The Influence of Air Cleaners on Indoor Particulate Matter Concentrations and Oxidative Potential in Residential Households in Beijing

By

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A thesis submitted in partial fulfillment of the requirements for the degree of

Master of Science

(Civil and Environmental Engineering)

at the

University Wisconsin-Madison

Acknowledge

This thesis is dedicated to my parents, Guoping Zhan and Zhuhui Wang. I would not be where I am today without their unconditional support through the years.

First, I would like to thank my adviser, Professor James Schauer, who has given me priceless academic guidance since my junior year at the University of Wisconsin-Madison. I thank him for his patience and expertise in leading me into air pollution research and for helping me to become a better engineer. I also appreciate his advice on my life decisions, which has impacted me a lot. Also, many thanks to Dr. Martin Shafer and Professor Matthew Ginder-Vogel who have lent me help and advice on my research and papers. I would also like to thank my committee member, Professor Greg Harrington.

In addition, I would like to thank Professor Mike Bergin's research group at Duke University, Professor Mike Bergin and his advice on my research and my paper, and Karoline Johnson and Christina Norris for collecting samples in Beijing for this project. I am also thankful for the help from professors at Tsinghua University. I would like to thank Underwriters Laboratories as well, who funded this research.

Finally, I am thankful for the people I worked with in the Civil and Environmental Engineering department and the Environmental Chemistry and Technology Graduate program. A special thanks to labmates in our group, Alex Lai, Ahlam Hanai, Mike Olson, and others past and present, who make a friendly research environment. I would like to thank Chris Worley for his assistance and endless patience as well as Megan McConville, Tsuyoshi Takahashi, Semi Ozler, and many others for their precious friendship at EC&T, and I am glad to be part of the fantastic EC&T community. I am very thankful for my Chinese friends here in Madison who make my life much more colorful and accompany me through every new challenge.

Table of Contents

Abstract

In many developing regions where poor air quality is endemic, there is a rapidly growing number of air filtration devices available to clean indoor air. However, few studies have been performed to assess their effectiveness. This thesis studies the effectiveness of air cleaner at removing indoor air pollutants including $PM_{2.5}$ and its components, as well as ozone (O_3) and nitrogen dioxide (NO_2); the effectiveness in reducing oxidative potential, which is expressed by reactive oxygen species (ROS) activity, is investigated as well. Filter-based $PM_{2.5}$, O_3, and NO_2 samples were collected for outdoor, indoor, and personal exposure during the summer of 2016 in Beijing. This study shows that air cleaners can provide a healthier indoor environment, but they do not reduce personal exposure to particles and gas phase air pollutants nor work at reducing personal exposure the oxidative potential of fine particulate matter. Therefore, it is not entirely clear whether air cleaners will provide health benefit to users, and future research should focus on quantifying high pollutant exposure microenvironments to reduce the personal exposure. This thesis will start with the introduction to air pollutions; next, it will be results and discussion on the changes of $PM_{2.5,}$ its component and the oxidative potential of PM with air cleaner installation; finally, it is the conclusion and implication of this study. Followed by the appendix for analysis methods of ozone and nitrogen dioxide as well as the result.

Chapter 1 will give an introduction of ambient and indoor air pollution, including air pollution sources, main air pollutants, and their health effects. Then, there will be a review on indoor air pollution control strategies and previous studies on air cleaners. The final part is a statement describing the purpose of this research.

Chapter 2 is the results and discussion of the effectiveness of air cleaners at reducing $PM_{2.5}$ and its components as well as the oxidative potential of $PM_{2.5}$. This chapter will start with indoor to outdoor correlations, and the indoor to outdoor ratio (I/O) is used to assess the effectiveness of air cleaners. Then, there is personal exposure to the outdoor ratio (P/O), and the microenvironmental method is adopted to evaluate personal exposure. At the end of this chapter, ROS activity results for indoor, outdoor, and personal exposure are analyzed and discussed.

Chapter 3 is the conclusion of this thesis. It will summarize the results from this study and point out its limitations. It will also state the significance and implications of this research, ending with future research directions.

Appendix will talk about the ozone and nitrogen dioxide chemical analysis methods developed at Water Science and Engineering Lab at UW Madison. They are obtained by using Ogawa O_3/NO_x passive sampler, and are used employed to assess the O_3 and NO_2 levels at different environments. The results will be presented and briefly discussed. The method is set up for the study but the data from the pilot study was not sufficient for a chapter discussion but these methods can be used in future studies.

Chapter 1 Introduction

Air pollution has become a worldwide problem; in fact, 92% of the world's population lives in places where outdoor air quality fails to meet WHO guidelines in 2014[1]. Ambient air pollution in both cities and rural areas was estimated to cause 3 million premature deaths worldwide in 2012, and 88% of those premature deaths occurred in low- and middle-income countries, mostly within Southeast Asia and the Western Pacific Region[1]. Indoor air pollution in rural areas is mainly related to the emissions from inefficient solid biomass fuels (e.g., wood, animal dung, and crop waste) burning by open fires and simple stoves[16]. Each year, approximately 4.3 million people die prematurely from illness attributable to the household air pollution caused by the inefficient use of solid fuels (2012 data) for cooking[16]. Urban indoor air quality issues are often affected by the ambient air pollution, and as people move from rural areas to cities, the urban indoor air pollution becomes a serious concern for residences in megacities. WHO estimates that in 2012, 72% of outdoor air pollution-related premature deaths were due to ischemic heart disease and strokes, while 14% of deaths were due to chronic obstructive pulmonary disease or acute lower respiratory infections, and 14% of deaths were due to lung cancer[1]. From 2008-2013, global urban air pollution levels increased by 8%, despite improvements in some regions; specifically, in the Eastern Mediterranean and Southeast Asia regions and low-income countries in the Western Pacific Region, levels of urban air pollution increased by more than 5% in more than two-thirds of the cities[2]. The situation is particularly serious in quickly developing countries like China and India, which is due to the increasing urban size and population as well as the expanding energy demand and emissions from transportation and industries. In 2014, 92% of the population in China experienced more than 120 hours of unhealthy air (US EPA standard), and the population-weighted average exposure to $PM_{2.5}$ from the study was 52 $\mu g/m^3$[3]. In 2014, 37 cities from India were featured in the top 100 world cities with the worst PM_{10} pollution, and the cities of Delhi, Raipur, Gwalior, and Lucknow were listed in the top 10[4]. Therefore, air pollution around the world is an urgent issue that needs solving.

1.1 Ambient air pollution sources and main air pollutants

There are three main types of air pollution sources: mobile sources, stationary sources, and natural sources[5]. Mobile sources include both on-road vehicles (i.e., cars, trucks, and buses) and off-road equipment (i.e., ships, airplanes, agricultural, and construction equipment[5]). Stationary sources, including factories, refineries, boilers, and power plants, emit a variety of air pollutants from a single location, and these are also known as point sources of pollution[6]. There are two types of stationary sources that generate routine emissions of air toxics defined by the EPA: major sources are defined as sources that emit 10 tons per year of any of toxic air pollutants[7], or 25 tons per year of a mixture of air toxics; area sources consist of smaller-sized facilities that release lesser quantities of toxic pollutants into the air[6]. Natural sources include wind-blown dust, wildfires, and volcanoes, but usually they do not create air pollution problems like other sources can.

The 2005 "WHO Air Quality Guidelines[8]" offer global guidance on key air pollutants including particulate matters, ozone, nitrogen dioxide, and sulfur dioxide. Each country sets different acceptable limits for air toxics, and the national air quality standards also vary by country according to the approach adopted for balancing health risks, technological feasibility, economic considerations, and various other political and social factors[8]. Particulate matter (PM) is a mixture of solid particles and liquid droplets found in the air, and it includes PM_{10} which describes inhalable particles whose diameters are 10 micrometers and smaller, and $PM_{2.5}$ which describes fine inhalable particles with diameters 2.5 micrometers and smaller[9]. These particles are emitted directly from sources like construction sites, unpaved roads, fields, smokestacks, or fires, but most particles form in the atmosphere due to complex reactions of chemicals such as sulfur dioxide and nitrogen oxides, which are pollutants emitted from power plants and automobiles[9]. The range of health effects due to PM is broad, but these health effects predominantly relate to the respiratory and cardiovascular systems[8]. The WHO guideline for 24-hour averages of PM_{10} and $PM_{2.5}$ are 50 $\mu g/m^3$ and 25 $\mu g/m^3$, respectively. Tropospheric (ground level ozone) is created by chemical reactions between nitrogen oxides (NO_x) and volatile organic compounds (VOC), which are emitted by cars, power plants, industrial boilers, refineries, chemical plants, and other sources chemically reactive in the presence of sunlight[10]. Excessive ozone in the air can cause breathing problems, trigger asthma, reduce lung function, and cause lung diseases[8]. The WHO guideline value for an 8-hour mean is 100 $\mu g/m^3$. Nitrogen dioxide (NO_2) is one of a group of highly reactive gases known as oxides of nitrogen or nitrogen oxides (NO_x), and it is used as an indicator for the larger group of nitrogen oxides[11]. The major sources of anthropogenic emissions of NO_2 are combustion processes like fuel burning and power generation. NO_2 and other NO_x will interact with water, oxygen, and other chemicals in the atmosphere to form acid rain which can harm sensitive ecosystems as well as infrastructures in urban areas[11]. For public health, long time exposures to elevated concentrations of NO_2 may aggravate asthma and potentially increase the risk of respiratory infections. In addition, people with asthma, as well as children and the elderly are generally at a greater risk of being affecting by NO_2[11]. WHO suggests 200 $\mu g/m^3$ for a 1-hour mean of the NO_2 concentration[8]. SO_2 is used as the indicator for the entire group of gaseous sulfur oxides (SO_x) which provoke concerns as air pollutants [12]. The main anthropogenic source of SO_2 is the burning of sulfur-containing fossil fuels (coal and oil) for domestic heating, power generation, and motor vehicles[8]. SO_2 is also a main component of acid rain when it combines with water and forms sulfuric acid; in addition, SO_x and NO_x particles both contribute significantly to particulate matter pollution[12]. SO_2 can induce inflammation effects within the respiratory system, affect the functions of the lungs, and causes irritation of the eyes[1]. The 24-hour mean of SO_2 suggested by WHO is 20 $\mu g/m^3$. The standards and regulations of air pollutants are being thoroughly researched and developed to further reduce public health risks as well as benefit social economics.

1.2 Indoor air pollution sources and health effects

Indoor air quality (IAQ) refers to the air quality within and around buildings and structures and is related to the health and comfort of building occupants[13]. Since the 1970s, the level of interest in indoor air quality and health research has continued to rise as more people realize the serious impacts of indoor air pollution[14]. A growing body of scientific evidence has indicated that exposure to poor quality indoor air will produce more harmful health effects than poor quality outdoor air in the largest and most industrialized cities. Furthermore, some research indicates that people spend approximately 90 percent of their time indoors[15]. Vulnerable groups of people including very young or elderly people as well as chronically ill people, especially those who suffer from respiratory or cardiovascular disease, will spend even more time indoors and their exposure to pollutants will have a much more serious impact on them[15]. Therefore, exposure to indoor air pollution is of greater risk and should receive intensive attention.

Indoor air pollution sources can be clustered into three categories: pollution due to the fabrics of buildings and interior designs; emissions from indoor activities including cleaning, cooking, heating, etc.; and infiltration and penetration from outdoor sources. Toxic pollutants coming from the construction of buildings and interior decorations include asbestos for fire retardant and insulation, formaldehyde from furnishings, radon due to soil, concrete and stones, and other organic substances from solvents and building materials as well as allergens from household dust, animals, insects, plants, and internal surfaces[14]. These chemical and biological pollutants have confirmed adverse health effects which can be found in many literatures. It is a worldwide problem for urban residences and building codes, and regulations could help with this problem. Emissions from cooking and heating are mainly referring to poor households in low- and middle-income countries that use open fires and simple stoves with very incomplete combustion burning biomass (e.g., wood, animal dung, and crop waste) and coal[16]. Consequently, women and young children are at high risk of being exposed to high levels of indoor air pollution every day. Three billion people around the world still cook and heat their homes using unprocessed solid fuels, but the inefficient burning technologies and poor ventilating systems result in high levels of household air pollution with a range of health-damaging pollutants, including soot particles that can be 100 times greater than acceptable levels for fine particles[16]. In addition, women responsible for cooking and their young children are most heavily exposed[17]. It is estimated that about 2.5 million deaths each year result from indoor exposures to particulate matter in rural and urban areas in developing countries, accounting for 4-5% of the 50-60 million global deaths that occur annually[18]. Unfortunately, it is not an easy problem to fix: a solution must take into account the many aspects of household energy supply and utilization and should assess pollution and exposure reductions, fuel efficiency, and their impacts on local and global environment, safety, capacity to meet household needs, affordability, and sustainability[28]. In recent years, indoor air pollution due to ambient source infiltration and penetration has become a serious concern for people living in heavily polluted big cities in developing countries. Many studies have shown the correlations between outdoor and indoor air pollution which means that the level of indoor pollutants will be largely affected by ambient concentrations, so indoor residents will be exposed to high levels of air pollutants on heavily polluted days[19]. This thesis is mainly concerned with particulate matter concentration and its components in residential housing due to unhealthy ambient PM levels. Currently, people address this problem by installing air cleaners in their homes, and the effectiveness of this control strategy is explored in this study.

1.3 Indoor air pollution control strategies

Indoor air pollutants can be categorized into two groups: particulate matters and gaseous pollutants, which have both been discussed in the previous section. There are three basic strategies to reduce pollutants' concentrations in indoor air, which are source control, ventilation, and air cleaning[20]. Source control reduces or cuts emissions from individual sources of pollutants, and it is usually the most effective pollution control strategy. For example, smokers can smoke outdoors, solid fuel combustion for heating can be replaced by electricity, etc. Ventilation means exchanging air between the inside and outside of a building, which brings good quality outdoor air indoors, circulates air throughout the home, and exhausts polluted air outdoors[20]. In addition, localized ventilation uses exhaust fans in bathrooms and kitchens to remove excess moisture and strong, local pollutants. However, this strategy requires an acceptable level of outdoor air quality which is not applicable for our study due to the heavily polluted ambient environment. Air cleaners use various types of technologies to control the levels of airborne particles and gaseous pollutants and are usually used along with source control and ventilation. Several technologies are available for air cleaning: filtration uses air filter and gas-phase filters to remove particles and gases respectively; ultraviolet germicidal irradiation (UVGI) removes biological pollutants, such as bacterial and mold spores; photocatalytic oxidation (PCO) uses catalysts to destroy gaseous pollutants; ozone-generating devices sold as air cleaners use UV light to control indoor air pollutants, but they are not usually safe and effective in removing pollutants[20]. Some air cleaning devices are designed to be installed in the HVAC system, and some are used in portable, stand-alone units. The study of this thesis uses a portable air cleaner to remove particulate matters in a single room, and there are generally two types of available particle removal air-cleaning devices: mechanical and electrical air filters. In mechanical air filters, particles either become trapped in the fibers of the filter or stick to the filter because of an electrostatic charge[21]. The high efficiency particulate air (HEPA) filter is one type of extended surface mechanical air filter made of submicron glass fibers and is used in this study. Electronic air cleaners use electrostatic attraction to trap charged particles, so they can only remove small particles and do not work for gases or odors[20].

