

Impacts of Air Cleaners on Indoor Air Quality in Residences Impacted by Wood Smoke

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Supporting Information

ABSTRACT: Residential wood combustion is an important source of ambient air pollution, accounting for over 25% of fine particulate matter (PM_{2.5}) emissions in Canada. In addition to these ambient contributions, wood smoke pollutants can enter the indoor environment directly when loading or stoking stoves, resulting in a high potential for human exposure. A study of the effectiveness of air cleaners at reducing wood smoke-associated PM_{2.5} of indoor and outdoor origin was conducted in 31 homes during winter 2009–10. Day 1, the residents' wood burning appliance operated as usual with no air cleaner. Days 2 and 3, the wood burning appliance was not operational and the air cleaner was randomly chosen to operate in "filtration" or "placebo filtration" mode. When the air cleaner was operating, total indoor PM_{2.5} levels were significantly lower than on placebo filtration days ($p = 0.0001$) resulting in a median reduction of 52%. There was also a reduction in the median PM_{2.5} infiltration factor from 0.56 to 0.26 between these 2 days, suggesting the air cleaner was responsible for increased PM_{2.5} deposition on filtration days. Our findings suggest that the use of an air cleaner reduces exposure to indoor PM_{2.5} resulting from both indoor and ambient wood smoke sources.



INTRODUCTION

Residential wood combustion is an important source of ambient air pollution, accounting for over 25% of fine particulate matter (PM_{2.5}) emissions in Canada.¹ Since wood smoke emissions often occur in residential areas and pollutants can also enter the indoor environment directly when loading or stoking stoves, they also have a high potential for human exposure.^{2–5} This is of concern since exposure to wood smoke has been associated with declines in lung function,⁶ increasing respiratory symptoms in children^{7,8} and increased emergency room visits.^{7,9}

Indoor and outdoor wood burning appliances, e.g., stoves, wall insert fire places, furnaces, and outdoor wood boilers, are widely used for space and water heating and to a lesser extent cooking in the wintertime in the Annapolis Valley, Nova Scotia. In a recent source apportionment analysis conducted in the Annapolis Valley, Nova Scotia, Canada, the mean wood smoke

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contribution to ambient $PM_{2.5}$ during winter 2008/09 was 56.2% (range 32.6% - 73%).¹⁰ This suggests a significant impact of wood smoke emissions on a local scale. As well, the Annapolis Valley's topography is conducive to trapping emissions especially during thermal inversions (typically 2 to 5 severe inversions occur per winter) leading to elevated mean 24-h $PM_{2.5}$ concentrations greater than $20 \mu\text{g}/\text{m}^3$.¹⁰

A small number of studies have identified air cleaners as an inexpensive method of reducing indoor $PM_{2.5}$.¹¹⁻¹⁴ Mechanistically, air cleaners contribute to reduced particle concentrations by increasing the decay rate (k). Therefore, air cleaner use in homes impacted by wood smoke could be reasonably expected to lower $PM_{2.5}$ of both indoor and ambient origin, as well as decrease the infiltration factor (F_{inf}), which is the fraction of the outdoor air particle concentration that penetrates the building envelope and remains suspended in indoor air.^{15,16}

In order to better understand the efficacy of air cleaners in improving indoor air quality in homes impacted by wood smoke, a study of indoor and outdoor $PM_{2.5}$ was conducted in the Annapolis Valley, Nova Scotia between December 2009 and April 2010. Days with the air cleaner in use were compared to days with placebo filtration. During both filtration and placebo filtration days, the wood stove was not in operation in order to assess the impact of $PM_{2.5}$ of ambient origin on indoor air quality. Days when the wood stove was operational were compared to days with placebo filtration in order to better understand the impact of the wood-burning appliance itself on indoor air quality. Total indoor $PM_{2.5}$, F_{inf} and the components of indoor $PM_{2.5}$ including levoglucosan were examined.

■ EXPERIMENTAL SECTION

The study design, methodology, and instrument validation have all been described in detail elsewhere.¹⁷ Briefly, between December 2009–April 2010 Health Canada and Dalhousie University conducted a study of residential outdoor and indoor air quality in the Annapolis Valley, Nova Scotia. Monitoring was completed for 31 homes for three consecutive days each, with two homes monitored concurrently per week. Indoor monitoring equipment was placed on the main floor, typically in the living room. Outdoor monitoring equipment was located in the backyard, several meters away from the home. All monitors were located at breathing height (1.5 m).

On day 1, participants used their indoor wood burning appliance as normal. On the second day, the indoor wood burning appliance was not operated. An air filtration unit (Filtrete Ultra Clean Air Purifier model series FAP02-RS, 3M, London, ON, Canada) with a 3M patented air filter was placed on the opposite side of the room in relation to the air quality monitors and was randomly assigned to operate in "filtration" or "placebo filtration" mode (where the internal filter was replaced with a placebo filter) on the "high" setting. On the third day, the wood burning appliance remained inactive, and the air cleaner was in the opposite mode to the previous day. On days 2 and 3, supplementary electric heating was provided to the home owners. Two technicians visited each home daily to change out the monitoring equipment and administer questionnaires. Ethics approvals to conduct the study were obtained from both the Health Canada and Dalhousie University Research Ethics Boards.

