FINAL REPORT

Assessing the Influence of Indoor Air Filtration on the Health of Asthmatic Children in China

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Sampling locations in Songjiang, China.

Executive Summary

Although filtration of indoor air is common in many regions of the world where there are harmful levels of air pollution, the health benefits of filtration are unclear. This is particularly true in China, where large urban regions including Beijing and Shanghai have air pollution concentrations well above guidelines for human health set by the World Health Organization (WHO). With this in mind, we conducted a pilot study in Beijing and a full study in Shanghai that involved assessing the influence of indoor air filtration on the respiratory health of asthmatic children. The Shanghai study involved the homes of 43 asthmatic children in a crossover approach that comprised a period of indoor filtration to remove fine particulate matter (PM₂₅) and volatile organic compounds (VOCs), as well as a period of sham filtration, when filters were removed from the filtration devices. Real-time, low-cost air pollution monitors were developed specifically for the project to measure PM $_{2.5}$ and ozone (O $_{3}$), and time integrated sampling was undertaken to measure the composition of PM $_{2.5'}$ as well as concentrations of specific VOCs. Overall, indoor filtration reduced concentrations of PM_{2.5} by ~68% in children's bedrooms. However, as exposures are dependent on the daily activity patterns of the children the reductions in overall PM_{2.5} exposure were, on average, ~27%. The chemical characteristics of PM_{2.5} indoors pointed to the infiltration of outside air (i.e., from mobile sources and re-suspended dust). PM_{2.5} indoors also included a relatively large fraction of particulate organic carbon associated with indoor sources such as cooking and cleaning products, to name a few. In addition, indoor air filtration reduced the oxidative potential of PM₂₅. Ozone concentrations indoors were substantially lower than outdoors during both true and sham filtration due to the deposition to indoor surfaces. The overall concentrations of VOCs were generally much higher indoors than outdoors, with indoor concentrations, on average, 30% lower during true than sham filtration. Unique indoor sources were identified for VOCs including cooking, consumer products, and furniture. Overall, indoor filtration reduced concentrations of both PM_{2.5} and VOCs indoors, with both having unique indoor sources, as well as contributions from outdoor sources.

Perhaps most importantly, the reduction in the exposure to indoor air pollutants due to filtration was associated with several health benefits including improved airway mechanics, increased peak expiratory flow and reduced pulmonary inflammation, and improved small airway function in some participants. This underscores the importance of reducing exposure to air pollution, in particular PM_{2.5}, in order to promote children's health. In addition, this study serves as a framework for future studies that will assess the impacts of air pollution interventions on the health of children.

1.0 Introduction

A substantial amount of research has been undertaken in China in the last few decades to better understand the mixture of ambient pollutants in large cities, their sources, and related health impacts. Ambient PM_{25} is the primary environmental risk factor influencing both morbidity and mortality, and in 2015 it was estimated to be responsible for 4.2 million deaths globally.**¹** It is the primary environmental pollutant influencing both morbidity and mortality in China. Although it should be pointed out that people spend the majority of their time indoors, and indoor pollutants may disproportionately affect health. This is especially true with respect not only to PM_{2.5} but volatile organic compounds (VOCs) as well, which tend to be measured at higher concentrations indoors than outdoors and which have a plethora of indoor sources. It should also be pointed out that the chemical composition of both PM₂₅ and VOCs, and the related source contributions to indoor chemical species, are relatively poorly characterized in indoor environments in China. Furthermore, the links between indoor and outdoor pollutant concentrations and personal exposure need to be better quantified to determine true pollutant exposures.

While the use of air purifiers can be a quick and simple way to minimize inhalation exposure, little is known about the components of PM_{2.5} and the VOCs that are removed by purifiers under real-world conditions in homes in highly polluted urban locales such as Beijing and Shanghai. Further lacking is an understanding of how these improvements in air quality translate to reductions in personal exposure. Lastly, little research has been conducted to determine how an intervention such as the use of an air purifier might affect respiratory health among individuals such as asthmatic children who are particularly susceptible to these pollutants. With this in mind, we brought together a multi-disciplinary team of scientists, engineers, and physicians and developed and implemented a study to assess the influence of indoor air filtration on the health of asthmatic children in Shanghai. This work involved: the development of novel, low-cost, real-time air quality sensors deployed as outdoor, indoor, and personal exposure monitors; detailed characterization of the chemical composition of PM_{2.5} and VOCs; and the measurement of key health endpoints in asthmatic children related to lung function. Our results illustrate clear benefits of indoor filtration for children due to the reduction in indoor air pollution and personal exposure. Furthermore, through this work we developed a clear and effective framework that will serve as a basis for future air pollution and health exposure studies; this framework will be critical for improving the health of children using interventions including filtration and activity modification.

