are expected to be highest.

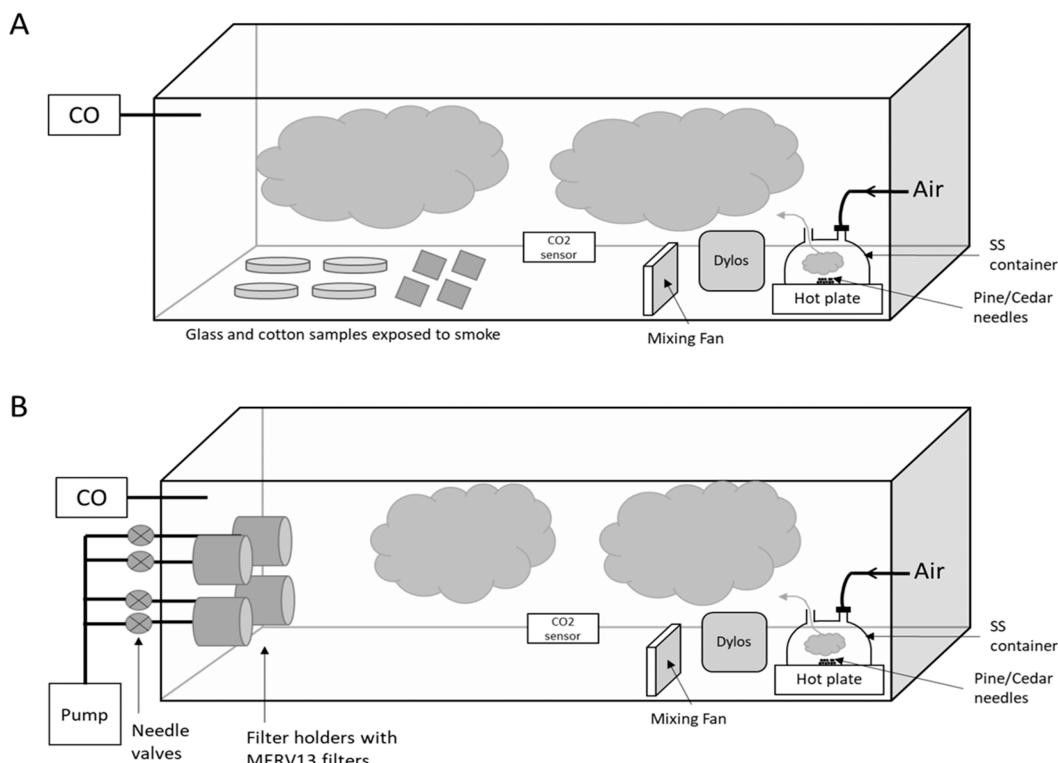
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**Figure 1.** Schematic of the smoke exposure chamber with fir and cedar needle combustion smoke for (A) cotton and glass materials exposed to and fir and cedar needle combustion smoke and (B) the filter holders with MERV13 filters exposed to a smoke generation process similar to that of cotton and glass.

Understanding the magnitudes and persistence of indoor surface-associated PAHs is necessary to evaluate exposures from smoke exposed materials and interventions that may be effective at reducing PAHs on smoke exposed materials. For example, the Red Cross<sup>24</sup> gives advice on removing smoke odor after a fire by using mild detergents on indoor surfaces such as painted drywall and furniture, and using laundry detergent and products containing trisodium phosphate on clothes. The U.S. EPA recommends misting indoor surfaces with water before sweeping them to keep dust on the surface and avoiding inhalation.<sup>25</sup> The efficacy of such interventions is not well-known; one of the few studies on this subject evaluated the impact of laundering firefighter's clothes to remove PAHs,<sup>26</sup> and showed a significant reduction of PAHs postlaundry for only 3 of the 16 samples areas. Studies of materials common to indoor environments<sup>27</sup> are warranted to understand what efficacy may be expected of processes that aim to remove PAHs from indoor materials.

In this study, we evaluate the accumulation and retention of PAHs on three indoor materials (glass, cotton, and mechanical air filter media) after PAH spiking, simulated wildfire smoke exposure, and aging indoors over a period of 4 months. These materials were chosen as common household materials that are present in meaningful quantities, are present in proximity to occupants, and are with which occupants physically interact. The effectiveness of cleaning processes on glass and cotton are determined. These data contribute to understanding the dynamics of PAHs persistence on indoor surfaces, supporting exposure assessment and decision making toward effective and accessible strategies for reducing indoor exposures to wildfire smoke.

## 2. MATERIALS & METHODS

**2.1. Selected Indoor Test Materials.** Glass, cotton, and mechanical air filter media were the test materials in this study. Petri dishes (100 mm diameter × 10 mm Pyrex, 7740 borosilicate glass) were used as glass samples. Squares of 25 cm<sup>2</sup> (5 cm × 5 cm) were cut from 100% organic cotton bedsheet (Mellanni, 400 thread count) and from MERV13 filters (Tex-Air filters, Air Relief Technologies, Inc.).

**2.2. Negative Controls and Spiked Materials.** Negative controls were test materials neither exposed to smoke nor spiked with a PAH mixture. Spiked materials were test materials loaded with a 16-PAH mixture (certified reference material, *TraceCERT*, Sigma-Aldrich Co. LLC) was diluted in dichloromethane (Optima, Fisher Scientific). Fifteen compounds in the mixture are present on the U.S. EPA PAH priority list;<sup>8</sup> see Table S1 in the Supporting Information for the list of compounds used in this study. The target concentration loaded was 26 µg/m<sup>2</sup> for glass and 40 µg/m<sup>2</sup> for filters and cotton. The spiking solution was uniformly deposited over the material by using a syringe.