Continuous, near real-time (1 min integration) $PM_{2.5}$ mass concentrations were measured inside and outside each home simultaneously using TSI DustTraks (Model 8520, TSI, St.

Paul, MN) operating at a flow rate of 1.7 L min^{-1} . The DustTrak monitors have been shown to overestimate compared to gravimetric $PM_{2.5}$ methods and as such will be referred to as fine particles (FP).¹⁸ MS&T area samplers (37 mm diameter, 2 μm pore size, ring supported PTFE filters, Air Diagnostics and Engineering Inc. Harrison, ME) and Thermo ChemCombs (47 mm diameter, Prefired Quartz filters, Model 3500, Thermo Scientific, Waltham, MA, U.S.A.) were also deployed at these locations to collect 24-h $PM_{2.5}$ samples. Both operated at a flow rate of 10 L min^{-1} using SKC Inc. Leland Legacy pumps (SKC Inc., Eighty Four, PA, U.S.A.) with flow rate checks performed pre and post sampling using a TSI 4140 digital mass flow meter (TSI, St. Paul, MN). An end flow rate change of $\pm 20\%$ was deemed acceptable. Pre and post sample gravimetric measurements of the 37 mm diameter PTFE filters were conducted in accordance with USEPA quality assurance guidelines.¹⁹ The quartz filters were analyzed for levoglucosan, a wood smoke chemical marker, at the University of Montana, following a method summarized in Bergauff et al.²⁰⁻²² Briefly, the quartz filter was spiked with a deuterated recovery standard, placed in a vial, and extracted by ultrasonication using ethyl acetate containing 3.6 mM triethylamine. The extract was filtered, and reduced in volume to approximately 500 μL before being evaporated to dryness and derivatized with N-Obis-(trimethylsilyl)trifluoroacetamide, trimethylchlorosilane, and trimethylsilylimidazole to convert the sugar anhydrides to its trimethylsilyl derivatives. Sample fractions were analyzed by gas chromatography/mass spectrometry on a Hewlett-Packard GC/MSD (GC model 6890, MSD model 5973, Hewlett-Packard Company, Palo Alto, CA, U.S.A.) using an HP-5 MS capillary column or equivalent. Following this analytical method, minimum detection limits for levoglucosan were 185.0 ng/mL .²⁰ The PTFE filters were analyzed by XRF and future manuscripts will utilize the data for source apportionment analyses.

Air exchange rates (AER) were measured inside the home by the perfluorocarbon tracer (PFT) technique.²³ Four PFT emitters were deployed on day 1 and taken down at the end of sampling on day 3. A daily capillary adsorption tube (CAT) placed in the main living area was used as the receptor for the PFT tracer gas in all homes. In a subset of 8 homes, a second CAT was placed in the basement. For each home, the air exchange rate, expressed as air changes per hour (ACH), was calculated by dividing the air flow rate by the technician-measured house volume. In the subset of homes with two CATs, the air exchange rate was determined using the volume-weighted measured concentrations in the basement and living area.

A 3M Filtrete FAP02-RS model air cleaner was used as they are designed to operate in rooms up to 15.8 m^2 , a condition met by the primary room sampled in each home. The published clean air delivery rates (CADRs) for this model are $3.6 \text{ m}^3/\text{min}$, $2.9 \text{ m}^3/\text{min}$, and $4.2 \text{ m}^3/\text{min}$ for dust, tobacco smoke, and pollen, respectively.²⁴ Prior to the sampling period in each home, a new 3M filter was installed. A Davis Vantage Pro II weather station (Davis Instruments Corp. Hayward, California 94545 U.S.A.) was deployed outside each home, which recorded temperature (T), pressure (mb), and relative humidity (RH) at 15 min intervals. Indoor temperature (T) and relative humidity (RH) were recorded at 1 min intervals using a YesTek monitor (Model 206 Falcon).

Descriptive statistics for daily FP, levoglucosan and air exchange are presented for each experimental day. Since no

differences were observed for T and RH by experimental day, descriptive statistics are presented on an aggregate basis only. Indoor/outdoor ratios for each experimental day were calculated for the daily FP and associated levoglucosan.

Estimates of F_{inf} were created for each experimental day by dividing the mean censored indoor concentration by the mean outdoor concentration (censored I/O ratio). This approach has been shown to agree well with other methods of estimating F_{inf} ¹⁶ and has the added benefit of maximizing the number of F_{inf} estimates available. (See also Supporting Information, SI, for results of applying 3 alternative methods for determining F_{inf} .) Indoor peaks were removed using a previously developed censoring algorithm.²⁵ The start of an indoor peak was identified when the indoor concentration increased at least $5 \mu\text{g}/\text{m}^3$ from the previous half hour, and either the indoor concentration increase exceeded a concurrent outdoor increase in concentration or the indoor concentration was greater than the outdoor concentration. The end of a peak was identified when the indoor concentration was within $2 \mu\text{g}/\text{m}^3$ of the background concentration. The background concentration was considered to be the concentration in the hour prior to the start of the peak. All half hours within the peak were assigned a censored value equal to the background concentration. Indoor, outdoor, and censored concentrations were then plotted to ensure that all indoor generated peaks were removed, and that the algorithm had not overcensored indoor levels that were likely due to outdoor fluctuations. Where necessary, data were manually censored to resolve these issues. An example of this censoring method output is shown (Figure 1).