Four standard methods are provided by the American Society of Heating, Refrigerating, and Air-Conditioning Engineers (ASHRAE) and the Institute of Environmental Sciences and Technology (IEST) to assess the effectiveness of air filters: the weight arrestance test, atmospheric dust spot efficiency test, dioctyl phthalate (DOP) penetration test, and particle size removal efficiency (PSE) test. The four tests are designed to evaluate the air filter with different levels of efficiency. The Association of Home Appliance Manufacturers (AHAM)[22] updates compilations of particle air cleaner performance quarterly, and Consumers Union[23], an independent product testing firm, periodically reports in the monthly Consumer Reports magazine results of their evaluations of air cleaners, which provide consumers valuable guidance on selecting air cleaning devices[24].

The Clean Air Delivery Rate (CADR) is the measure of portable room air cleaner's delivery of contaminant-free air and used as guidance for customers to choose the right product according to their room size[20]. It tests the effectiveness of portable air cleaners in a room size test chamber for three types of particles in indoor air: dust, tobacco smoke, and pollen. However, the CADR test only measures the ability to remove 80% of particles (i.e., dust, tobacco smoke, and pollen) assuming one air exchange per hour, and it does not show the effectiveness of removing ultra-fine particles which are the most numerous and the most dangerous[25]. Therefore, lots of studies contribute to testing the effectiveness of portable air cleaning devices. Effectiveness, ε, has been defined by Nazaroff[26] as the difference in indoor concentration due to air cleaning ($C_{noAC} - C_{AC}$,) compared with the "no cleaning" case, C_{noAC}:

$$\varepsilon = \frac{C_{noAC} - C_{AC}}{C_{noAC}}$$

Combined with the definition of CADR, the effectiveness can be rewritten as:

$$\varepsilon = \frac{CADR}{V(\lambda v \ + \ \lambda d) \ + \ CADR}$$

where λv is the air infiltration rate (in units of inverse time), and λ_d is the removal rate due to deposition onto surfaces. Therefore, the closer the effectiveness is to 1, the more ideal the performance of the air cleaner is in removal of the contaminant[26]. A review study concludes that the effectiveness is strongly dependent on the particle-size category for a given CADR, which means many of the particle air cleaners tested by AHAM[22] meet the 80% effectiveness criterion for small particles, but the criterion for larger particle sizes (e.g., particles with diameters 2 µm and greater) is rarely reached[24]. Another study found that steady-state concentration reductions could reach 90% and higher with three or more air cleaners in residential dwellings[27], and the model indicates that a high air exchange rate by any method including HVAC could diminish the relative effectiveness when outdoor concentration remains high for long periods of time.

1.4 Purpose of the thesis

This thesis studies the effectiveness of air cleaner in reducing fine particles concentration in residential houses in heavily polluted areas. The air cleaner selected for this study uses activated carbon for gaseous pollutants and HEPA for particles. The study focuses on the ability of air cleaners to remove $PM_{2.5}$ particles, and at the same time, results from chemical and biological analyses help to obtain a better understanding of the effectiveness of air cleaning devices. A large amount of studies has been done to test the performances of various types of air cleaners, and several studies have indicated that the HEPA air filter is effective in reducing fine particles which have the most dangerous adverse health effects; however, most existing studies are based on modeling or aiming to help a small group of people suffering from allergy or respiratory diseases. Studies rarely done in developing countries, like China and India, who suffer from unhealthy levels of air pollution especially high concentration of fine particulate matter in recent years. The continuous heavily polluted atmospheric environment stimulates the market of air cleaner in big cities of relevant developing countries, but not enough data is available to show the ability of air cleaners in providing healthy indoor environments for residents. So, our research is being done to fill gaps in the literature and to study the performance of air cleaners in protecting residents in heavily polluted areas with high PM concentrations. Therefore, this thesis will talk about the study on the effectiveness of installing air cleaners in residential houses which took place in Beijing, China during the summer of 2016; in addition, it will show the results from chemical analysis and toxicity studies to help reach the conclusion. This study intends to serve as pilot research in air cleaner studies for places with high levels of air pollution and to contribute to the building of an air pollutant profile with/without air cleaning devices working in the indoor environment.

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Chapter 2 The Influence of Air Cleaners on Indoor Particulate Matter Components and Oxidative Potential in Residential Households in Beijing

2.1 Abstract

In many developing regions with poor air quality, the use of air filtration devices to clean indoor air is growing rapidly. However, few studies have been performed to assess their removal efficiency. In this study, we collected indoor, outdoor and personal exposure filter-based samples of fine particulate matter (PM2.5) with both properly operating, and sham air cleaners in five Beijing residences from July 24th to August 17th, 2016. Mass concentrations of PM2.5 and several health relevant components of PM2.5 including organic carbon, elemental carbon, sulfate, nitrate, ammonium, and 21 selected metals were analyzed to evaluate the effectiveness of air cleaners. The effect of air purification on PM2.5 reactive oxygen species (ROS) activity, a metric of the oxidative potential of the aerosol, was also evaluated. The average indoor PM2.5 concentration during true filtration was 8.47 µg/m3, compared to 49.0 µg/m3 during sham filtration; thus, air cleaners can significantly reduce the indoor PM2.5 concentration well below WHO guideline levels and significantly lower all major components of PM2.5. However, the utility of air cleaners in reducing overall personal exposure to PM2.5 and its components was marginal in this study: the average personal exposure PM2.5 concentration was 67.8 and 51.1 µg/m3 during true and sham filtration, respectively, and it is likely due to the activity patterns of the subjects. Short-term exposure contributions from environments with high PM2.5 concentrations, including exposure to traffic related emissions as well as uncharacterized indoor microenvironments, likely add substantially to the total PM2.5 exposure burden. The toxicity assay indicates that the air cleaners can also significantly reduce ROS activity in the indoor environment; however, this decrease did not translate to a reduction in personal exposure. Elemental carbon, lead, and arsenic were well-correlated with the ROS activity, thus adding to the knowledge base of drivers for ROS activity.

2.2 Introduction

Air pollution is a serious problem in developing countries worldwide as it can cause harm to humans and the natural environment (Kinney et al., 2008; Kim, Jahan, & Kabir, 2013). It is believed to kill more people worldwide annually than AIDS, malaria, breast cancer, and tuberculosis combined (WHO, 2014). Airborne particulate matter (PM) is especially harmful to human health, and several studies have shown that long-term exposure to PM is associated with mortality from cardiovascular diseases (Hoek et al., 2013). In China, a country suffering from high concentrations of particulate matter, a study of the hourly air pollution data from over 1500 sites found that the population-weighted average exposure to fine particulate matter ($PM_{2.5}$) was 52 µg/m^3, and 38% of the population experienced average concentrations that were above the US EPA 24-hour standard of 35 µg/m^3 (Rohde & Muller, 2015). The indoor environment may protect people from harmful exposure to outdoor pollutants to some extent; however, many studies have shown that outdoor PM can enter an indoor environment with little attenuation unless filters or other PM-removing devices are used in the building ventilation systems or within a room. This is especially concerning for the elderly, the very young, and those of ill health who spend a greater amount of time indoors and who are also more susceptible to the adverse health effects of exposure to air pollution (Byrne et al., 1998). Given these concerns, there is a fast-growing market for air cleaners to improve indoor air quality in countries such as China that suffer long-term air pollution issues, although there are few studies documenting their effectiveness and, in particular, the extent to which they reduce overall human exposure to specific pollutants.

Many studies have concluded that there are correlations between the outdoor, indoor, and personal exposure concentrations of particulate matter and its components (Janssen et al., 2005; Jones, Thornton, Mark, & Harrison, 2000; Williams et al., 2000; Janssen, Hoek, Harssema, & Brunekreef, 1999), and several studies have found that pollutant concentrations decrease from outdoor to indoor environments with indoor/outdoor (I/O) ratios of less than 1 (Cho et al., 2005; Chen & Zhao, 2011). However, personal exposure is higher than outdoor concentrations in most studies (Williams et al., 2000; Meng et al., 2005; Wallace et al., 2000) and may be in part explained by the existence of excess mass near the person, a "personal cloud" related to personal activities (Ozkaynak et al., 1995).

Several studies have found that the use of high efficiency particulate air (HEPA) filters can reduce symptoms for asthma sufferers and lead to clinical benefits for asthma patients (Reisman et al., 1990; Sublett et al., 2011). There are also studies showing that HEPA filters decrease airborne allergens in indoor environments and reduce personal exposure to allergens (e.g., allergens from cats) (Gore et al., 2003), pollens, and fungal spores (Sublett et al., 2011) as well as relieve seasonal allergic rhinitis symptoms (Morris et al., 2006; Reisman et al., 1990). Additional studies found that HEPA filters reduced concentrations of particulate matter (Myatt et al., 2008), VOCs including toluene and formaldehyde (Xu et al., 2010), CO_2, and environmental tobacco smoke (ETS) levels (Myatt et al., 2008).

However, there are limited data available to determine how effective air cleaners are at reducing the concentrations of chemical components of $PM_{2.5}$. Components of $PM_{2.5}$ include elemental carbon (EC) and organic carbon (OC), which are indicators of carbonaceous species and account for up to 40% of $PM_{2.5}$ mass in urban atmospheres (Seinfeld et al., 1998); sulfate, nitrate, and ammonium, which account for a significant portion of the overall $PM_{2.5}$ mass concentration (Chan & Yao, 2008) and can be used to approximate the contribution of the power, industry, transportation, and residential sectors to overall emissions (Zhang, et al., 2012; Hodan & Barnard, 2004); EPA regulated hazardous metal air pollutants (Sb, As, Cd, Cr, Pb, Mn, and Ni), and other toxic metals documented in the literature. Moreover, the biological toxicology of PM has been well studied but the effectiveness of HEPA filters at reducing the activity of reactive oxygen species (ROS), which may be implicated in adverse health outcomes, to our knowledge, has not been studied. Similarly, little research has been done to investigate personal exposure to pollutants after the installation of an air cleaner. This is especially problematic in developing countries with high ambient particulate matter concentrations and where the market of air cleaners is growing rapidly. Therefore, in this study, the effectiveness of air cleaners at removing the chemical components and reducing the oxidative potential of particulate matter in the indoor environment was assessed. The relationship between air cleaner use and personal exposure to the above species was also addressed. It is the first study to investigate both the detailed chemical species and ROS activity of $PM_{2.5}$, and it serves as a pilot study for further research on the effectiveness of air cleaners at removing $PM_{2.5}$ and its specific chemical components as well as its ROS activity.

2.3 Sampling and Analysis Methods

2.3.1 Sampling summary

Sampling took place in Beijing, China, from July 24th to August 17th, 2016. The typical average daily $PM_{2.5}$ levels over recent years have ranged from 60 to 100 µg/m^3 in Beijing (Li et al., 2015; Liu et al., 2015). Samples were collected in the apartments of six participants in locations spread across Beijing. Physical characteristics and locations of the apartments of participants (i.e., size, floor level, layout) are described in Table 1 in the supplementary material section. At the beginning of the study, an Atmosphere® Air Purifier was placed in the bedroom of the participant.

During sampling, indoor and outdoor $PM_{2.5}$ samples were collected on 37 mm quartz and Teflon filters using SKC Personal Environmental Monitors (PEMs) and AirChek XR5000 pumps set to run for 48 hours at 4 L/min (lpm). The indoor sampler was set up in the same room as the air purifier start one day before in almost all cases to allow for an equilibrium to be reached in the room. Outdoor samples were collected outside another window of the apartment to ensure that the window in the bedroom with the purifier could be kept fully closed during sampling.

Personal exposure samples were collected on 37 mm Teflon filters using the same sampling equipment but at a flow rate of 2 lpm simultaneously with indoor and outdoor sampling. Pumps were placed in a small backpack, with the filter holders attached to the strap of the bag closest to the participants' breathing zone. Participants were asked to keep the bag with them at all times except when showering, etc., at which point the bag was to be placed outside the bathroom door. While sleeping, the participants were instructed to put the sampler on a shelf or table at approximately breathing height. During the 48-hour personal sampling period, participants recorded their hourly activities in a survey, including where they were (e.g., in the room with the purifier, outside) and what type of activities they were doing, whether or not there were people smoking or cooking, and whether the windows were open or closed.

Sampling occurred twice in each home – once during a "true" filtration period and once during a "sham" filtration period: true filtration means all three stages of filtration (i.e., a coarse pre-filter, a replaceable HEPA filter, and a replaceable carbon filter) are used while sham filtration only uses the pre-filter. In total, 36 Teflon and 24 quartz-fiber filters were collected in homes, with five trip blanks and five loading blanks of each filter type collected throughout the study. Trip blanks are filters taken from the lab to the sampling locations, and loading blanks are additionally loaded into the samplers and then removed after a few minutes. The filters of outdoor, indoor, and personal exposure of one home in a 48-hour sampling period form one group, so each group contains three Teflon filters and two quartz filters; thus, there are 12 groups overall and six of them are under the true filtration condition while the other six groups are under the sham filtration condition.

2.3.2 Mass, Organic and Elemental Carbon and Ionic Analyses

PM mass concentration (gravimetric mass) was determined from the Teflon filters. Filters are equilibrated under controlled temperature (22-24 ◦C) and relative humidity (40-50%), and pre- and post-PM masses are obtained using a microbalance (0.001 mg) (Shirmohammadi et al., 2015). Elemental carbon (EC) and organic carbon (OC) were determined by the NIOSH

Thermal Optical Transmission method (Birch & Cary, 1996). Nitrate and ammonium were quantified by extracting the Teflon filters with 10 mL of high purity water and the water-soluble ions determined by ion chromatography (ICS-5000+, Dionex).

2.3.3 Elemental and ROS Analyses

The total concentrations of 50 elements in PM were quantified by magnetic-sector ICPMS (Thermo Finnigan Element 2, Thermo Scientific, U.S.A.) after solubilization of PM with a mixed-acid microwave-aided digestion protocol (von Schneidemesser et al., 2010). The total measurement uncertainty of each quantified element was estimated by propagating the uncertainty (i.e., ± one standard deviation) of the components from the SF-ICPMS analysis (three analytical replicates), blank subtraction (multiple blanks), and the uncertainty in extraction recovery (Heo et al., 2015). Elements with low signal/noise (not statistically different than zero) were removed from analysis, and all remaining metals with enrichment factor (EF) larger than 50 are presented in this paper, since they are likely to have significant anthropogenic contributions.

Oxidative potential (ROS activity) of the $PM_{2.5}$ was measured by exposing (in 96-well plates) rat alveolar macrophage cells (NR8383, American Type Culture Collection) to aqueous PM extracts (Landreman et al., 2008). Prior to sample exposure, the macrophage cells are loaded with the fluorescent probe DCFH-DA (2’,7’-dichlorofluorescein diacetate). DCFH-DA is membrane permeable and is deacetylated by cellular esterases to DCHF (2’,7’-dichlorodihydrofluorescein) in the cytoplasm of the cell. DCFH is oxidized by ROS to highly fluorescent 2,7-dichlorofluorescein (DCH), then it can be detected by a micro-plate reader. The ROS activity of samples was compared to untreated control cell activity and is reported in units of Zymosan equivalents (μg ZYM/mg PM and μg ZYM/m^3) (Shirmohammadi et al., 2015).

