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Measuring, modelling and mitigating particulate matter in indoor environments

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Abstract

With modern populations spending on average greater than 90% of their time indoors, a large proportion of total exposure to air pollution occurs within indoor environments. Exposure to air pollution is linked to numerous health effects and mortality, with indoor air quality estimated to be the ninth-largest risk to public health. One of the main components of indoor air quality is particulate matter (PM), the sum of all solids and liquids suspended within the air. PM includes dust, pollen and combustion products. PM is linked to mortality from respiratory and cardiovascular diseases, with the smallest sized particles believed to be the most significant to health.

This thesis sits at the nexus between measurement, modelling, and mitigation of indoor PM. It (a) outlines the utility of different PM metrics and sensors, and the challenges facing PM measurement; (b) characterises the efficacy of HEPA filtering technology in both the laboratory and the real world and (c) outlines the development and deployment of an effective integrated air quality monitor. Together this provides a holistic contribution to managing PM in indoor environments.

PM measurement uses three key metrics, Pmass, Pnum and Psize. Pmass is a robust, low-cost measurement, and is easy to compare and benchmark against other Pmass measurements. Pnum, the number of suspended particles in air, is suggested to be more important in a health context, as it is disproportionately weighted by fine and ultrafine particles, which are thought to have a greater impact on health. In the future, metrics that better link PM to health effects, such as surface area, particle length concentrations or compositional measurements, are likely to become more important, as better measurement technologies are developed. Particulate mass is measured using gravimetric methods, photometers and beta attenuation methods and is suited to compliance monitoring and trend analysis, as measurements are easily compared. Particle number is measured using condensation particle counters, optical particle counters and diffusion chargers, and is suited to source characterisation, trend analysis and ultrafine particle investigations. Particle size distribution measurements can be made by a scanning mobility particle sizer or fast mobility particle sizer, and are typically limited to research applications.

High-efficiency particulate air (HEPA) type purifiers are effective at removing “real-world” PM from air. Although ultrafine particles are removed effectively, particles between 200-300nm are removed least effectively, and this is noteworthy as particles of this size can penetrate buildings effectively, remain airborne for extended periods and are especially important in a health context. HEPA purifiers are effective in the real world, but a single air purifier is insufficient to purify an entire residence, so it should be placed where residents spend the most time. Air change will also reduce air purifier efficiency when ambient levels of PM exceed those indoors.

With low-cost sensors becoming increasingly accurate and available, measurements of air quality are likely to be increasingly available, with individuals being better able to understand their exposure. With this, it seems likely that future “smart homes” will contain “internet of things” indoor air quality sensors, which can give recommendations and make interventions (activating ventilation and purification), all in real-time. As measurements of the properties of PM become more diverse and well established, epidemiologists will be better equipped to understand the relationship between PM properties (mass, size, composition, surface area) and health. This will allow for better regulation of PM, to improve health as the correct properties of PM can be regulated.

Contents

List of papers

This thesis contains several papers that are published, submitted, and in preparation for submission to appropriate journals. These papers are represented in Chapters 2-5 of the thesis. They are listed below, with a brief description of the contribution made by the candidate and co-authors.

I. **Lowther, S.D**., Jones, K.C., Wang, X., Whyatt, J.D., Wild, O. and Booker, D., 2019. Particulate matter measurement indoors: A review of metrics, sensors, needs, and applications. *Environmental Science & Technology*, *53* (20), pp.11644-11656. **(Chapter 2)** *Lowther, S.D. proposed the review topic, collated the research and wrote the manuscript, with supervision from Jones, K.C., Wang, X., Whyatt, J.D., Wild, O. and Booker, D.*

II. **Lowther, S.D**., Deng, W., Fang, Z., Booker, D., Whyatt, D.J., Wild, O., Wang, X. and Jones, K.C., 2020. How efficiently can HEPA purifiers remove priority fine and ultrafine particles from indoor air? *Environment International*, *144*, p.106001. **(Chapter 3)** *Lowther, S.D. completed the experiments, with equipment provided by Wang, X and technical support from Deng, W and Fang, Z. Lowther, S.D designed the experiments and wrote the manuscript with supervision from Jones, K.C., Wang, X., Whyatt, J.D., Wild, O. and Booker, D.*

III. **Lowther, S.D.**, Deng, W., Fang, Z., Booker, D., Whyatt, D.J., Wild, O., Wang, X. and Jones, K.C, Factors affecting real-world applications of HEPA purifiers in improving indoor air quality. *To be submitted to Environmental Science: Advances* **(Chapter 4)** *Lowther, S.D. completed the experiments, with equipment provided by Wang, X and technical support from Deng, W and Fang, Z. Lowther, S.D designed the experiments and wrote the manuscript with supervision from Jones, K.C., Wang, X., Whyatt, J.D., Wild, O. and Booker, D.*

List of Appendices

I. **Lowther, S.D**., Jones, K.C., Wang, X., Whyatt, J.D., Wild, O. and Booker, D., 2019. Particulate matter measurement indoors: A review of metrics, sensors, needs, and applications. *Environmental Science & Technology*, *53*(20), pp.11644-11656.

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Chapter 1. Introduction

1.1 Indoor air quality

Indoor air quality (IAQ) is becoming increasingly important as modern populations spend ~90% of their time indoors. Exposure to indoor air pollutants (IAPs) can have adverse effects on health, and therefore, concentrations of IAPs should be minimized.

IAQ consists of solid, liquid and gaseous components. Important gaseous components of IAQ include volatile organic compounds (VOCs), which can be generated through combustion, furniture or household products, carbon dioxide (CO_2), which can be generated through human respiration, and nitrogen dioxide (NO_2), which is mainly generated by vehicular sources and later penetrates indoors. Particulate matter (PM) is the sum of all liquids and solids suspended in the air and this includes pollen, dust, smoke, combustion particles and cough droplets.

Modern populations spend ~50% of their time in residential environments where IAPs are generated through cooking or combustion, household products, furniture or penetration inwards from outdoors (Kornartit *et al.*, 2010). They also spend ~30% of their time at work where they may be exposed to a variety of IAPs depending on the type of work (Kornartit *et al.*, 2010). The other 20% of the time is spent in transit, outdoors or in other indoor environments (Kornartit *et al.*, 2010). With so much time spent in indoor environments, any improvements to IAQ are likely to impact significantly on an individual's total exposure to air pollutants.

With buildings in the UK becoming increasingly energy-efficient to meet sustainability targets (Geller *et al.*, 2006), indoor residential environments are becoming increasingly sealed to minimise heat loss and the subsequent energy expenditure. However, this increase in sealing, although useful in reducing the amount of ambient (outdoor) pollution transferring inwards, can lead to the accumulation of dangerous levels of IAPs from indoor sources (Adgate, Ramachandran and Pratt, 2002). With the COVID-19 pandemic ongoing, individuals are spending more time than ever in their homes and awareness of the importance of IAQ is growing.

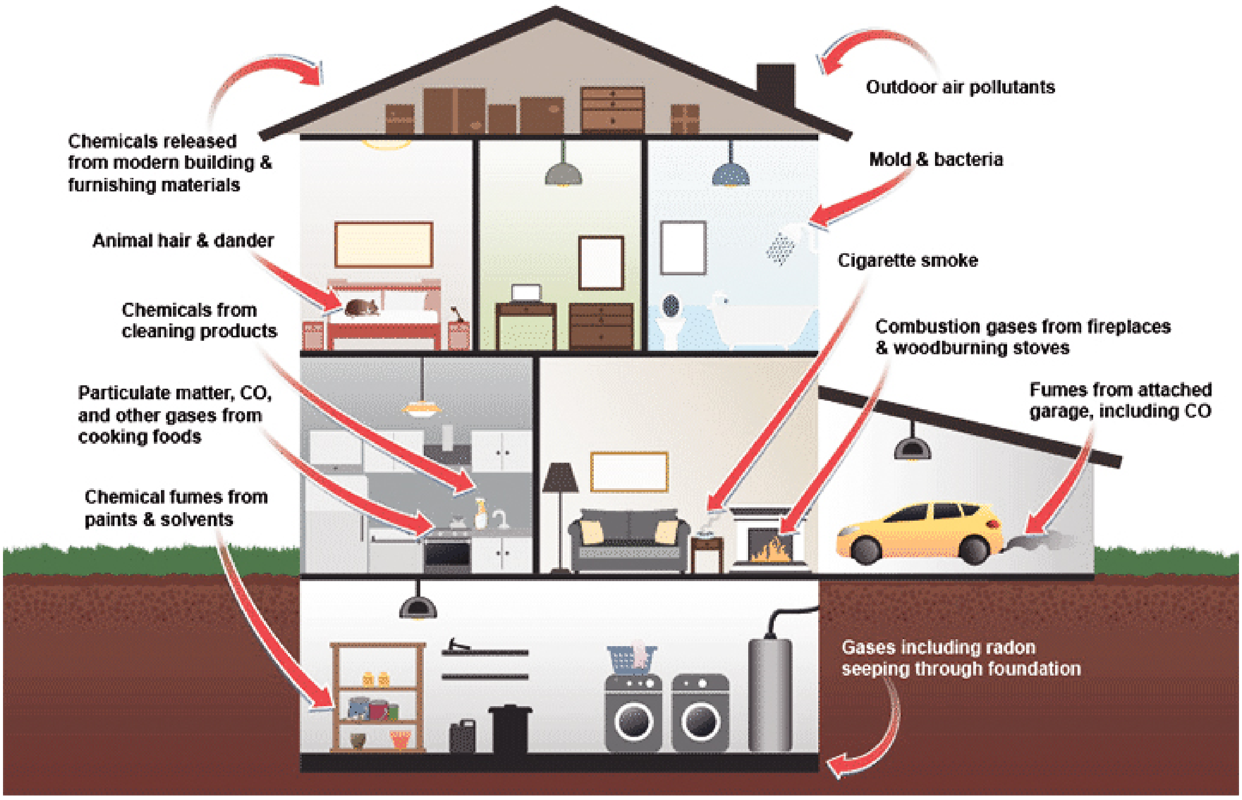

Figure 1. Sources of IAPs in residential environments (EPA, 2021).

1.2 VOCs, CO_2 and NO_2

VOCs are compounds with a high vapour pressure and low solubility. They commonly used in or produced by the manufacturing of paints, varnishes and furniture. VOCs can be released from a variety of liquids or solids, sources of VOCs in homes include, aerosol sprays, furnishings, paints, varnishes, cleaning products, air fresheners or incense (United States Environmental Protection Agency, 2021). Currently, VOCs of the highest importance include, acetaldehyde, a-pinene, benzene, formaldehyde, naphthalene, styrene, tetrachloroethylene and trichloroethylene (Shrubsole *et al.*, 2019). The EPA found that VOCs are 2-5 times as abundant in homes as outdoors incense (United States Environmental Protection Agency, 2021). VOC exposure can be reduced by having high ventilation when using paints varnishes or cleaning products, by purchasing increasingly common low-VOC products and using activated carbon filtration technologies. Because there are hundreds of VOCs, each with different sources, behaviours and levels to toxicity, it makes researching any individual VOC difficult, slowing our understanding.

CO_2 is a colourless gas present at all times in the air. Global average CO_2 levels are currently 412 ppm , and increasing concentrations receive large recognition because of their impact on climate change (NOAA, 2018). However, CO_2 is also largely important within an indoor air quality context. CO_2 in households can be generate by combustion or respiration, and can easily reach concentrations several times greater than background. With respiration as a major source of CO_2, occupancy can have a major effect on the CO_2 concentration in any given environment. As CO_2 is inert, and levels outdoors are always at a background level, it can also be used as a general indicator of IAQ and ventilation. Therefore, by controlling CO_2 indoors you will also largely be controlling other indoor-generated indoor air pollutants.

NO_2 is one component of the group of reactive gases known as nitrogen oxides (NO_x), it is predominantly generated through combustion sources, so is often associated with vehicular emissions and heavy traffic. Nitrogen dioxide can be removed from the atmosphere through the processes of wet and dry deposition. Nitrogen dioxide can be found at high concentrations indoors, especially when there are potent sources, for example, gas appliances (WHO, 2010a).

1.3 IAQ, PM and health

Poor IAQ has been estimated to be the ninth-largest global burden of disease risk (Forouzanfar *et al.*, 2015), with the WHO associating 3.7 and 4.3 million deaths to ambient and indoor air pollution respectively, in 2012 (World Health Organization, 2014). When combined, indoor and outdoor air pollution is responsible for the fourth-largest number of deaths per disease factor, behind high blood pressure, smoking and high blood sugar (Roser and Ritchie, 2018). Deaths associated with poor IAQ are highest in low-income countries across Sub-Saharan Africa and Asia (Roser and Ritchie, 2018).

More specifically, PM exposure increases the morbidity of and mortality from cardiovascular and respiratory diseases (World Health Organization, 2013). Ultrafine particles (particles <100nm in diameter) are thought to be particularly important as they can penetrate the lungs, enter the bloodstream and are found in the organs, for example, the brain (Maher *et al.*, 2016). Additionally, there are associations between PM exposure and heart attacks, aggravated asthma and decreased lung function (EPA, 2022).

However, there are still many unknowns when it comes to the effects of PM on health. PM is a aggregate of many different sizes, shapes and compositions, each with different surface properties. So although it is known that PM of different size bands (PM_{10}, $PM_{2.5}$ and PM_1) as a whole have adverse effects on health, it still remains largely unknown which sizes, shapes and compositions are most impactful on health. This would be extremely useful to determine as it would allow for mitigation of health outcomes by targeting the most damaging sources of pollution as a priority. Furthermore, PM act as a transport mechanism for other chemicals, which can become bound onto the particles surface, which adds another layer of complexity to what is actually responsible for causing negative health outcomes.

1.4 VOCs, CO_2 and health

There are many VOCs, each with independent effects on health. They generally cause irritation to the eyes and respiratory system and can have carcinogenic effects. Research efforts have been made to capture the individual health effects associated with individual VOCs, and accordingly VOCs can be ranked in importance based on a combination of their abundance and health importance. Concentration guidelines for a number of VOCs have been generated based on their perceived health importance (Shrubsole *et al.*, 2019).

Above 1000 ppm, CO_2 has been reported to have impacts on comfort, cognitive function and the respiratory system. A full review of the effects of CO_2 on health, written by myself, can be found here (Lowther *et al.*, 2021).

1.5 Particulate matter indoors

PM in indoor environments can be generated by solid fuel use, cooking, candles, smoking and cleaning products, and can be later resuspended through processes such as hoovering, cleaning or walking around. Depending on the outdoor to indoor air change rate of the environment, penetration of PM can contribute to indoor PM concentrations (Huang *et al.*, 2015).

PM can be naturally removed from the environment through deposition and ventilation. Generally, the largest particles are deposited the fastest because of their greater mass, with the smallest particles remaining airborne for longer durations of time. Increasing air change rate (a measure of ventilation) can reduce PM indoors, however, this is only when indoor PM concentrations exceed ambient PM concentrations. PM can also agglomerate, which means smaller particles fusing into larger particles or structures.

Whereas the effect of gaseous pollutants on health can largely be quantified by concentration, PM is not so simple. Many parameters, for example, mass, size, shape, composition, number and surface area can control the potency of PM. Additionally, many of these parameters are difficult to measure without large expensive equipment, meaning there is still a lot unknown about the effect of changing these parameters on measured health effects. Additionally, as each of the parameters will influence PM behaviour, each particle behaves differently, which can make modelling behaviours complex. Put simply, research has only scratched the surface of the complexity of PM, especially in its relationship with health.

Further context surrounding PM is explained in Chapter 2.

1.6 IAQ in China and the UK

Indoor air quality is highly variable in different contexts as it is a function of a multitude of factors, including - but not limited to - building design and use, city design, social practices, etc. To illustrate this point, it is informative to contrast the differences between the UK and China. Guangzhou, China (population ~ 15 million) where the experimental work for this thesis was conducted, will be contrasted against Lancaster, UK (population ~ 50000).

Firstly, ambient air quality is generally poorer in China than in the UK, due to the rapid urbanisation and industrialisation that has occurred in China over the past three decades. In 2017, 73% of China cities failed to meet the national $PM_{2.5}$ standard of 35 µg/m^3 (Ministry of Ecology and Environment the People's Republic of China, 2018). However, it should be noted that rapid reductions in PM concentrations are being made, for example, Beijing's $PM_{2.5}$ concentration has dropped from 90 µg/m^3 in 2013 to 58 µg/m^3 in 2017 (Ministry of Ecology and Environment the People's Republic of China, 2018). Urban areas in the UK have average $PM_{2.5}$ concentrations of 10-15 µg/m^3 (DEFRA, 2012).