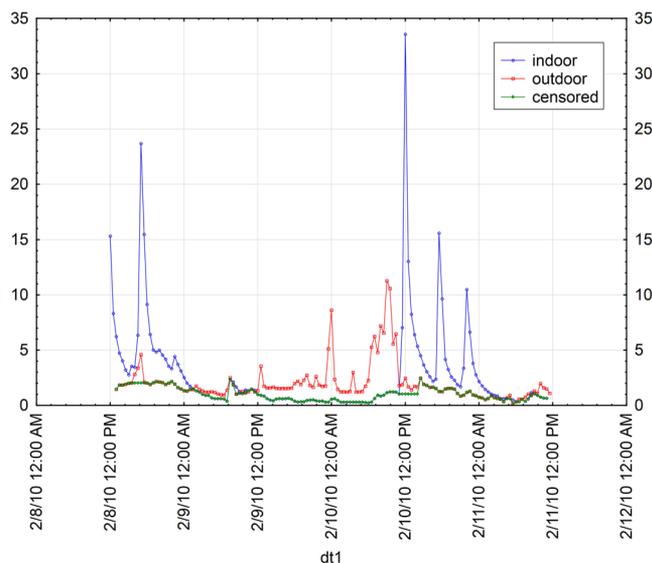


Figure 1. DustTrak measurements indoors (blue), outdoors (red), and censored indoors (green). At times, the indoor measurements are not censored (no apparent sources) and the green curve overlays (obscures) the blue one.

In order to calculate a valid estimate of F_{inf} at least 75% of the daily data were required to create both the censored indoor mean and outdoor mean.

As shown in eqs 1 and 2, the home specific estimates of F_{inf} were used to estimate the ambient component ($C_{in(a)}$) and the indoor-generated component ($C_{in(ig)}$) of the total indoor FP concentration. In cases where the ambient component exceeded the total indoor concentration, the ambient

concentration was set to the total measured indoor concentration, and the indoor-generated component was set to zero.

$$C_{in(a)} = F_{inf} \times C_{out} \quad (1)$$

$$C_{in(ig)} = C_{in} - C_{in(a)} \quad (2)$$

A Wilcoxon signed rank test was performed to assess differences between experimental days. All data analyses were completed using SAS EG V4.2 (SAS Institute Inc., NC, U.S.A.). All graphs were prepared using Statistica v10 (Statsoft, Inc., OK, U.S.A.).

RESULTS AND DISCUSSION

Housing Characteristics. Housing characteristics for all homes are presented in Table 1. Approximately half of the

Table 1. Housing Characteristics

category	N (homes)	% total
primary wood burning appliance		
free standing wood stove	21	67.7
wall insert	3	9.7
pellet stove	1	3.2
wood furnace	3	9.7
free standing and wall insert	1	3.2
wood furnace and wall insert	1	3.2
outdoor wood boiler	1	3.2
garage type		
no garage	16	51.6
detached garage	8	25.8
attached garage with no door	2	6.5
attached garage with connecting door	5	16.1
number of days per week wood burning appliance in use		
5 to 7 days	28	90.3
3 to 5 days	2	6.4
1 to 2 days	1	3.2
construction year		
1945 and before	14	45.2
1946 to 1960	3	9.7
1961 to 1980	7	22.6
1981 to 2000	4	12.9
2001 or later	3	9.7

homes included in the study were built prior to 1945 ($n = 14$). The primary heating fuel source was wood, or a combination of wood with oil or wood with electricity ($n = 24$). The size of the homes ranged from <200 to 350 m^2 with only 7 homes having attached garages.

QA/QC. Prior to the commencement of the study, DustTraks were assessed for precision and bias by running them side by side to determine bias-corrected precision.^{18,26} This resulted in a bias-corrected mean precision of 6% (std. dev. 3%; range 3–10%). The limit of detection (LOD) of DustTrak monitors has been estimated at 3–5 DustTrak units, corresponding to $1\text{--}2 \mu\text{g}/\text{m}^3$ gravimetric equivalent.¹⁸ Similarly, the LOD of the $\text{PM}_{2.5}$ gravimetric method has been reported to be $7.5 \mu\text{g}/\text{filter}$.²⁷ Our contract laboratory reports an MDL of $20 \mu\text{g}/\text{filter}$ (personal communication, Xintang Feng, Alberta Innovates). For our 10 Lpm pumps, this corresponds to about $1.3 \mu\text{g}/\text{m}^3$. More than 80% of the DustTrak and gravimetric $\text{PM}_{2.5}$ samples exceeded these nominal LODs. A regression of the outdoor DustTrak