**2.3. Exposed Materials.** Glass and cotton were exposed to smoke in a 62 × 62 × 152 cm steel, sealed exposure chamber (Figure 1A) for a period of 1 to 5 days with 9 h of smoke exposure per day. A mix of Douglas Fir and Western Red Cedar needles, common trees in the Northwestern U.S., was used as fuel. The smoke generator was similar to Zhang et al.<sup>28</sup> Three aluminum cups (6.3 cm diameter), each containing three grams of mixed needle biomass, were placed in a sealed stainless steel container (1.5 L) with combustion gas (N<sub>2</sub> or filtered air) flow of 200 mL/min. A hot plate (Elite Gourmet, model ESB-301BF) heated the stainless-steel container with temperature measured by a thermocouple (Tektronix,

DTM920). At 500 °C, necessary for the formation of most PAHs,<sup>29</sup> the hot plate was turned off. The container was refueled and heated again every 45 min to elevate smoke for ~9 h each day. A mixing fan (Nidec Beta, V TA300DC) was installed, and well-mixed conditions confirmed via separate studies using CO<sub>2</sub> injection; concentrations in three locations were consistent within 10%. Inside the exposure chamber, carbon monoxide (Ecotech, Serinus 30), CO<sub>2</sub>, relative humidity, temperature (HOBO onset, MX 1102) and particulate matter (Dylos, DC1700) were measured.

Four MERV13 filters were exposed to smoke in the same conditions as cotton and glass but during a separate exposure event (Figure 1B), because filters were loaded with forced airflow through the media, as in an air cleaner or air-conditioning system. One vacuum pump pulled air through four filters, with needle valves installed to ensure similar flow rates across each replicate filter (4.4 ± 0.1 L/min). Pieces of MERV13 filters (5 cm × 5 cm) were uniformly cut from the four MERV13 filters after smoke exposure.

Fir and cedar needles were collected a week prior to each experiment in July 2022 and stored in a paper bag in the laboratory. Biomass moisture content was calculated by weighing a separate, representative sample of biomass before and after drying, following the method described by Huang et al.<sup>30</sup> Moisture content was measured as prior studies indicate it influences PAH emission factor.<sup>31</sup> Smoke exposure conditions and fuel moisture content for retention and cleaning experiments are shown in Table S2 in the Supporting Information.

**2.4. Retention Experiments.** Following spiking and exposure to smoke, PAH surface concentrations were determined at seven points in time by placing seven sets of each material studied in an office on the Portland State University campus (see Figure S1 of the Supporting Information). Materials aged in place over ~4 months, with sampling on days 0, 14, 28, 37, 58, 90, and 133. Each set of materials included duplicates for each of the negative controls, spiked samples, and exposed samples. Environmental data in the office were measured during the 133 days of aging: ozone (2B Tech, model OEM-106-L), CO<sub>2</sub> (Aranet4), particulate matter (PurpleAir, PAII), relative humidity, temperature, and visible light intensity (HOBO, U12-012) (Figure S2 of Supporting Information).

Negative controls and spiked materials, previously defined in section 2.2, for cotton and glass were prepared and placed on a horizontal desk exposed to direct sunlight through a window (natural light location, Figure S1) in the office. Negative controls and spiked materials for filters and a second set of glass materials were prepared and placed on a horizontal desk in the same office but exposed only to artificial light from overhead LEDs (artificial light location, Figure S1). The aim of this experimental design was to allow comparison of decay rates for glass with two distinct lighting conditions, as PAHs may be removed through photodegradation.<sup>32,33,27</sup> Smoke exposed materials, as defined in section 2.3, were prepared and placed in a natural light location for cotton and glass and in an artificial light location only for the filter material.

**2.5. Cleaning Experiments.** Triplicate sets of cotton and glass materials were, respectively, spiked or exposed to smoke in the same conditions as the retention experiments (Figure 1A). Glass materials were cleaned using a wipe soaked with 2 mL of a commercial glass cleaner (corresponding to approximately 2 sprays) or ethanol 95% (EtOH). Sampling

wipes soaked in isopropanol (IPA) were used to sample PAHs that remained postcleaning on glass. PAHs were also extracted from the wipes used to clean the surfaces and from the sampling wipes. Cotton samples were washed in a washing machine (Maytag, Legacy series, Quiet series 100), temperature hot (~60 °C) and cycle “regular” using a common laundry detergent and dried in a dryer (GE, model DHDSR46EE0WW), temperature: cottons, regular heat (~60 °C). PAHs were directly extracted from the cotton samples after the dryer cycle.

**2.6. Measurement and Calculations.** The sampling, extraction, and analysis are detailed in Laguerre and Gall<sup>34</sup> and Supporting Information to this paper (including Table S3 for compounds and associated GC-MS parameters). In brief, we collect surface associated PAHs that may partition and deposit using solvent-soaked wipes (glass materials) followed by dichloromethane (DCM) extraction or direct extraction (cotton, filters) of the material in DCM. Samples were analyzed via gas chromatography–mass spectrometry. Two surrogate standards, naphthalene-D8 and chrysene D-12 were added to the sample prior to extraction; chrysene D-12 extraction efficiencies were used to determine correction factors to account for sample loss during extraction. Laguerre and Gall<sup>34</sup> previously developed and critically evaluated the analytical method used here, and showed chrysene D-12 surrogate recovery is representative of recoveries for the PAHs studied here within ~5%. Benzo[b]fluoranthene recoveries were previously observed to be lower (~50%) than chrysene D-12 (~70%), and as a result, concentrations of this compound may be underestimated here.