High levels of ambient pollution within Chinese mega-cities have been linked to negative health outcomes by several studies. Shang *et al.*, 2013 showed that daily mortality in various Chinese mega-cities could be attributed to ambient pollution exposure, Cao *et al.*, 2012 linked particulate matter with cardiopulmonary mortality in highly polluted cities and Mokoena *et al.*, 2019 related Xian pollution to respiratory mortality (Ethan, Mokoena and Yu, 2021).

IAQ can often be poor in the mega-cities of China, with a combination of high levels of ambient pollution and potent sources of IAPs, including gas stoves, incense burning and fry cooking. Therefore, it is common for indoor air quality in China to exceed the national guidelines, and it is suggested that levels of formaldehyde and benzene may put occupants at risk of cancer (Huang *et al.*, 2018). Although China has implemented some effective control strategies on IAQ, including, improved stoves, ventilation improvements and indoor smoking avoidance, there are still persistent problems in the control of IAQ. Lacking mandatory standards for IAQ, limited product emission labelling and failure to maintain air cleaners are all areas that could use improvement (Hao, Zhu and Fan, 2018).

Figure 2. Light smog in Guangzhou, China, taken by Scott Lowther.

Secondly, due to larger population densities in Guangzhou (2100/km^2) than Lancaster (250/km^2), Chinese residences are smaller, with greater occupancy per m^2. As occupants generally generate IAPs, greater occupancy can lead to worse concentrations indoors. The sources of PM in homes of China and the UK are also substantially different. In China, major sources include solid fuel use, gas-stove cooking, smoking and incense burning. These are combustion sources and are therefore potent sources of PM (Tse *et al.*, 2011; Apte and Salvi, 2016). In the UK, where smoking is illegal indoors, where incense is not commonly used, and where gas stoves are being phased out, these sources are far less important. Southern China flats are more scarcely furnished than in the UK, with mostly wooden furniture and wood or laminate flooring, whereas, carpets and fabric furnishings are more common in the UK. This is because the climate in Guangzhou is warmer and more humid relying heavily upon air conditioning, whereas the UK relies more heavily on central heating.

Also, since Guangzhou flats have few potent IAP sources, it is common for ambient PM to exceed indoor PM (Chapter 4). Therefore, ventilation (a common method of improving IAQ in non-urban areas of the UK), can act as a source of PM into the indoor environment.

With both Chinese and UK populations spending ~80/90% of their time in indoor environments (Klepeis *et al.*, 2001; Duan *et al.*, 2015), any improvement in IAQ will lead to significant reductions in total PM exposure levels and possible improvements in respiratory or cardiovascular health.

1.7 Ventilation

Ventilation acts as an important determinant of IAQ. When ambient levels of pollution are high, and indoor levels are low, ventilation can act as a source of pollution into indoor environments. Reversely, if indoor levels are high and outdoor levels are low, ventilation is an important removal mechanism. Poor ventilation has historically been linked to a number of health outcomes, for example, sick building syndrome (Dimitroulopoulou, 2012). Occupants are often able to perceive poor ventilation and accordingly it can have effects on productivity (Dimitroulopoulou, 2012). Although, high rates of ventilation in European residences are ideal for IAQ, this needs to balance against thermal comfort and energy efficiency. Because of the importance of appropriate ventilation, a number of standards and regulations exist for European dwellings (Dimitroulopoulou, 2012). In Asian mega-cites, ventilation needs to be used in tandem with good filtration technologies, to ensure a high rate of air exchange, whilst mitigating against incoming pollution.

1.8 IAQ standards and regulation

As the importance of IAQ has only been recognised recently, limited standards are governing its control. For example, although the WHO recognises the health importance of exposure to PM in indoor environments, they do not have specific guidelines for PM concentrations in indoor environments (WHO, 2010b). For less complex pollutants, where concentrations are the main factor affecting health impacts, the WHO, independent bodies such as WELL or Leadership in Energy and Environmental Design (LEED) and national bodies such as Public Health England have developed IAQ guidance. Typically, these guidelines pertain to pollutants that are more important in the context of IAQ rather than ambient air quality, e.g. VOCs (benzene, formaldehyde and naphthalene etc.), radon and carbon monoxide. More practically, WELL and LEED have recommended ventilation strategies and guidance, since ventilation is an important tool in reducing exposure. Given that PM toxicity is extremely complex, it is difficult for these bodies to define how toxic PM in indoor environments is compared to ambient PM, and how to set appropriate guideline concentrations accordingly. Several research articles have summarised the existing IAQ standards for PM (Li, Wen and Zhang, 2017), VOCs (Shrubsole *et al.*, 2019) and CO2 (Lowther *et al.*, 2021).

1.9 Indoor PM filtration technologies

Several filtration technologies exist to reduce PM within indoor environments (Cheek *et al.*, 2021). These include electronic and mechanical type filters. Electrostatic purifiers pass particles through an ionization section, where they are charged and then pass through a series of plates with the opposite charge, where the particles are then deposited. Conversely, high-efficiency particle air (HEPA) type filters are mechanical and rely on particles becoming physically trapped within the filter medium. When removing particles is the priority, HEPA type air purifiers are often the most common and robust solution (Zhang *et al.*, 2011). For example, HEPA type purifiers occupied 53% of the air purifier market share for China in 2019 (Associated Press News, 2021). HEPA filters were commercialised in the 1950s, and remain a leading technology in particle filtration today.

1.10 Measurement of IAQ

A variety of measurement technologies exist to quantify the concentrations of air pollutants in air. PM can be sensed in a variety of ways, which are outlined in detail in Chapter 2. Gaseous components of IAQ have to measured using different means. Metal oxide sensors are a popular measurement technique, in these, target gases react with the surface of the metal oxide, which changes the electrical resistance of the metal oxide, which can be measured. Increases and decreases in the resistance can then be attributed to the concentration of the target gas (Barsan and Weimar, 2020). Electrochemical sensors are a close alternative, in these an electrochemical reaction on the surface of sensor, between the target gas and surface, generates an electrical current, that can then be measured (Manjavacas and Nieto, 2016). Nondispersive infrared (NDIR) sensors are an effective technology for measuring CO_2 concentrations, they shine infrared light down a cylinder with a light detector at the end. As the wavelength of light is chosen to be absorbed by CO_2 at a high rate, the amount of infrared measured by the light detector, is proportional to the concentration of CO_2 in the air. Gaseous pollutants are commonly measured in parts per million (ppm) or parts per billion (ppb), 1ppm equates to 1 part mass in 1 million parts of mass.

1.11 Research Aims

In view of the above, this thesis aims to,

1. Identify the key metrics, measurement techniques and issues facing PM measurement indoors.
2. Examine, in a laboratory setting, how effective HEPA type air purifiers are in removing PM from indoor air.
3. Identify, in a real-world setting, which factors are important to using an air purifier effectively in a Chinese residential environment.
4. Discuss the design, development, testing, and applications of a novel online multi-sensor indoor air quality monitoring technology.
5. Discuss future trends in the field of indoor air quality research.

1.12 Thesis Outline

This PhD program was in collaboration with the Guangzhou Institute of Geochemistry and Chinese Academy of Sciences, with the support of Professor Wang Xinming. The author spent two years in China

(August 2017 – September 2019) conducting the experimental portion of this thesis, with the assistance of Professor Wang's air quality laboratory and team.

The following literature review in **Chapter 2** provides an explanation of PM measurement in indoor environments, detailing metrics, measurement techniques and challenges. The findings showed that different measurement techniques and metrics all had useful applications, depending on the research being conducted. Some metrics and measurement techniques were better equipped for looking at smaller or larger particle fractions, with others being better for comparisons against historical or similar measurements. Understanding the difficulties associated with PM measurement indoors, and the measurement equipment used, provided a strong foundation for the following experimental work.

Chapter 3 details the first experimental work conducted as part of the thesis. It identified that air purifiers (HEPA type in particular) were a key method to reduce an individual's exposure to PM. Although their performance for removing standard particle types (pollen, dust and smoke) is routinely tested, it appeared unclear how effectively they might be in removing heterogeneous "real-world" particles – those in the ambient air of a Chinese mega-city. Therefore, this experiment aimed to determine how effective air purifiers are at removing these "real-world" particles, relative to the standard particle types. This was conducted in an environmentally controlled chamber, the largest in Asia, located at the Guangzhou Institute of Geochemistry.

After Chapter 3, where it was proved that HEPA type air purifiers were effective in removing "real-world" particles from indoor air, **Chapter 4** aimed to better understand which factors affected the performance of air purifiers in Chinese residences, something that had not been previously researched. After having understood that air purifiers perform well under controlled conditions (Chapter 3), it was important to determine if any mistakes in use could inhibit their performance in the real world. In short, this paper aimed to determine what advice could be given to the owner of an air purifier, to help them reduce their exposure to PM, to the greatest degree possible.

Chapter 2. Particulate matter measurement indoors: a review of metrics, sensors and experimental applications

Abstract

Many modern populations spend ~90% of their time indoors, with household particulate matter being linked to millions of premature deaths worldwide. Particulate matter is currently measured using particle mass, particle number and particle size distribution metrics, with other metrics, such as particle surface area, likely to be of increasing importance in the future. Particulate mass is measured using gravimetric samplers and photometers and is best suited to use in compliance monitoring, trend analysis and high spatial resolution measurements. Particle numbers are measured by Condensation Particle Counters, Optical Particle Counters and Diffusion Chargers. Particle number measurements are best suited to source characterisation, trend analysis and ultrafine particle investigations. Particle size distributions are measured by gravimetric impactors, Scanning Mobility Particle Sizers, Aerodynamic Particle Sizers and Fast Mobility Particle Sizers. Particle size distribution measurements are most useful in source characterisation and particulate matter property investigations. Most measurement options remain expensive and intrusive. However, we are on the cusp of a revolution in indoor air quality monitoring and management. Low-cost sensors will facilitate personalized information about IAQ, allowing citizens to reduce exposures to PM indoors and to resolve issues such as IAQ vs energy efficiency. Indeed, the low cost will put this simple technology in the hands of citizens who wish to monitor their own IAQ in the home or workplace, to inform lifestyle decisions. Low-cost sensor networks also look promising as the solution to measuring spatial distributions of PM indoors, however, with regard to current low-cost sensors, there are important sensor/data quality, technological and ethical barriers to address. An improved understanding of epidemiology is essential to understanding which metrics correlate most against health effects, allowing indoor specific PM standards to be developed and to inform the future of experimental applications.

Introduction

Poor indoor air quality (IAQ) has been estimated to be the 9th largest global burden of disease risk,[1] with the World Health Organisation (WHO) attributing >4.3 million premature deaths to household air pollution in 2012, compared to 3.7 million deaths attributed to ambient (outdoor) air pollution.[2] The Institute for Health Metrics and Evaluation attributed 2.57 million premature deaths to household air pollution in 2016,[3] with Asia, Africa, Europe and the Americas contributing 74%, 23%, 1% and 2% respectively.[4] Particulate matter (PM), the sum of all solid and liquid particles suspended in the air, is a major metric of IAQ. PM is strongly associated with myocardial infarction, strokes, heart failure, asthma, chronic obstructive pulmonary disease and lung cancer.[5] PM <200nm in diameter has also been observed in the brain, and this may be causally linked to neurodegenerative diseases, for example, Alzheimer's disease.[6] It is well established that modern populations on average spend >90% of their time indoors, and on a daily basis indoor air contributes 19-76% of an individual's exposure to particles <100nm in diameter.[7,8] Brunekreef[9] summarises the short and long term effects of PM on health.

Historically, the focus of measurement has been on outdoor PM. However, indoor PM is being increasingly identified as an area that requires more research. Understanding the sources, sinks and behaviour of PM within indoor environments is important to accurately predict personal exposures and population health burdens, as well as to design practical and effective mitigation strategies.

Atmospheric PM is generally measured using two main metrics; particle mass concentration (Pmass) and particle number concentration (Pnum). Pmass is the mass of particles within a given volume (usually $\mu g/cm^3$) and Pnum is the number of particles within a given volume (particles/cm^3). Another important characteristic of PM is the particle size distribution (Psd); this is the particle concentration (either mass or number) measured over a range of different particle sizes. Finally, the chemical composition of particles can also be measured, although chemical composition is not covered in detail by this paper. It is still largely unknown how each of these metrics relate to health effects and therefore which metric is the best suited to measuring health risk.

The challenges associated with measuring PM in the indoor environment are distinctly different to those of measuring PM in the outdoor environment. Indoor environments can have much more variable PM levels than outdoors,[10] with PM levels much lower than outdoors when there are no dominant indoor sources; however, in the presence of indoor sources, PM levels can rapidly rise to several orders of magnitude greater than outdoor levels. The confined nature of indoor environments allows PM to accumulate, but can also lower PM below ambient concentrations through removal of particles during building shell penetration. With pressure placed on developing energy efficient buildings with low heat losses and therefore low air exchange rates, accumulation of indoor generated PM pollution is a significant problem.[11] This means that PM monitoring equipment used indoors needs to be able to accurately measure PM over a wide range of concentrations. Indoor particle events are frequently (but not always) time and space specific; brief, intermittent and highly variable.[8] This means that high spatial and temporal resolution measurements are necessary to further understand the controls and influences on PM. High temporal resolution measurements are common practice, with any “real time” measurement instrument providing sufficient resolution in most cases; however, spatially varying measurements are less common with campaigns rarely deploying multiple sensors within a single indoor environment.

There are many other more practical issues associated with measurement of indoor PM. There are constraints of size, noisiness and intrusiveness placed on equipment. For example, in an occupied school classroom it would be inappropriate to deploy large noisy equipment. This is much less of an issue with outdoor measurements. Some of the most severe indoor air pollution is experienced in urban and rural, middle and low-income homes in developing countries, often with high levels of ambient pollution and use of coal or biomass stoves. Many of these locations lack access to electricity, and hence, measurements must be made with either battery-operated devices or passive samplers. The increasing focus on indoor PM has driven researchers to develop smaller, lighter, inexpensive battery-operated sensors. However, developing sensors that are sufficiently sensitive when measuring low concentrations and small size ranges of PM is challenging.

A wide range of technologies and sensors hasbeen developed to measure PM. These vary in precision, accuracy, sensitivity (detection limits), time resolution and cost. However, the appropriate ranges for these criteria can vary substantially when considering the size, cost and experimental applications. For example, when measuring in unoccupied spaces (as opposed to occupied spaces), what is reasonable in terms of size, weight and noisiness of equipment is very different. It is important to understand how these sensors vary in nature in order to be able to correctly select the most effective for any given measurement scenario.

This paper critically reviews PM metrics, the sensors and measurement techniques associated with these metrics, and the sensors and techniques that are most appropriate for each experimental application. The future of IAQ and PM measurement, the challenges, solutions and anticipated shifts in focus are also discussed. This will aid future studies in selecting the most appropriate metrics, measurement techniques, and sensor technology, as well as highlighting the challenges associated with measuring PM indoors.

The PM sensors selected for review are based on the following criteria;

a. The sensor must operate within sensible ranges for the criteria listed above, whilst considering their applications.
b. There must be evidence that the sensor has been used to measure PM within indoor environments, with substantial benefits compared to other sensor types.

PM is currently measured using gravimetric, optical, oscillating microbalance, beta-attenuation and electrical current techniques. Some of the sensors can measure one or more of the metrics listed. Indoor measurements are usually collected in the centre of the environment, at 0.75-1.8m height to simulate the breathing zone, with a height of 1.5m recommended.[12]

Current developments in PM monitoring are being influenced by: a) PM sensors that have traditionally been used in occupational health; and b) developments in miniaturisation & wireless technology. Low-cost, portable sensors, capable of measuring high temporal resolution concentrations have been at the forefront of meeting the rapid increase in public awareness and interest in quantifying personal exposure to indoor air pollutants It is important to consider how these sensors may play a role in measuring IAQ specifically.[13] Many of the sensors used in indoor environments were initially developed to measure vehicular emissions or industrial environments which tend to have high concentrations of pollutants compared to indoor environments.[14] Therefore, it is important to establish which role each sensor can most effectively play in IAQ measurement. Table 1 outlines the sensors reviewed in this paper and the abbreviations used where appropriate.

Measurement Method	Sensor	Abbreviation
Gravimetric	Gravimetric Filters	N/A
	Impactors	N/A
Optical	Photometers	N/A
	Optical Particle Counter	OPC
	Condensation Particle Counter	CPC
	Scanning Mobility Particle Sizer	SMPS
	Fast Mobility Particle Sizer	FMPS
	Aerodynamic Particle Sizer	APS
Electrical	Diffusion Size Classifier	DiSC
	Nanoparticle Surface Area Monitor	NSAM

Table 1. A list of reviewed sensors and their abbreviations categorised by their measurement method.