Figure 1: Sampling locations in Songjiang, China. All sampling sites from the Shanghai study are shown; sensor calibration was done at the EPA monitoring station, and additional sampling was done on the rooftop of the Shanghai First People's Hospital to understand temporal trends in PM_{2.5} during the study.

2.0 Approach

This project comprised two field campaigns; the first, in Beijing, China, in July/August 2016 (the "Beijing pilot"), was conducted to evaluate methodologies prior to the full study. During this pilot we tested our equipment and methods in the homes of nine adults. The second campaign (the "Shanghai project" or the "full study") was undertaken in Songjiang (a suburb of Shanghai), China, in February-April 2017. Here we expanded upon the Beijing pilot study to sample in the homes of 43 asthmatic children (**Figure 1**). While no health measurements were made during the Beijing pilot, the asthmatic children enrolled in the Shanghai study underwent measurements of airway mechanics, spirometry, and fractional exhaled nitric oxide (FeNO) at the beginning and the end of each period of filtration, and they, along with their caregivers, completed a childhood asthma control test (C-ACT) at each visit. Participants also measured their peak expiratory flow (PEF) twice daily at home. By pairing C-ACT results and measurements of lung function, we were able to assess whether within-person changes in the C-ACT score are associated with changes in measurements of lung function, airway mechanics, and lung inflammation – something that had not previously been done.

Figure 2: Overview of the sampling approach for the Shanghai study. This approach was used for each group of households, with half undergoing sham filtration first and the other half undergoing true filtration first. This schedule was applied to four groups of homes (~10 homes/group) with homes in each group undergoing sampling at approximately the same time. Twenty homes participated in VOC sampling.

During the field campaigns, stand-alone air purifiers were used in homes for one period of true filtration (i.e., with pre-, activated carbon, and high-efficiency particulate air [HEPA] filters installed) and a second period of sham filtration (i.e., with coarse pre-filter only). Participants were asked to keep doors and windows closed during periods of filtration. We collected stationary, time-integrated measurements of PM₂₅, ozone, and VOCs indoors and immediately outdoors (e.g., on a balcony, patio) at homes. We also collected data and samples to understand personal exposure to PM₂₅ and ozone, with these data collected for 48 hours during each period of filtration (**Figure 2**). During personal exposure sampling, participants were asked to complete an hourly time-activity diary indicating their approximate location during that time (e.g., indoors in the room with the purifier, outdoors, in the classroom/office). VOC measurements were made for 90-minute periods during each intervention.

Time-integrated sampling allowed us to investigate the complexity of the mixture of pollutants in these environments. For PM_{2.5}, we assessed the oxidative potential of the components (i.e., their ability to generate reactive oxygen species [ROS], which are precursors to oxidative stress), which has been linked to some of the adverse health effects associated with exposure to PM. For both PM_{2.5} and VOCs, we identified potential sources of pollutants, investigated differences in the contributions of sources indoors and outdoors to air quality, and assessed the impact of purifier use on pollutants and source contributions. We complemented time-integrated sampling with the collection of minute-resolved data for PM_{2.5} and ozone for stationary and exposure measurements; these data were used to investigate temporal and smallerscale spatial differences in pollutants. The combined results – of the mixture of pollutants, their oxidative potential, and temporal trends in pollution – can be leveraged to identify the best solutions for improving indoor air quality, personal exposure, and health in these environments.