2.3.4 Air Cleaner Technical Information

This study used the Atmosphere® Air Purifier sold by Amway^{TM}. The air purifier’s external dimensions are 38.1 cm wide x 27.9 cm deep x 73.7 cm tall, and it contains three-stages of filtration: a washable pre-filter that captures large particles and fibers; a replaceable HEPA filter that removes the airborne contaminants that pass through it; and a replaceable activated carbon filter that reduces household odors. During both the true and sham periods of this study, the flow rate on the purifier was set to level 2 (2.8 m^3/min). This was the highest setting still quite enough to not disturb the participants.

2.3.5 Enrichment Factor and Dust Content

One approach to evaluate the relative contribution from non-crustal sources to particulate elemental concentrations is to express the metal concentrations in the form of crustal enrichment factors (EF). EF is defined as the concentration ratio of a given metal to that of aluminum (Al) (or any other element which derives mainly from rock and soil dust) normalized to the same concentration ratio characteristic of the upper continental crust (i.e., the composition of the continental crust) (Wedepohl et al., 1995). Low EF values (less than 10) indicate that an element is likely not enriched by non-crustal sources; if EF values are significantly larger than 10, it indicates an important contribution from other natural or anthropogenic sources (Barbante et al., 2003). In this study, EF is calculated by Equation (1) (Xu et al., 2004):

$$EF = \frac{Element(sample)/Al(sample)}{Element(UC)/Al(UC)} \tag{1}$$

where UC refers to upper continental crust concentrations. Crustal composition data is obtained from the study of Wedepohl (1995). Due to the large average EF of all samples in this study, metals with EF larger than 50 are considered as elements significantly affected by anthropogenic sources and $PM_{2.5}$ components resulting from human activity.

The dust concentration is calculated with Equation (2) (Hueglin et al., 2005).

$$Dust = 1.89\,Al\ +\ 1.43\,Fe\ +\ 1.4\,Ca\ +\ 1.66\,Mg\ +\ 1.7\,Ti\ + 2.14\,Si \tag{2}$$

The dust concentration and all of the elements are measured in $\mu g/m^3$. Silicon (Si) is estimated from Al using a factor of 3.41 (Hueglin et al., 2005). Other studies also include manganese (Mn) and potassium (K) in the equation. Since the enrichment factor of Mn in the samples is larger than 50, suggesting that anthropogenic sources are significant contributors to its total concentration, it is dropped from the equation. There was no insoluble K measured, so K was not used in the dust calculation.

2.3.6 Correlations and infiltration factor

Many studies have examined relationships between outdoor and indoor PM concentrations, with the infiltration factor being used to parameterize and characterize these relationships. The effective infiltration factor used in this study and many others is derived from Equation (3), which shows the mass balance between outdoor and indoor pollutants:

$$V\frac{dC_{in}}{dt} = aPVC_{out} -\ aVC_{in} -\ KVC_{in} + \dot{S} \tag{3}$$

where V is the volume of the room; t is time; a is the air exchange rate due to infiltration; P is the particle penetration factor; K is the particle deposition rate; and $\dot{S}$ is the indoor particle emission rate. All of the parameters, with the exception of V and a, are functions of both time and particle size (Li & Chen, 2003).

The ideal infiltration factor represents the equilibrium fraction of ambient particles that penetrate indoors and remain suspended (i.e., avoid mixing with particles from indoor sources) (Chen & Zhao, 2011), and is defined under an assumption of the steady-state and an indoor particle emission rate of 0 (Chen & Zhao, 2011):

$$F_{in} = \frac{aP}{a+K} \tag{4}$$

Effective infiltration factor in this study is used to assess the outdoor $PM_{2.5}$ concentration effects indoors and it can be calculated by Equation (5) with well-mixed room assumptions, where the parameter $C_{\dot{s}}$ is the indoor particle concentration contributed by indoor sources. F_{in} and $C_{\dot{s}}$ can be solved from the regression of indoor concentration against the outdoor concentration (Ott, Wallace, & Mage, 2000).

$$C_{in} =\ F_{in}C_{out} + C_{\dot{s}} \tag{5}$$

In this study, the variation of effective infiltration factors due to physical differences between homes is ignored since the apartments are similar across Beijing. When the air cleaner is set to work in the indoor environment, the particle loss due to the HEPA filter needs to be added to Equation (3). Therefore, the mass balance will be adjusted to Equation (6) (Ma et al., 2015):

$$V\frac{dC_{in}}{dt} = aPVC_{out} - aVC_{in} - KVC_{in} + \dot{S} - CADR \times C_{in} \tag{6}$$

where CADR is the clean air delivery rate with the unit of volume per time. Then, the infiltration factor is adjusted to:

$$F_{in} = \frac{aP}{a + K + \frac{CADR}{V}} \tag{7}$$

Therefore, the infiltration factor under the true filtration condition will be smaller than that of the sham filtration condition.

2.4 Results and Discussion

2.4.1 Overview

Table 1 summarizes the $PM_{2.5}$ mass concentration, organic carbon (OC), elemental carbon (EC), sulfate, nitrate, ammonium, dust, and selected metals concentration data from the study. Metals with enrichment factors greater than 50, hazardous air pollutants as defined by the EPA, and the main inorganic components of dust are included in the table. The first three columns present the absolute average concentrations for the outdoor, indoor, and personal exposure samples. The outdoor concentration is typically two-fold or larger than the parallel indoor sample, while the personal exposure concentrations are comparable to outdoor concentrations. The personal exposure concentrations of certain metals (e.g., Sb, Zn, Mo, Cu, Fe) are higher than the outdoor concentrations, with the outdoor to personal exposure ratio as low as 0.22 for Mo. The next two columns in Table 1 present the outdoor to indoor removal percentages for the true and sham filtration conditions, respectively. The removal percentage during true filtration ranges from 70% to 95% (except for OC), while, not unexpectedly, no removal rates higher than 70% were measured during the sham filtration conditions. For a few elements (e.g., Sn and B), there are negative percentages in the sham filtration column and comparatively low percentages in the true filtration column (like OC). This is very likely due to the contribution of indoor sources to the pollutants being measured and will be discussed in greater detail later. Figures 1 & 2 show the indoor, outdoor, and personal exposure concentrations of major PM descriptors ($PM_{2.5}$ mass, sulfate, dust) and reactive oxygen species (ROS) activity under the sham and true filtration conditions. It is clear that the use of the air cleaner substantially reduced indoor concentrations of $PM_{2.5}$ mass, sulfate, dust, and ROS activity, but there was no comparable reduction in personal exposure concentrations.

2.4.2 Indoor to outdoor changes due to air cleaner installation

Indoor to Outdoor Correlations, and Infiltration Factors

The effective infiltration factor and indoor sources were estimated under both the sham and true filtration conditions for all species measured in the study by applying linear regression using Equation (5), where the slope of the line indicates the effective infiltration factor and the intercept represents the indoor sources. Since these are generated using data from all five homes, they are the linear fit average effective infiltration factor for similar homes in Beijing, which represent the outdoor contributions of $PM_{2.5}$ to indoors. Correlation coefficients (and infiltration factors) for mass, EC, OC, sulfate, nitrate, ammonium, and dust are shown in Table 2.

Species with a correlation coefficient larger than 0.8 and P-value smaller than 0.05 are bolded in the table, indicating that a linear regression fit of indoor versus outdoor concentrations is significant. The indoor mass, elemental carbon, sulfate, and ammonium concentrations under the sham filtration condition are well-correlated with outdoor sources; and infiltration factors of 0.49, 1.07, 0.83 and 0.58 were estimated, respectively. These outcomes indicate that the indoor concentrations of the above species are mainly attributable by outdoor sources. The results are consistent with previous studies as summarized in a review paper that that measured $PM_{2.5}$ infiltration factors from 21 large-scale studies (i.e., more than 20 homes and found infiltration factors ranging from 0.3 to 0.82) (Chen & Zhao, 2011). Another study on retirement homes in the Los Angeles Basin found a high infiltration factor for EC, likely because EC is capable of infiltrating through the building envelope with great efficiency (Polidori et al., 2007). At the same time, correlation coefficients are lower for OC, dust, and nitrate and they are supported by previous studies: several studies have shown a considerable contribution of indoor sources of OC to total PM, including the retirement homes study reporting that 13%-26% of the measured indoor OC is emitted or formed indoors (Polidori et al., 2007). Meng et al. found that outdoor OC contributes to only 21.7% of indoor OC (Meng et al., 2009). These findings have been explained by the fact that cooking is one of the most significant indoor sources of OC (Abdullahi at al., 2013). The very low correlation of dust between indoors and outdoors under sham filtration conditions may be due to house cleaning since a previous study found soil dust tracers (Ca, K) are associated with housework such as vacuuming (Baxter et al., 2007). Low values of nitrate may be due to the nitric acid absorbed on particles volatilizing back to gas phase HNO_3 due to the temperature and humidity changes (Patterson & Eatough, 2000). Also, the lack of significance of the estimates may be due, in part, to the small sample size as well as the variations of indoor sources between homes.

Indoor $PM_{2.5}$ concentrations during true filtration do not show a strong correlation with outdoor PM concentrations due to the varied purifier performance in different homes (such as room size – Equation (7). At the same time, the estimated infiltration factor is very small due to the existence of air cleaner and can be explained by the introduction of CADR to the infiltration factor calculation shown in Equation (7). Similar observations have been made in other studies showing that central air conditioning system operation, which has a similar function to air cleaners, reduces the infiltration factor (Meng et al., 2009).

Table 2 of the supplemental material presents a summary of the linear regression fits of indoor and outdoor concentrations for the measured metal data. They follow the same trend that the estimated infiltration factors of metals under true filtration are much lower than those estimated for sham filtration.

Indoor/outdoor (I/O) ratio

The relationship between indoor and outdoor particle concentrations (I/O), under the sham and true filtration conditions can be used to assess the effectiveness of an air cleaner to reduce PM and PM components. Figure 3 presents the I/O ratio for the main species under both the sham and true filtration conditions. The I/O ratio is much smaller with true filtration than sham filtration for nearly every species, indicating that the concentrations of these main species indoors are effectively reduced by the air cleaners. During true filtration, the I/O ratios of all main species do not exceed 0.55 with the exception of OC. The higher I/O ratio for OC is likely due to indoor OC emission sources, as discussed in the previous section.

The I/O ratio mean values and standard deviations are shown in Table 3, where ratios statistically different from 1 are bolded, and ratios with standard deviations equal or larger than the mean value are underlined. The ratio is affected by many factors including indoor particle sources, room size, outdoor particle concentration, etc., which results in a large range of measured values in the literature (Chen & Zhao, 2011). In this study, the mass concentration ratio is 0.68 and 0.19 under the sham and true filtration conditions, respectively, indicating a quite substantial air cleaner influence on $PM_{2.5}$ mass. In previous studies that excluded smoking indoors, $PM_{2.5}$ mass I/O ratios ranged from 0.71-1.5 (Chen & Zhao, 2011). The ratio during sham filtration falls at the lower end of these reported values, perhaps due to the the lack of HVAC system and high outdoor concentrations of $PM_{2.5}$ in Beijing compared to the cities in the US and Europe where the aforementioned studies were conducted. In this study, the OC I/O ratio is relatively high for both filtration conditions which ranges from 1.1-1.8 and 0.78-1.4 for sham and true filtration, respectively, which may indicate sources of OC indoors, potentially from the continuous emissions of organic compounds like VOCs from cooking. Previous studies have obtained similar results: a study in the UK reported I/O ratios of OC in the range of 2.2-7.7 while ratios of other components in $PM_{2.5}$ such as EC, sulfate, lead, and zinc were all under 1.5 (Jones et al., 2000). In the case of true filtration, the I/O ratio for all main species are statistically smaller than 1 (except B). In addition, several components have I/O ratios around or larger than 1 during sham filtration, such as Sn, W, and Ni, indicating significant indoor sources like fabrics, glass, and plastics, but all of them are removed by the air cleaner effectively, as evidenced by I/O ratios of less than 0.30 for these same components during true filtration.

The average indoor $PM_{2.5}$ mass concentration during true filtration was 8.47 $\mu g/m^3$ (all values are under 10.5 $\mu g/m^3$) – well below the World Health Organization (WHO) air quality guideline for $PM_{2.5}$ of 25 $\mu g/m^3$ (24-hour average) (WHO, 2006). However, the average indoor mass concentration during sham filtration was 49.0 $\mu g/m^3$ – nearly double the WHO standard. This indicates that the air cleaner may effectively reduce $PM_{2.5}$ particles, providing a healthier indoor environment.

2.4.3 Personal exposure to $PM_{2.5}$

The personal exposure of each participant to $PM_{2.5}$ concurrent with the 48-hour indoor and outdoor sampling at their homes was measured, as detailed previously. Personal exposure to outdoor concentration ratios is calculated in the same way as the I/O ratio in the previous section and is shown in Figure 4. The specific P/O values are given in Table 3 as well – ratios statistically different than 1 are bolded and ratios' standard deviations equal or larger than the mean value are underlined. With the exception of nitrate (NO_3^-), tin (Sn), and nickel (Ni) under sham filtration, none of the P/O ratios are statistically different than 1 – thus significant differences between outdoor concentrations and average personal exposure concentrations were not observed. Similarly, there were no P/I ratios statistically different than 1 under the sham filtration, although several components of $PM_{2.5}$ have P/I ratios significantly larger than 1 during true filtration. Although participants spent substantial time indoors in the reduced PM environment, this did not, on average, translate into a reduction in personal exposure.

Relatively high personal exposure concentrations to $PM_{2.5}$ have been found in many previous studies (Meng et al., 2005; Williams et al., 2000), and there are several approaches that can be used to better understand why personal exposures are higher than one would predict from measured reductions in indoor air concentrations. The microenvironmental method is one of the common ways used to estimate personal exposure from a participants' time-activity records and microenvironment $PM_{2.5}$ concentration data. The microenvironmental method is given by (Meng et al., 2005):

$$E_t = \sum_j C_j t_j \tag{9}$$

where E_t is the time-averaged personal $PM_{2.5}$ exposure concentration (µg/m^3); C_j is the $PM_{2.5}$ concentration in the j^{th} microenvironment (µg/m^3); and t_j is the time fraction a person spends in the j^{th} microenvironment (dimensionless). In this study, on a one-hour time interval, participants recorded time spent in four microenvironments in the survey: a room with an air cleaner, other rooms in the apartment, an office with a central air conditioning system, and the outdoor environment. Participants' activity during sampling periods is summarized in Table 3 and Figure 1 in the supplemental materials, and the information is used to estimate the personal exposure using the microenvironmental method. Without $PM_{2.5}$ concentration data for indoor environments lacking air cleaners, two estimates were calculated using the assumption that the other rooms and office $PM_{2.5}$ concentrations were the same as 1) the indoor concentration and 2) the outdoor concentration. The estimated personal exposure results are shown in Table 4 with measured personal exposure included for comparison. Both estimates resulted in personal exposure concentrations less than the measured values, indicating that there are microenvironments with high $PM_{2.5}$ concentrations that have not been included in the calculation.