Oscillating microbalance, beta attenuation, and low-pressure impaction measurement methods are not included within this review. Detailed descriptions of these methods are outlined by this international standard.[15]

Review of metrics

In the atmosphere there are three broad particle modes; the fine/nuclei mode (<0.1µm), the accumulation mode (0.1-2.5µm) and the coarse mode (>2.5µm); in short, these modes are the consequence of physical processes such as; emission, nucleation, accumulation and scavenging. Pmass is usually measured as $PM_{2.5}$ or PM_{10} and this is the mass of all particles with an aerodynamic diameter of ≤2.5µm and ≤10µm, respectively. PM_1 is also used but is not yet widely implemented and there are discussions currently on whether $PM_{0.5}$ or $PM_{0.1}$ should be introduced in the future.[16] PM_{10} and PM_1 are arbitrarily selected as size cut-offs out of convenience, whereas $PM_{2.5}$ is selected more purposefully to include accumulation and fine particles (which remain suspended for longer), but to exclude the coarse particles (which are deposited more rapidly). These cut-offs are also indicative of historic progress; for example, the UK Department of Environment, Food and Rural Affairs (DEFRA) introduced PM_{10} and $PM_{2.5}$ in 1997 and 2007, respectively, as sensing technologies and understanding improved. Other measurement ranges include total suspended particulate (TSP), a measurement of the mass of all the particles present in the air, and inhalable, thoracic and respirable size fractions, used in health-related sampling. The inhalable, thoracic and respirable fractions represent the particles that can enter the respiratory system through the mouth and nose (50% penetration efficiency (D_{50}) at 100µm), pass through the larynx and enter the bronchial region of the lungs (D_{50}=10µm) and enter the deepest part of the lungs, the ciliated alveoli (D_{50}=4µm), respectively.[17] It should be noted that these definitions were developed in a workplace exposure context. The EPA[18] provide a useful visualisation of the sizes of $PM_{2.5}$ and PM_{10} and Nazaroff[19] outlines the sources, compositions and behaviours of different particle size fractions in indoor environments.

Particle mass

Pmass is the most commonly used metric to measure PM, because it is the easiest metric to measure and can be measured accurately at a relatively low cost. Due to the characterisation as PM_{10}, $PM_{2.5}$ and PM_1, it is convenient to draw comparisons between different indoor environments and measurements collected by different equipment. Easy comparisons allow us to develop sensible Pmass concentration standards. Currently there are no universal standards for Pmass specifically for indoor environments. The WHO recommends outdoor Pmass guidelines for $PM_{2.5}$ of 10µg/m^3 (annual mean) and 25µg/m^3 (24 hour mean) and for PM_{10} of 20µg/m^3 (annual mean) and 50µg/m^3 (24 hour mean).[20] However, these standards are not indicative of where adverse health effects begin to occur (for $PM_{2.5}$, this is just above background concentration at 3-5µg/m^3).[20] Rather, they are the lowest levels at which "total, cardiopulmonary and lung cancer mortality have been shown to increase with more than 95% confidence in response to long-term exposure to $PM_{2.5}$".[20] The WHO investigated whether it was necessary to introduce specific indoor Pmass standards.[21] They concluded that because there was no significant difference between the hazardous nature of PM in indoor and outdoor environments, and because indoor PM levels are often greater than those outdoors (in the presence of indoor sources) it was not necessary to introduce any specific, more stringent indoor Pmass standards. WHO outdoor Pmass guidelines are therefore assumed to apply to indoor environments.[21] This assumption makes sense for now, given our currently limited understanding of the nature of IAQ; however, in the future it is likely that specific indoor standards will be established, when the effect of composition and concentration on health are better understood. For example, if it is discovered that acute exposure to

PM has more detrimental health outcomes than chronic exposure to PM, the standard might change from something based on the average to something that reflects the number of times a certain risk threshold is exceeded and the duration of that exceedance.

The smallest particle modes tend to make a large contribution to Pnum and a very low contribution to Pmass, this is illustrated well by NARSTO[22] which visualises typical number and volume concentrations (indicative of PMass) of atmospheric particles.

Particle number

As measurement technologies have advanced and as focus moves towards the importance of UFPs, Pnum is becoming an increasingly used metric for measuring PM within indoor environments. Although the Pnum metric is being used increasingly in outdoor and indoor PM studies, it was originally used to characterise vehicular exhaust emissions.[23] Unlike Pmass, Pnum varies by several orders of magnitude - from $<10^3$ particles/cm^3 in relatively clean environments to $>10^6$ particles/cm^3 when there are dominant sources of indoor pollution.[24] In contrast to Pmass, Pnum is mostly made up of smaller particles. For example, Pnum concentrations have been measured to be 2 orders of magnitude greater within the <0.5µm size range than in the 0.5-18µm size range.[25] This means that when fine or UFPs are the focus of a study the Pnum metric is often employed. Currently there are no standards for Pnum in indoor or outdoor environments,[26] likely due to the relatively recent adoption of ambient Pnum measurements, the highly variable nature of the Pnum metric, and the difficulty of comparing studies measuring Pnum. Epidemiologists have suggested that Pnum is a more important predictor of health than Pmass.[27,28] This is because Pnum better represents the smaller particle size fractions, which penetrate further into the respiratory system, potentially causing more damage.[29]

Figure 3 illustrates how the variable nature of Pmass and Pnum measurements differ, with the measurement range of Pnum being much greater and therefore requiring equipment with sensitivity over a very wide range of concentration - spanning several orders of magnitude.

Particle size distributions

Understanding the Psd in indoor environments is important for several reasons. The size of particles determines how far they can penetrate the respiratory tract. Smaller particles often have a higher toxicity per unit mass, due to a larger surface area to mass ratio.[30] The Psd metric is commonly used in studies which attempt to identify the major PM sources within indoor environments, and to understand the important removal processes and residence times of PM.

The future of ultrafine particle metrics

There is currently much debate on what should be the flagship metric for the measurement of UFPs. The metrics mainly considered are Pnum, surface area, “active” surface area, and particle reactivity (which relates to chemical composition). There is general agreement that the metric should provide insight into how PM interacts with the body through intake, uptake and transport. Therefore, there is some consensus that a surface area-related metric should be introduced, since these better correlate with the biological and toxicological activity of particles than either Pmass or Pnum.[31] However, there are concerns over whether it is plausible to achieve accurate measurements with relatively simple equipment. It seems likely that the UFP maximum size cut-off will be moved from 100nm to between 200-500nm and that diffusion chargers may be the most effective way of measuring the proposed “active” or “lung deposited surface area (LDSA)”.[31] Measuring compliance or exposures using the Pmass and Pnum metrics can be of limited use, given that physiochemical properties of particles give rise to varying levels of toxicity and this toxicity is also an important determinant of health effects. Instead, identifying and quantifying the sources generating the most reactive species of UFPs is suggested.[31]

The future of epidemiological metrics

There is strong agreement that future metrics should provide more information on the health impacts of particles than just physical properties. Particle length concentration and active or geometric surface concentrations might be more indicative of a particle’s effect on health than Pmass or Pnum.[32] Whereas Pnum takes no account of particle size, particle length concentration is Pnum multiplied by the diameter of particles within a given size range. The geometric surface area concentration equals the particle number concentration multiplied by the particle’s diameter squared, within a certain size range. Although the geometric and active surface areas both relate to particle diameter, the geometric surface area makes the assumption that particles are spherical whilst active surface concentration does not.[33] These may eventually become more important metrics for PM in epidemiological studies.

Characterisation of measurement approaches and sensors

Here, principles of measurement techniques and sensors associated with each PM metric are briefly outlined, and provides references to more detailed descriptions.

Particle mass

Pmass is measured using gravimetric and optical methods.

Gravimetric methods are simply based on weighing a filter sample before and after a sampling period and calculating the Pmass from the difference in weight. Filters have a collection substrate on which particles of all sizes are deposited, unless there is a cyclone or impactor used to remove larger particles. Conventionally this is active sampling, with a pump pulling air through the filter at a known flow rate, however, passive samplers that do not require a pump have also been developed.[34] Passive samplers are much lighter, smaller and less noisy than pump-operated active samplers, but must be deployed for longer periods of time to be effective and – depending on the design - can be influenced substantially by wind speed and particle size. The University of North Carolina (UNC) passive sampler, has been shown to correlate well against active samplers within indoor environments.[35] After a sample has been collected, it can be examined using an optical or electron microscope to determine the number, size, shape and structure of the particles collected. This data can then be used to calculate the Pnum and Psd that the sampler was exposed to.[34,36–38] This method uses a scanning electron or optical microscope and automated image analysis. Unfortunately, gravimetric sample analysis is labour intensive and cannot be used to measure UFPs (<0.1µm) which are too small to be observed. There are also inherent errors in particle size and surface area measurements due to estimating 3D properties of particles from 2D images.

Passive gravimetric samplers are extremely small (1.5-5cm diameter) and light (1-5 grams) and require no maintenance. These samplers can be deployed for both long and short periods, however, this is subject to the ambient concentrations as the sensors can have too few or too many particles deposited for the automated image analysis to be effective. If the samplers are operated in especially low PM environments, or for very short periods, this requires more scanning electron microscope (SEM) images to be analysed to determine an accurate concentration. With gravimetric samplers the chemical composition of particles can be acquired using processes such as instrumental neutron activation analysis (INAA) and particle-induced X-ray emissions (PIXE) as well as many other processes, although this can be a time-consuming. In simple terms, the sample is bombarded with ions or neutrons and the generated radiation is indicative of the original nuclides.

Impactors can separate particles based on their inertia; and this allows for measurement of a mass-based size distribution. The most commonly used type of Impactor is the Cascade Impactor, in which particles flow through a series of sections (typically 3 to 15),[15] each containing an impaction plate. In each section, particles above a certain size cut-off will be deposited onto the impaction plate. Between each stage the diameter of orifices decreases, which leads to increased velocity of the aerosol and impaction of progressively smaller particle sizes. The Cascade Impactor is made up of several stages followed by a final filter, which collects any particles that were not deposited in any of the previous stages. Conventional Cascade Impactors cannot size particles <400nm, however some Low-Pressure Cascade Impactors can size particles from 30nm upwards.[15,39] The collection substrates can be removed from the Cascade Impactor and be processed in the same ways as standard gravimetric filters. Figure 1 illustrates a cascade impactor.

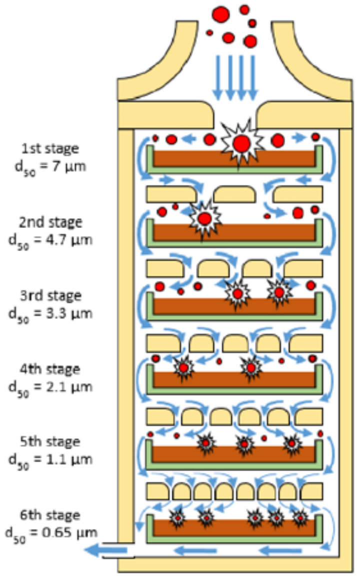

Figure 1. A illustrative diagram of a cascade impactor (Jensen and Shafer, 1998).

Optical methods are based on the interactions of light with particles. When light hits a particle, it is either scattered or absorbed. Optical methods are based on the principle of measuring scattering, absorption and extinction (the sum of scattering and absorption) to determine the particle concentrations of an aerosol.[39] Scattering Laser Photometers measure the intensity of scattered light in one or more directions using a photometer detector; the combined intensity of scattered light is directly proportional to the volume concentration of the aerosol within the optical volume.[40] Photometers collect real time measurements with a frequency of 1s, and measure particles from ~40-100nm upwards, however, measurement efficiencies are significantly lower at smaller particle sizes. They are commonly used with impactors or cyclones to measure PM_1, $PM_{2.5}$, PM_{10} or the respirable aerosol fraction and are extremely popular due to their small, portable, robust and reliable nature. Photometers can measure accurately over a larger range of Pmass concentrations (0.001-200 mg/m^3), making them suitable for both clean and highly polluted environments. Examples of commercial photometers include the DustTrak, DataRAM 4 and UCB-PATS. Figure 2 shows a diagram of the internals of a photometer.

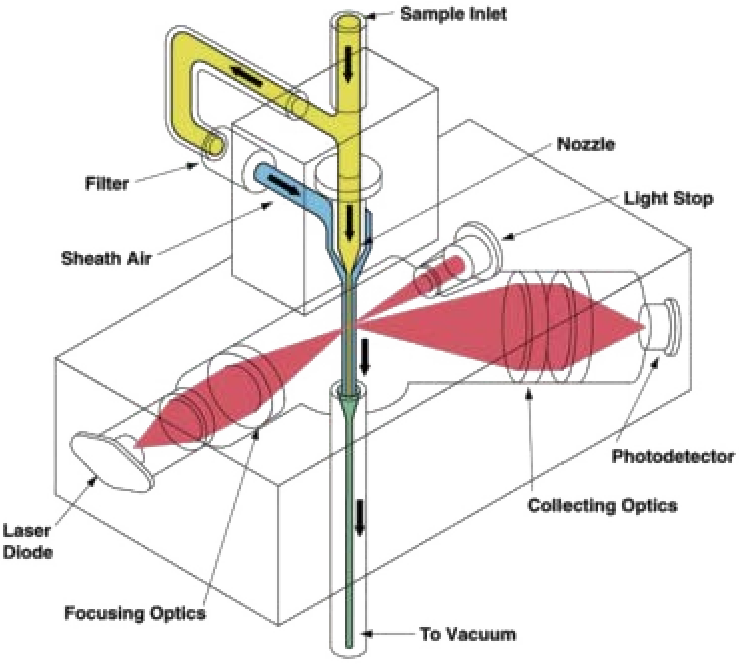

Figure 2. A illustrative diagram of a photometer (TSI, 2022).

Gravimetric samplers are more accurate than optical methods since they measure PM directly rather than indirectly. Therefore, at the start of campaigns, optical Pmass sensors are often co-located with gravimetric samplers to be calibrated.[41] Optical measurements vary depending on the optical properties of particles, and therefore are not synonymous with exact mass concentrations.[42] However, there is a significant trade-off with gravimetric filters being much more labor intensive than real time measurements. When using gravimetric filters to measure Pmass, the samples need to be dried and weighed and then chemical testing requires additional labour. In addition, the process of drying the filters can remove health-related volatile compounds, affecting the mass measurement. Collecting data on Pnum, Psd, particle shape, particle surface area and particle structure requires use of a scanning electron microscope, however, automation has made this process less labour intensive.

Particle number

Pnum is measured using optical and current methods. Optical Particle Counters (OPCs) work similarly to a scattering laser photometer with a diode laser shining on the optical volume with the scattered flash being measured by a photodetector; unlike photometers, only one particle is illuminated at once. The photodetector converts the flash of light into an electrical current, and as each electrical current corresponds to a different particle, the number of particles can be counted. The size of the particle is proportional to the intensity of the flash and electrical current generated, so using a calibration curve particles can be sized based on the amplitude of the current generated.[39] For example, the Alphasense OPC can count particles with a diameter of 0.35-<40µm and place them into 24 size categories, with a sampling frequency of 1s upwards. Other OPCs include the TSI Optical Particle Sizer 3330 and GRIMM OPC. OPCs are light, portable, rugged and quiet, however their main disadvantage is their inability to count particles <300nm. Particles smaller than this cannot be counted by the optics, hence are grown to a size where they can be counted in the CPC (described below). Particles <300nm in size make a substantial contribution to total Pnum concentrations.

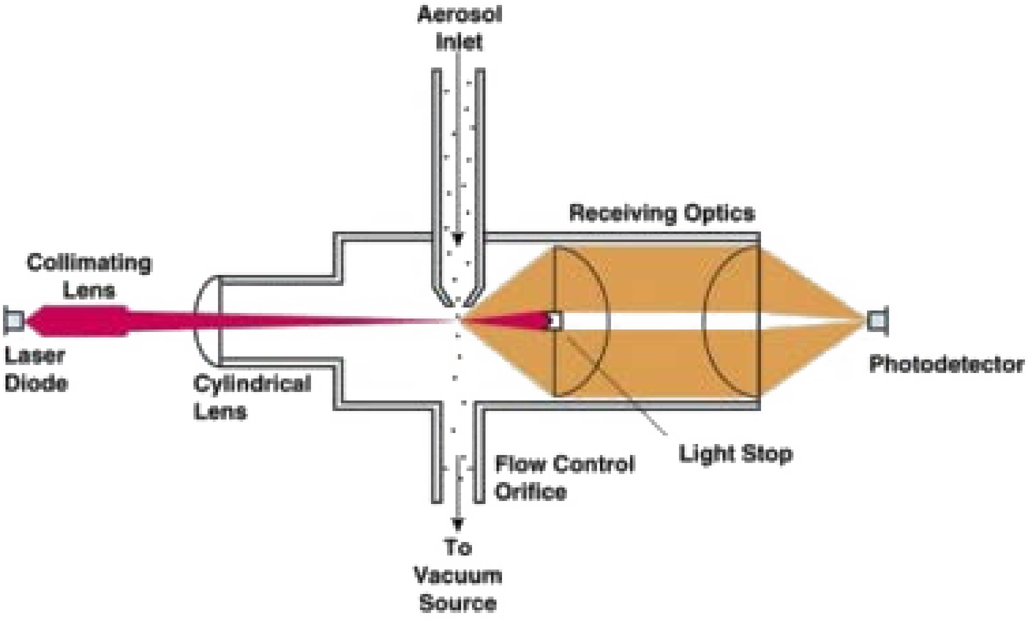

Figure 3. A illustrative diagram of a OPC (TSI, 2022).