Table 2. Descriptive Statistics for Environmental Measurements

measurement	experiment condition	location	N	min	25th	median	75th	max	median I/O ratio (min–max)
FP ($\mu\text{g}/\text{m}^3$) (daily averages)	wood burning appliance on	indoor	31	4.63	7.74	9.88	14.13	246.32	1.11 (0.17–117.8)
		outdoor	29	2.09	5.67	8.73	11.68	61.97	
	filtration	indoor	31	0.90	1.81	3.17	6.13	21.26	0.42 (0.08–2.29)
		outdoor	29	0.91	6.35	7.66	14.10	65.28	
	placebo filtration	indoor	29	2.60	4.16	8.58	13.57	64.42	1.42 (0.24–19.72)
		outdoor	31	0.51	2.93	5.90	11.57	35.66	
PM _{2.5} (gravimetric) ($\mu\text{g}/\text{m}^3$)	wood burning appliance on	indoor	29	0.43	3.35	4.15	6.31	83.71	1.24 (0.05–20.7)
		outdoor	31	0.64	2.09	2.90	3.05	14.43	
	filtration	indoor	19	0.35	0.81	1.92	4.63	11.28	0.63 (0.08–5.78)
		outdoor	30	0.42	1.75	2.51	5.22	22.60	
	placebo filtration	indoor	32	0.37	1.56	3.87	8.17	30.19	1.20 (0.03–24.0)
		outdoor	22	0.34	1.79	3.67	5.78	115.97	
levoglucosan (ng/m^3)	wood burning appliance on	indoor	27	ND*	0.059	0.085	0.134	0.689	0.49 (0.06–10.80)
		outdoor	29	ND	0.071	0.151	0.260	1.557	
	filtration	indoor	19	ND	ND	0.034	0.078	0.189	0.17 (0.00–0.77)
		outdoor	18	0.055	0.113	0.169	0.416	3.258	
	placebo filtration	indoor	19	ND	0.036	0.050	0.102	0.448	0.36 (0.00–14.62)
		outdoor	20	0.021	0.068	0.096	0.282	1.458	
temperature ($^{\circ}\text{C}$)	all days	indoor	95	15.24	19.21	20.76	21.85	24.64	n/a
		outdoor	92	−14.37	−1.39	1.40	4.56	11.29	
relative humidity (%)	all days	indoor	95	26.00	30.16	33.04	38.13	45.07	n/a
		outdoor	95	41.44	74.35	85.75	90.67	96.47	
AER (air exchanges/h)	wood burning appliance on	indoor	32	0.13	0.34	0.46	0.66	1.23	n/a
	filtration	indoor	30	0.08	0.24	0.36	0.55	1.06	n/a
	placebo filtration	indoor	32	0.13	0.27	0.39	0.67	1.16	n/a

*Not detected.

Table 3. Descriptive Statistics for the Infiltration Factor and the Indoor-Generated and Ambient Components of Indoor Fine Particle Concentration

components by experimental condition	n	min	25 th	median	75 th	max	median ambient (min–max) (%)
wood burning appliance on							
F_{inf}	26	0.14	0.48	0.58	0.71	0.96	51 (0.5–94)
indoor-generated component ($\mu\text{g}/\text{m}^3$)	26	0.62	2.83	3.97	7.72	245.01	
ambient component ($\mu\text{g}/\text{m}^3$)	26	1.26	3.66	4.82	7.36	11.96	
filtration							
F_{inf}	28	0.07	0.15	0.26	0.40	0.84	79 (11–100)
indoor-generated component ($\mu\text{g}/\text{m}^3$)	28	0.00	0.30	0.65	2.23	18.16	
ambient component ($\mu\text{g}/\text{m}^3$)	28	0.31	1.49	2.26	3.39	7.57	
placebo filtration							
F_{inf}	28	0.16	0.39	0.56	0.72	1.00	59 (2–100)
indoor-generated component ($\mu\text{g}/\text{m}^3$)	28	0.00	1.06	2.38	10.97	63.19	
ambient component ($\mu\text{g}/\text{m}^3$)	28	0.76	2.14	3.52	6.07	13.76	

concentrations on the PM_{2.5} values resulted in a slope of 0.94 (standard error 0.05) and an intercept of 0.24 (SE 0.30) $\mu\text{g}/\text{m}^3$ ($n = 78$) with an adjusted R^2 of 0.79. The corresponding regression for indoor concentrations gave a slope of 0.87 (SE 0.02) and an intercept of -0.71 (SE 0.26) $\mu\text{g}/\text{m}^3$ ($n = 75$) and adj. $R^2 = 0.96$. Since the DustTrak and gravimetric PM_{2.5} samples agreed quite well, the DustTrak data presented in this manuscript have not been corrected to their gravimetric equivalent. The LOD for levoglucosan was estimated to be 185 ng/filter.²⁰ Nine indoor and two outdoor samples were below the LOD. Limited data were available for the levoglucosan results with only 14 residences having valid comparable data across the different sampling days, primarily as a result of pump failures.

Descriptive Statistics. When the air cleaner was in operation, indoor FP levels were significantly lower (median = 3.17 $\mu\text{g}/\text{m}^3$) than on days when the placebo filtration (median = 8.58 $\mu\text{g}/\text{m}^3$) was employed ($p = 0.0001$) (Table 2).

Overall, the air cleaner resulted in a significant reduction of 52% in median indoor FP concentrations compared to days with placebo filtration (Table 3). The air cleaner use also was associated with reductions of 42% and 30% in the median concentrations of levoglucosan on the other 2 days, but neither reduction achieved significance at the $p = 0.05$ level.