High outdoor $PM_{2.5}$ concentration microenvironments that were not modeled or included likely relate to transportation activities. The literature indicates that transportation activities such as exposures resulting from road or railway dust and vehicle emissions can result in extremely high personal exposure to particles (Steinle et al., 2015; Vallejo et al., 2004; Xu Y. et al., 2010). Participants in this study indicated that they had walked, biked, or taken the subway to work, and recorded this as time outside in the time-activity surveys; however, the estimates for outdoor concentrations were based solely on the samples collected directly outside the participants' homes, and this certainly does not capture differences in outdoor exposure levels across the city and during a commute. In addition, high $PM_{2.5}$ concentrations in kitchens during cooking are not included in the calculation and may contribute substantially to personal exposure if the participants cook in the kitchen or spend time in the kitchen when cooking occurs during the sampling period. Also, some studies have found that exercising both indoors and outdoors can result in high exposure to $PM_{2.5}$ (Xu Y. et al., 2010) probably due to greater generation or stirring-up of particles. Moreover, the air cleaner was placed in the bedrooms of the participants' homes and indoor samples were collected in the bedrooms but participants spent considerable time in other rooms where the $PM_{2.5}$ concentration may have been higher than in the bedrooms. Time spent in these other microenvironments is not considered in the estimation. Other personal activities such as house cleaning, watching TV, and computer use can also have a large influence on personal exposures, and future studies should include more detailed questionnaires of activity profiles and assessment of PM levels in these environments and during these activities.

2.4.4 Reactive Oxygen Species Measurements

Many epidemiologic studies have shown associations between ambient fine particulate matter (PM) and adverse health outcomes including cardiovascular and respiratory diseases, increased mortality, emergency room visits, and time lost from school and work, after both short- and long-term exposure to ambient air pollution (Pope et al., 2004; Cho et al., 2005; Johnson et al., 2004; Sørensen et al., 2003). The mechanisms of PM-induced adverse health effects are many, and a fundamental biochemical understanding of these processes is still in its infancy. However, there is increasing evidence that many of the adverse health effects derived from oxidative stress are initiated by reactive oxygen species (ROS) within affected cells (Cho et al., 2005). A growing body of literature documents that PM is able to induce proinflammatory effects in the nose, the lung, and the cardiovascular system (Li et al., 2003; Donaldson et al., 2001; Baulig et al., 2003), an effects cascade initiated in-part by ROS generation. Ongoing research shows that ROS are formed in cells through the reduction of oxygen with the catalytic assistance of electron-transfer enzymes and redox-active chemical species such as redox-active organic chemicals and metals (Cho et al., 2005; Squadrito et al., 2001; Dellinger et al., 2001). Therefore, assessing the influence of air cleaners on the ROS activity of PM may be an important predictor of impacts on human health.

The ROS activity of $PM_{2.5}$ under the true and sham filtration conditions for outdoor, indoor, and personal exposure are shown in Figures 2 and 3, respectively. The same trend as documented with measured chemical species is observed in ROS activity: indoor concentrations show clear decreases with true filtration while personal exposure samples show little change compared to outdoor concentrations with the air cleaner working in participants' bedrooms. The ROS activity reduction follows a similar trend as $PM_{2.5}$ and components of $PM_{2.5}$ that the air cleaner is effective in reducing indoor ROS activity but does not work to reducing personal exposure ROS activity, and it is likely because the ROS activity is correlated with some sources indicated by the chemical components of $PM_{2.5}$. To investigate this, the correlation between ROS activity and PM components was examined using both absolute concentrations and the removal percentage. Results are shown in Table 5. The first two columns in Table 5 are the correlations of ROS with absolute concentrations expressed both on a linear and LOG-scale (sample size is 30). Statistically significant ($p < 0.05$) correlation coefficients larger than 0.7 are in bold. Elemental carbon has the highest correlation coefficient (0.86) with ROS activity. Similar strong correlations between ROS and EC have been observed in several studies, including in the Los Angeles Basin where Cho et al. (2005) observed a correlation of $R^2 = 0.79$. Elemental carbon is emitted mainly from incomplete combustion (Duan et al., 2012) and would most likely come from motor vehicle emissions during the summertime in Beijing; therefore, the results indicate that ROS activity may be closely related to motor vehicle emissions during the summertime in Beijing. A study focusing on the relationship between redox activity and specific chemical species concluded that metal-containing sources may be actively involved in the resulting health effects through ROS formation and oxidative stress or direct electrophilic reactions (Ntziachristos et al., 2007). Results show that most of the measured metals correlate well with ROS activity, especially As and Pb, with correlation coefficients of 0.80 and 0.85, respectively. OC shows relatively low correlation in the analysis.

The third and fourth columns in Table 5 are the removal percentage (indoor to outdoor and personal exposure to outdoor) correlation coefficients between ROS and the PM component species. Reductions in ROS activity from outdoor to indoor correlate highly with changes in measured levels of many $PM_{2.5}$ components ($r > 0.7$). However, the removal percentage of outdoor to personal exposure does not show very strong correlations between ROS activity and PM components due to the relatively small changes in concentrations of most species. The linear regression fit of ROS removal percentage with PM mass, EC, OC, and dust removal percentage are selected as examples to be shown in Figure 4. Consistent with the statistics summarized in Table 5, the linear fits in Figure 4 are reasonably strong for the outdoor to indoor removal percentage and much less so for the outdoor to personal exposure removal percentage. Among the four analyses, removal percentage of elemental carbon has the best linear regression fit with that of ROS activity ($R^2 = 0.88$), suggesting that ROS activity is more closely related to the EC component than other $PM_{2.5}$ components measured in this study. These results importantly indicate that air cleaners have the potential to provide substantial health benefits to homeowners; however, microenvironment exposures must be controlled. The detailed chemical speciation outcomes suggest roles for both EC and specific metals in driving ROS activity – results consistent with available literature. The general approach detailed in this study when applied to larger sample sizes will provide the statistical power to further refine the roles of specific $PM_{2.5}$ components in ROS activity and measured human health outcomes.

2.5 Conclusion

Several conclusions can be drawn from the results of this study. The I/O ratio of $PM_{2.5}$ mass concentration and all measured components (except organic carbon) is reduced significantly with an indoor PM filtration device. We observed a reduction in average indoor $PM_{2.5}$ concentration from 49.0 $\mu g/m^3$ under the sham filtration condition to 8.50 $\mu g/m^3$ under true filtration, a decrease of over 80%. The 8.50 $\mu g/m^3$ value is substantially lower than the 25 $\mu g/m^3$ (24-hour average) WHO guidelines; therefore, it can be concluded that air cleaners are effective in removing $PM_{2.5}$ particles and are able to provide a healthier indoor environment. However, the air cleaner was not effective in reducing personal exposure $PM_{2.5}$, due to contributions of exposures that occurred in microenvironments with high $PM_{2.5}$ concentration both outdoors (e.g., during transportation) and indoors (e.g., while cooking occurred). The ROS activity of the indoor PM is well-correlated with most of the measured components of $PM_{2.5}$; however, similar to that observed with $PM_{2.5}$ mass and its components, air cleaner use did not significantly reduce personal exposure to PM-induced ROS. In summary, air cleaners, as implemented in this study, are effective in reducing $PM_{2.5}$ total mass concentration as well as its components', and their use can also reduce the toxicity of the particles in the indoor environment. To effectively reduce the personal exposure to $PM_{2.5}$, it is likely that controls on personal exposure during uncharacterized activities with high PM will need to be implemented. Data from these microenvironments/activities should be collected in future studies to quantitatively assess the complete exposure profile. Based on the results presented above, it is not entirely clear that indoor filtration has a health benefit given the dominance of high concentration non-indoor sources to personal exposure of $PM_{2.5}$. Subsequent research would benefit from having a larger sample size in different seasons and including a broader range of housing types with contrasting infiltration rates.

Acknowledge

The authors gratefully acknowledge the participants in this study for opening up their homes and for cooperating in the data collection process. Thank you also to Linchen He and Meichen Liu of Duke University, and Wei Jingya of Tsinghua University for their assistance with logistics and data collection in the field.

This work was supported by funding from Underwriters Laboratories.

2.6 Figures

Figure 1 Fine particulate matter mass concentration, sulfate concentration, dust concentration and reactive oxygen species activity under sham filtration.

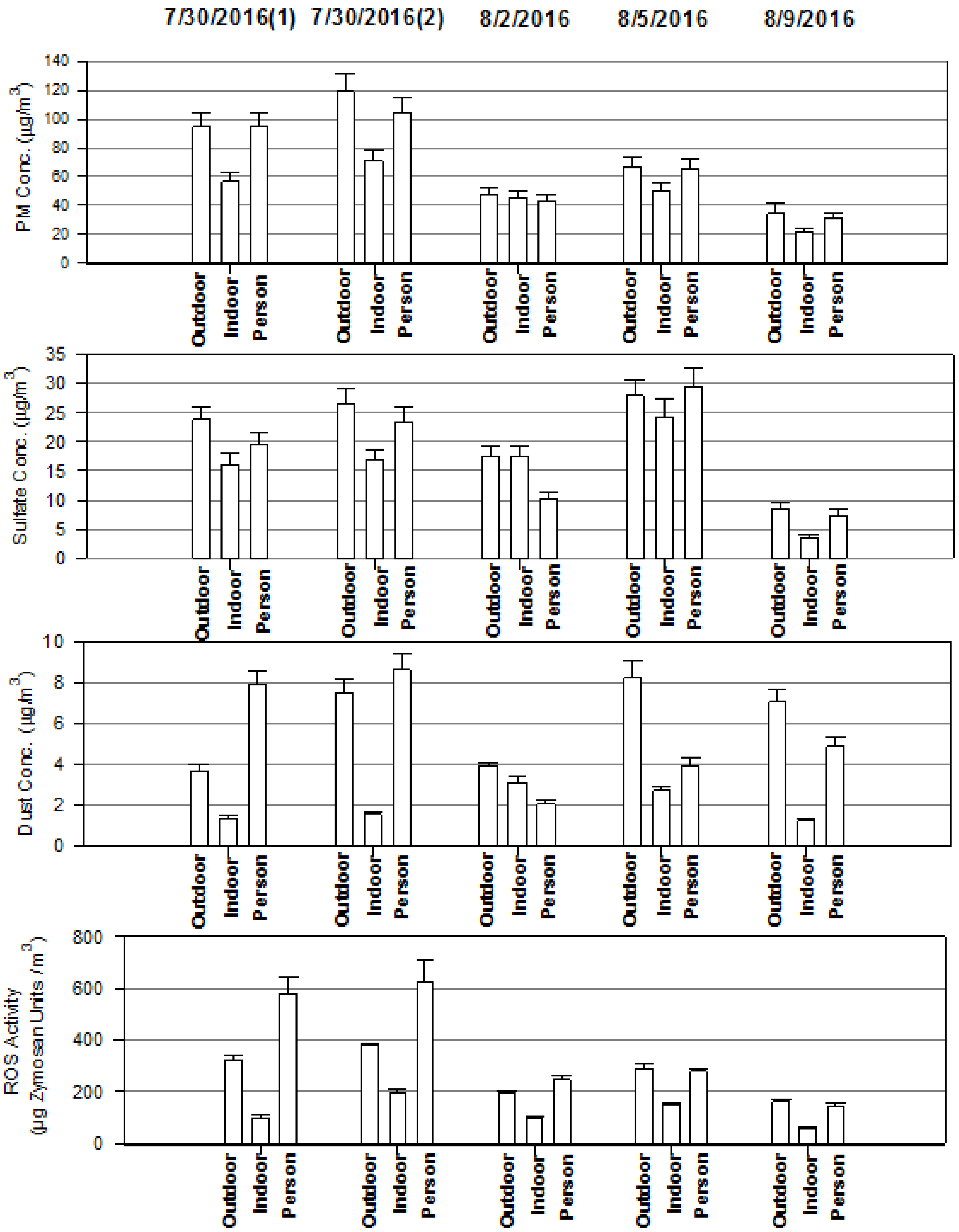


Figure 2 Fine particulate matter mass concentration, sulfate concentration, dust concentration and reactive oxygen species activity under true filtration.