Condensation Particle Counters (CPCs) share similar operating principles to OPCs but can count particles of much smaller sizes. CPCs condense a solvent (typically butanol, isopropyl alcohol, or more recently water) onto the surface of particles to grow them to a size where they can be counted. Once grown, the particles pass through the focal point of the laser beam and are individually counted. Vapor around the particles needs to reach a certain degree of super-saturation before condensation occurs, the magnitude of this super-saturation determines the minimum countable size of the CPC. There are two types of CPCs; Full Flow CPCs (sometimes called Continuous Flow Laminar CPCs) and Mixing CPCs (sometimes called Fast CPCs). In Full Flow CPCs the aerosol is drawn through a conditioner where it is saturated with vapor and brought to thermal equilibrium. The aerosol then passes into a cooler growth tube where the liquid is condensed onto the surface of particles. Full Flow CPCs require higher regulation of temperature control than other CPCs. In aerosol research the Full Flow CPC is the most commonly used due to its robust and reliable nature,[43] however, they have a relatively low sampling frequency (several seconds) due to zones of recirculation and time needed to establish super-saturation.

Many of the atmospheric processes measured by CPCs are rapid and therefore there is a need to develop a CPC capable of higher frequency sampling. Mixing CPCs were developed in the 1980s to increase the temporal resolution of measurements.[44] In a Mixing CPC, a cold aerosol flow is mixed with a warm saturated gas flow; this dilutes the aerosol flow allowing the CPC to cope with the dynamic range of indoor Pnum. When this was designed it was able to achieve mixing times as fast as 0.6s, 10 times faster than any commercially available full flow CPC.[43] Therefore, Mixing CPCs are sometimes referred to as Fast CPCs. One of the main uses of a Mixing CPC is as part of a Scanning Mobility Particle Sizer (SMPS) (described below), which measures particle size distributions. SMPS's commonly have a scan time of 2-4 minutes; mainly limited by the slow temporal resolution of the CPC. Under certain conditions a "Fast CPC" could capture data at 3s.[45] However, realistically scans are unlikely to be reduced below 30s. Currently, Full Flow CPCs and Mixing CPCs have temporal resolutions of 0.25-3s and 16-100ms respectively.[43] The detection limit, or "cut-off", of a CPC is described by its 50% detection efficiency diameter (d_{50}); the size at which <50% of particles passing through the CPC are counted. Changing the temperatures within the condenser and saturator can change the D_{p50} of a CPC. Handheld CPCs can make counting errors when multiple particles are located together in the optical detection region, and this is a common occurrence for concentrations >250000 particles/cm^3. CPCs need to be maintained level (specifically full-flow CPCs), to prevent the working fluid entering the optical circuitry and this makes them difficult to use for personal exposure monitoring.[46]

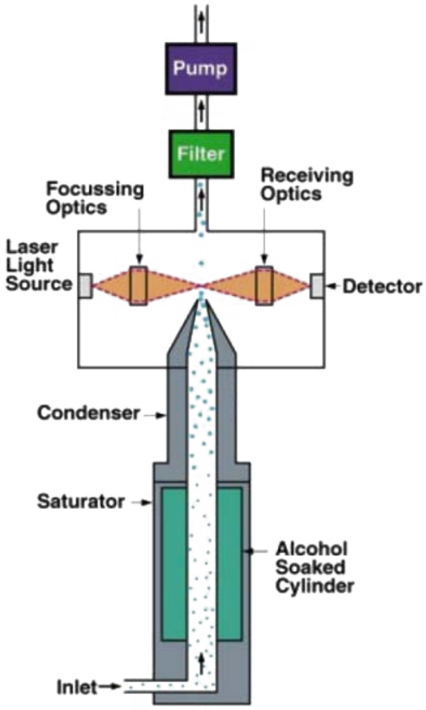

Figure 4. A illustrative diagram of a full flow CPC (TSI, 2022).

The Diffusion Size Classifier (DiSC) can estimate Pnum, the average particle diameter in the size range of 10-700nm and LDSA. In the DiSC the aerosol is charged in a unipolar diffusion charger and then passes through two electrometer stages. The DiSC is one type of Diffusion Charger, primed to be important in the future of UFP measurement due to its ability to measure surface-area related metrics. The first stage or "diffusion stage" consists of a stack of stainless-steel screens connected to sensitive electrometers; the second stage is a HEPA filter connected to an electrometer. Deposition of particles in each of these areas generates a current; $I_{diffusion}$ and I_{filter}. The relationship between these generated currents can be used to calculate the Pnum and average particle diameter.[47,48] The DiSC is small, portable and battery operated, and this makes it highly suitable for field measurements. The DiSC performs very well for its size and cost but is significantly less accurate than the larger and more expensive CPC and SMPS. Although the accuracy is generally good, the DiSC can perform poorly under certain conditions; for example, measuring monodisperse aerosols of specific compositions.[46] When compared to the SMPS and CPC the mean particle size and Pnum were within ±30% and ±50% of reference values, respectively.[49] They identified that the presence of particles >400nm drastically bias the mean particle size measurement. An example of this piece of equipment is the TESTO DiSCmini. Traditionally, DiSCs are cheaper than CPCs; however new periodic technical inspection regulations for vehicle emissions are being introduced in Germany, with 35,000 garages mandated to have Pnum sensors by the 1st January 2021.[50] This large demand will drive the market to produce low-cost (<5000$), reliable and easy to use Pnum measurement devices. The likely result will be that DiSCs and CPCs will be forced to become competitive on price, which will likely increase their widespread use in other sectors, for example, IAQ measurement.

Particle size distribution

Psd can be obtained from SMPS, Fast Mobility Particle Sizer (FMPS), OPCs, APSs and passive air samplers. In Differential Mobility Analysers (DMA) particles are given a unipolar corona charge and are passed through an electric mobility analyser; from here the particles of a given diameter are selected based on their electrical mobility.[40] The size selected by the DMA is determined by the magnitude of the voltage being applied. Exponentially increasing the voltage allows scanning through a particle diameter size range in several minutes. This is the concept behind the operation of a SMPS. An

Electrostatic Classifier is a particle neutralizer combined with a DMA; it can generate and subsequently size select particles. An SMPS is essentially an Electrostatic Classifier connected in-line with a CPC; in this system the Electrostatic Classifier selects particles of a given size, and the CPC counts these particles. The SMPS is the most precise instrument for measuring particle size distributions. The FMPS differs from the SMPS by using an Electrostatic Classifier and multiple low noise electrometers to measure Pnum and Psd in the 5-600nm range. The benefit of this over the SMPS is a 1s sampling frequency; making it more suitable for measuring rapid aerosol processes. However, the SMPS will measure the very smallest particles with a higher accuracy than the FMPS. Examples of this equipment include TSI's SMPS 3938 and FMPS 3091. The SMPS with a range of 1-1000nm is often paired with an Aerodynamic Particle Sizer (APS) with a range of 0.5-20µm when Pnum and Psd of particles >1000nm are also of interest.

The APS uses the inertia of particles to determine their size; firstly, the aerosol flow is constricted through a nozzle and this accelerates the aerosol. The velocity of the particle can then be related to the particle's surface area and mass and therefore aerodynamic diameter. The aerodynamic diameter is determined assuming spherical particles and uniform density. Secondly, particles then pass through 2 laser beams separated by 200µm; as the particle passes through each beam, light is scattered onto a photodetector. The time difference detected between the time of the two pulses of scattered light can be used to determine the velocity and therefore aerodynamic diameter. The magnitude of the electrical current generated by the scattered light also provides a secondary estimate of the particle size. APSs are commonly operated alongside an SMPS but can be used exclusively when particles <500nm are not of interest. For example, APS measurements are suitable for source characterisation of resuspended material.

Particle surface area

Active particle surface area is an important metric in the future of UFP measurement and can be measured by Diffusion Chargers, specifically NSAMs.

Diffusion Chargers use corona discharge to create unipolar ions that diffuse onto the active surface of particles, an electrometer then measures the charge that is transferred from the ions to the particle.

This charge can be related to the active surface concentration; a fraction of geometric surface area. This is a similar measurement premise to the DiSC. The nanoparticle surface area monitor (NSAM) measures particles between 20-400nm using the principle of unipolar diffusion charging.[51] It can measure lung-deposited particle surface area concentrations, based on lung particle deposition models.

Table 2 summarises the key properties of each of these sensors with their advantages and disadvantages.

Table 2. A comparison of the properties of PM sensors.

	Equipment	Real Time (Time Resolution)	Portability	Size Range	Detection Limits	Price Catagory (1 Lowest - 5 Highest)	Advantages	Disadvantages
Particle Mass	Gravimetric Filters	No	High	150nm<	10µg/m^3<	1	Filters can be used to determine mass and number concentrations, size distribution and composition. Cheap and simple to deploy indoors.	Processing the filters is highly time consuming
	Photometer	Yes (1s)	High	(40-100nm)-10µm	0.001-200mg/m^3	1-2	Portable, reliable, accurate and relatively cheap.	Not a direct measure of PM
	Low-cost photometers	Yes (1s)	Very High	N/A	0-600µg/m^3	1	Low-cost and high portability	Is not a self-sufficient sensor, it needs to be built into a system with a computer. Low sensitivity, especially at low concentrations.
Particle Number	CPC (Full Flow)	Yes (3s)	Medium	2.5-15nm<	<1x10^4-1x10^6 particles/cm^3	2-3	Highly robust and reliable equipment. Regulatory compliant in vehicle emission measurement due to a longer standing history of use.	Lower time resolution than mixing CPCs. More likely to have optics contaminated by working fluid than mixing CPC
	CPC (Mixing)	Yes (0.5s)	Medium	2.5-15nm<	<1x10^4-1x10^6 particles/cm^3	2-3	Higher time resolution than mixing CPCs, important for rapid atmospheric processes. Can measure higher concentrations due to inbuilt dilution.	More complicated to accurately measure sample flow,
	OPC	Yes (1s)	High	0.3-20µm	<1x10^4 particles/cm^3	1-2	Lower cost and more portable than conventional CPCs.	Unable to measure the smallest particles (<0.3µm).
	DiSC	Yes (1s)	High	10-700nm	<5x10^2-1x10^6 particles/cm^3	1-3	Portable, reliable, robust and lightweight. Accuracy is within 15-20% of a reference CPC. Can determine average diameter of measured particles (accuracy with 30% of SMPS).	Less accurate than CPCs, is not directly counting particles. Accuracy is good, but can be poor for certain particle compositions and shapes.
Particle Size Distribution	Impactors	No	Variable	1µm-10µm	N/A	1-2	Useful when looking at the size specific chemical characteristics of aerosols.	Not useful for smaller, or ultrafine particles. Sample analysis is time consuming.
	SMPS	Yes (1-4 mins)	Low	2.5-1000nm	1-1x10^7 particles/cm^3	4-5	Provides the highest resolution size distribution of particles .	Much lower time resolution than an FMPS
	APS	Yes (10s)	Medium	0.5-20µm	1000 particles/cm^3	3-4	High Temporal resolution, and can be used to suppliment equipment unable to measure at the larger sizes	Unable to measure the smallest particles (<0.5µm).
	FMPS	Yes (1s)	Low	5-560nm	N/A	4-5	No radioactive source. Much higher time resolution than an SMPS.	Size distribution generated has a lower resolution than a SMPS (30 channels vs 190 channels). Electrometers provide les accurate particle number concentrations tan the CPC used in the SMPS
Particle Surface Area	NSAM	Yes (1s)	High	10-1000nm	<10000 µm^2/cm^3	2-3	Portable, able to measure LDSA.	High sensitivity to environmental variables

Experimental applications

This section outlines the experimental applications of the previously described technologies, with aim to provide insights about why sensors were chosen for a given application, and the practicalities of deploying senor types indoors.

Compliance measurement, temporal trends and source apportionment

Compliance measurements are commonly collected when it is necessary to understand how the severity of pollution relates to national, international standards and historical measurements, usually making Pmass the focal metric. Many of these studies use real-time measurements and therefore often contain sections devoted to temporal variations in PM and apportioning these variations to potential sources.

One cheap and robust method for testing compliance is by using gravimetric aerosol samplers; however, these are not real time and therefore cannot be used to identify short-term temporal variation. Passive aerosol samplers have been used within primary schools, with a mixture both of polycarbonate and quartz filters deployed at child breathing height (1.2m).[52] They demonstrate that long-term temporal variations can be measured using gravimetric samplers deployed and collected over several seasons.[52] They were also able to apportion sources using chemical analysis, with particular sources having distinctive chemical compositions.[52] Passive samplers are smaller and quieter than larger active samplers; so appropriate for use in schools, many workplaces and homes.

Cascade Impactors can be used upstream of traditional gravimetric samplers when the mass-based particle distribution is of interest. For example, cascade impactors were used to simultaneously sample particles of >10µm, 10-2.5µm, 2.5-1µm and 1µm> in 8 different indoor locations in France.[53] This included private residences, a school and a restaurant across a range of urban, suburban and rural settings. The particles were chemically analysed to determine the concentrations of 16 US-EPA priority Polycyclic Aromatic Hydrocarbons (PAHs). Gravimetric samplers are also a relatively unobtrusive method of sampling, for example, size segregated Pmass was measured in a well-ventilated primary school gym during PE lessons, using personal cascade impactors to minimise intrusiveness and health and safety risks.[54]

NSAMs are used to account for UFP exposure by measuring LDSA. When measuring UFP exposure and dosing, LDSA is often chosen because it is likely a better indicator of health effects than other metrics. For example, exposures were measured in four elderly care centres, with spatial and temporal distributions of LDSA being assessed.[55]

Major sources are typically identified through chemical composition, but sources can also be apportioned through temporal measurements, although this is less accurate, and speculative. For example, apportioning increases in PM during rush hour traffic periods to vehicular emissions. A DustTrak was used to measure PM_{10} in Hong Kong schools to measure compliance against the Hong Kong Air Quality Objectives, with increased levels of PM attributed to sources such as traffic and construction.[56]

When measuring compliance, it is important to consider whether a device is suitable for the levels of PM being measured, this is because some sensors cannot detect low levels of PM whilst others cannot detect high levels of PM. For example, photometers were used to measure Pmass in highly polluted, densely populated, low income housing in urban Dhaka, Bangladesh.[57] The photometers were converted from smoke detectors and were developed specifically to measure in high pollution environments, with a lower detection limit of $50\mu g/m^3$. However, this caused difficulties with 49% of $PM_{2.5}$ measurements falling at or below $50\mu g/m^3$.

Indoor vs outdoor comparisons

Indoor vs Outdoor studies are usually focused on understanding the contributions of outdoor air to IAQ, the penetration rates of particulates and how ventilation and building design may improve or worsen IAQ. In addition, in developing countries especially, air pollution generated within households can have significant influence on ambient air pollution concentrations. These studies are not limited to use of any specific metric, in fact, each metric adds different value and should be selected based on the specifics of the investigation.

For example, the influence of outdoor air pollution and smoking on indoor $PM_{2.5}$ and Black Carbon levels, was quantified in 21 industrial community homes.[58] The Personal Exposure Monitors (gravimetric filterss) needed to be replaced every 3 days to prevent particle overload on the impaction plate or perturbation of the size cut-off inlet. The samplers were placed in the main activity room away from the windows, and heating and combustion sources to try to ensure the samples were representative of the whole room. Gravimetric samplers were chosen because industrial particle emissions are mainly composed of larger sized particles, contributing more to particle mass.

Indoor, outdoor and personal exposure samples were collected for 6 Beijing residences whilst operating air purifiers.[59] Gravimetric sampling was chosen because the researchers wanted to chemically quantify various health-related chemical components. Therefore, they compared the indoor/outdoor ratios of 27 chemical species.