FP F_{inf} . FP F_{inf} estimates can be found in Table 4 and Figure 2. Median FP F_{inf} estimates were 0.58 (wood burning appliance on), 0.26 (filtration), and 0.56 (placebo filtration). Corresponding estimates using the sulfur indoor/outdoor ratio were nearly identical: 0.55, 0.25, and 0.52 (Table S1 in Supporting

Table 4. Change in Environmental Concentrations Related to Experimental Conditions

comparisons across experimental conditions ^{a,b}	<i>n</i>	median % change	median difference	<i>p</i> ^c	# of homes with decrease
indoor FP ($\mu\text{g}/\text{m}^3$)					
placebo filtration vs wood burning appliance on	28	-16	-1.65	0.5	18
filtration vs placebo filtration	29	-52	-3.48	0.0001	23
ambient component of indoor FP ($\mu\text{g}/\text{m}^3$)					
placebo filtration vs wood burning appliance on	25	-25	-1.31	0.2	17
filtration vs placebo filtration	27	-40	-1.44	<0.0001	21
indoor-generated component of indoor FP ($\mu\text{g}/\text{m}^3$)					
placebo filtration vs wood burning appliance on	25	-37	-1.08	0.9	15
filtration vs placebo filtration	27	-71	-1.44	0.01	20
outdoor FP ($\mu\text{g}/\text{m}^3$)					
placebo filtration vs wood burning appliance on	29	-35	-2.58	0.2	20
filtration vs placebo filtration	28	-2	-0.22	0.7	14
indoor levoglucosan (ng/m^3)					
placebo filtration vs wood burning appliance on	16	-42	-0.06	0.6	10
filtration vs placebo filtration	14	-30	-0.02	0.2	9
outdoor levoglucosan (ng/m^3)					
placebo filtration vs wood burning appliance on	17	0	0.00	0.9	8
filtration vs placebo filtration	13	28	0.02	1.0	5

^a% change for placebo filtration vs woodburning appliance on = (placebo filtration - woodburning appliance on)/(woodburning appliance on) \times 100. ^b% change for filtration vs wood burning appliance on = (filtration - placebo filtration)/(placebo filtration) \times 100. ^cWilcoxon Ranked Sum Test.

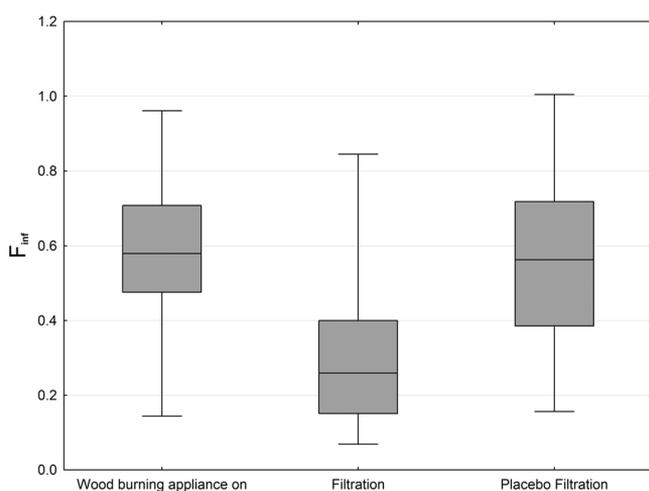


Figure 2. Boxplot of F_{inf} estimates by experimental day. Boxplots show medians, 25–75 percentiles (box) and min–max (whiskers).

Information, SI). Also, despite a lack of detailed agreement ($R^2 \leq 50\%$), all three methods of estimating F_{inf} agreed in showing that F_{inf} was reduced by about a factor of 2 by filtration. F_{inf} estimates were not significantly different between days with the wood burning appliance on and days with placebo filtration ($p = 0.8$).

F_{inf} was significantly different between days with filtration and placebo filtration ($p < 0.0001$). This suggests that the reduction in F_{inf} on filtration days (0.26) is a result of the air cleaner increasing the deposition rate (k) of particles. A rough estimate of this increase in k due to the air filter is on the order of $0.6\text{--}0.7\text{ h}^{-1}$, compared to a value of $0.2\text{--}0.3\text{ h}^{-1}$ when the filter was off. (Note that k appears in the denominator of the expression for $F_{\text{inf}} = Pa/(a + k)$, where P is the penetration factor and a the air exchange rate.) This calculation is described further in the SI.

A recent study in British Columbia, Canada reported that mean indoor $\text{PM}_{2.5}$ concentrations were reduced from 5.8 to

$3.9\text{ }\mu\text{g}/\text{m}^3$, and mean F_{inf} estimates were reduced from 0.28 to 0.10, when using a HEPA filter in 19 homes in a wood smoke impacted community.¹¹ A Montana study of two homes measured PM with a different method than was used in this study; they were however able to demonstrate a 61% reduction in $\text{PM}_{2.5}$ when comparing operating and nonoperating Filtrete air filters, which compares with our findings.¹² In a Danish study of 27 residences impacted by heavy traffic emissions, the impact of a particle filtration unit was assessed in an indoor PM intervention study. The authors were able to estimate during the active filtration that the median $\text{PM}_{2.5}$ concentration was 54.5% lower (57.4% and 51.7% for bedroom and living room, respectively) compared with the median during the placebo filtration. For both rooms, the difference was statistically significant ($p < 0.01$, independent sample t test).¹⁴ A Swedish personal, indoor and outdoor study conducted to identify the impacts of domestic wood burning on exposure compared homes with a wood stove to a set of reference homes. They found elevated, but not significantly different, $\text{PM}_{2.5}$ in homes with wood burning (median = $12\text{ }\mu\text{g}/\text{m}^3$ vs $9.5\text{ }\mu\text{g}/\text{m}^3$ $p = 0.278$).²⁸