PAHs selected for calculations for each material were screened for quality via the extraction recovery estimated during the method development;<sup>34</sup> among the 16 PAHs, only PAHs with total recovery percentages (as defined and with numeric values provided in Laguerre and Gall<sup>34</sup>) ≥ 40% were selected (Table S4). Therefore, 9, 10, and 7 PAHs were selected, respectively, for glass, MERV13 filter, and cotton for analysis. Figure S3 shows the decision tree for the application of a correction based on surrogate recovery.

Spiked materials were used to calculate the decay rate of the PAHs on each surface. Decay rates were estimated using eq 1:

$$-\ln\left(\frac{C_t}{C_0}\right) = kt \quad (1)$$

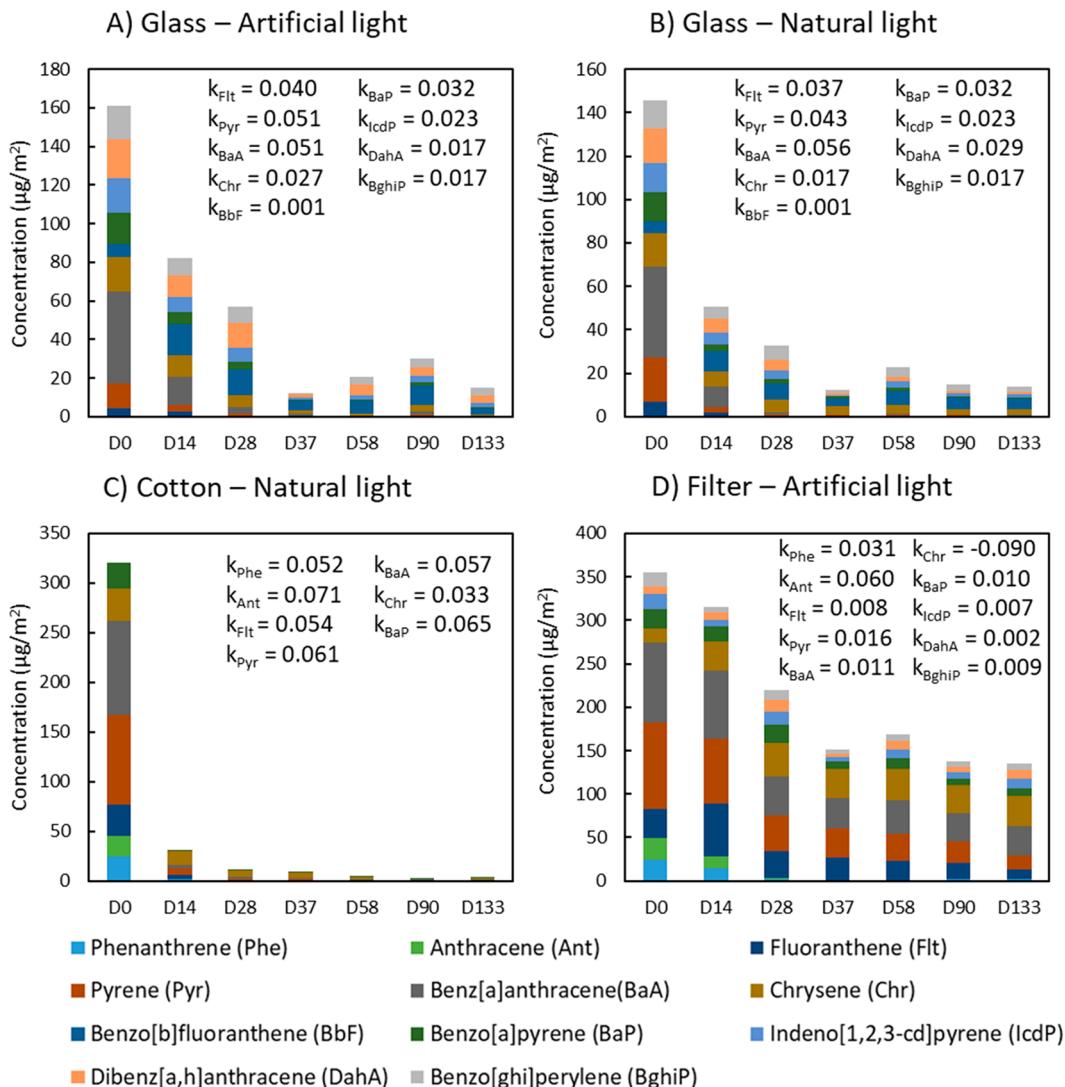
where  $C_t$  and  $C_0$  are the measured surface concentrations of PAHs ( $\mu\text{g}/\text{m}^2$ ) at time  $t$  and  $t = 0$ , respectively, and  $k$  is the decay constant ( $\text{day}^{-1}$ ).

The efficiency of cleaning processes was calculated with eq 2:

$$R(\%) = \frac{C_i - C_f}{C_i} 100 \quad (2)$$

Where  $R$  is the percentage removal (%),  $C_i$  is the initial concentration of PAHs from the spiked or exposed surface precleaning ( $\mu\text{g}/\text{m}^2$ ), and  $C_f$  is the concentration of PAHs postcleaning ( $\mu\text{g}/\text{m}^2$ ).