3.0 Findings

3.1 CHARACTERIZING THE INDOOR ENVIRONMENT - POLLUTANT CONCENTRATIONS, FILTRATION EFFECTS, AND SOURCES

$PM_{2.5}$

During both field campaigns, PM_{2.5} concentrations were higher outdoors than indoors. An examination of the timeresolved data and indoor-to-outdoor ratio (I/O) of pollutants, which accounts for differences in ambient concentrations over time and can point to whether pollutants are of indoor or outdoor origin, illustrated that much of the PM_{2.5} measured indoors was infiltrating from outside. Differences in concentrations and I/O by filtration status were illustrative of a benefit of filtration for air quality indoors.

In Beijing, PM_{2.5} concentrations were, on average (± standard deviation), 49±6 μ gm⁻³ indoors during sham filtration (outdoors: 60±4 μgm-3) and 13±2 μgm-3 indoors during true filtration (outdoors: 63±5 μgm-3).**²** The extent of infiltration varied by home. In Shanghai (Figure 3), 40 of the 42 homes experienced lower concentrations of PM_{2.5} indoors during true than sham filtration, with reductions in I/O ranging from 31% to 94% across homes. For many homes, filtration meant the difference between concentrations exceeding the WHO's 24-hour guideline value of 25 μgm⁻³ and meeting these guidelines;³ in all microenvironments except the bedroom during true filtration, mean concentrations of PM_{2.5} exceeded the WHO guidelines.**⁴**

Figure 3: Summary of hour-averaged PM₂₅ and ozone concentrations for all homes in the Shanghai study. These data collected using low-cost monitors illustrate the higher concentrations of PM_{2.5} and ozone outdoors as compared to indoors and highlight the benefit of filtration for PM_{2.5} indoors and, to a lesser extent, for personal exposure to PM $_{\textrm{\tiny{2.5}}}$. $^{\textrm{\tiny{3}}}$

As expected, concentrations of ozone were substantially higher outdoors than indoors and followed a diurnal trend, with higher concentrations measured later in the day. The low concentrations indoors are likely due to rapid natural loss to surfaces in the indoor environment, especially as doors and windows remained closed. Additionally, concentrations often fell below or near the limit of detection of the monitors (mean limit of detection: 7.0 ppb); there was no noticeable difference in ozone by filtration status.**3-5**

The composition of the PM_{2.5} differed indoors and outdoors, with particulate organic matter - which has many indoor sources including cooking, use of cleaning products, and dust – dominating PM₂₅ indoors (Figure 4).⁶ Indoor to outdoor differences in pollutant composition are of great importance as these differences underpin the health effects related to exposure. In Beijing, components of PM_{2.5} - with the exception of organic carbon - were lower during true than sham filtration.⁷ Similarly, in Shanghai, components of PM₂₅ including organic matter, elemental carbon, sulfate, trace elements and dust were generally lower during true than sham filtration indoors.**6**

Based on elements quantified from filter samples and using variable reduction techniques to pinpoint those that grouped together, sources of indoor PM_{2.5} in Shanghai were identified as resuspended dust, regional aerosol, roadway emissions, residual oil combustion, alloy steel abrasion and a La and Ce source. With the exception of the latter two, these are ambient sources. A number of the elements quantified are hazardous or transition metals hypothesized to be of health-relevance (i.e., Cr, Mn, Co, Ni, As, Cd, Sb, Pb, Ti, V, Fe, Cu, Zn and Mo). Roadway emissions, regional aerosol and residual oil combustion were most strongly associated with oxidative potential of the PM_{2.5}, and elements associated with the other three sources had low oxidative potential. Importantly, there were significant differences in the contributions of the six sources of PM_{2.5} to indoor pollution for all sources except resuspended dust and the La and Ce source under the two filtration scenarios, and filtration was associated with lower total oxidative potential indoors indicating that filtration may reduce the particulate pollution indoors and in doing so, remove some of the components of PM that are likely to be the most toxic.**6,8**

Figure 4: PM2.5 component mass concentrations. Results are separated by filtration status; **EC**=elemental carbon; **dust**=sum of dust oxides; **OM**=organic matter (1.4 x organic carbon); **trace elements**=sum of elements quantified by ICP-MS. *Indoor or outdoor data did not pass QA/QC.