Total Indoor FP Concentrations and Indoor Components. Similar results to the total indoor FP were observed for the corresponding ambient-generated and indoor-generated components (see Tables 3 and 4). That is, both the ambient-generated and indoor-generated components were higher on days when the wood burning appliance was in operation (indoor-generated median = $3.97\text{ }\mu\text{g}/\text{m}^3$; ambient-generated median = $4.82\text{ }\mu\text{g}/\text{m}^3$) compared to days when placebo filtration was employed (indoor-generated median = $2.38\text{ }\mu\text{g}/\text{m}^3$; ambient-generated median = $3.52\text{ }\mu\text{g}/\text{m}^3$). These differences were not statistically significant, possibly a result of the limited sample size. However, when the air cleaner was in operation, both components of the indoor FP decreased significantly compared to placebo filtration (indoor-generated median = $0.65\text{ }\mu\text{g}/\text{m}^3$, $p = 0.01$; ambient-generated median = $2.26\text{ }\mu\text{g}/\text{m}^3$, $p < 0.0001$). Overall, the air cleaner resulted in a

71% median reduction in the indoor-generated component and a 40% median reduction in the ambient-generated component.

Levoglucosan. Levoglucosan, a sugar anhydride from the incomplete combustion of cellulose, has been identified as a selective organic marker of biomass combustion.^{29–32} Ambient levoglucosan concentrations have been measured in multiple airsheds, including Seattle, WA (13–760 ng/m³),³³ Spokane, WA (2–327 ng/m³),³⁴ Fresno and Bakersfield, CA (23–7590 ng/m³),³⁵ and Libby, MT (1980–3700 ng/m³).³⁶ Indoor residential levoglucosan concentrations have been measured from 238.1 ± 310.1 to 1050 ± 1027 ng/m³ in wood burning communities.^{21,37} As demonstrated in Table 2, the concentrations measured in this study were much lower in both the indoor and ambient environments compared to these other published studies.

I/O Ratios. Table 2 presents the I/O ratios by sample day for fine particles and levoglucosan concentrations. Median I/O ratios for FP were 1.11, 0.42, and 1.42, while the median I/O ratios for levoglucosan were 0.49, 0.17, and 0.36 when the wood burning stove was on, during filtration and during placebo filtration, respectively. This suggests that a large contribution of wood smoke indoors results from both ambient sources and from the wood stove when it is operating. The comparatively lower median I/O ratio found when the air cleaner was in use suggests that the air cleaner was effective at removing some of the infiltrated FP and levoglucosan from the indoor air. Results were also highly variable by home, as seen in Table 2, suggesting that for some homes there were significant indoor or outdoor sources influencing the ratios.

Limitations of the study include only having 1 day per scenario in each home. This was a result of the need to complete all sampling during the winter season and having limited access to monitoring equipment. An additional day of monitoring to compare when the woodstove was operational with an air cleaner running would have been beneficial for explaining the ambient contribution. The lack of available levoglucosan data to compare across the different experimental days made it difficult to identify the true sources of particulate matter. It is also unknown how the gas-particle partitioning for levoglucosan changed across the samples collected, and how the semivolatile nature of levoglucosan under different temperatures influenced our sample results.³⁸ This should be investigated in future studies. A factor contributing to the possibility for error was the extremely low level of outdoor PM concentrations, sometimes dropping below 1 μg/m³ for a daily average as measured by the gravimetric samplers. At such low levels, the DustTrak instruments can display zero offsets and zero drift, leading to errors of about the same magnitude as the measurements.¹⁸

■ ASSOCIATED CONTENT

● Supporting Information

Calculation of infiltration factor; regressions of measured (blue) and censored indoor DustTrak concentrations on measured outdoor values (Figure S1); distribution of F_{inf} estimates for three experimental conditions using three methods (Table S1); comparison of two methods for determining F_{inf} (Figure S2); calculation of deposition rate for homes and for the filtration unit; estimates of deposition rates k and k_{filter} for 3 values of the penetration factor P (Table S2); and additional references. This material is available free of charge via the Internet at <http://pubs.acs.org/>.