The SMPS is often the equipment of choice in investigations aiming to understand indoor penetration and deposition behavior. This is because these processes are determined by particle size, which can be measured by the SMPS. For example a SMPS and APS were used to measure both indoor and outdoor Psd simultaneously by alternating between indoor and outdoor air inputs using a specially designed sampling manifold.[60] A similar study was conducted in modern offices.[25] In this investigation they were able to determine differential infiltration and deposition as a function of size. Indoor and outdoor Pnum and Psd were measured simultaneously in a school using 2 SMPS units; they were able to identify the main sources influencing indoor PM and determine indoor/outdoor ratios as a function of size.[61]

Another study used an OPC, CPC and gravimetric samplers simultaneously; the OPCs size distribution was used to supplement the absolute Pnum measurements of the water-based CPC.[62] With the water-based CPC also having a maximum size cut-off of 3µm, the OPC was able to extend this range up to 10µm. This study aimed to understand indoor and outdoor source contributions to indoor air over different size ranges, so the OPC was important to measure the size distributions. OPCs provides a more affordable and portable measurement of Psd than the SMPS, but lack in the ability to measure <300nm.

Our current understanding of the relationship between indoor and outdoor PM is summarised by Chen[63].

Source characterisation

Source Characterisation investigations aim to quantify the particle generation of various PM sources within the indoor environment. This can be studied in a real-world, laboratory or chamber environment, predominantly using Pmass, Pnum and Psd metrics and real-time measurement equipment.

In a chamber setup fine and UFP emissions were measured from 13 particle sources using an OPC and CPC.[64] A total-capture dilution tunnel systemwas used to investigate the Pnum and Psd emissions for 11 household cook stove-fuel systems using a SMPS.[65] Conversely, cooking was characterized using a SMPS and APS in the real-world namely 15 homes in Brisbane, Australia.[66] Elevated Pnum was linked to 21 other indoor activities using occupant logs and these events were measured using a CPC and Dustrak. Controlled environments allow for more accurate and repeatable measurement of particle generation; but real-world measurement can provide a more realistic basis for investigating processes; for example, dispersion, ageing and deposition of PM.

OPCs are ideally suited to resuspension studies because they negate the OPCs biggest weakness, the inability to measure particle <300nm. This is because the process of resuspension by human activities contributes most to the coarse particle fraction (>1um).[66] OPCs are commonly used when the UFP fraction is not of interest or to supplement the measurements made by other equipment. OPC measurements can add Pnum and Psd data to data collected by CPCs and photometers. Alternatively, at additional cost; the OPC could be paired with a diffusion size classifier to measure a much wider range of 10nm-20µm. Of 13 studies measuring resuspended particles and how they varied in size between different human activities, 8 utilized OPCs and 3 utilised APS.[67]

When high time resolution data is important, a FMPS may be used rather than a SMPS. For example, a FMPS (5.6-560nm) and OPC (0.3-20um) were used to characterise the emissions from 7 wood burning fireplaces in German homes.[68] Although these fireplaces largely have airtight seals, the combustion chamber needs to be opened regularly to put more wood in and this led to increased Pnum concentrations within the room. As the chamber only remains open for a few seconds, a shorter time resolution is required than the >1-minute resolution of the SMPS. Therefore, the researchers used an FMPS to measure Pnum and Psd at 1s resolution. The nanoparticle emissions of burning incense were characterised using a FMPS within a chamber.[69] High time resolution data was important to understand periods of rapid change; for example, the post-burning period after the incense was extinguished.

High resolution spatial and temporal measurements

It is becoming increasingly important to understand the lateral PM variations within rooms and buildings. Therefore, there is a need for low-cost, portable real-time sensors. It appears like the flagship measurement type for this will be photometry, with the majority of developed low-cost sensors working on the light scattering principle.[70] These investigations are currently still largely testing the premise, whether these instruments are accurate and sensitive enough to measure accurately remains to be seen. For example, several of these photometers were deployed in a single room to determine lateral variations in PM.[71] Similarly, these sensors have been deployed in households in Raipur, India to understand the spatiotemporal resolution of PM generated by cookstoves.[42] In lab tests the sensors agree well with reference grade equipment, however, these sensors become saturated at 4-5mg/m^2 compared to the 20 mg/m^3 of the SidePak, making them less suitable for high pollution environments.[72]

Discussion

Whether it is important to measure multiple metrics in an investigation depends upon what the study design and focus is. Using a variety of instruments, measuring the same or different metrics will improve confidence in results and understanding of the nature of indoor PM. For example, in photometer-based studies photometers are regularly co-located with reference grade gravimetric samplers to ensure the data collected is of suitable accuracy or to calibrate the photometer if necessary. With Pnum and Psd measurement, it is important to consider whether it is possible to expand the particle diameter range measured by using multiple pieces of different equipment. For example, supplementing the SMPS (2.5-1000nm) or FMPS (5-560nm) with use of an APS (0.5-20µm) or OPC (0.3-20µm). Combining use of high cost, less portable and more accurate equipment with low-cost, highly portable equipment can also improve the spatial resolution of results. This is important, because air pollution will not be homogeneous throughout a single room or building. Given the disparity between the concentrations of PM indors in developing and developed countries (responsible for ~97% and 3% of premature deaths due to household air pollution in 2016),[3] it is essential to be aware of the range limitations of sensors. For example, many low-cost photometers are able to measure effectively in high concentration environments, but are extremely insensitive in at low concentrations.[72] Conversely, mixing CPCs and photometers are extremely versatile in measuring a wide dynamic range; with sensitivity at low concentrations and with inundation only at very high concentrations. This is due to the mixing CPCs dilution of the aerosol flow and the photometers historical use for occupational health monitoring in high pollution environments, for example, saw and flour mills.

Challenges of PM metrics

A challenge of the Pnum/Psd/Surface area metric is the difficulty in comparing measured values across literature. Different investigations often operate equipment with differing size ranges or operate the same piece of equipment at different size ranges. Unfortunately, comparing measurements of different size ranges is not meaningful.[8] In this review paper, even the difference in minimum diameter measurement between 6-15nm made reliable comparisons difficult and therefore only 2 out of the 8 studies included in this review were directly comparable. Having no standardised size range is one of the reasons why universal standards have not yet been developed. Different types of Pnum and Psd sensor have highly variable size ranges depending on the method being implemented; this adds further complexity to the task. Without regulation, expecting developers to comply to standardised size ranges is difficult as developers will want freedom to change and increase their size ranges as technology improves. On the other hand, developers like regulation-backed markets because it mandates the need for their products. It will be important to allow developers to contribute to discussions around standardisation. Being unable to directly compare to other literature or universal standards, investigations instead must rely on internal comparisons, for example, the levels were *x* times greater than background levels or Site A was *x* relative to Site B. Although there are standards issued for the Pmass metric, very few are specific to indoor environments and this is a problem because the variability in concentration, chemical composition and Psd can all be significantly different to outdoor environments. This is because indoor air is made up of contributions from both indoor and outdoor sources; this means that although they share many physio-chemical properties, they are often far from homogenous.[8] Future campaigns should aim to deploy high grade sensors to measure the physio-chemical properties of indoor air pollution under a variety of scenarios; one such campaign is

HOMEchem.[73] Once the nature of IAQ is better understood it can be compared to outdoor air quality in order to make informed decisions on whether health standards should differ between them.

Standardising the field of PM measurement

Given the difficulty in making comparable measurements, there is a need for the community to come together and to work towards standardized measurements. Accordingly, there are two types of guidelines that need to be defined: 1) standardized size ranges or D_{50s} for individual measurement techniques e.g. CPCs, OPCs, DiSCs and NSAMs, and 2) regulatory guidelines for what are allowable concentrations of PM for these standardized size ranges.

However, setting these guidelines is not an easy task (especially No. 2), and therefore, a model for developing improved guidelines is illustrated in Figure 5.

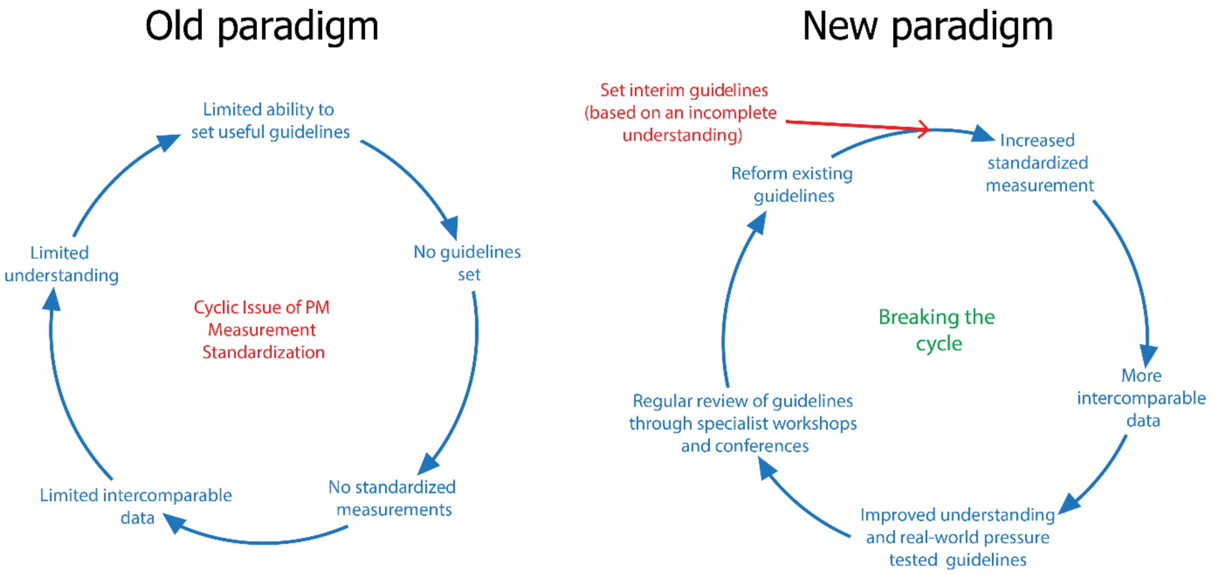

Figure 5. The cyclic issue of PM Measurement standardization and breaking the cycle.

Initially, due to our limited understanding of the epidemiology of Pnum and surface area concentrations, regulatory guidelines are difficult to relate to health effects, as is the case for existing Pmass guidelines. Instead, good, satisfactory, poor and extremely poor standards, could be based on the frequency of measurement, i.e. based on measurement frequency percentiles as a posed to a relationship to health effect. As the “new paradigm” cycle continues and epidemiological understanding improves, we could start to implement the more useful health related guidelines.

Development of these guidelines are beneficial to a variety of stakeholders. Firstly, PM researchers and epidemiologists will have more inter-comparable measurements, which will improve understanding on their respective fields. Regulators will be better informed, allowing for development of more robust guidelines. This will expand the usefulness of Pnum, Psd and surface area metrics for regulatory purposes, and this intern will benefit equipment manufacturers by increasing demand for their products.

IAQ conflicting with building energy efficiency

In recent years improvements to the energy efficiency of buildings has reduced air exchange rates between indoors and outdoors, to improve energy efficiency through reducing heat losses. However, this has created indoor environments where pollution can accumulate to much higher levels than before.[10] There must be a balance between improving energy efficiency, reducing operation costs and improving IAQ.[74] However, in some environments such as in the mega-cities of many developing countries; reducing the exchange between indoor and outdoor air may improve the IAQ, especially in indoor environments without dominant sources of indoor air pollution. Improved energy efficiency may motivate increased research on IAQ, how it relates to outdoor air quality and the nature of indoor air pollutants ageing indoors. It is worth noting that indoor air chemistry will have similar complexity to atmospheric chemistry. Some of the most important questions relating to IAQ and “green buildings” are addressed by Steinemann[75].

Low-cost sensors

Low-cost sensors are becoming increasingly important in the modern world and are essential to improving the spatial and temporal resolution of urban air pollution measurements.[13,76] Static and scarce air pollution monitoring stations are not sufficient to fully understand the behaviour of pollution in urban areas. Existing networks could be supplemented with a low-cost high-density network. The same concept applies indoors, but on a smaller scale; by taking a single measurement in a room or building the assumption is that the air is homogenous and the spatio-temporal variation is neglected, introducing errors into exposure intake estimations.[71] It is worth noting however, that even high resolution spatio-temporal measurements may not be fully indicative of individual exposure given the “personal cloud” effect,[77] therefore, in the future low-cost sensors will also be used extensively for personal exposure measurements. Switching from wired to wireless data transmission has been shown to reduce initial investment and annual operation cost by 3x and 5x, respectively, in the US.[78] Wireless data transmission also allows for unattended large-scale monitoring. Wireless data transmission is especially important in indoor data collection because minimising intrusiveness is important. Intrusion into residences and workplaces can be disruptive; and being able to minimise intrusion will increase the participation in projects. Furthermore, in unoccupied environments, intrusions can influence indoor PM through resuspension. Some of the issues currently associated with the future of low-cost sensor networks are; the consistency and durability of sensors, the reliability of data and the high costs associated with data processing. Moreover, most low-cost sensors have a short lifetime of 6 months to a few years, which will need to be improved for these sensors to be viable in the future. The costs of calibration, servicing (replacing batteries), integrating the sensor into a device and data processing often exceeds the cost of the sensors themselves; these costs need to be reduced. Sensor performance will need to be tested under a wide range of environmental conditions before they are ready to be deployed on a large scale. Gas sensors are being miniaturized at a much faster rate than particle monitors, largely because it is difficult to develop particle monitors that are very sensitive to small concentrations.[78] Moreover, with gas sensors, the focus is purely on concentration, with PM it is important to understand Pnum, Pmass, Psd and chemical compositions. This makes PM a lot more difficult to effectively quantify when compared to ambient gas concentrations. Another shortcoming of low-cost devices is their signal to noise ratio, making them much less useful in low pollution environments. This can be negated to some extent by recording for longer periods and averaging the results, or by co-locating several sensors and taking an average. Digital filters are commonly applied to the data from these sensors to remove high-frequency noise.[71]

Applications of and Considerations for Low-cost sensors

In cities, creating networks like this may be made easier due to existing infrastructure, mainly access to fast reliable internet. However, some of the worst air pollution is experienced in rural low-income areas where networks would be difficult to set-up and maintain. In addition, low-cost sensors are increasingly important in making measurements in the developing world due to their nature of being easily deployed and operated and having low power requirements. Moreover, with PM concentrations usually higher within the developing world, the decreased sensitivity at low concentrations is less of a problem. A summary of low-cost sensors and monitors is available, however, these are neither specific to indoor or outdoor environments.[70]

If low-cost sensor networks are introduced nationally with a large database being generated, several issues need to be considered. It is important that the data collected is easily accessible to various stakeholders; and that the database is not held by a private company that might abuse the database for financial gain.[78] It is necessary to consider who would own such a network and database; public bodies, research bodies, commercial entities or citizens. Minimizing the need for data processing and analysis through automation could make air pollution data from portable sensors more accessible to the public. If the data can be interpreted into an easily accessible form, then citizen uptake will increase. Fortunately, low-cost sensors require very little knowledge to be deployed and require less technical maintenance than more complicated equipment, which may also increase citizen uptake. Low-cost sensors could also provide immediate value, by warning occupants of high pollution levels and offering suggestions to mitigate these, for example, opening the window or automatically turning on an air purifier. It is also worth considering whether it is essential for these low-cost sensors to perform well at low-concentrations; if their primary purpose is to inform users in real time of dangerously high levels. People may be more inclined to remediate IAQ problems based on personalised measurements of IAQ, rather than city wide pollution measurements and warning systems.

Ethical concerns of real-time measurement

Real time measurement within residences also raises various ethical concerns. For example, IAQ data can be used to infer activity, such as when the house is occupied or unoccupied, whether residents are awake or asleep and whether activities such as cooking, or bathing are occurring. Therefore, personal data is inadvertently being collected alongside the IAQ data. This needs to be considered when viewing the data in real-time and ensuring anonymity when viewing data not in real time. A further ethical concern occurs if dangerously high pollutant levels are observed in real time. What level of responsibility and duty of care should residents expect? Should looking at results in real-time be avoided completely, even though this is an important part of maintaining sensors and troubleshooting?

Conclusions and recommendations

Pmass is the most widely used metric of PM currently with measurements being robust, reliable, and easily compared to well defined standards and a long-standing literature. However, specific indoor health related standards are still to be developed. Pmass will always remain useful due to its long history of use in compliance testing, however, future epidemiological studies may conclude that Pmass itself has less correlation to human health effects than other metrics. Pmass is likely to continue its evolution from PM_{10} and $PM_{2.5}$ down to PM_1 or $PM_{0.5}$ as technology improves. The dominant contribution to Pmass comes from larger particles (>100nm), whereas Pnum is largely influenced by the smallest particles (<100nm). The Pnum metric is becoming increasingly popular as focus is shifting towards measurement of UFPs. Unfortunately, with Pnum it is very difficult to make comparisons to previous studies and it seems unlikely that any air quality standards will be developed soon. The Pnum metric is considered a better determinant of health effects than Pmass,[27] but Pmass sensors are being reduced in size at a much faster rate than Pnum sensors. This might make Pmass sensors the preferable choice for the low-cost sensor networks of the future. Although Pnum sensors are being reduced in size, for example, OPCs; they cannot measure the very smallest particle sizes. The Psd metric can allow for observations that cannot be determined from Pmass or Pnum alone and will become increasingly implemented as sensors improve. Psd will likely remain as a "research grade" measurement, solely for understanding the nature of PM and applying that to developing better metrics and sensors for compliance monitoring. There is consensus that future UFP metrics should provide insight into how PM interacts with the body through intake, uptake and transport. Therefore, many are favouring a surface area related metric, which correlates better with toxicological or biological activity than either Pmass or Pnum.[31] NSAMs are a likely to become an increasingly important measurement option, as interest in measuring surface area related metrics increase.