**2.7. Exposure Assessment.** Exposures to surface-associated PAHs post simulated wildfire smoke event in a hypothetical bedroom was quantified for two pathways: inhalation exposure from PAHs re-emitted from surfaces and dermal contact with cotton materials in bedding. Our model is focused on doses received over 1 week due to nightly 8-h



**Figure 2.** Mean concentration of the PAHs for (A) spiked glass samples in an artificial light location, (B) spiked glass in a natural light location, (C) for spiked cotton samples, and (D) for filter samples with a decay constant ( $k$ ) for each PAH ( $\text{day}^{-1}$ ). Values presented have subtracted the negative control measurement, which are reported in Figure S5 in the Supporting Information. The standard error for each decay constant is presented in Table S7 in the Supporting Information.

sleeping periods in a bedroom; we calculate PAH doses due to re-emission from cotton bedsheets, glass, and four MERV13 filters and dermal contact with cotton bedsheets. Materials are all assumed to be initially contaminated with PAHs at loadings after 9 h of smoke exposure (Section 3.3). The concentrations of surface-associated PAHs for each day from day 1 to day 7 of the exposure period were modeled using the initial concentration and decay constants determined from spiked materials. Daily doses do not consider exposures received in other settings, or from other sources in the bedroom. This scaling analysis explores the potential for contribution to PAH exposure after a wildfire smoke event through only re-emission and contact with PAHs from select surfaces. It is known PAHs are transformed and may be degraded into transformed PAH products (TPP) with toxicities and transport that differ from parent compounds;<sup>35</sup> such factors should be included in future studies of PAH and TPP exposure due to indoor wildfire smoke. Further model assumptions and details are listed in the Supporting Information.

For inhalation exposure, the change in indoor air concentrations of PAHs due to re-emission is calculated by assuming that surface decay is due solely to partitioning to indoor air, resuspension, or partitioning to respirable airborne particles. This approach likely represents an upper-limit estimate of inhalation exposure to PAHs from materials considered, as it does not account for processes that generate, or resulting exposures to, TPP. The change in indoor steady-state PAH air concentration due to re-emission is calculated with eq 3:

$$\Delta C_{\text{PAHs}} = \frac{\sum_i \frac{(C_{t=0} - C_t)A}{t}}{\lambda V} \quad (3)$$

where  $\Delta C_{\text{PAHs}}$  is the change in indoor air concentrations of PAHs above background due to re-emission ( $\mu\text{g/m}^3$ ),  $i$  refers to the material, either glass, cotton, or MERV 13 filter,  $C_{t=0}$  is the surface concentration of PAHs at day 0 ( $\mu\text{g/m}^2$ ),  $C_t$  is the surface concentration of PAHs at a specific day ( $\mu\text{g/m}^2$ ),  $A$  is the total surface area contaminated ( $\text{m}^2$ ) that re-emits PAHs,  $t$  is the time elapsed between day 0 and the specific day (h),  $\lambda$  is

the assumed outdoor air change rate ( $0.5 \text{ h}^{-1}$ ),<sup>36</sup> and  $V$  is the volume of an average bedroom ( $31 \text{ m}^3$ ).<sup>37</sup>

The  $\Delta C_{\text{PAHs}}$  was calculated for each day from day 1 to day 7 and averaged ( $\bar{\Delta C}_{\text{PAHs}}$ ). The change in inhalation daily dose due to re-emission from materials is

$$DD_{\text{i.e.}} = \bar{\Delta C}_{\text{PAHs}} V_{\text{inhaled air}} \quad (4)$$

where  $DD_{\text{i.e.}}$  is the daily dose calculated for inhalation exposure ( $\mu\text{g}$ ), and  $V_{\text{inhaled air}}$  is the average volume of inhaled air in 8 h of sleep for an adult ( $2.4 \text{ m}^3$ ).<sup>38</sup>

For the dermal contact route, the daily dose was calculated using eq 5, based on and adapted to our scenario from Bernard and Dudler.<sup>39</sup> For this calculation, exposure to cotton material in sheets, duvet, and pillowcase was taken into account, as the most obvious dermal exposure route of materials studied.

$$DD_{\text{d.e.}} = \frac{C_{\text{PAHs}} \times S \times t \times T \times A}{SDAF \times 24} \quad (5)$$

where  $DD_{\text{d.e.}}$  is the average daily dose over the first week for dermal exposure ( $\mu\text{g}$ ),  $C_{\text{PAHs}}$  is the average surface concentration of PAHs over a week ( $\mu\text{g}/\text{m}^2$ ),  $S$  is the surface of cotton that the body is exposed to during the night ( $9.4 \text{ m}^2$ , a summation of sheet, duvets, and pillowcase area in possible contact with the subject; this parameter is likely variable across subjects),  $t$  is exposure time each day (8 h),  $T$  is the fraction transferred to the skin (7%),<sup>39</sup>  $A$  is the fraction absorbed by the skin, set at 100%,<sup>39</sup> SDAF is the solvent-dependent adjustment factor, taken as 10 as in Bernard and Dudler,<sup>39</sup> and 24 corresponds to 24 h/day.

### 3. RESULTS AND DISCUSSION

**3.1. Smoke Exposure and PAH Accumulation on Glass.** We performed smoke generation tests to evaluate the impact of burning conditions on gas, particle, and surface concentrations of pollutants. The impact of fuel type, pyrolysis ( $\text{N}_2$ ) vs combustion (air), and feed rate (i.e., number of smoke generation events) on pollutant levels is shown in Figure S4 of the Supporting Information. Wood powder incense (Indo Lao Shan, Sandalwood) and a mixture of fir and cedar needles from local trees were tested as fuel. Concentrations of pollutants (CO and  $\text{PM}_{2.5}$ ) are the levels in the smoke exposure chamber; surface concentrations of PAHs are determined from measurements made from samples of glass surfaces.