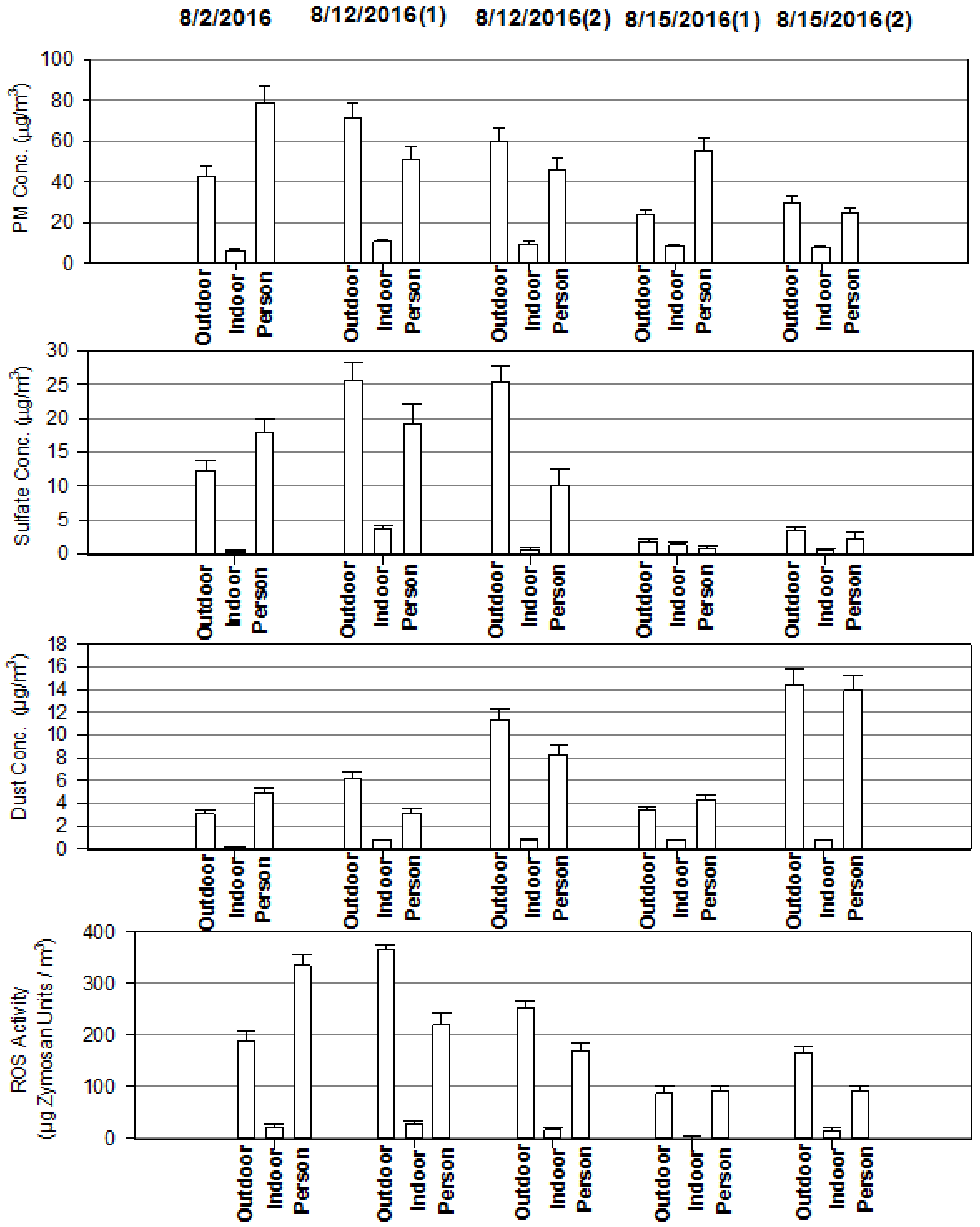
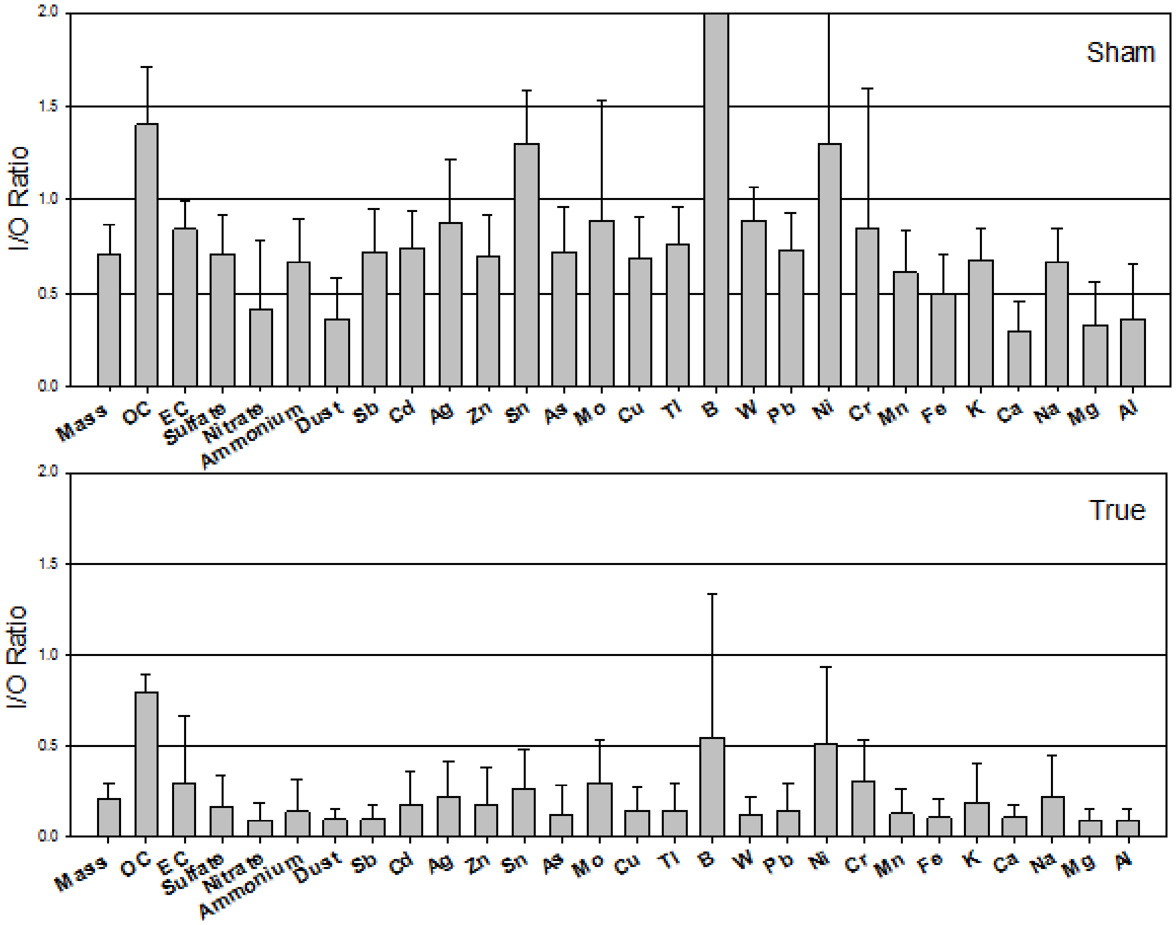
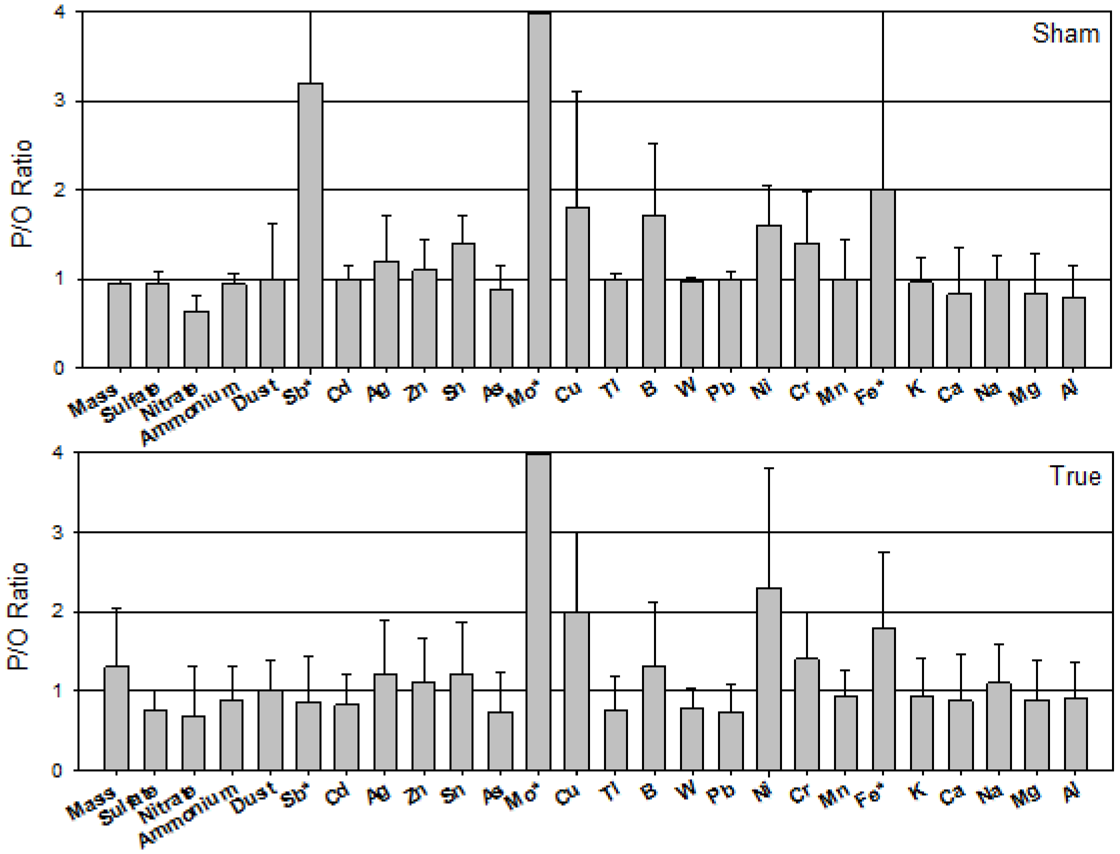


Figure 3 Indoor to Outdoor Concentration Ratio of Main Chemical Species



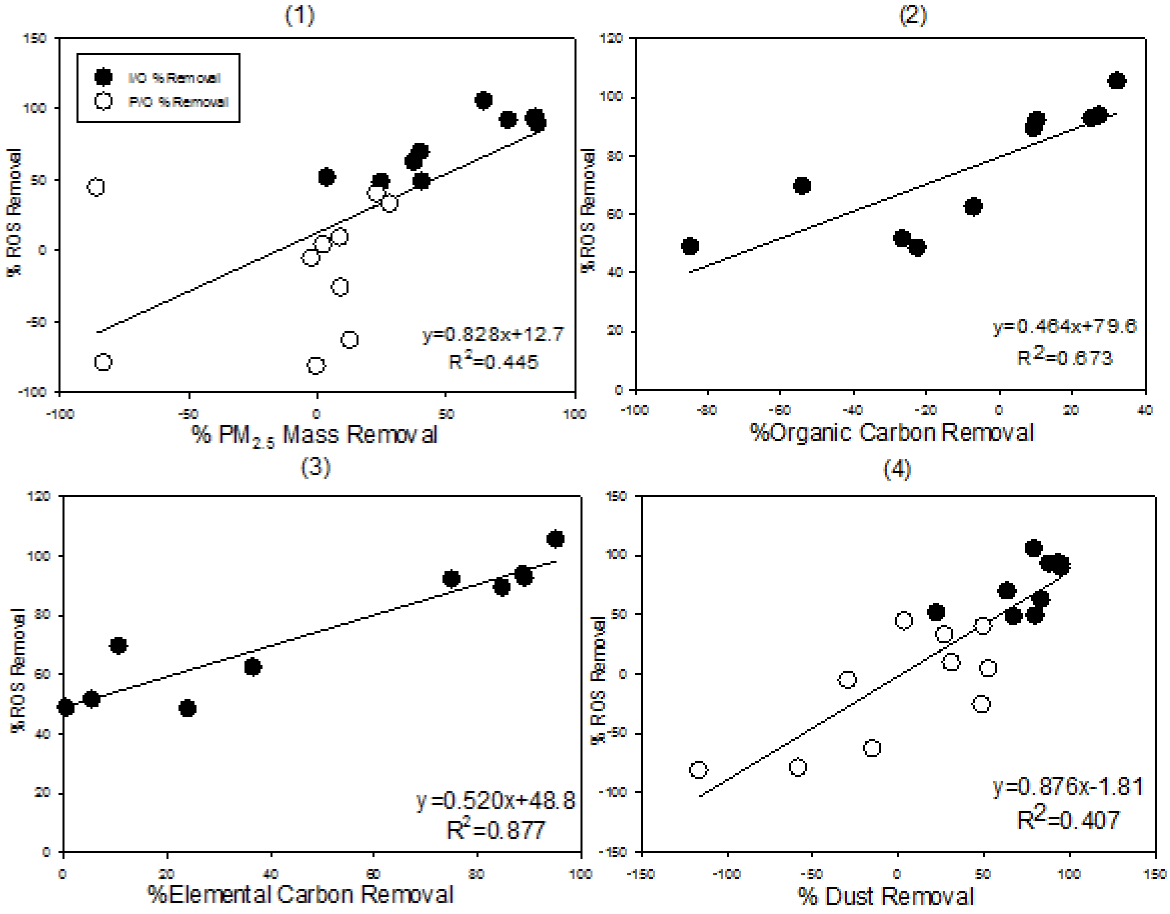
The data range is set to 0.0-2.0 for both sham and true conditions, values of boron(B) is out of range and can be found in Table 3.

Figure 4 Personal Exposure to Outdoor Concentration Ratio of Main Chemical Species



The data range for the two figures are set to 0-4. Elements with star have values that are out of range and can be found in Table 3.

Figure 5 Correlation of percent removal of reactive oxygen species with percent removal of (1) fine particulate matter mass, (2) organic carbon, (3) elemental carbon, and (4) dust for both percent removal indoor to outdoor (I/O % removal) and personal exposure to outdoor (P/O % removal).



2.7 Tables

Table-1 Main species' average concentration and enrichment factor for outdoor, indoor, and personal exposure, and indoor to outdoor removal percentage during sham and true filtration.

	Outdoor Concentration*		Indoor Concentration*		Personal Exposure Concentration*		I/O % Removal (Sham)		I/O % Removal (True)		Enrichment Factor
Species	Value	STD	Value	STD	Value	STD	Value	STD	Value	STD	Value
Mass	59	30	29	25	59	26	29	16	79	9	*
OC	11	1.0	12	4.4	*	*	-40	31	21	10	*
EC	2.5	0.75	1.4	1.1	*	*	16	15	70	36	*
Sulfate	18	10	9.1	9.1	14	9.4	29	21	83	17	*
Nitrate	7.7	7.0	2.1	2.6	4.5	4.3	59	37	91	10	*
Ammonium	8.1	5.9	3.9	4.6	7.6	6.0	33	23	86	18	*
Dust	6.8	3.5	1.4	0.90	6.1	3.4	64	22	94	98	*
Sb	3.7	0.20	1.2	0.080	5.9	9.2	28	9	90	1	4,200
Cd	0.68	0.048	0.35	0.025	0.69	0.41	26	9	82	2	2,300
Ag	0.15	0.053	0.094	0.037	0.20	0.14	12	56	78	14	1,100
Zn	100	14	45	5.6	110	62	30	15	82	3	620
Sn	2.5	0.17	2.0	0.14	3.3	1.6	-32	16	74	3	470
As	3.2	0.93	1.4	0.76	2.7	1.8	28	42	88	16	460
Mo	0.76	0.080	0.38	0.043	3.5	4.3	11	15	70	4	320
Cu	12	0.96	5.3	0.41	20	12	31	10	85	2	300
Tl	0.39	0.016	0.19	0.0086	0.39	0.20	24	5	85	2	180
B	5.7	0.55	4.5	0.49	7.0	5.9	-98	20	45	9	140
W	0.47	0.034	0.31	0.023	0.45	0.36	11	11	88	2	120
Pb	35	2.9	16	1.4	33	17	27	11	85	2	99
Ni	2.6	0.33	1.7	0.24	4.2	1.7	-26	22	49	7	65
Cr	5.0	0.39	2.3	0.24	6.4	2.4	15	12	69	4	53
Mn	28	2.2	8.8	0.72	25	9.0	40	8	90	2	14

Fe	630	46	160	14	1,000	850	39	6	87	2	6.1
K	550	62	220	29	480	170	50	15	89	4	5.7
Ca	910	79	130	15	570	400	32	5	81	2	5.5
Na	250	28	110	14	260	110	70	14	89	5	3.1
Mg	230	19	34	4.2	140	96	33	6	78	2	2.8
Al	470	39	70	7.7	290	200	67	6	91	2	1.0

Unit of PM mass, OC, EC, sulfate, nitrate, ammonium, dust is µg/m^3, and all metals are in the unit of ng/m^3.

Table 2 Correlation coefficients between outdoor and indoor concentration of mass, OC, EC, dust, sulfate, nitrate and ammonium as well as estimated Infiltration factor for these species.

	Sham Filtration				True Filtration			
	Correlation Coefficient	P-Value	Infiltration factor	Indoor Source	Correlation Coefficient	P-Value	Infiltration factor	Indoor Source
Mass	**0.93**	**0.02**	**0.49**	**13.81**	0.67	0.22	0.06	5.83
OC	0.56	0.33	0.33	0.67	0.27	0.66	0.20	6.21
EC	**0.88**	**0.05**	**1.07**	**-0.58**	0.42	0.48	-0.19	0.94
Dust	0.09	0.88	-0.04	2.18	0.55	0.33	0.03	0.40
Sulfate	**0.88**	**0.05**	**0.83**	**-1.66**	0.46	0.43	0.06	0.44
Nitrate	0.27	0.66	0.09	2.88	0.36	0.55	0.03	0.14
Ammonium	**0.91**	**0.03**	**0.58**	**1.16**	0.42	0.48	0.04	0.09

Species with correlation coefficient larger than 0.8 is bolded in the table, which means their outdoor and indoor concentration are considered as well-correlated.

Table 3 Indoor to outdoor, personal exposure to outdoor and personal exposure to indoor ratio of main species during sham and true filtration.