Many countries and organisations are having to decide whether to solely legislate using the historical and conventional Pmass metric or to adopt other metrics. However, this is an extremely difficult task given the current lack of understanding of PM and its associated health effects, even though PM is one of the best understood forms of air pollution, there is still much that needs to be understood. Several essential epidemiological questions still need to be answered.[79] Firstly, which properties, or combination of properties, are the most important determinants of potency; for example, size, surface area, or chemical composition. Secondly, are chronic background pollution levels or acute high-level pollution events a greater determinant of health effects. This will largely inform what type of standard should be enforced; for example, daily average or “should not exceed x for longer than y duration”. It will also inform the needs of regulatory compliance measurements, for example, if high level pollution events are deemed the most important determinant of health effects, then having PM monitors which are sensitive to low concentrations is far less important. Until these questions are answered, we should continue to experiment using a wide range of metrics to better understand the nature of PM.

With low-cost real-time sensors being at the forefront of meeting rapidly increasing public interest, and with citizen science projects becoming more frequent, personalized information about IAQ is likely to become increasing available.[13,80] Eventually this could lead to real-time IAQ warning systems in homes; allowing residents to more easily mitigate IAQ issues.[80] This level of technology would also allow us to resolve many of the issues related to IAQ and energy efficiency.As real-time low-cost sensors become increasingly common, high spatial distribution measurements which were previously difficult are also likely to become more common. However, it is unlikely that the current generation of low-cost PM sensors will be able to detect subtle variations in the indoor environment. But this will likely change as sensors are periodically improved. To fully understand the future of PM measurement, it is essential to understand how PM interacts with health, and which metrics are best able to capture this interaction.[79] A better understanding of epidemiology will inform the future of compliance measurement and source characterization in indoor environments.

Indoor PM is attributed to many premature deaths worldwide. Several essential metrics and measurements techniques are available that practitioners and scientists can use to better understand and reduce indoor PM. In the future it is essential to better understand what PM properties most strongly effect health and to channel this information into the development of improved metrics, measurement techniques, legislative standards and new experimental applications.

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Chapter 3. How efficiently can HEPA purifiers remove priority fine and ultrafine particles from indoor air?

Highlights

1. It is currently unclear how particles of different sizes are removed by air purifiers
2. Three popular models were tested in China's largest indoor smog chamber
3. Particles <100nm were removed efficiently
4. 200-250nm particles were least efficiently removed
5. Ambient air particles were removed at a similar rate to standard particle types

Abstract

More than 1 million premature deaths in Asia annually are estimated to be associated with indoor air quality. HEPA (high-efficiency particulate air) filter air purifiers (APs) are widely used in urban Chinese residences by the growing middle class, as public awareness of air pollution increases. Currently, understanding of how particle size affects particle removal is inconsistent, and the rate at which different particle types are removed remains largely unknown. Therefore, this investigation aimed to determine the relationship between particle size and the removal efficiency of particles, and how efficiently ambient air is filtered compared to standard particle types which are typically used for such tests (tobacco smoke, dust and pollen). Three of the most popular AP models in China were tested in China's largest indoor controlled chamber laboratory and the removal efficiencies of particles in the 18-514nm range were identified. Each AP had a distinct profile of removal efficiency against particle size, but the three APs shared similarities in performance, with removal efficiency consistently lowest at 200-250nm. This size fraction is important in an exposure context as these particles are abundant in ambient air in mega-cities, can penetrate through building shells effectively, remain airborne for long periods of time and can penetrate the deepest areas of the lungs. Ambient air particles were removed at a similar rate to test particles; this confirms that the Association of Home Appliance Manufacturers' (AHAM) standards are a suitable proxy for "real world" performance.

Introduction

An estimated 4.2 million premature deaths globally were attributed to indoor air pollution in 2016, compared to 3.8 million from outdoor air pollution (WHO, 2018). It is estimated that 90% of people breathe air that does not comply with the World Health Organization Air Quality Guidelines (WHO, 2016). Poor indoor air quality is estimated to be the 9th largest global burden of disease risk (Forouzanfar et al., 2015). The Institute for Health Metrics and Evaluation (2017) attributed 2.6 million premature deaths to indoor air pollution in 2016; Roser and Ritchie (2018) partitioned this estimate by continent with Asia, Africa, Europe and the Americas contributing 74%, 23%, 1% and 2% respectively, demonstrating the significance of premature deaths in Asian countries. On average, modern populations spend more than 80% of their time indoors (Duan et al., 2015; Klepeis et al., 2001), with the indoor environment contributing 19-76% of an individual's ultrafine particle (UFP) exposure (Morawska et al., 2013).

Particulate matter (PM) is defined as the total of all solid and liquid particles suspended in air and is a major determinant of indoor air quality (IAQ) (Lowther et al., 2019). PM is strongly associated with negative health outcomes including strokes, heart failure, asthma and lung cancer (Lim et al., 2012). Size is an important property of PM with regard to its potential health effects. Therefore, PM is commonly categorized based on its aerodynamic diameter into the commonly regulated standards of <10µm (PM_{10}), <2.5µm ($PM_{2.5}$), and <100nm (UFPs). Smaller particle size fractions are able to penetrate further into the respiratory tract and are thought to have a higher toxicity per unit mass due to a larger surface area to mass ratio (Harrison and Yin, 2000; HEI Review Panel, 2013).

In China, more than 1 million premature deaths were attributed to long-term exposure to $PM_{2.5}$ in 2016 (Health Effects Institute, 2018). In 2017, the average annual ambient $PM_{2.5}$ concentration across 338 Chinese cities was 44 µg/m^3, with 73% of these cities failing to meet the national air quality standard of 35 µg/m^3 (Ministry of Ecology and Environment the People's Republic of China, 2018). Furthermore, in China it is estimated that 66-87% of total exposure to $PM_{2.5}$ of outdoor origin occurs within indoor environments (Xiang et al., 2019). It should, however, be noted that although PM levels in China are severe, rapid reductions in PM concentrations are being observed. For example, the average $PM_{2.5}$

concentration in Beijing dropped from 90 µg/m^3 in 2013 to 58 µg/m^3 in 2017 (Ministry of Ecology and Environment of the People's Republic of China, 2018).

The two fundamental sources of PM in indoor environments are: (i) PM generated by indoor sources and activities and (ii) PM generated by outdoor (ambient) sources penetrating indoors. Important indoor PM sources in China include solid fuel use, cooking, smoking and incense burning (Apte and Salvi, 2016; Tse et al., 2011). Solid fuel use is especially dangerous in China from a health perspective (Zhang and Smith, 2007), with solid fuel combustion generating high levels of PM with substantial concentrations of carbon, iron, lead, cadmium and silica (Apte and Salvi, 2016). However, in the absence of major indoor sources, outdoor to indoor air exchange is the most significant source of PM indoors. In a study of 41 Beijing residences, a strong correlation (R≥0.90) was found between ambient and indoor $PM_{2.5}$, with ambient levels accounting for ≥84% of the variance of indoor levels (Huang et al., 2015). In a summary of 77 studies involving over 4000 homes, indoor/ambient $PM_{2.5}$ ratios were found to vary substantially, from 0.5-3.5 (Chen and Zhao, 2011). Additionally, buildings in China are often ineffective in preventing ambient fine particles from entering indoor environments, with standards for air tightness of residential buildings being less restrictive than in the United Kingdom or United States (Hu et al., 2018). Given that ambient air strongly influences indoor air in China, the composition and properties of the air are likely to be very similar, in contrast to conditions where indoor sources dominate. Therefore, this study focuses on ambient particles that contribute significantly to indoor environments, to estimate the performance of HEPA type air purifiers in real world indoor environments.

High-efficiency particulate air (HEPA) filters are an effective technology for improving IAQ when removing PM is the priority (Zhang et al., 2011). To be defined as such, a HEPA filter must be able to remove 99.97% of particles greater than or equal to 0.3 µm. In a HEPA Air Purifier (AP), air is forced through the HEPA filter and particles are physically captured. The four key mechanisms through which particles are captured are diffusion, interception, inertial impaction and sieving. Diffusion causes the smallest particles to be removed, whereas interception, inertial impaction and sieving processes are more effective at removing the largest particles (Yang, 2012). This means that particles of an intermediate size (100-400nm) are the least efficiently removed (Kowalski et al., 1999). Particle size, charge and shape are the controlling factors determining how effectively particles are removed by the

HEPA medium. Studies have shown that HEPA filters can reduce particulate mass and particle number concentrations by >50% (Batterman et al., 2012, 2005; Kelly and Fussell, 2019; Ward et al., 2017; Wheeler et al., 2014). There is also limited evidence to suggest that these reductions lead to improvements in cardio-respiratory health (Fisk, 2013; Morishita et al., 2015). Collectively, studies have reported that use of indoor APs may be associated with reductions in blood pressure, oxidative stress, systematic inflammation and improved lung function (Kelly and Fussell, 2019). Health benefits are most consistently observed in Asian mega-city homes, likely due to higher baseline indoor concentrations and therefore more significant absolute reductions (Kelly and Fussell, 2019).

The Chinese AP market stood at $ 2 billion in 2017 and is predicted to surpass $ 4.3 billion in 2023 (BIS Research, 2018). HEPA AP technology held ~40% of market share in 2016 and is the fastest growing segment of the market (BIS Research, 2018). This growth in the market can be attributed to the growing Chinese middle class and improved awareness of IAQ, with APs mainly used by more affluent members of Chinese society.

The Association of Home Appliance Manufacturers (AHAM) is the main body which verifies the performance of HEPA APs and although they are based within the United States, they produce certified ratings for AP brands all over the world. They measure the filtering efficiency of HEPA APs using the Clean Air Delivery Rate (CADR) metric - the flow rate of particle-free air output in cubic feet per minute (ft^3/min; note: 1 ft^3/min = 0.028 m^3/min). AHAM test the CADR of HEPA APs for three particle types, tobacco smoke (0.09-1 µm), household dust (0.5-3 µm) and pollen (5-11 µm) (AHAM, 2002). However, within a laboratory context it is currently unknown how efficient HEPA APs are in removing “real world” particles, i.e. those found in ambient air. Therefore, it is valuable to investigate how well ambient air particles are removed in comparison to AHAM standard particle types, to see whether the selected particle types are representative of real-world performance.

Combustion-generated particles can penetrate National Institute for Occupational Safety and Health (NIOSH) N95 filtering face-piece respirators more efficiently than standard sodium chloride particles (Gao et al., 2015). Peck et al. (2016) investigated whether this applied to HEPA APs, concluding that diesel combustion particles were removed more efficiently than both NaCl and AHAM test particles, with lowest and highest removal efficiencies at 42-100 nm and 100-700 nm respectively. For standard particle types Sultan et al. (2011) and Waring et al. (2008) both observed erratic CADR performance below ~40 nm (potentially due to instrument sensitivity), and consistent performance above 40 nm. Mølgaard et al. (2014) tested two HEPA APs between 12-660 nm; one performed consistently with increasing size whilst the other experienced a peak in removal efficiency at ~200 nm. Furthermore, Lee et al. (2015) found the lowest filtration efficiencies for three APs to fall within the UFP size range. The findings of these studies contradict our current understanding of the filtration efficiency of HEPA filters - a minimum efficiency of around 200-300 nm varying from filter to filter (Kowalski et al., 1999). However, it is worth noting that a filter may not perform as efficiently within an AP as it does in laboratory tests, given processes like filter bypassing (a result of AP design) and short circuiting of filtered air (Shaughnessy and Sextro, 2006). Therefore, it is currently unclear how effectively "real world" particles of different sizes are removed by commonly available household APs, and why some measurements of performance do not align with the current understanding of the removal processes of HEPA APs. This paper aims to resolve these uncertainties.

Using the Guangzhou Institute of Geochemistry's state of the art chamber laboratory, the largest indoor chamber in China (Wang et al., 2014), this investigation aimed to determine: (a) which particle sizes from ambient air are most and least efficiently removed by APs and explain how this might be important in a real-world context; and (b) whether ambient air particles are removed more or less efficiently than AHAMs standard particle types (tobacco smoke, dust and pollen) and whether AHAM should therefore consider adjusting their CADR measurements accordingly.

Methodology

Selection of air purifiers

For this investigation, three HEPA APs were selected to represent the small (CADR 100-200), medium (CADR 200-300) and large (CADR >300) AP sizes (Table 1). All three APs were purchased on the Chinese market and were certified by AHAM. They were selected as popular models that represent different filter, AP design types and sizes. AHAM certification provides a means of allowing performance comparisons to be made between models for the removal of different particle types and their associated size fractions. The reason that tobacco smoke (90-1000 nm), dust (500-3000 nm) and pollen (5000-11000 nm) CADRs for the same AP are different is due to differential removal based on their respective particle sizes.

		CADR (ft^3/min)	
	100-200	200-300	300<
Model	Blueair 203	Midea KJ400G-E33	Philips AC6608
Referred to as	AP(Small)	AP(Medium)	AP(Large)
AP Type	Compact	Tower	Cube
Filter type	Single Filter	Circular Filter	Dual Filter
Tobacco Smoke CADR (ft^3/min)	155	226	369
Dust CADR (ft^3/min)	155	229	389
Pollen CADR (ft^3/min)	155	236	451
Purchase Cost RMB (USD)	2000 (200)	1700 (250)	4000 (700)
Filter Replacements RMB (USD)	200 (50)	600 (90)	600 (90)
Recommended Room Size (sq ft)	240	350	572

*RMB costs represent the cost on the Chinese market, USD represents price on the US market

* 1 ft^3/min = 28.3 litres/min

Table 1. Summary of selected HEPA APs (AHAM, 2018).

Experimental setup

The atmospheric chamber laboratory at the Guangzhou Institute of Geochemistry, Chinese Academy of Science was used for this investigation. The properties of this chamber are described in detail by Wang et al. (2014). It consists of a 30m^3 fluorinated ethylene propylene Teflon film reactor (hereon referred to as a Teflon reactor) housed within a temperature-controlled enclosure (hereon referred to as the chamber enclosure). The Teflon reactor can be filled and vented using pumps with a flow rate in excess of 1m^3/min, meaning that it may be filled entirely within 30 minutes. A blower motor from a high-volume air sampler using a tube with a 6 cm bore was used to minimize particle losses. Figure 1a illustrates the layout of the chamber laboratory and 1b displays an image.