One purpose of the measurements presented in Figure S4 was to compare our PAH loading conditions to an expectation of an indoor space to a major wildfire smoke event. During the 2020 Labor Day Fires in the Western U.S.,<sup>40</sup> outdoor  $\text{PM}_{2.5}$  and CO concentrations measured in Portland, OR, USA were  $>500 \mu\text{g}/\text{m}^3$  and 15 ppm, respectively.<sup>41</sup> Other studies during major wildfire events report  $\text{PM}_{2.5}$  and CO concentrations on this order.<sup>17,42</sup> Infiltration factors during recent wildfire events are  $\sim 0.2\text{--}0.5$ ,<sup>17</sup> corresponding to  $\sim 100\text{--}250 \mu\text{g}/\text{m}^3$  (similar to Figure S4) as an expected range of indoor  $\text{PM}_{2.5}$  of outdoor origin during the 2020 Labor Day Fires.

The surface PAH concentration on glass ranged from 2.1 to  $13.3 \mu\text{g}/\text{m}^2$  when wood powder is pyrolyzed while it ranged from 0.9 to  $2.2 \mu\text{g}/\text{m}^2$  when burning needles, in both cases increasing with increasing duration of smoke exposure. Fir and cedar needle combustion under air flow produces a different composition of PAHs on surfaces than pyrolysis under nitrogen flow (Figure S4). Similar behavior is observed for the formation of volatile organic compounds (VOCs) with

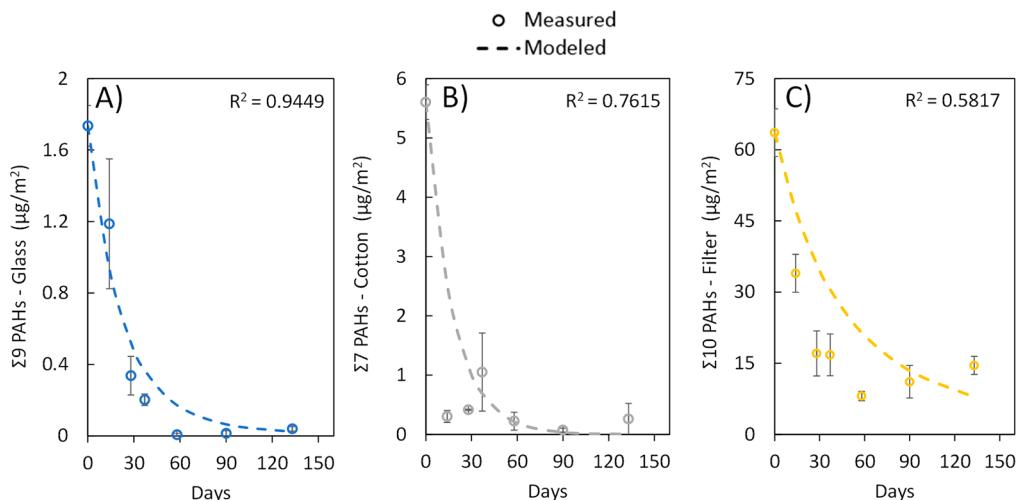
wood powder as fuel under nitrogen conditions or air conditions.<sup>28</sup> Subsequent experiments to load materials with PAHs used fir and cedar under air flow because (1) it is assumed more representative of real wildfire burn conditions than other tested conditions and (2) results show 1 day, with 9 h of smoke exposure, creates a surface loading 10× the method detection limit (MDL, Table S6 in the Supporting Information).

### 3.2. Retention of PAHs on Spiked Surfaces and Decay Rates.

Test materials (glass, cotton, and mechanical filters) were spiked with a known quantity of PAHs and aged undisturbed in an office environment as described in Section 2.4. Spiked samples allowed for more confident calculation of decay constants for each material than smoke-exposed samples due to substantially higher initial concentrations and a simpler matrix. Decay constants are listed in Figure 2 (and Table S7) and surface concentrations shown have subtracted negative controls (Figure S5). In general, PAH concentrations substantially decreased from the initial spiked concentration over the first  $\sim 40$  days. By the third sampling event (37 days after spiking), total PAH concentrations were reduced by 92–97% for glass and cotton and by 58% for filters. It appears that all materials tested reach a near-steady-state surface concentration within the sampling time frame of 133 days. As noted in Figure S2, sampling on D58 and D90 occurred during or after the presence of wildfire smoke in the region; we speculate that increases in observed PAHs on glass and filter samples are a result of wildfire smoke penetrating into the office. Figure S2 shows that indoor  $\text{PM}_{2.5}$  concentrations were elevated during and prior to these sampling events.

In general, the highest decay constants are calculated for cotton (Flt, Pyr, BaA, Chr, and BaP, being the only PAHs to overlap between the three materials), while the lowest were for the filter material. Concentrations of PAHs on cotton decreased by nearly an order of magnitude within the two first weeks. We speculate the cotton bedsheet may have contained unsaturated  $\text{C}=\text{C}$  bonds that could have interacted with ozone in the office (present at measurable concentrations, see Figure S2 in Supporting Information) to generate hydroxyl radical that contributed to the observed PAH decay. It is also possible that the organic cotton bedsheet harbored a microbial community that differed from other materials; microbes may contribute to PAH degradation.<sup>35</sup> However, it must be noted that only one replicate of cotton was available for day 14, the second replicate on this day having low recovery of the surrogate standard (chrysene-D12, < 40%). Figure 2, panel D shows that decay is slower for filter material, which may be explained by the fact that the filter traps particles in a complex porous matrix that also provides substantial surface area for partitioning of PAHs. The filter has a higher background PAH concentration; this may be a result of manufacturing processes which involve heating of organic raw materials, e.g., polypropylene, to create the fibrous matrix.<sup>43</sup>

This study explored the possibility of photoenhanced degradation of surface PAHs on glass, since PAHs can absorb light in the UV and visible light wavelengths, leading to excited energy states that can initiate electron transfer reactions.<sup>44</sup> Environmental conditions measured in each location over the aging period are shown in Figure S6. The natural light location received, on average, more visible light and had periods of much higher light intensity than those placed in artificial light; however, the light sensor used could not distinguish the wavelength of light and only measured in the visible spectrum.