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Notes

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■ REFERENCES

- (1) Lepage, M.; Boulton, J. W. Source Apportionment of Particulate Matter in Canada. 2000. RWDI Group, Guelph, Ontario.
- (2) Allen, R.; Leckie, S.; Millar, G.; Brauer, M. The impact of wood stove technology upgrades on indoor residential air quality. *Atmos. Environ.* **2009**, *43*, 5908–5915.
- (3) Glasius, M.; Ketzel, M.; Wahlin, P.; Jensen, B.; Monster, J.; Berkowicz, R.; Palmgren, F. Impact of wood combustion on particle levels in a residential area in Denmark. *Atmos. Environ.* **2006**, *40*, 7115–7124.
- (4) Larson, T.; Su, J.; Baribeau, A. M.; Buzzelli, M.; Setton, E.; Brauer, M. A spatial model of urban winter woodsmoke concentrations. *Environ. Sci. Technol.* **2007**, *41*, 2429–2436.
- (5) Sexton, K.; Spengler, J. D.; Treitman, R. D. Effects of residential wood combustion on indoor air-quality—A case-study in Waterbury, Vermont. *Atmos. Environ.* **1984**, *18*, 1371–1383.
- (6) Koenig, J. Q.; Larson, T. V.; Hanley, Q. S.; Rebolledo, V.; Dumler, K.; Checkoway, H.; Wang, S. Z.; Lin, D. Y.; Pierson, W. E. Pulmonary-function changes in children associated with fine particulate matter. *Environ. Res.* **1993**, *63*, 26–38.
- (7) Larson, T. V.; Koenig, J. Q. Wood smoke: Emissions and non-cancer respiratory effects. *Ann. Rev. Public Health* **1994**, *15*, 133–156.
- (8) Norris, G.; Young Pong, S. N.; Koenig, J. Q.; Larson, T. V.; Sheppard, L.; Stout, J. W. An association between fine particles and asthma emergency department visits for children in Seattle. *Environ. Health Perspect.* **1999**, *107*, 489–493.
- (9) Norris, G.; Larson, T.; Koenig, J.; Claiborn, C.; Sheppard, L.; Finn, D. Asthma aggravation, combustion, and stagnant air. *Thorax* **2000**, *55*, 466–470.
- (10) Gibson, M. D.; Ward, T. J.; Guernsey, J. R.; Wheeler, A. J.; Seaboyer, M.; King, G. H.; Stieb, D. M. Wood smoke source apportionment and home infiltration study in the Rural Annapolis Valley, Nova Scotia, Canada. Extended abstract in the *Conference Proceedings of the 103rd Air & Waste Management's Annual Conference & Exhibition*, Calgary, Alberta, Canada, June 22–25, 2010.
- (11) Barn, P.; Larson, T.; Noullett, M.; Kennedy, S.; Copes, R.; Brauer, M. Infiltration of forest fire and residential wood smoke: An evaluation of air cleaner effectiveness. *J. Expo. Anal. Environ. Epidemiol.* **2008**, *18*, 503–511.
- (12) Hart, J. F.; Ward, T. J.; Spear, T. M.; Rossi, R.; Holland, N.; Loushin, B. Evaluating the effectiveness of a commercial portable air purifier in homes with wood burning stoves—A preliminary study. *J. Environ. Public Health.* **2011**, Article ID 324809, 1–7.