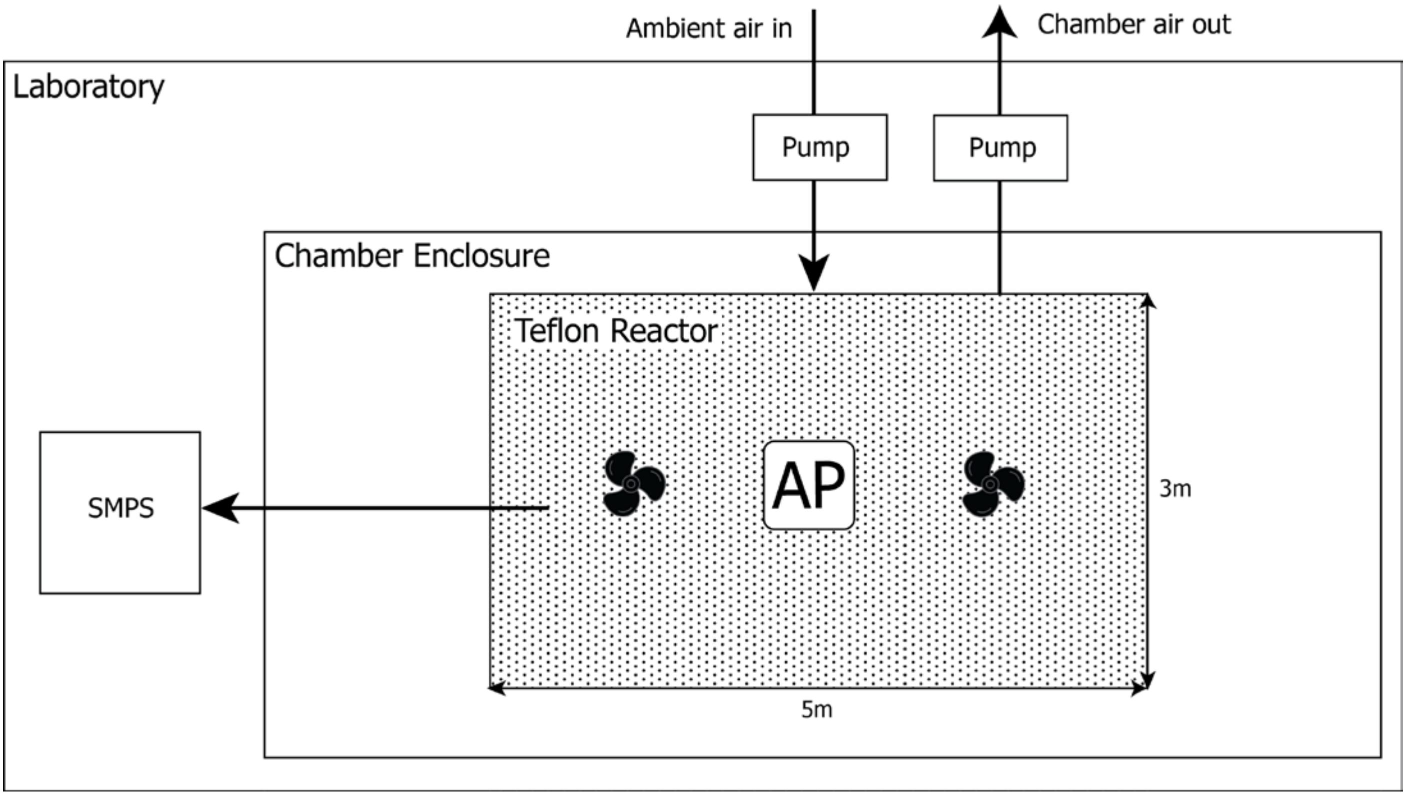

Figure 1a. Layout of the chamber laboratory, chamber enclosure and Teflon reactor at the Guangzhou Institute of Geochemistry.

Figure 1b. An image of the chamber laboratory, chamber enclosure and Teflon reactor at the Guangzhou Institute of Geochemistry.

During the experiments, the Teflon reactor was filled entirely with ambient air from outside the laboratory. It is important to understand the composition of this ambient air. Liu et al. (2014) have previously reported that on the Guangzhou Institute of Geochemistry site, carbonaceous aerosols (which contribute a large fraction of $PM_{2.5}$) could be attributed to fossil fuel (46%), non-fossil fuel (51%) and biomass burning (3%). In a larger study of the city, in the dry season, when this investigation was conducted, ambient PM was largely composed of emissions from vehicular (21%), industrial (20%), residential (4%), power generation (2%) and other unknown sources (53%) (Cui et al., 2015). In 2017 Guangzhou had an annual average $PM_{2.5}$ concentration of 35 $\mu g/m^3$ (Ministry of Ecology and Environment the People's Republic of China, 2018), with $PM_{2.5}$ in the dry season of 2013 composed of secondary organic aerosol (23%), primary organic aerosol (14%), sulphate (14%), nitrate (11%), ammonium (7%), elemental carbon (4%) and an unidentified fraction (28%) (Cui et al., 2015). The atmospheric chamber laboratory is located ~250m from an 8-lane highway, and therefore UFPs will likely be of vehicular origin. Air was sampled at a height of 1m, directly outside the atmospheric chamber laboratory.

Before each test the Teflon reactor was evacuated, and ambient air was drawn in from directly outside the laboratory. Two Teflon coated fans located within the reactor gently mixed the air during filling and throughout the duration of each experiment. The Teflon reactor was not a fixed volume or shape like a stainless-steel chamber, and so there was some variation in the reactor volume between experiments. This is addressed in more detail later. Given that the air was purged entirely from the reactor before it was refilled, no additional cleaning was required between test runs. A TSI SMPS (Scanning Mobility Particle Sizer) consisting of a Differential Mobility Analyzer (DMA - classifier model 3080) and Condensation Particle Counter (CPC - model 3775) was used to measure the total particle number concentration (PNC) and particle size distribution (PSD) between 18-514nm in 94 size bins, with a full scan completed once every minute. Once the Teflon reactor was filled with ambient air, the AP was started. Experiments were repeated a minimum of four times for each AP, at each of three fan speeds (low, medium and high). A new HEPA air filter was used for each AP for the duration of the repeats, therefore, filter loading had a minimal effect on performance given that a single filter was used for no more than 24 hours in total (filters are rated for roughly ~1-2 years of regular use).

Clean air delivery rate

CADR was calculated using the equation $CADR = V(Dm - Dn)$ where V is the volume of Teflon reactor in ft^3, Dm is the particle number decay rate when the AP is active and Dn is the natural particle number decay rate in the reactor when the AP is inactive (AHAM, 2002). Dm and Dn are first-order loss rates (min^{-1}), the decay constants of an exponential decay in particulate number concentrations, as measured by the SMPS. Initial total particle number concentrations within the reactor varied between $8x10^4$ – $2x10^5$ #/cm^3, depending on the ambient conditions at the time.

If a decay series met either of the following criteria, then it was excluded; (a) if the decay series contained less than 9 points, meaning that the minimum test duration was less than 9 minutes (AHAM, 2002), and (b) if greater than 30% of values in the decay series exceeded their previous values.

Criteria (a) was responsible for identifying failed decay series on the largest AP at the highest fan speed, with the AP cleaning the reactor too quickly (<10 minutes), making calculations of decay rate and therefore CADR unreliable. Criteria (b) was mainly used to identify failures on the smallest AP at the lowest fan speed, with some decay series being difficult to identify amongst variability caused by mixing. Failure to meet these criteria is illustrated in Table 2.

Calculating natural decay rate was essential to determine how much particle removal was due to the AP and how much was due to other processes including agglomeration, wall loss and deposition. Natural Decay rates in the Teflon reactor were calculated with an AP present, but not actively running, using the SMPS for each of the 94 size bins from 18-514nm. In this way, both the measured decay rate and natural decay rate were specific to the particle size. The decay rates were measured five times within a single day, see Figure 2.

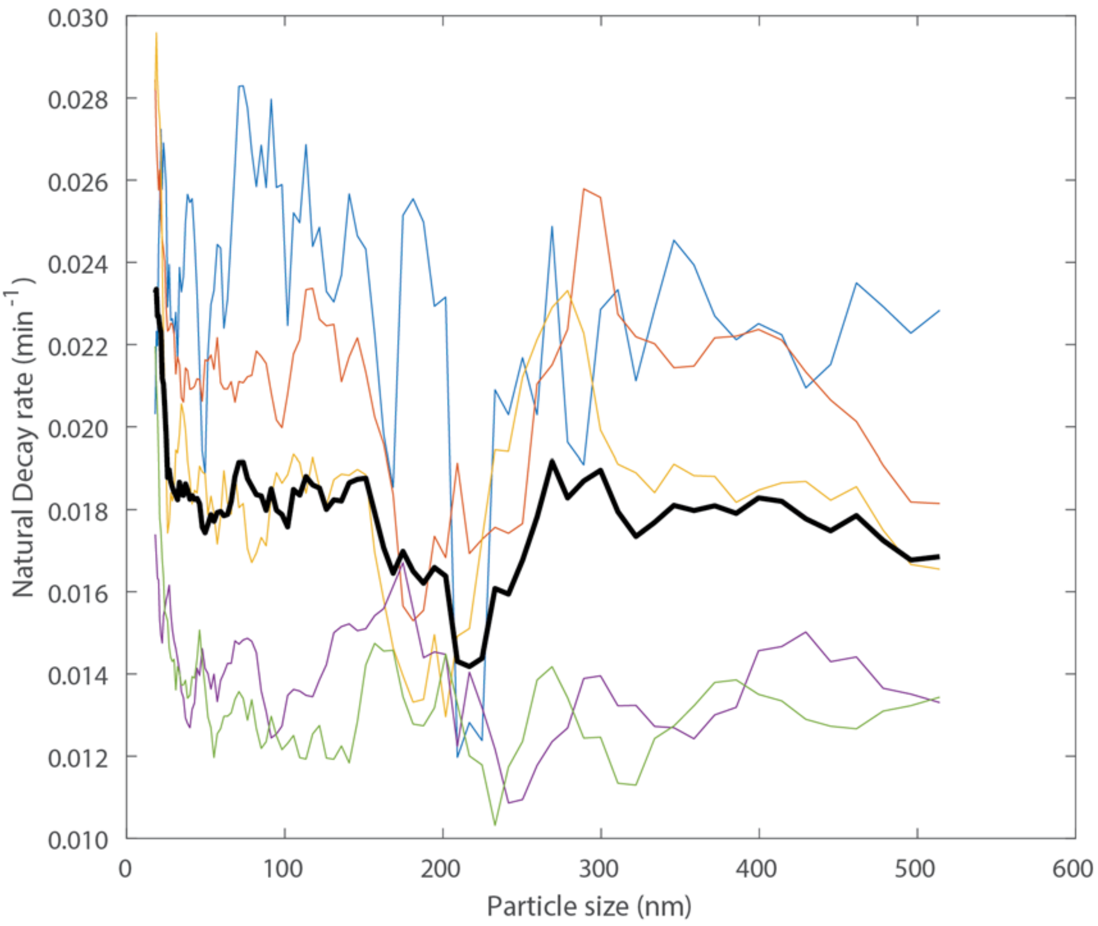

Figure 2. Natural decay rate (min^{-1}) in Teflon reactor without use of APs, n=5. Solid black line represents the average natural decay rate (min^{-1}).

Given that the Teflon reactor was inflated using ambient air, the reactor was not a fixed volume at the start of every experiment. The minimum and maximum volumes for the reactor at the fixed roof height were therefore calculated using the trace gas injection method (Mazzeo, 2012). The minimum and maximum volumes were 24.5 m^3 and 27.2 m^3 respectively, however, the reactor was inflated to an intermediate volume between the minimum and maximum volume. A volume of 25.9 m^3 (the midpoint between maximum and minimum volumes) was therefore used in the calculations.

In comparable chamber studies, high concentrations of tobacco smoke, vehicle exhaust, sodium chloride or pollen were released into the chamber and mixed (Mølgaard et al., 2014; Peck et al., 2016; Sultan et al., 2011; Waring et al., 2008). In this experiment, the reactor was filled with much lower particle concentrations in ambient air. The challenges associated with this investigation were likely larger than that of comparable chamber studies; given the nature of using ambient air, which varies temporally in composition, humidity and temperature. Furthermore, given that the reactor needed to be inflated to an approximate size, this limited the ability to use a fixed volume of air. However, measuring ambient air (with complex compositions) under real world conditions is likely more indicative of real-world performance than laboratory tests utilizing standardized particle types.

Results and discussion

Air purifier performance statistics

Air Purifier	Fan Speed	CADR (ft^3min^{-1}) Statistics								Electrical Power Draw (W)	Energy efficiency (CADR/W)	Noise (dB)	Noise rating (CADR/dB)
		Min	Max	Mean (s.d.)	Coefficient of Variation (%)	Median	N	R	S				
AP(Large)	High	130	440	316 (58)	18	330	6	564	64	60.5	5.2	51.4	6.1
	Medium	123	346	251 (35)	14	251	6	564	7	35.0	7.2	44.2	5.7
	Low	58	279	151 (36)	24	152	6	564	1	20.0	7.6	34.5	4.4
AP(Medium)	High	130	288	230 (23)	10	232	4	376	1	36.3	6.3	49.9	4.6
	Medium	75	221	154 (25)	17	161	4	376	1	16.1	9.6	40.2	3.8
	Low	57	112	95 (10)	11	98	4	376	2	6.9	13.8	N/A	N/A
AP(Small)	High	25.9	327	172 (52)	31	160	7	658	4	61.5	2.8	47.0	3.7
	Medium	98	308	155 (29)	19	156	5	470	1	45.2	3.4	37.3	4.2
	Low	34	79	55 (10)	18	56	4	376	23	16.6	3.3	34.5	2.3

Table 2. Air Purifier Statistics, R is the number of repeats, N is the number of decay series measured (R multiplied by the number of size bins = 94), S is the number of runs that failed to meet the selection criteria. The CADR values presented were averaged over the 94 size bins from 18-514 nm.

Our results show that larger APs and higher fan speeds generate larger average CADRs than smaller APs and lower fan speeds, as expected. This aligned with electrical power draw, which also increased with increasing AP size and fan speed. The coefficients of variation measured over 18-514nm were comparable to those of Waring et al. (2008), who measured 16% and 14% for the two HEPA APs tested. Table 2 also shows that the APs were most noise and energy efficient when running on their lowest fan speeds. On lower fan speeds AP (Medium) was substantially more energy efficient than AP (Large) or AP (Small).

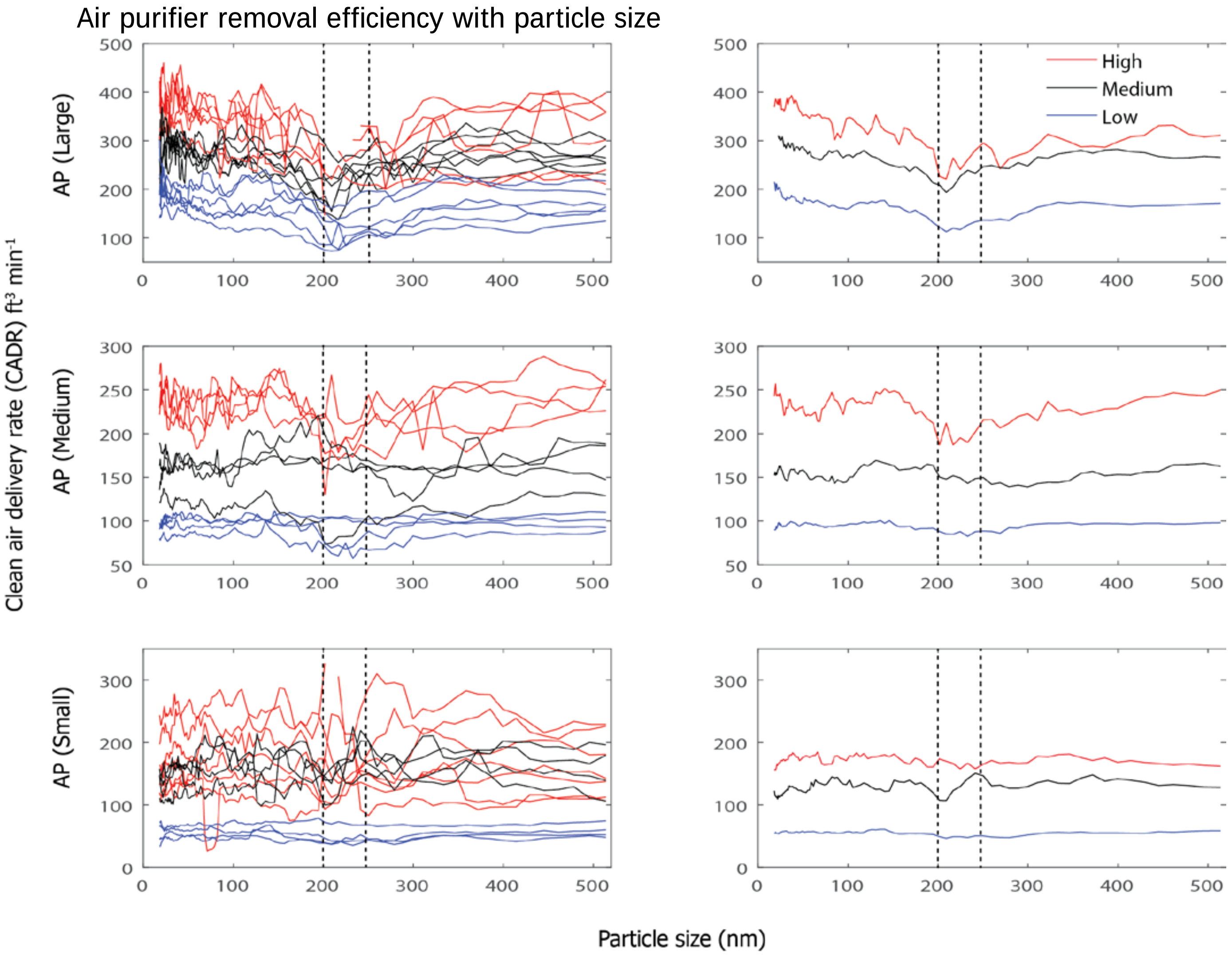

Figure 3. CADR as a function of particle size for three APs and for three fan speeds. Each line on the left plots represents a single AP decay series whilst the lines on the right plots show the average for each fan speed.