**Figure 3.** Time series values of PAH concentrations for smoke-exposed samples. PAH concentrations are summed for the suite of compounds quantified for each material: (A) exposed glass samples (9 PAHs, blue), (B) exposed cotton (7 PAHs, gray), and (C) exposed filters (10 PAHs, yellow). The experimental values represent the average  $\pm$  range of the two replicates for each day. The dashed lines is the summed PAH concentration, calculated using the decay constant for each PAH taken from the spiked sample experiments and the experimentally measured concentration of the smoke-exposed sample on Day 0. Values presented have subtracted the negative control measurement, which are reported in Figure S5 in the Supporting Information.

The decay of the sum of nine PAHs detectable above the MDL on glass at each location is similar across the two sets of samples ( $0.025 \pm 0.005 \text{ day}^{-1}$  vs  $0.023 \pm 0.005 \text{ day}^{-1}$ , Table S7). Looking at individual PAHs, 7 of 9 PAHs have similar decay constants (within uncertainty, in this case the standard error of the regression, shown in Table S7) when comparing natural light to artificial light. One compound, chrysene, had slightly lower decay in the natural light location, while dibenz[*a,h*]anthracene had a greater decay rate in natural light. These results imply that photoenhanced removal of PAHs from glass, under the lighting conditions studied here, was generally within measurement and model uncertainty. This may be due to attenuation of light, especially in the UV spectrum, by the windows in the indoor environment studied here. Future work should investigate windows common to households that may allow differing UV light penetration to the PAH loaded surfaces than those windows studied here.

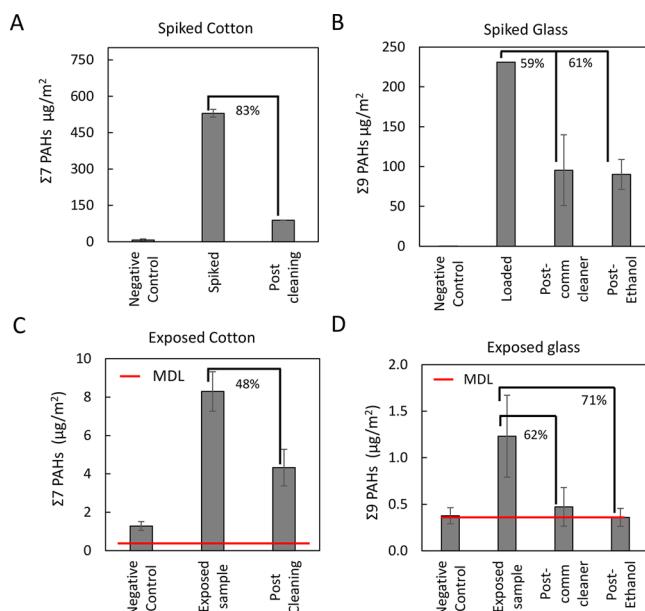
**3.3. Retention of PAHs on Smoke-Exposed Surfaces.** Glass, cotton, and filter samples were exposed to smoke and aged in the artificial light location (filters) or natural light location (glass and cotton) for up to 133 days as previously described. As shown in Figure 3, levels of PAHs decrease dramatically within the first  $\sim 40$  days, similarly to the spiked samples. After 37 days, levels decreased by 74% for the filter material, by 88% for glass material, and by 81% for cotton. Experimental values for each individual PAH measured are provided in Tables S8, S9, and S10 for glass, cotton and filters, respectively. Note that the low value observed for cotton on day 14 is due to a high background and a single replicate on this specific day.

The decay curves (dashed lines, Figure 3) were predicted using first-order constants calculated from the spiked samples (Figure 2) and the initial measured concentration of each PAH on the smoke-exposed sample over 133 days of aging. In general, our model shows good fit to the experimental values, implying our smoke exposed samples are behaving similarly to the spiked samples, especially for glass ( $R^2 = 0.94$ ), and cotton ( $R^2 = 0.76$ ). Our model for filter material explains less variance than that for glass and cotton ( $R^2 = 0.58$ ), generally

underestimating the decay. This may be due to the filter media trapping a larger fraction of the smoke matrix than other tested materials, resulting in more pronounced differences in PAH behavior for spiked vs smoke exposed filters than other materials. Goodness of fit would be improved for cotton (to  $R^2 = 0.97$ ) if day 14 were removed, as this day represents a single replicate. Our results indicate decay constants calculated using spiked samples appear reasonably representative of the persistence occurring on smoke-exposed samples.

**3.4. Efficacy of Cleaning Processes.** Cleaning processes were evaluated on spiked and exposed surfaces for two materials: cotton and glass. Filters studied here are disposable, and there is no relevant cleaning process. Generally, the cleaning processes studied show relatively high effectiveness, with some differences between cleaning of spiked and smoke exposed materials. Figure 4A and B show high removal of PAHs on spiked cotton (83%) and spiked glass ( $\sim 60\%$ ) respectively. The cleaning process has a lower efficacy for smoke-exposed cotton (48%) compared to spiked cotton (83%) (panels A and C, Figure 5). Concentrations of each PAH are listed in Tables S11 and S12 for spiked and exposed samples, respectively. We hypothesize that smoke-exposed cotton samples contain particle-bound PAHs and that these particles are trapped by the cotton, reducing the ability of the cleaning agents to physically remove the PAHs that are retained by the material. It is possible that spiked samples may more closely represent PAH accumulation that would result from vapor-phase accumulation while smoke exposed materials also include PAHs present from particle deposition; there may exist the need for tailored interventions depending on the pathway of PAH accumulation.