	I/O(Sham)		I/O(True)		P/O(Sham)		P/O(True)		P/I(Sham)		P/I(True)	
Species	Value	STD	Value	STD	Value	STD	Value	STD	Value	STD	Value	STD
Mass	0.71	0.16	**0.21**	**0.091**	0.94	0.054	1.3	0.74	**1.4**	**0.27**	**6.4**	**3.7**
OC	**1.4**	**0.31**	**0.79**	**0.10**	*	*	*	*	*	*	*	*
EC	**0.84**	**0.15**	**0.30**	**0.36**	*	*	*	*	*	*	*	*
Sulfate	**0.71**	**0.21**	**0.17**	**0.17**	0.95	0.14	0.75	0.27	1.5	0.49	*17*	*25*
Nitrate	**0.41**	**0.37**	**0.094**	**0.096**	**0.63**	**0.19**	0.68	0.63	2.4	1.5	*37*	*69*
Ammonium	**0.67**	**0.23**	**0.14**	**0.18**	0.94	0.12	0.88	0.42	1.5	0.36	*94*	*190*
Dust	**0.36**	**0.22**	**0.097**	**0.061**	0.98	0.63	1.00	0.39	3.4	2.2	**14**	**9.8**
Sb	**0.72**	**0.23**	**0.10**	**0.076**	*3.2*	*3.9*	0.86	0.58	*4.5*	*5.0*	12	11
Cd	**0.74**	**0.20**	**0.18**	**0.18**	1.0	0.14	0.83	0.39	1.5	0.64	*14*	*19*
Ag	0.88	0.33	**0.22**	**0.20**	1.2	0.52	1.2	0.69	1.4	0.41	*13*	*13*
Zn	**0.70**	**0.22**	**0.18**	**0.20**	1.1	0.33	1.1	0.57	1.7	0.72	*15*	*17*
Sn	**1.3**	**0.28**	**0.26**	**0.22**	**1.4**	**0.31**	1.2	0.67	1.1	0.18	*10*	*12*
As	**0.72**	**0.24**	**0.12**	**0.17**	0.88	0.27	0.74	0.49	1.3	0.52	*15*	*18*
Mo	0.89	0.64	**0.30**	**0.23**	*5.3*	*6.7*	*9.5*	*12*	*8.2*	*11*	*25*	*19*
Cu	**0.69**	**0.22**	**0.15**	**0.12**	1.8	1.3	2.0	1.0	2.7	1.8	**18**	**11**
Tl	**0.76**	**0.20**	**0.15**	**0.15**	0.99	0.077	0.76	0.42	**1.4**	**0.37**	*15*	*18*
B	**2.0**	**0.96**	0.55	0.78	1.7	0.82	1.3	0.81	0.98	0.61	4.9	4.4
W	0.89	0.18	**0.12**	**0.097**	0.96	0.049	0.79	0.24	1.1	0.23	**7.1**	**5.7**
Pb	**0.73**	**0.20**	**0.15**	**0.15**	0.99	0.082	0.74	0.34	1.4	0.41	6.4	5.4
Ni	1.3	1.0	**0.51**	**0.43**	**1.6**	**0.45**	2.3	1.5	1.8	0.82	**5.0**	**1.7**
Cr	0.85	0.74	**0.31**	**0.22**	1.4	0.59	1.4	0.60	2.3	1.5	4.8	1.8
Mn	0.61	0.22	**0.13**	**0.13**	1.0	0.43	0.92	0.35	1.8	0.74	**9.0**	**7.4**
Fe	**0.50**	**0.21**	**0.11**	**0.098**	*2.0*	*2.2*	1.8	0.96	4.3	4.1	24	23

K	**0.68**	**0.17**	**0.19**	**0.21**	0.96	0.27	0.92	0.49	1.5	0.54	**6.0**	**4.0**
Ca	**0.30**	**0.15**	**0.11**	**0.068**	0.83	0.52	0.88	0.59	3.3	2.0	**8.1**	**6.8**
Na	**0.67**	**0.18**	**0.22**	**0.23**	1.0	0.27	1.1	0.48	1.6	0.54	**6.3**	**4.0**
Mg	**0.33**	**0.23**	**0.091**	**0.063**	0.83	0.46	0.89	0.50	3.7	2.6	**9.1**	**7.7**
Al	**0.36**	**0.29**	**0.096**	**0.059**	0.79	0.36	0.91	0.46	3.3	2.1	**8.5**	**6.2**

Bolded number means the ratio is statically different than 1; underlined number means their standard deviation is equal or larger than the value.

Table 4 Estimation of PM personal exposure concentration from human activities record (two assumptions: office concentration same as 1) indoor or 2) outdoor), comparing to measured PM personal exposure concentration (48-hour measurement period).

Date	Hours in room with filtration	Hours in other rooms	Hours in office	Hours outside	Filtration	Measured Indoor	STD	Measured Outdoor	STD	Estimated PE-1*	STD	Estimated PE-2*	STD	Measured PE	STD
7/30/2016(1)	15	20	4	9	sham	57	6	94	9.6	64	6.7	82	8.5	95	9.9
7/30/2016(2)	20	14	2	12	sham	71	7.4	120	12	83	8.6	99	10	100	11
8/2/2016(1)	18	30	0	0	sham	46	4.8	47	5	46	4.8	47	4.9	43	4.7
8/2/2016(2)	19	12	15	2	TRUE	6.2	0.83	43	4.5	7.7	0.98	28	3.1	79	8.4
8/5/2016	28	0	16	4	sham	50	5.2	67	6.9	52	5.4	57	5.9	66	7.1
8/9/2016	34	5	9	0	sham	21	2.4	34	6.9	21	2.4	25	3.7	31	3.5
8/12/2016(1)	2	36	8	2	TRUE	9.4	1.2	60	6.2	12	1.4	58	6	46	5.5
8/12/2016(2)	38	8	0	2	TRUE	11	1.3	71	7.3	13	1.5	23	2.5	51	6
8/15/2016(1)	17	17	0	14	TRUE	8.4	1.1	24	2.6	13	1.5	18	2	55	5.9
8/15/2016(2)	18	5	0	25	TRUE	7.7	0.98	30	3.2	19	2.1	21	2.4	24	2.9

*Estimated PE-1 assumes that PM2.5 concentration in other rooms and office is the same as measured indoor value; estimated PE-2 assumes that PM2.5 concentration in other rooms and office is the same as measured outdoor values.

Table 5 Correlation of ROS concentration (both linear and LOG scale) and ROS percent removal with measured species.

	Correlation with ROS Concentration		Correlation with ROS Concentration (LOG Scale)		Correlation with ROS removal (I/O)		Correlation with ROS removal (P/O)	
Species	Value	P-Value	Value	P-Value	Value	P-Value	Value	P-Value
Mass	**0.87**	**0.00**	**0.88**	**0.00**	**0.87**	**0.00**	0.14	0.69
OC	0.2	0.40	0.51	0.02	**0.82**	**0.00**	*	*
EC	**0.86**	**0.00**	**0.96**	**0.00**	**0.93**	**0.00**	*	*
Sulfate	**0.72**	**0.00**	**0.76**	**0.00**	0.64	0.05	0.54	0.11
Nitrate	**0.73**	**0.00**	**0.81**	**0.00**	0.61	0.06	0.59	0.07
Ammonium	**0.84**	**0.00**	**0.79**	**0.00**	0.79	0.01	0.69	0.03
Dust	0.44	0.02	**0.71**	**0.00**	0.61	0.06	**0.73**	**0.02**
Sb	0.64	0.00	0.66	0.00	**0.87**	**0.00**	0.65	0.04
Cd	**0.78**	**0.00**	**0.80**	**0.00**	**0.8**	**0.01**	0.64	0.05
Ag	**0.73**	**0.00**	**0.74**	**0.00**	**0.81**	**0.00**	0.28	0.44
Zn	0.66	0.00	**0.77**	**0.00**	**0.74**	**0.01**	0.38	0.27
Sn	**0.71**	**0.00**	**0.74**	**0.00**	**0.81**	**0.00**	**0.86**	**0.00**
As	**0.80**	**0.00**	**0.75**	**0.00**	**0.79**	**0.01**	0.64	0.05
Mo	0.3	0.11	0.55	0.00	0.56	0.09	-0.02	0.96
Cu	**0.75**	**0.00**	**0.77**	**0.00**	**0.83**	**0.00**	0.36	0.31
Tl	**0.79**	**0.00**	**0.77**	**0.00**	**0.83**	**0.00**	**0.72**	**0.02**
B	0.04	0.82	0.30	0.11	0.52	0.12	-0.39	0.27
W	0.41	0.02	**0.74**	**0.00**	**0.83**	**0.00**	**0.74**	**0.01**
Pb	**0.85**	**0.00**	**0.76**	**0.00**	**0.83**	**0.00**	**0.74**	**0.01**
Ni	0.24	0.21	0.58	0.00	0.43	0.22	-0.37	0.29
Cr	0.35	0.06	0.59	0.00	0.47	0.17	-0.01	0.98
Mn	0.67	0.00	**0.74**	**0.00**	**0.76**	**0.01**	**0.75**	**0.01**
Fe	0.57	0.00	**0.70**	**0.00**	**0.73**	**0.02**	0.45	0.19

K	0.61	0.00	**0.72**	**0.00**	**0.72**	**0.02**	**0.72**	**0.02**
Ca	0.38	0.04	0.67	0.00	0.54	0.11	**0.74**	**0.01**
Na	0.55	0.00	**0.75**	**0.00**	0.67	0.04	0.52	0.12
Mg	0.36	0.05	0.69	0.00	0.62	0.05	**0.77**	**0.01**
Al	0.34	0.06	**0.70**	**0.00**	0.57	0.08	**0.75**	**0.01**

Correlation coefficient larger than 0.7 and with p-value less than 0.05 is bolded in the table.

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2.9 Supplemental Materials

Table 1 Sampling Home Description

Home	Setting	Location	Floor	Additional Information
1	2-bedroom	Back of road; Close to second ring road	3rd	Indoor porch
2	1-bedroom	Close to a busy road	ground	Large balcony elevated above the ground by 8 feet
3	multi-bedroom	Tsinghua Campus	6th	2 enclosed porches, no air conditioner
4	1-bedroom	Tsinghua Campus	3rd	Indoor porch, no air conditioner
5	1-bedroom	N/A	N/A	Construction occurring, indoor porch, smoking in balcony
6	multi-bedroom	N/A	19th	New apartment, large indoor porch
7	2-bedroom	N/A	N/A	Old building, construction occurring right behind the building
8	2-bedroom	Back of busy roads in the military complex	3rd	Indoor porch
9	1-bedroom	N/A	12th	Indoor porch

Table 2 All Species' Concentration and Enrichment Factor under Outdoor, Indoor and Personal Exposure Conditions, and Indoor to Outdoor Removal Percentage in Sham and True Filtration

	Outdoor Conc.	STD	Indoor Conc.	STD	Personal Exposure	STD	I/O %Removal (sham)	STD	I/O %Removal (true)	STD	Enrichment Factor
Mass	59	30	29	25	59	26	29	16	79	9.1	N/A
OC	11	1.0	12	4.4	N/A	N/A	-40	31	21	10	N/A
EC	2.5	0.75	1.4	1.1	N/A	N/A	16	15	70	36	N/A
Sulfate	18	10	9.1	9.1	14	9.4	29	21	83	17	N/A
Nitrate	7.7	7.0	2.1	2.6	4.5	4.3	59	37	91	9.6	N/A
Ammonium	8.1	5.9	3.9	4.6	7.6	6.0	33	23	86	18	N/A
Dust	6.8	3.5	1.4	0.90	6.1	3.4	64	22	94	98	N/A
Rh	0.0011	0.00050	0.00036	0.00044	0.0020	0.0016	47	55	91	38	N/A
Pd	0.016	0.0031	0.0021	0.0022	0.015	0.0053	67	24	99	13	N/A
Sb	3.7	0.20	1.2	0.080	5.9	9.2	28	9	90	1	4,200
Cd	0.68	0.048	0.35	0.025	0.69	0.41	26	9	82	2	2,300
Ag	0.15	0.053	0.094	0.037	0.20	0.14	12	56	78	14	1,100
Zn	100	14	45	5.6	110	62	30	15	82	2	620
Sn	2.5	0.17	2.0	0.14	3.3	1.6	-32	16	74	3	470
As	3.2	0.93	1.4	0.76	2.7	1.8	28	42	88	16	460
Mo	0.76	0.080	0.38	0.043	3.5	4.3	11	15	70	4	320
Cu	12	0.96	5.3	0.41	20	12	31	10	85	2	300
Tl	0.39	0.016	0.19	0.0086	0.39	0.20	24	5	85	2	180
B	5.7	0.55	4.5	0.49	7.0	5.9	-98	20	45	9	140
W	0.47	0.034	0.31	0.023	0.45	0.36	11	11	88	2	120
Pb	35	2.9	16	1.4	33	17	27	11	85	2	99
Ni	2.6	0.33	1.7	0.24	4.2	1.7	-26	22	49	7	65
Cr	5.0	0.39	2.3	0.24	6.4	2.4	15	12	69	4	53
Cs	0.27	0.018	0.12	0.0086	0.27	0.13	26	9	89	1	15

V	2.6	0.22	1.2	0.11	2.4	2.0	33	9	91	2	14
Mn	28	2.2	8.8	0.72	25	9.0	4000	8	9000	2	14
P	27	2.5	10	1.2	25	12	35	10	86	4	12
Li	1.0	0.069	0.24	0.031	0.37	0.29	43	8	97	2	7.2
Fe	630	46	160	14	1,000	850	39	6	87	2	6.1
Ba	13	0.95	2.8	0.28	24	29	59	6	90	2	6.1
Co	0.28	0.027	0.068	0.012	0.26	0.13	55	10	91	4	6.0
K	550	62	220	29	480	170	50	15	89	4	5.7
Ca	910	79	130	15	570	400	32	5	81	2	5.5
Rb	2.0	0.17	0.79	0.085	1.8	0.76	31	11	89	3	5.3
U	0.034	0.0025	0.0087	0.0010	0.025	0.011	47	9	88	3	3.5
Na	250	28	110	14	260	110	70	14	89	5	3.1
Ti	46	4.0	8.3	1.2	31	21	67	6	90	3	3.0
Mg	230	19	34	4.2	140	96	33	5	78	2	2.8
Sr	3.5	0.31	0.86	0.091	2.9	1.6	61	7	87	3	2.8
Eu	0.0095	0.0010	0.0017	0.00039	0.013	0.014	64	10	93	4	2.7
Dy	0.033	0.0030	0.0072	0.0015	0.024	0.011	51	12	94	3	2.5
La	0.30	0.021	0.077	0.0069	0.36	0.29	58	6	92	1	2.4
Sc	0.076	0.013	0.0090	0.0059	0.043	0.033	72	18	96	7	1.6
Nd	0.19	0.012	0.031	0.0037	0.14	0.088	64	6	92	2	1.5
Yb	0.014	0.0029	0.0019	0.00074	0.0079	0.0055	68	14	95	4	1.5
Ce	0.52	0.042	0.061	0.015	0.59	0.64	74	6	100	2	1.4
Ho	0.0048	0.00052	0.00069	0.00025	0.0031	0.0017	69	11	94	5	1.4
Lu	0.0018	0.00030	0.00030	0.00014	0.0013	0.00077	69	16	93	8	1.4
Th	0.067	0.010	0.010	0.0023	0.064	0.043	66	12	94	3	1.4
Y	0.20	0.011	0.022	0.0019	0.089	0.048	70	4	95	1	1.3
Sm	0.030	0.0034	0.0046	0.0013	0.022	0.016	68	10	93	4	1.2
Al	470	39	70	7.7	290	200	67	6	91	2	1.0
Nb	0.098	0.012	0.018	0.0029	0.074	0.041	65	9	90	3	0.82