VOCs

In Shanghai, VOCs were measured at higher concentrations indoors than outdoors, and total VOCs and numerous individual VOCs were measured at lower concentrations during true filtration than sham filtration, further illustrating the benefit of filtration on indoor air quality (**Figures 5** and **Figure 6**). Some compounds (e.g., octanal, heptanal, α-cedrene) were specific to the indoor environment, a few were ubiquitous (e.g., acetaldehyde, formaldehyde), and others were detected infrequently. Six source groupings – namely, vehicle emissions, cooking-related emissions, a hexane source, consumer products, solvents, and wooden products/materials (e.g., furniture, structural materials) – were identified using

Figure 5: Concentrations of total volatile organic compounds (TVOC). All outdoor data are combined, indoor data are separated by filtration status.

Figure 6: Volatile organic compounds measured at least 5 times in the Shanghai study. (*Left*) Patterns of compounds vs. underlying factors identified (n=6) using non-negative matrix factorization for all samples (n=79). (*Right*) Differences in concentrations (%) indoors calculated as the concentrations for (true-sham)/sham*100% for all compounds with statistically significant differences. The vertical blue line indicates no change in concentration between filtration periods. Positive values indicate higher concentrations during true than sham filtration.

variable reduction techniques. Some of these emissions were uniquely from indoor sources, while others (e.g., vehicle emissions) appeared to infiltrate from ambient sources.**⁹** Total VOCs were substantially higher in our work than in previous work in the US,**¹⁰** although on a compound-by-compound basis our values were sometimes higher and sometimes lower than those measured previously in China and elsewhere, likely indicating differences in building practices and materials used in construction, participant behaviors, and differences in study design (e.g., sampling time, location).

3.2 EFFECTS OF FILTRATION ON PERSONAL EXPOSURES AND HEALTH

In spite of marked differences in air quality indoors, differences in personal exposure to PM_{2.5} were less remarkable under the two filtration scenarios; some participants experienced lower personal exposures during true than sham filtration, but this was not the case for all participants.**⁴** This variation in filtration effects across participants was observed in both field campaigns. Personal exposures to PM_{2.5} and ozone often exceeded the concentrations measured indoors, and sometimes exceeded concentrations measured directly outside the home, indicating contributions to personal exposure from other highly polluted microenvironments (e.g., transit environments, school). In Shanghai, personal exposure to PM₂₅ ranged from 19 to 92 μgm-3 during sham filtration and 6 to 65 μgm-3 during true filtration.**⁴**

Pairing time-resolved data from the low-cost monitors carried for personal exposure with hourly time-activity data provided by the participants we examined the contributions of different environments to time-weighted personal exposure.**⁴** Although the concentrations in the bedroom were often relatively low compared to those in other environments (e.g., outdoors) (**Figure 7**), the high number of hours spent in this environment resulted in the bedroom, along with the classroom, being the greatest contributor to personal exposure to PM_{2.5} during sham filtration. On average, PM_{2.5} contributions from the bedroom were 55% lower during true than sham filtration. Similarly, despite low concentrations of ozone indoors compared to outdoors, the bedroom was the greatest contributor to ozone exposures. Understanding these

Figure 7: PM_{2.5} concentrations in micro-environments (*left***) and micro-environmental contributions to personal exposure (***right***). Although PM_{2.5}** concentrations in the bedroom were typically lower than in other micro-environments measured, they contributed disproporionately to personal exposures due to the overwhelming amount of time spent in this environment. As such, reductions in PM₂₅ that may be attributed to filtration proved beneficial for personal exposures.

contributions is of great importance when considering where, when and how to implement interventions to produce the greatest reductions in exposure and the largest gains for health.

For PM₂₅, filtration was associated with lower source contributions to personal exposure from regional aerosol and roadway emissions; differences for the remainder of the sources were not significant.**⁶** Perhaps most importantly, filtration was associated with lower total oxidative potential for personal exposure,**⁸** improved small airway function and mechanics, increased peak expiratory flow and reduced pulmonary inflammation**11, 12** (**Figure 8**).