Figure 4B shows that, on spiked glass, the two cleaning agents (a commercial glass cleaner and ethanol) show a similarly high removal efficiency (59% and 61% respectively). As shown in Figure 4D, removal efficiency is similar for exposed materials (62% and 71% respectively for the commercial glass cleaner and ethanol). Overall, cleaning processes are generally efficient, with 48–83% removal. This is in contrast with results from the study of Banks et al.,<sup>26</sup>



**Figure 4.** Mean concentration of the sum of PAHs pre- and postcleaning for (A) spiked cotton, (B) spiked glass, (C) exposed cotton, and (D) simulated wildfire smoke exposed glass with the percentage removal postcleaning. The red line shows the MDL and “comm cleaner” refers to a commercially available glass cleaner tested.

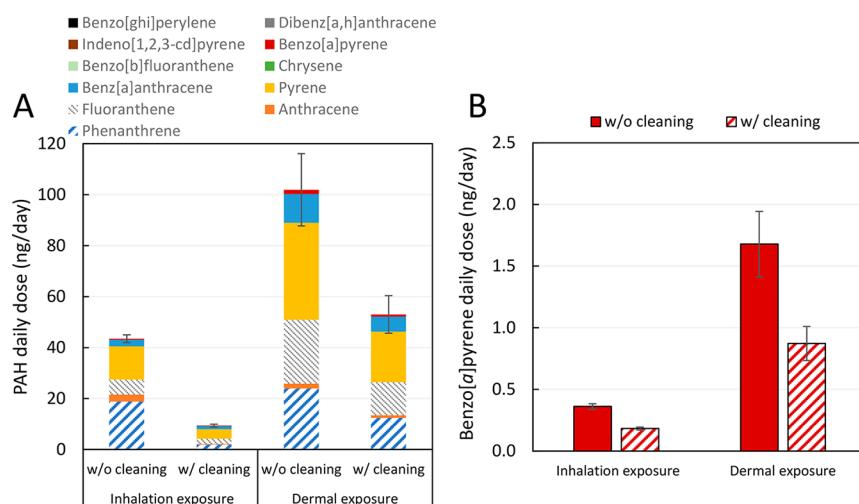
where no statistically significant reduction in measured PAHs was observed when comparing firefighters’ t-shirts after engaging a house fire and postlaundry. For a container burn (diesel pan fires set in a shipping container), a 23% reduction in the sum of 13 PAHs postlaundry t-shirts was measured. The higher efficacy for cleaning cotton determined in the present study of cotton bedsheets may be due to differences in the materials; firefighter t-shirts likely differ from the cotton bedsheets used here in physical characteristics and chemical composition. There also exist differences in analytical methods to recover PAHs from materials as well as laundering methods across the studies. Finally, the origin and exposure conditions to smoke differ dramatically; firefighters are in close

proximity to the source and PAH emission factors and size distributions of particulate-phase PAHs vary with combustion conditions and fuel.<sup>45</sup> Future studies should explore such complex interactions to further understand the efficacy of laundering of smoke-impacted indoor materials.

**3.6. Exposure Assessment.** We conducted a scaling analysis to estimate the magnitudes of exposures to PAHs postsmoke event due to retention and assumed re-emission of PAHs on select indoor materials. This analysis allows comparison of the relative contribution of inhalation vs dermal exposures in a simulated room. The average daily doses for inhalation and dermal exposure were predicted for a week in a model bedroom during 8 h sleeping periods following a wildfire smoke event. We then compare the daily doses that would occur if cleaning materials and replacement of filters were immediately performed after the smoke event ends, with the initial PAH concentration calculated using the cleaning efficiencies shown in Figure 4. Cleaning and replacement include wiping glass with a commercial cleaner, laundering of cotton, and changeout of the MERV13 filters for new filters. Figure 6 shows a summary of this exposure analysis for the sum of PAHs studied in this work and for benzo[a]pyrene, specifically, a known carcinogenic PAH.

Daily doses for inhalation and dermal exposures for total PAHs (Panel A, Figure 5) are 43 and 102 ng/day, respectively, in the week following a smoke event, absent cleaning of materials, and removal of filters. Total PAHs includes those with different IARC classifications (Table S1, Supporting Information), with some being classified as known or possibly carcinogenic. There exist comparatively fewer studies of health impacts of dermal exposure to PAHs than inhalation or oral pathways; dermal exposure is often considered through soil contamination. With other subsequently noted limitations of this assessment, exposure and dose calculations should be interpreted with caution. However, we believe this analysis implies post smoke event exposure to surface-associated PAHs may warrant attention.