Table 3 Indoor to Outdoor, Personal Exposure to Outdoor and Personal Exposure to Indoor Ratio of all Species with Both Sham and True Filtration Conditions

	I/O(Sham)		I/O(True)		P/O(Sham)		P/O(True)		P/I(Sham)		P/I(True)	
	Value	STD	Value	STD	Value	STD	Value	STD	Value	STD	Value	STD
Mass	0.68	0.010	0.19	0.0055	0.94	0.0093	1.1	0.019	1.4	0.017	6.0	0.23
OC	1.4	0.20	0.78	0.11	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
EC	0.86	0.20	0.23	0.018	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
Sulfate	0.75	0.16	0.090	0.037	0.86	0.19	0.73	0.21	1.1	0.27	8.1	3.9
Nitrate	0.34	0.069	0.065	0.021	0.59	0.12	0.57	0.12	1.8	0.36	8.9	2.9
Ammonium	0.69	0.14	0.054	0.012	0.97	0.20	0.87	0.18	1.4	0.29	16	3.8
Dust	0.32	0.058	0.081	0.017	0.90	0.16	0.90	0.17	2.8	0.52	11	2.4
Rh	0.53	0.55	0.089	0.38	2.0	2.2	1.6	1.4	3.7	5.3	19	78
Pd	0.33	0.24	0.0081	0.13	1.2	0.71	0.99	0.43	3.5	3.3	120	1,900
Sb	0.74	0.086	0.11	0.014	3.4	4.6	0.87	0.87	4.6	6.2	7.7	7.9
Cd	0.75	0.088	0.11	0.016	1.0	0.51	0.73	0.54	1.4	0.69	6.4	4.9
Ag	0.88	0.56	0.19	0.14	1.3	1.1	1.2	1.1	1.4	1.3	6.3	6.2
Zn	0.71	0.15	0.10	0.025	1.1	0.64	0.82	0.60	1.5	0.91	8.0	5.9
Sn	1.3	0.16	0.22	0.030	1.4	0.64	1.1	0.56	1.1	0.49	5.1	2.5
As	0.74	0.42	0.069	0.16	0.93	0.76	0.58	0.50	1.3	1.2	8.4	22
Mo	0.81	0.15	0.19	0.039	4.0	5.1	5.1	6.0	4.9	6.3	27	32
Cu	0.70	0.096	0.13	0.023	1.7	1.3	1.7	0.72	2.4	1.8	14	6.2
Tl	0.76	0.053	0.12	0.018	1.00	0.29	0.80	0.58	1.3	0.37	6.4	5.2
B	1.4	0.20	0.31	0.086	1.5	1.4	1.1	0.86	1.1	1.0	3.6	3.0
W	0.90	0.11	0.098	0.015	0.94	0.59	0.79	0.42	1.0	0.66	8.1	4.5
Pb	0.73	0.11	0.13	0.020	1.0	0.41	0.75	0.46	1.4	0.57	5.9	3.7
Ni	1.1	0.22	0.26	0.070	1.4	0.46	1.4	0.73	1.4	0.46	5.4	3.1
Cr	0.85	0.12	0.22	0.043	1.3	0.47	1.1	0.57	1.5	0.57	5.1	2.8

Cs	0.74	0.093	0.11	0.015	1.0	0.29	0.84	0.71	1.4	0.39	7.6	6.4
V	0.67	0.094	0.092	0.018	0.87	0.78	0.75	0.37	1.3	1.2	8.2	4.4
Mn	0.60	0.078	0.10	0.018	0.99	0.43	0.82	0.35	1.7	0.75	8.0	3.4
P	0.65	0.10	0.14	0.038	1.0	0.61	0.86	0.41	1.6	0.96	6.3	3.5
Li	0.57	0.082	0.026	0.023	0.51	0.35	0.28	0.29	0.89	0.61	11	20
Fe	0.48	0.064	0.096	0.018	1.8	1.7	1.9	1.4	3.8	3.7	19	14
Ba	0.41	0.065	0.10	0.019	2.9	3.6	1.5	1.3	7.0	8.8	15	14
Co	0.45	0.10	0.093	0.035	0.99	0.60	1.1	0.60	2.2	1.4	11	8.4
K	0.67	0.15	0.15	0.037	0.94	0.37	0.85	0.41	1.4	0.58	5.7	2.8
Ca	0.26	0.048	0.082	0.018	0.78	0.46	0.71	0.55	3.0	1.8	8.7	7.1
Rb	0.69	0.11	0.11	0.027	0.91	0.36	0.89	0.52	1.3	0.53	8.3	5.6
U	0.53	0.090	0.12	0.026	0.94	0.51	0.80	0.37	1.8	1.0	6.6	3.5
Na	0.67	0.14	0.16	0.046	1.00	0.49	1.00	0.56	1.5	0.76	6.4	4.0
Ti	0.33	0.064	0.10	0.028	0.69	0.32	0.87	0.65	2.1	1.0	8.5	7.1
Mg	0.30	0.055	0.073	0.018	0.76	0.40	0.75	0.60	2.6	1.4	10	8.8
Sr	0.39	0.074	0.13	0.029	1.0	0.75	0.89	0.46	2.6	1.9	6.8	3.8
Eu	0.36	0.10	0.073	0.037	2.1	2.4	1.2	1.0	5.8	7.0	16	18
Dy	0.49	0.12	0.063	0.031	0.91	0.41	0.70	0.48	1.9	0.94	11	11
La	0.42	0.057	0.077	0.014	0.94	0.75	1.7	1.3	2.2	1.8	22	18
Sc	0.28	0.18	0.038	0.067	0.68	0.42	0.69	0.73	2.4	2.1	18	44
Nd	0.36	0.059	0.080	0.018	0.92	0.59	0.86	0.62	2.6	1.7	11	8.7
Yb	0.32	0.14	0.047	0.045	0.81	0.68	0.68	0.65	2.5	2.6	15	18
Ce	0.26	0.056	0.0033	0.021	0.82	0.88	1.8	1.7	3.2	3.6	540	3,800
Ho	0.31	0.11	0.058	0.047	0.86	0.54	0.73	0.48	2.7	2.1	13	15
Lu	0.31	0.16	0.070	0.076	0.86	0.51	0.66	0.66	2.8	2.0	9.4	16
Th	0.34	0.12	0.057	0.025	1.3	0.96	1.0	0.93	3.9	2.9	18	18
Y	0.30	0.043	0.051	0.0089	0.76	0.35	0.52	0.37	2.5	1.2	10	7.6
Sm	0.32	0.098	0.065	0.039	0.92	0.77	0.90	0.72	2.9	2.7	14	16
Al	0.31	0.059	0.079	0.016	0.74	0.39	0.78	0.65	2.4	1.3	9.9	8.4

Nb	0.35	0.091	0.098	0.030	0.89	0.63	0.91	0.54	2.6	1.9	9.2	5.8

Table 4 Reactive Oxygen Species Activity Value (Unit: µg Zymosan Units /m^3)

	Outdoor		Indoor		Personal Exposure	
Sham Filtration	Value	STD	Value	STD	Value	STD
7/30/2016	320	23	97	11	580	64
7/30/2016	381	8	195	12	622	84
8/2/2016	195	9	94	7	245	16
8/5/2016	290	18	150	6	278	11
8/9/2016	161	9	61	6	146	11
True Filtration						
8/2/2016	187	19	20	8	336	19
8/12/2016	367	8	27	6	220	22
8/12/2016	253	13	16	6	170	16
8/15/2016	88	13	0	6	92	7
8/15/2016	165	12	13	6	92	8

Chapter 3 Conclusion and Implications

People have studied the particulate matter in developing countries for a long time (i.e., since the early 2000s), including both indoor and ambient air pollution. Indoor air quality is a critical issue to study since people spend considerable amounts of time indoors, especially sensitive elderly and young people. Rural and urban indoor pollution are both critical issues, but they are different, as mentioned in the introduction. As more people move to cities, the urban pollution becomes a greater issue for larger populations and the indoor air pollution will become a critical problem from people in megacities. It was not until the most recent 5 years that various types of air cleaning devices became available, and the market has grown rapidly in megacities in response to unhealthy levels of air pollution; thus, not many studies have been done to test the effectiveness of air cleaners, so there is not adequate data available to show that air cleaners could provide a healthy indoor environment for occupants in residential homes. This thesis is one of the first few studies on the effectiveness of air cleaners for residential housing in developing countries with serious particulate matter pollution problems. The sampling was taken place in Beijing, China, which is one of the big cities suffering from serious particulate matter pollution. The study obtained the indoor, outdoor, and personal exposure concentrations of $PM_{2.5}$; chemical analysis provides data of $PM_{2.5}$ components, which include organic carbon, elemental carbon, sulfate, nitrate, ammonium, dust, and 21 selected metals; oxidative potential (ROS activity) of $PM_{2.5}$ is measured by macrophage-based assays which add toxicological information to the research. The data analysis uses infiltration factor and I/O ratio to describe outdoor to indoor changes, and microenvironment method is used to explain the high personal exposure concentration. This thesis also used the Ogawa passive O_3 and NO_2 sampler to measure the indoor, outdoor, and personal exposure concentrations; analyses methods are developed in the Water Science and Engineering Lab at UW-Madison which is described in the appendix.

Our results obtained during sham filtration agree well with previous research showing that the concentrations of all PM components except organic carbon were lower indoors than in the outdoor environment (i.e., I/O ratios < 1). During true filtration, the I/O ratio reduced significantly for each of the measured components of PM (except OC), indicating that the air cleaner effectively reduces the total PM mass concentration and its components. Low removal of OC has been discussed in the literature and is likely a result of the continuous emissions of cooking residuals and VOC from furniture in the indoor environment. The average indoor PM mass concentration during true filtration was 8.47 $\mu g/m^3$ (all values were under 10.50 µg/m^3) – well below the World Health Organization (WHO) air quality guideline for $PM_{2.5}$ of 25 µg/m^3 (24-hour average) (WHO, 2006). However, the average indoor mass concentration during sham filtration was 49.02 µg/m^3 – nearly double the WHO standard. This indicates that the air cleaner may effectively reduce $PM_{2.5}$ particles and provide a healthier indoor environment.

We did not find any significant differences in personal exposure to PM, and its components dependent on filtration status; thus, the air cleaner may not effectively reduce average personal exposures. This may be because the air cleaner was installed in the bedroom, but participants spent one third to half of their time outdoors or in an office during sampling. Also, literature suggests that some microenvironments such as transportation (i.e., motor vehicles, subways, buses) and activities such as cooking and exercising can result in high $PM_{2.5}$ exposure over short periods of time and may disproportionately contribute to high average personal exposures (Steinle et al., 2015). This is particularly relevant to our study as most participants reported that they commute by subway or bike to their offices. To better understand and estimate personal exposure concentrations and to reduce average personal exposure, data on $PM_{2.5}$ concentrations in different microenvironments, such as workplaces, transportation, restaurants, etc., should be collected; moreover, time-activity surveys completed by participants should be updated accordingly with specific microenvironments.

The results from the Reactive Oxygen Species activity analyses show that the air cleaner reduces the ROS activity of the indoor environment, but this reduction is not observed for personal exposure. The correlation analysis indicates that ROS activity is well-correlated with most of the measured PM components, and many of these correlations have been found in previous studies. The percentage removal of ROS also has a high correlation coefficient with the percentage removal of PM components (both I/O and P/O). These results can inform further research into the mechanisms of ROS increases. Additionally, the results of this study can be applied to future studies on the effectiveness of air cleaners to provide health benefits to patients with respiratory diseases (e.g., asthmatics).

For the gas phase pollutants, ozone and nitrogen dioxide, they did not follow the trend of particles. The indoor ozone concentration varied by home and date, and personal exposure to ozone was well below the safety standard (EPA). Based on the summer sampling data, air cleaner does not effectively reduce ozone concentration for either indoor or personal exposure. The air cleaner is effective in reducing the indoor NO_2 concentration, but the reduction in personal exposure is not effective. However, outdoor, indoor, and personal exposure are far below the WHO NO_2 standard, which should not cause concern.

This thesis offers valuable data in air cleaner effectiveness studies in polluted areas and can be used as a pilot study for further research on air cleaning devices. It also suggests that people should try to avoid outdoor activities and stay at home with the air cleaner working to reduce personal exposure to $PM_{2.5}$ on heavily polluted days, especially very young and elderly people as well as patients with respiratory problems. However, there is much that can be done to improve this study. First, additional research is needed to better understand the contribution of microenvironments to personal exposure to $PM_{2.5}$, so that strategies can be found to reduce personal exposure concentration; then, the next phase of this study (if applicable) should have a larger sample size to make the conclusion more persuasive, and there will be more available and robust statistical analysis methods, such as source apportionment; finally, data should be collected in other seasons across more geographical locations, since pollution levels and PM components will be different. This thesis can also be used as a pilot study for further research on air cleaners' effects in relieving symptoms of asthma and respiratory diseases, and data in this study could help with the experimental design. Also, results of our studies could lead to innovative research in the air cleaning manufacturing industry and provide consumer guidance in using air cleaners to reduce PM concentrations in homes.

Appendix

Ozone and Nitrogen Dioxide Analytical Methods

The concentration of ozone and nitrogen dioxide were measured in this study as well, and they were sampled by Ogawa O_3/NO_2 passive sampler. The sampling schedule was the same as $PM_{2.5}$ with air cleaner under sham and true filtration conditions, and there were measured values for outdoor, indoor and personal exposure. This section introduces the analytical methods for ozone and nitrogen dioxide developed at water science and engineering lab at University of Wisconsin Madison, and the results are shown. The data is not sufficient for chapter discussion in this thesis, but they can be used in the future studies.

A.1.1 Analytical method of Ozone by Ogawa sampler

In the study, Ogawa ozone passive sampler badge is used to measure the O_3 concentration in air. The filter for ozone badge is coated with a nitrite-based solution, and ozone oxidizes the nitrite to nitrate. Therefore, after exposure, the filter is extracted with ultra-pure (Milli-Q) water and the extract is analyzed by ion chromatography (IC) to determine the nitrate ion concentration, which is used to calculate the ozone concentration.

The sampling period in the study is 48 hours, and after exposure, filters are kept in Teflon bottles in freezer before analyses. There are two filters per sampler for duplicates uses, and one of them is used for analyses in our study. Since nitrite on coated filters is slowly converted to nitrate even without exposure to ozone, 10% of blank filters have been collected including trip blanks and loading blanks.

Step 1. Use pre-cleaned tweezer to transfer ozone filters (blanks and samples) from storage bottles to 15 mL plastic vials which have been washed and dried.