Improvement (%) in physiological response to true filtration compared to sham filtration

Figure 8: Effects of filtration (true vs. sham) on respiratory markers. For all markers except PEF, mean differences and 95% confidence intervals for afterbefore intervention changes between true and sham filtration are shown, adjusted for covariates. For PEF, the point and the bar show mean difference and 95% confidence interval between average PEF values measured daily during true filtration and those similarly measured during sham filtration.

FeNO = fractional exhaled nitric oxide; R₅= airway resistance measured at 5Hz; R₂₀= airway resistance measured at 20Hz; R₅-R₂₀= small airway resistance; X₅= airway reactance measured at 5Hz; Fres = resonant frequency; PEF = peak expiratory flow; FEF₂₅₋₇₅= forced expiratory flow during 25% to 75% of FVC; FEV₁= forced expiratory volume during the 1st second; FVC = forced vital capacity.

These differences in microenvironmental contributions, PM composition, oxidative potential, and health measurements highlight the potential benefit of this intervention. These findings are of importance for the asthma community as conventional inhaled medications are unable to penetrate the small airways but play an important role in asthma and its management; use of an air purifier like the one used in this research may proffer health benefits that cannot be achieved using conventional asthma control strategies such as inhalers.

For the examination of the relationship between C-ACT scores and markers of lung function the results were mixed, with a two-point decrease in the C-ACT score – the minimal change of clinical importance – significantly and beneficially associated with some markers (i.e., forced expiratory volume in the first second [FEV₁], forced vital capacity [FVC], airway resistance at 5 Hz [R $_{\rm s}$]), but not others. In addition, three of the four questions answered by the children on the C-ACT, but none of the questions answered by caregivers, were significantly associated with indicators of lung function.**¹³** This indicates the potential utility of the C-ACT responses – a tool that can be easily used at home – over time as a marker of changes in large airway function and resistance when it is not possible to obtain hospital-based measurements.

Lastly, although we did not make measurements of personal exposure to VOCs, we assessed the inhalation health risks presented by VOCs in the home during nighttime sleep and estimated the removal effectiveness of the air purifier on these VOCs. Seven compounds presented an inhalation cancer risk above the acceptable risk of 1x10⁻⁶ and four compounds exceeded the non-cancer risk threshold in the majority of homes.**¹⁴**

3.3 PERFORMANCE OF LOW-COST MONITORS

A key innovation of this study was to design, fabricate, and deploy low-cost monitors comprising sensors to measure PM $_{2.5}$ and O $_3$ for indoor/outdoor and personal exposure monitoring. With this in mind a great deal of effort involved laboratory and field evaluations, as well as the development of calibration procedures and quality assurance/quality control methodologies. The low-cost monitors developed at Duke University and used in this work performed well in side-by-side comparisons with reference monitors for both PM₂₅ and ozone concentrations.^{2, 3, 5} Hour-averaged ozone measurements from low-cost monitors (sensor: Alphasense OX-A4 and NO2-A4 series) were highly correlated (R²=0.9) with measurements from a reference analyzer during colocation in Shanghai and after a correction for relative humidity, PM_{2.5} measurements (sensor: Plantower PMS3003) also had strong correlations (R²=0.7).³ Over the Beijing pilot, sensors did not appear subject to drift, although this was not the case for the longer Shanghai study. Ozone sensors proved more variable than PM sensors, with a clear need for sensor-specific calibration constants, metrics of error, and determination of the limit of detection.**⁵** In contrast, PM sensors agreed well with both reference analyzers and each other. Our work was the first to develop, test, and implement low-cost sensors into a human health study and will serve as the framework for many future studies by our research team and others.

4.0 Summary

In conclusion, this timely work begins a thorough characterization of air pollutants in indoor, residential environments in urban China. It both illustrates and quantifies the potential benefit of using an air purifier in these environments to improve indoor air quality, to reduce personal exposure, and to improve the health of some of those most susceptible to the illeffects of air pollution: asthmatic children. Given the large population living in these types of environments and the poor air quality in these mega-cities, an understanding of how well these interventions can work can have broad impacts for health and provide individuals with some agency over the air quality in the spaces they occupy most frequently.TVOC and aldehyde levels are tens to over a hundred times higher relative to outdoors and that the sample duration of 1-hour vs. 4-hour results in a similar concentration.

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PUBLICATIONS

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