Figure 5A shows that summed across all PAHs studied here, doses are greater for dermal contact than inhalation. We note that this comparison is limited to the materials studied here;



**Figure 5.** Comparison of daily doses calculated for inhalation exposure and dermal exposure pre and post cleaning for (A) sum of the total PAHs and (B) for benzo[a]pyrene for 1 week after material exposure to smoke. Doses calculated use an initial PAH loading consistent with nine h of fir and cedar combustion, i.e., day 0 shown in Figure 3. Cleaning (replacement, in the case of filters) is assumed to occur immediately after the wildfire smoke event ends. Inhalation exposure is based on all materials tested while dermal exposure is based on cotton only.

there exists substantial additional surface area indoors that would alter the magnitudes shown in Figure 5. However, these results show potential for previously unrecognized PAH exposure pathways due to indoor wildfire smoke. For example, in the case of fluoranthene, if loss from surfaces is due to re-emission to indoor air, a dose of 6 ng/day is present, due to an average increase in bedroom air concentrations of 2.5 ng/m<sup>3</sup> from re-emission over a 1-week period. This concentration is greater than a tentative guideline value of 2 ng/m<sup>3</sup>.<sup>46</sup> Figure 5A shows the inhalation of re-emitted benzo[a]pyrene (B[a]P), known to be carcinogenic for humans,<sup>47</sup> from filters, cotton and glass alone contributes a dose of ~0.4 ng/day. This is 8% of the 4.8 ng/day that would result from an 8-h exposure to the U.S. EPA-determined reference concentration for inhalation exposure (2 ng/m<sup>3</sup>).<sup>48</sup> Bostrom et al. suggest a health-based guidelines for B[a]P exposure of only 0.1 ng/m<sup>3</sup>,<sup>46</sup> noting lifetime exposure to this concentration would result in 1 excess cancer case in 100 000 individuals. Epidemiological studies show air concentrations of B[a]P greater than 1 ng/m<sup>3</sup> can damage DNA.<sup>49</sup> Again, assuming that observed surface losses of B[a]P are due solely to re-emission, an increase of 0.15 ng/m<sup>3</sup> would be observed in air in the simulated bedroom.

Figure 5B shows that dermal contact to B[a]P results in an ~5× greater daily dose than does inhalation of B[a]P re-emitted from materials. Combining B[a]P re-emission and dermal contact results in a daily dose of ~2.1 ng/day, or nearly half of the dose that would result from 8 h exposure to the U.S. EPA reference concentration for this compound. Health outcomes of exposures are likely to differ for dermal vs inhalation pathways, e.g., lung cancer risks are likely associated with inhalation exposure while skin cancer associated with dermal exposure.<sup>50</sup> The results, however, indicate that further investigation into multiple exposure routes is warranted as there exists a potential for negative health outcomes associated with dermal exposures.

Results shown in Figure 5 demonstrate that daily doses are substantially reduced (48–78%) when surfaces have been cleaned (glass and cotton) and replaced (filters) immediately following the event. In total, daily doses for inhalation exposure are estimated to be 43 ng/day without cleaning and 9.4 ng/day with cleaning and removal, a reduction of ~80%. A reduction of ~50% is observed for dermal contact daily doses, from 102 ng/day without cleaning to 53 ng/day with cleaning. In the case of B[a]P (Figure 5B), daily doses for inhalation exposure are estimated at 0.36 ng/day without cleaning and 0.18 ng/day with cleaning, ~50% reduction. Given the increasing impact of wildfire smoke across major population centers, these straightforward cleaning activities may play a role in alleviating long-term health impacts due to exposure to PAHs after wildfire smoke events. Future studies should incorporate airborne PAH measurements in parallel with surface PAH measurements, to compare magnitudes of exposures to PAHs during wildfire smoke events to those received after the smoke event. Such studies are motivated, in concept, by increasing recognition of thirdhand cigarette smoke exposure;<sup>51</sup> this study indicates a similar exposure pathway may exist for wildfire smoke.

It is important to note that estimation of daily dose is based on PAH loadings assumed to be consistent with those resulting from 9 h of material exposure to a wildfire smoke event proxy. Wildfire smoke events are variable in duration and are derived from diverse fuels, especially at the wildland-urban interface; study of different biogenic and anthropogenic fuels is

warranted. Here, surface PAHs increased with increasing smoke exposure from a specific biomass fuel under lab conditions (Figure S4). This implies that postsmoke event daily doses for inhalation and dermal exposures will increase with the duration of the wildfire smoke event. It also must be noted that the exposures and doses estimated here consider only a narrow subset of materials, activities, and periods of the day that may contribute to personal PAH exposures. Further, our model assumed that surface decay was due solely to re-emission to air; surface transformations likely occurred that resulted in the production of PAH reaction products in air and on surfaces, with differing toxicities, which are not considered here and should be a focus of subsequent experiments. Further study of the aforementioned factors on inhalation and dermal exposures to PAHs in indoor spaces impacted by wildfire smoke is warranted during and after the smoke event.

## ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.est.3c05547>.

Additional details concerning the PAHs used in this study, conditions during wildfire smoke proxy generation, sample placement and environmental conditions during aging, GC-MS methodological details, PAH recovery, method detection limits, and values of PAH surface concentrations for each species included in the studies reported here ([PDF](#))

## AUTHOR INFORMATION

### Corresponding Author

Elliott T. Gall – *Department of Mechanical and Materials Engineering, Portland State University, Portland, Oregon 97201, United States; [orcid.org/0000-0003-1351-0547](#); Email: [gall@pdx.edu](mailto:gall@pdx.edu)*

### Author

Aurélie Laguerre – *Department of Mechanical and Materials Engineering, Portland State University, Portland, Oregon 97201, United States; [orcid.org/0000-0002-8167-3586](#)*

Complete contact information is available at: <https://pubs.acs.org/10.1021/acs.est.3c05547>

### Author Contributions

A.L. contributed data curation, formal analysis, investigation, methodology, visualization, writing - original draft, and writing - review and editing; and E.T.G. contributed conceptualization, funding acquisition, investigation, methodology, project administration, resources, supervision, validation, visualization, writing - original draft, and writing - review and editing.

### Notes

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