Step 2. Pipette 5 ml of Milli-Q water into each extract vial using the calibrated automatic pipet, and make sure filters in each extract vial are completely immersed in the aqueous solution.

Step 3. Place the vials with extractions on a rack in the ultrasonic bath. Adjust the level of water in the bath to be high enough to immerse the bottom of each vial.

Step 4. Sonicate the extract vials for 5 minutes, rotate the rack 90 degrees clockwise; repeat the rotation twice for a total sonication time of 20 minutes.

Step 5. Use 0.45 μm Whatman filter to filtrate extracts into cleaned 15 mL plastic vial by using plastic syringe. Whatman filter and syringe are disposable for each extract.

Step 6. The extractions are measure by Ion Chromatography for nitrate concentration, so standards are prepared with only nitrate ion.

An example calculation of ozone concentration is attached.

PSD Sampling Time = 48 hours

Sample Extraction Volume = 5ml

Ion Chromatographic analysis of extract = 0.1254 µg/ml nitrate

Total nitrate = 0.1254 µg/ml x 5ml = 0.627 µg

PSD Sampling rate for Ozone is 21.8 mls/min

$$O3\ ppmV = \frac{0.627\mu g\ NO3^{-}}{21.8\text{ml O3 / min x 2880 min}} \times \frac{1\mu\text{mol } O_3}{62\mu\text{g NO3}^{-}} \times \frac{1\mu\text{mol } O_3}{1\mu\text{mol NO3}^{-}} \times \frac{24.45\ \mu\text{L } O_3}{1\mu\text{mol } O_3} \times \frac{10^{-6}\text{M}^3}{1000\ \mu\text{l}} \times \frac{10^{6}\mu\text{l}}{\text{L}} \times \frac{10^{6}\text{ml O}_3}{\text{M}^3\ \text{O}_3}$$

= 0.003938 µL/L or 0.003938 ppm

=3.938 ppb

A.1.2 Quality Assurance /Quality Control (QAQC)

The analytical instrument (Ion Chromatography, ICS-5000 +, Dionex) is reliable and well-maintained. The ultrasonic water bath is working properly as well.

There are two ways of quality control: working standard nitrate solution is prepared from 1000 ppm standard solution, and the concentration is 10, 20, 30, 40and 50 mg/L. In the first QC method, 10 µL of each working standard solution is directly transferred onto the blank Ogawa filter by calibrated pipette. Then, the filters as well as the standard solution residuals are extracted in 5mL of Milli-Q water and followed the same procedure indicated above. For the second QC method, blank Ogawa filter is put into 5 mL Milli-Q water extract vial and the prepared nitrate working standard solution is added to the extraction. Then, it follows the same steps as the analysis method. Both QC methods have blank filter and blank extract, which means no nitrate solution is added. The calculated results indicate the method can limit the error ranges within 20%.

A.1.3 O_3 Results from the summer sampling

Figure A.1 Ozone Concentration with Sham Filtration

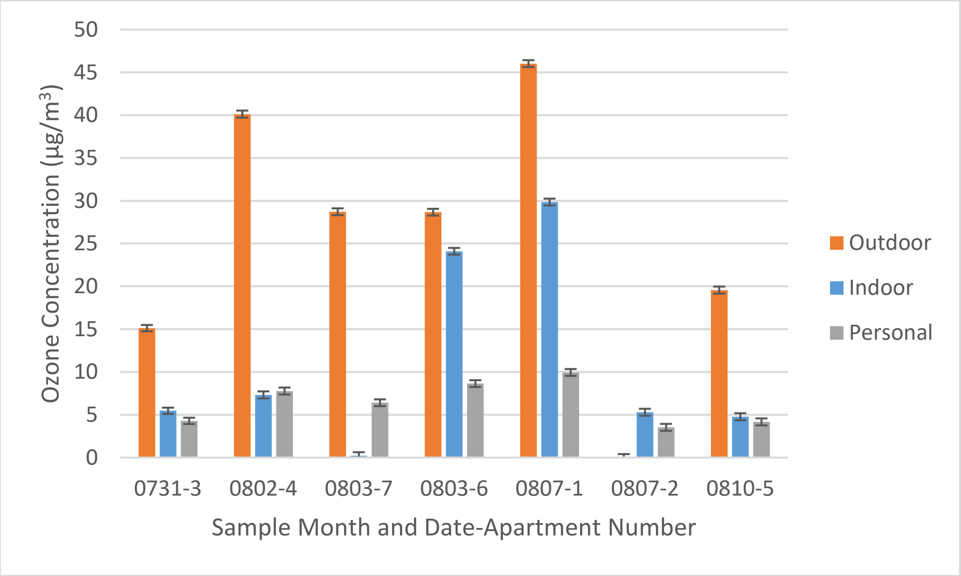
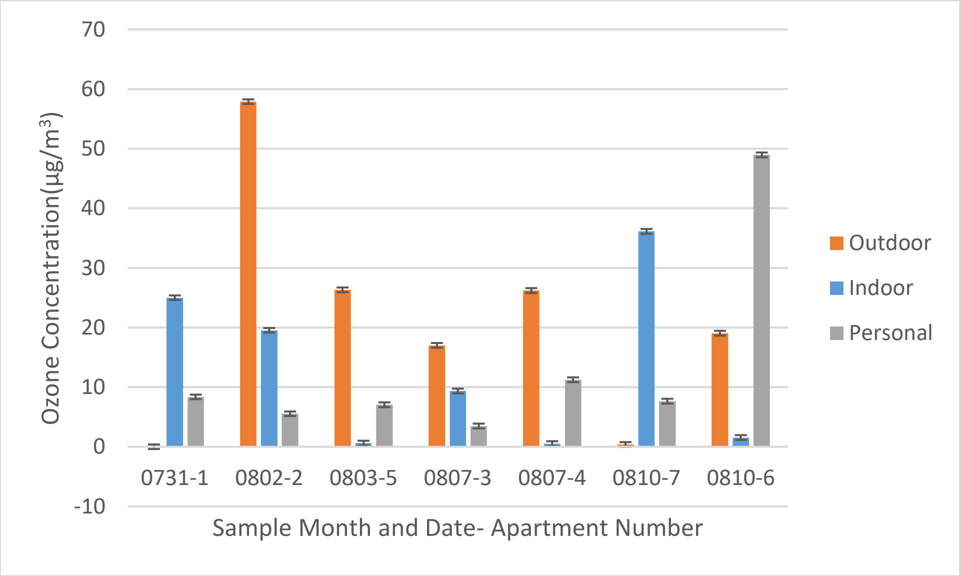


Figure A.2 Ozone Concentration with True Filtration



A.2.1 Analytical method of NO_2 by Ogawa sampler

The Ogawa sampler and filters can be used to monitor the concentration of NO, NO_2, NO_x and SO_2, and we only use it for NO_2 measurement in our study. The sampling period in the study is 48 hours, and after exposure, filters are kept in Teflon bottles in freezer before analyses. There are two filters per sampler for duplicates uses, and one of them is used for analyses in our study.

Step 1. Prepare solutions.

Sulfanilamide solution is prepared by dissolving 80 g of reagent grade sulfanilamide in a mixture of 200 ml concentrated phosphoric acid and 700 ml water. This solution is then diluted with water to make a total of 1000 ml.

The NEDA solution is prepared by dissolving 0.56 g N-(1-Naphthyl)-ethylenediamine dihydrochloride into 100 ml water.

Nitrite working standard solutions of 0 - 1.0 μg nitrite/mL by using 1000 ppm standard nitrate solution.

Step 2. Put cellulose fiber filter into a 25-mL glass vial, containing 8mL water, and then shake immediately.

Step 3. Over the next 30 minutes, occasional shaking of the vials.

Step 4. Prepare the color-producing reagent before using: mix the sulfanilamide solution and the NEDA solution in a 10:1 ratio. (10 parts Sulfanilamide solution to 1 part NEDA solution).

Step 5. At the end of the first 30-minute period, vials are cooled to 0 ˚C by using water and ice mixture, and 2 mL of color producing reagent are added. Shake quickly, after adding the color-producing reagent, and continue to keep vials cool for an additional 30 minutes.

Step 6. Vials are then allowed to equilibrate at room temperature, for about 20 minutes, and the amount of colored derivative is determined with a spectrophotometer at a wavelength of 545 nm.

Step 7. Mix 8 mL of each of the nitrite working standards with 2 ml of color reagent, shaking immediately, to prepare a standard curve with spectrophotometer at a wavelength of 545 nm. Chill standard solution, like samples, before adding color reagent.

An example calculation of NO_2 concentration is attached.

1. Obtain Standard Curve for NO, NO2 and NOx

Concentration of Standard NO_2 Solution (µg/ml)		Absorbance			Slope
x	x^2	y	y - y_0	x (y - y_0)	$\frac{\Sigma x(y-y_0)}{\Sigma x^2}$
(1)	(1) x (1)		A	(1) x A	(9) / (8)
0	0	(2)			
0.1	0.01	(3)	(3) – (2)		
0.2	0.04	(4)	(4) – (2)		
0.4	0.16	(5)	(5) – (2)		
0.6	0.36	(6)	(6) – (2)		
0.8	0.64	(7)	(7) – (2)		
	Σx^2			$\Sigma x(y-y_0)$	(9) / (8)
(8)	1.21			(9)	G

2. Calculation of Concentration Conversion Coefficients for NO2

$$\alpha_{NO_2} = \frac{10000}{(0.677x[P]x[RH])+(2.009x[T])+89.8}$$

$$[P]=\left\{\frac{2P_N}{P_T+P_N}\right\}^{2/3}$$

[T]=Temperature in degree centigrade

[RH]=Humidity in %

PN=17.535(water vapor pressure in mm Hg at 20 C)

PT=vapor pressure at ambient temperature

Sample Calculation:

$$[T]=9$$
$$[RH]=60$$
$$P_N=17.535$$
$$P_T=8.609$$

$$[P]=\left\{\frac{2P_N}{P_T+P_N}\right\}^{2/3}$$

$$=\left\{\frac{2x17.535}{8.609+17.535}\right\}^{2/3}$$

$$=1.216$$

$$\alpha_{NO_2} = \frac{10000}{(0.677x[P]x[RH])+(2.009x[T])+89.8}$$

$$= \frac{10000}{(0.677x1.216x60)+(2.009x9)+89.8}$$

$$= 63.58$$

3. Calculate the NO2 Concentration

Sample Absorbance (9)	(9)
Absorbance (10)	(9) – (2)
Solution Concentration (μg/ml) (11)	(10) / G
Collected Weight (ng) (12)	(11) x 8 x 1000
Concentration (PPB) (13)	αNO2 x (12) / (3)

A.2.2 Quality Assurance /Quality Control (QAQC)

All chemicals used in this method are recently bought, and the spectrophotometer is regularly maintained.

To control the quality of the analytical method, nitrite working standard solutions (0.2,0.4,0.6,0.8 and 1 μg /mL) are transferred onto Ogawa NO_x filters, and then follow the same procedures to extract in 8 mL water and further steps. Blank filter is also included as a part of QC. The calculated results indicate the method can limit the error ranges within 20%.

A.2.3 NO_2 Results from the summer sampling

Figure A.3 Nitrogen Dioxide Concentration with Sham Filtration

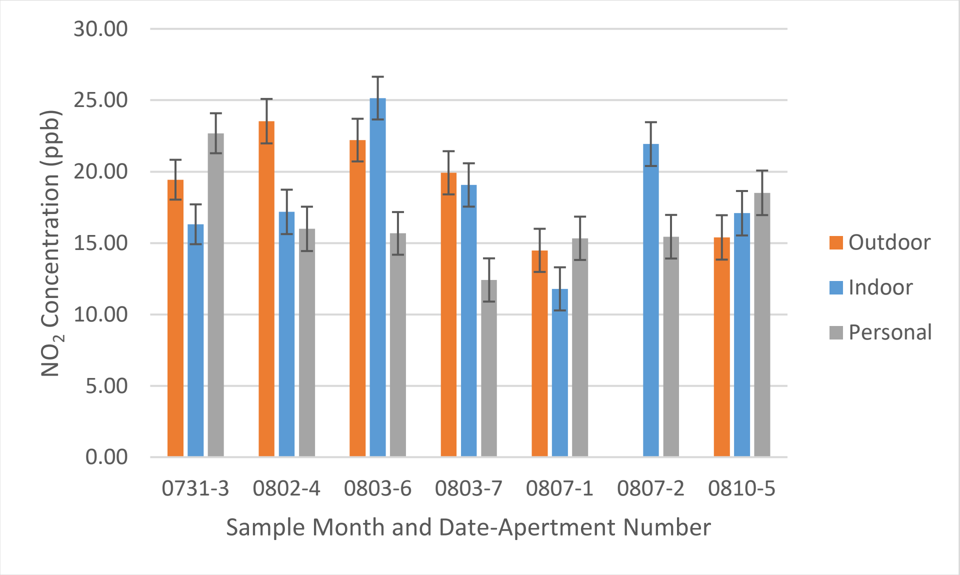
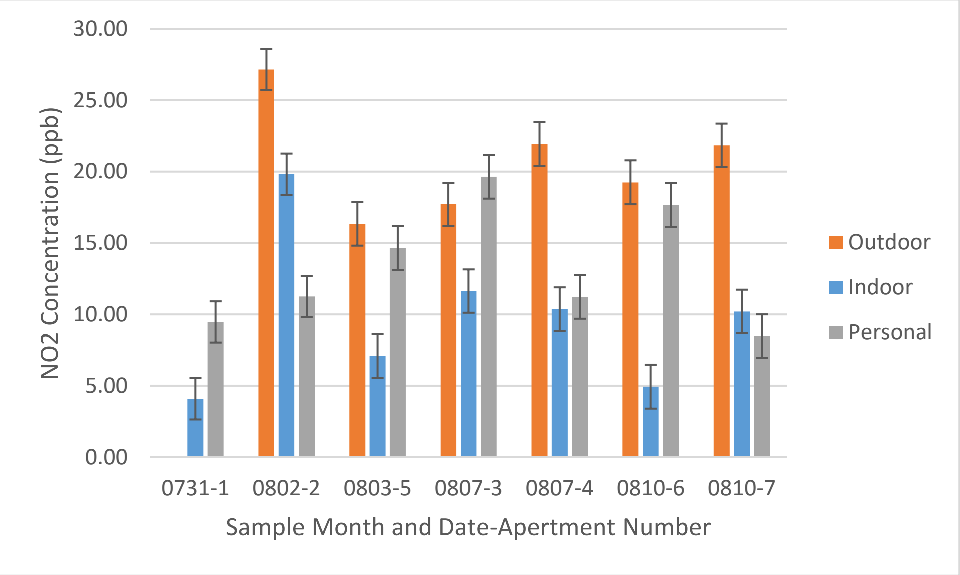


Figure A.4 Nitrogen Dioxide Concentration with True Filtration



Reference

Protocol for Ozone Measurement Using the Ozone Passive Sampler Badge. Harvard School of Public Health, Environmental Science and Engineering Program, Department of Environmental Health; Modified by Ogawa & Co., USA, Inc. January-2001.

NO, NO_2, NO_x and SO_2 Sampling Protocol Using the Ogawa Sampler. Developed by Yokohama City Research Institute of Environmental Science, Yokohama, Japan, Report No. 128, March 1997