- (13) Skulberg, K. R.; Skyberg, K.; Kruse, K.; Eduard, W.; Levy, F.; Kongerud, J.; Djupesland, P. The effects of intervention with local electrostatic air cleaners on airborne dust and the health of office employees. *Indoor Air* **2005**, *15* (3), 152–159.
- (14) Spilak, M. P.; Karotki, G. D.; Kolarik, B.; Frederiksen, M.; Loft, S.; Gunnarsen, L. Evaluation of building characteristics in 27 dwellings in Denmark and the effect of using particle filtration units on PM_{2.5} concentrations. *Building Environ.* **2014**, *73*, 55–63.
- (15) Allen, R.; Larson, T.; Sheppard, L.; Wallace, L.; Liu, L. J. Use of real-time light scattering data to estimate the contribution of infiltrated and indoor-generated particles to indoor air. *Environ. Sci. Technol.* **2003**, *37*, 3484–3492.
- (16) MacNeill, M.; Wallace, L.; Kearney, J.; Allen, R.; Van Ryswyk, K.; Judek, S.; Xu, X.; Wheeler, A. Factors influencing variability in the infiltration of PM_{2.5} mass and its components. *Atmos. Environ.* **2012**, *61*, 518–532.
- (17) Wheeler, A. J.; Gibson, M. D.; Ward, T.; Allen, R. W.; Guernsey, J. R.; Seaboyer, M.; Kutcha, J.; Gould, R.; Stieb, D. *Reductions in Residential Wood Smoke Concentrations and Infiltration Efficiency using Electrostatic Air Cleaner Interventions*. 12th International Conference on Indoor Air Quality and Climate, June 5–10, 2011, Austin, TX, USA.
- (18) Wallace L.; Wheeler A. J.; Kearney J.; Van Ryswyk K.; You H.; Kulka R.; Rasmussen P.; Brook J. Xu X. Validation of Continuous Particle Monitors for Personal, Indoor, and Outdoor Exposures. *J. Expo. Anal. Environ. Epidemiol.* **2011**, Advance online publication, 26 May 2010; doi:10.1038/jes.2010.15.
- (19) USEPA, Quality Assurance Guidance Document 2.12. Monitoring PM_{2.5} in Ambient Air Using Designated Reference or Class 1 Equivalent Methods, Human Exposure and atmospheric Sciences Division, U.S. Environmental Protection Agency: Research Triangle Park, NC, 1998.
- (20) Bergauff, M.; Ward, T.; Noonan, C.; Palmer, C. P. Determination and evaluation of selected organic chemical tracers for wood smoke in airborne particulate matter. *Int. J. Environ. Anal. Chem.* **2008**, *88*, 473–486.
- (21) Ward, T.; Palmer, C.; Bergauff, M.; Hooper, K.; Noonan, C. Results of a residential indoor PM_{2.5} sampling program before and after a woodstove changeout. *Indoor Air* **2008**, *18*, 408–415.
- (22) Gibson, M. D.; Kuchta, J.; Chisholm, L.; Duck, T.; Hopper, J.; Beauchamp, S.; Waugh, D.; King, G.; Pierce, J.; Li, Z.; Leitch, R.; Ward, T. J.; Palmer, P. I. Receptor modelling of boreal wildfire associated PM_{2.5} in Halifax, Nova Scotia, Canada. 10th EGU General Assembly, Vienna, April 7–12 2013. *Geophys. Res. Lett.*, Vol 15. EGU2013–1604. <http://meetingorganizer.copernicus.org/EGU2013/EGU2013-1604-4.pdf>. Accessed June 4, 2014.
- (23) Dietz, R. N.; Goodrich, R. W.; Cote, E. A.; Wieser, R. F. Detailed description and performance of a passive perfluorocarbon tracer system for building ventilation and air exchange measurements. *Am. Soc. Testing Mater.*, Special Technical Publication, Philadelphia. 1986. 904 203–264.
- (24) Association of Home Appliance Manufacturers (AHAM). AHAM Directory of Certified Products, Room Air Cleaners, 2010. http://www.ahamdir.com/aham_cm/site/pages/search_airCleaners.html.
- (25) Kearney, J.; Wallace, L.; MacNeill, M.; Xu, X.; Van Ryswyk, K.; You, H.; Kulka, R.; Wheeler, A. J. Residential indoor and outdoor ultrafine particles in Windsor, Ontario. *Atmos. Environ.* **2011**, *45*, 7583–7593.
- (26) McNamara, M. L.; Noonan, C. W.; Ward, T. J. Correction factor for continuous monitoring of wood smoke fine particulate matter. *Aerosol Air Qual. Res.* **2011**, *11*, 315–322 NIHMSID: 344195.
- (27) Annual data summary report for the chemical speciation of PM_{2.5} filter samples project: January 1 through December 31, 2011. Prepared for US EPA by RTI International. 3040 Cornwallis Road, Research Triangle Park, NC 27709. RTI 0212943/3.
- (28) Molnár, P.; Gustafson, P.; Johannesson, S.; Boman, J.; Barregård, L.; Sällsten, G. Domestic wood burning and PM_{2.5} trace elements: Personal exposure, indoor and outdoor levels. *Atmos. Environ.* **2005**, *39*, 2643–2653.
- (29) Caseiro, A.; Bauer, H.; Schmid, C.; Pio, C. A.; Puxbaum, H. Wood burning impact on PM₁₀ in three Austrian regions. *Atmos. Environ.* **2009**, *43*, 2186–2195.
- (30) Jordan, T. B.; Seen, A. J.; Jacobsen, G. E. Levoglucosan as an atmospheric tracer for woodsmoke. *Atmos. Environ.* **2006**, *40*, 5316–5321.
- (31) Schauer, J. J.; Kleeman, M. J.; Cass, G. R.; Simoneit, B. R. T. Measurement of emissions from air pollution sources. 3. C1-C29 organic compounds from fireplace combustion of wood. *Environ. Sci. Technol.* **2001**, *35*, 1716–1728.
- (32) Schmid, C.; Marr, I. L.; Caseiro, A.; Kotianova, P.; Berner, A.; Bauer, H.; Kasper-Giebl, A.; Puxbaum, H. Chemical characterization of fine particle emissions from wood stove combustion of common woods growing in mid-European Alpine regions. *Atmos. Environ.* **2008**, *42*, 126–141.
- (33) Simpson, C. D.; Paulsen, M.; Dills, R. L.; Liu, L. J.; Kalman, D. A. Determination of methoxyphenols in ambient atmospheric particulate matter: Tracers for wood combustion. *Environ. Sci. Technol.* **2005**, *39*, 631–637.
- (34) Jimenez, J.; Wu, C. F.; Claiborn, C.; Gould, T.; Simpson, C.; Larson, T.; Liu, L. J. S. Agricultural burning smoke in eastern Washington: Part I. Atmospheric characterization. *Atmos. Environ.* **2006**, *40*, 639–650.
- (35) Schauer, J. J.; Cass, G. R. Source apportionment of winter-time gas-phase and particle-phase air pollutants using organic compounds as tracers. *Environ. Sci. Technol.* **2000**, *34*, 1821–1832.
- (36) Ward, T. J.; Rinehard, L.; Lange, T. The 2003/2004 Libby, Montana PM_{2.5} Source Apportionment Research Study. *Aero. Sci. Technol.* **2006**, *40*, 166–177.
- (37) Ward, T. J.; Boulafentis, J.; Simpson, J.; Hester, C.; Moliga, T.; Warden, K.; Noonan, C. W. Results of the Nez Perce woodstove changeout program. *Sci. Total Environ.* **2011**, *409*, 664–670.
- (38) Xie, M.; Hannigan, M.; Barsanti, K. Gas/Particle Partitioning of 2-methyltetrols and levoglucosan at an urban site in Denver. *Environ. Sci. Technol.* **2014**, *48* (5), 2835–2842.