Figure 3 shows that AP (Large), (Medium) and (Small) are all effective at removing UFPs from ambient air. Each AP showed a distinctive removal profile which was consistent across the fan speeds, most likely attributed to the design of the HEPA filter and sealing. These profiles, although distinct, share some common themes. Generally, the APs performed least well between ~200-250nm, which is consistent with the understanding of the removal processes of HEPA filters (Kowalski et al., 1999; Stafford and Ettinger, 1972). This can be seen more clearly in Figure 4.

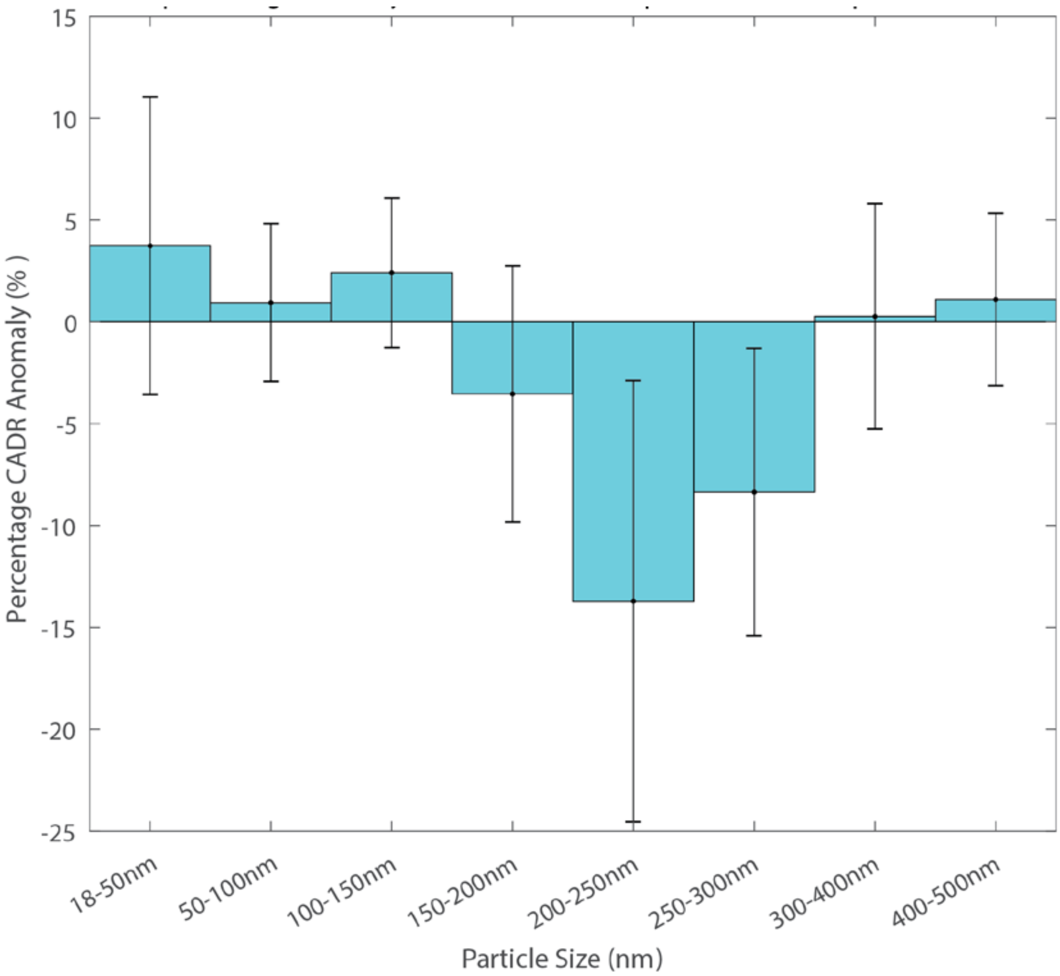

Figure 4. Percentage change in CADR relative to mean CADR for particle sizes between 18-500nm averaged over all tested air purifiers and fan speeds. Each value is the average value for the size bin. Percentage anomaly was calculated for the average of each of the APs for each given fan speed (n=9) and was divided into size bins. The standard deviation was calculated across the 9 arrays and is shown with the error bars indicating one standard deviation.

Figure 4 is consistent with the typical performance of a HEPA filter (minimum efficiency 200-300nm) (Kowalski et al., 1999; Stafford and Ettinger, 1972), and aligns with the understanding that diffusion primarily removes the smallest particles and that interception, inertial impaction and sieving primarily remove the largest particles, with particles in the intermediate size range (~100-400nm) least efficiently removed. However, this is contrary to the findings of Peck et al. (2016) who observed peak performance between 100-700nm and Sultan et al. (2011), Waring et al. (2008) and Lee et al. (2015) who observed lowest performance for particles <100nm and consistent performance above this. In Sultan et al. (2011) and Waring et al. (2008), these unexpected performances were attributed to non-uniform mixing in the chamber, with air flows short circuiting the APs and isolated flows forming due to particle size and flow dynamics. However, our results, based on the use of a Teflon reactor, may be more reliable than those generated in stainless-steel chambers, as our reactor was specifically designed to mix uniformly and reduce particle deposition. In addition, given that the Teflon reactor is more rounded than a stainless-steel chamber, this will promote mixing, reducing the likelihood of isolated flow-pathways forming. Waring et al. (2008) also attributed lower performances for UFPs due to particles within the APs bypassing the filter medium. Differences between our measurements and those of Waring et al. (2008) could be due to AP housing and filters being designed to be sealed more tightly during the past 10 years, in order to force particles through the filter medium. Alternatively, the subset of APs selected in this study could be especially well sealed; this may be linked to the bias towards selecting APs that were popular on the Chinese market and were therefore likely effective.

Given that the lowest removal efficiencies were observed within this 200-300nm range, it is worth considering the real-world importance of this size fraction. Firstly, these particles are relevant in a health context. Particles of <300nm can penetrate into the alveolar region of the lungs (Heyder, 2004) and pass into the circulation system, with particles <200nm being found in the brain (Maher et al., 2016) and it is thought that particles <240nm can cross the placental barrier, potentially impacting upon fetuses (Wick et al., 2010). Secondly, because the removal properties of building shells are similar to those of a HEPA filter, the particle size that most effectively penetrates cracks in building shells is ~200nm, similar to the 200-250nm for our HEPA APs (Hänninen et al., 2013; Liu and Nazaroff, 2001). Thirdly, the deposition velocity (m/s), the rate at which particles are deposited onto surfaces, is also lowest at ~200 nm which is consistent with the reactor deposition rates in this investigation (Lai, 2002). This means that particles within this size range can effectively penetrate building shells and will have longer airborne residence times, due to lower depositional velocities.

Particles within the 200-300nm size range are usually found at low concentrations in the atmosphere, typically falling between the Aitken (10-50 nm) and Accumulation (50-1000 nm) particle modes, subject to controls such as composition, humidity and turbulence. Irrespective, Cai et al. (2017) showed that there are still a significant number of particles found within this size range in Guangzhou, in fact, a second accumulation mode was observed with peak number concentrations within the 200-300nm range. Another investigation across 60 Hong Kong residences concluded that particles <400nm contributed the most to total particle mass (Chao et al., 2002). This is unusual, given that the smallest particles usually contribute the least to total mass measurements. The large concentrations of these particles in megacities could be attributed to secondary aerosols, vehicular and industrial emissions, which generate smaller sized particles (Zhang et al., 2018).

In summary, within Asian mega-cities, particles within the 200-300nm range are abundant in ambient air, can penetrate building shells effectively, can remain airborne for long periods, and are able to penetrate the deepest and most sensitive regions of the body. This means that the population are more likely to be exposed to particles of this size fraction than particles of other fractions in the indoor environment, which may have important health consequences. It is therefore important to note that HEPA APs currently are least efficient at removing this size fraction. It would be beneficial to design another filter media which could remove these 200-300nm particles without dramatically changing the pressure gradient across the filter medium.

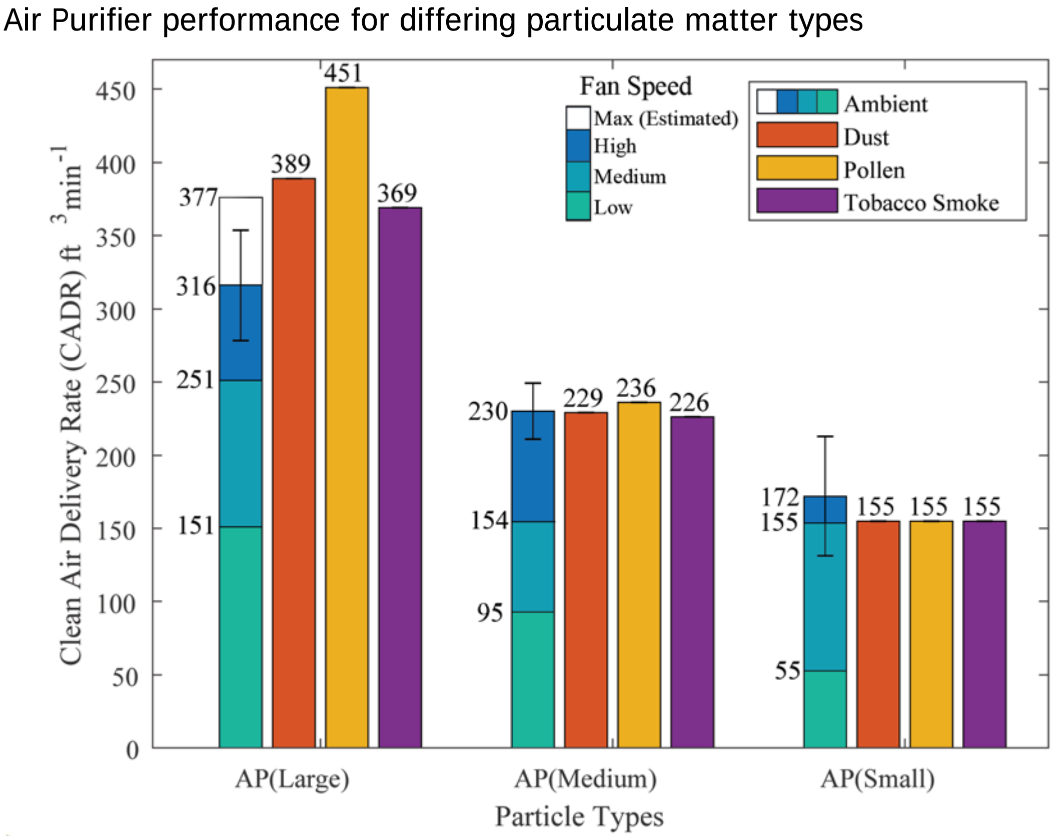

Figure 5. The CADR (ft^{3}min^{-1}) for different particle types for three APs. Ambient measurements collected in this study were compared against AHAM dust, pollen and tobacco smoke CADRs for the same APs. Error bars represent 95% confidence intervals around the means. For AP (Large), performance on maximum fan speed is estimated based on energy consumption ≈ fan rpm ≈ CADR Performance. Given that APs are only tested by AHAM at max speed, this should be used for comparison with AHAM measurements.

In this investigation, ambient particles, despite representing a smaller size fraction (18-514nm) than tobacco smoke (90-1000nm), dust (500-3000nm) and pollen (5000-11000nm), were removed with similar (or greater) efficiency than AHAM's standard particle types, as seen in Figure 5. Therefore, the AHAM standards appear indicative of how efficiently ambient air particles are removed by APs, and hence seem an appropriate proxy for "real world" AP performance.

Our results support Peck et al. (2016) who found that particles generated by diesel combustion were removed at a greater rate than AHAMs "standard" particle types. This similarity could be due to the strong influence of vehicular emissions (~20%) in ambient air in Guangzhou. Given the size of the diesel combustion generated particles used by Peck et al. (2016), and the ambient particles used in this experiment, we would expect them to be removed less efficiently than AHAM standard particle types. Peck et al. (2016) attributed this higher removal efficiency of diesel particles to differences in the measured size ranges between AHAM and those measured within their investigation. However, we hypothesize that this is likely due to smaller ambient and diesel particles having higher charge to mass ratios compared to tobacco, dust or pollen particles, which increases removal through the process of diffusion. As the filter media becomes more saturated with charged particles, this will more effectively remove particles with higher charge to mass ratios (Hanley et al., 1994).

Applying the relationship between particle size and AP removal efficiency as identified in Figure 5, we can estimate how efficiently different particle types commonly generated in indoor environments may be removed.

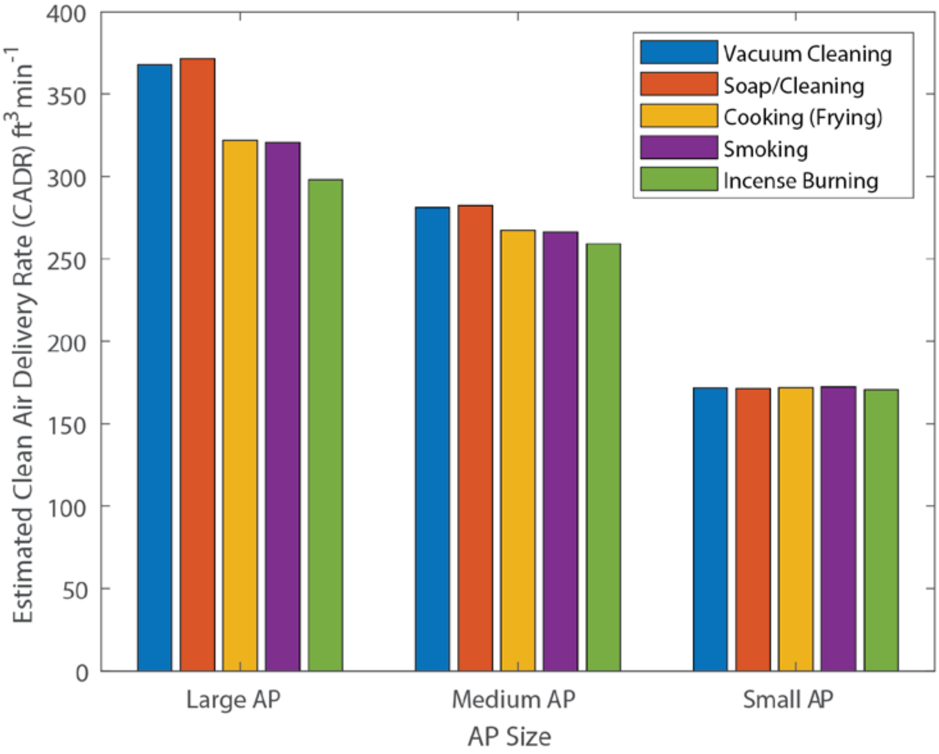

Figure 6. Estimated CADR values for different particle types for three APs running on high fan speed. The particle size distributions utilized to estimate CADR were adapted from Vu et al., (2017), assuming a log normal distribution of particle size generation. This estimation of CADR is based on particles being differentially removed based on particles size; it therefore does not account for other factors affecting removal, for example, particle shape, composition and electrostatic charges. Particle types are ordered in increasing mode particle size, with vacuum cleaning particles being the smallest and incense burning being the largest.

By applying PSDs for different particle types adapted from Vu et al. (2017), we can estimate CADR values for different particle types for each of our APs, as shown by Figure 6. For the largest AP, a 20% difference in CADR can be seen between the most and least efficiently removed particle types. The particle types with the lowest CADR scores were those with high particle number concentrations in the

200-250 nm range, where particles are least effectively removed. It is especially noteworthy that fry cooking, smoking and incense particles are removed less efficiently, given that these are common practices in Chinese households (Apte and Salvi, 2016).

Conclusions

Using the largest indoor smog chamber in China, this investigation aimed to determine (a) which particle sizes from ambient air were most and least efficiently removed by APs and explain how this may be important in a real world context and (b) whether ambient air particles were removed more or less effectively than AHAMs standard particle types.

This investigation found that although UFPs were effectively removed by each of the APs, a reduced removal efficiency was observed within the 200-250nm size range. This is important in a health context, with particles within that size range being present in significant concentrations in mega-cities (Cai et al., 2017), able to effectively penetrate the shells of buildings (Hänninen et al., 2013; Liu and Nazaroff, 2001), remain suspended (Lai, 2002), and penetrate into the deepest areas of the body (Heyder, 2004; Maher et al., 2016; Wick et al., 2010). Furthermore, this investigation found that ambient air particles were removed at a similar rate to AHAMs standard particle types, suggesting that these standards are representative of “real world” performance.

Further investigations should try to identify technologies which may improve the removal of 200-250 nm particles by HEPA filters without dramatically affecting the pressure drop. Additionally, it is necessary to understand the degree to which other properties of particles, apart from size, affect their removal rates. This could be used to further identify key particle types that may be important within a health context and which are more difficult to remove through filtering. Furthermore, some aspects of HEPA AP use should be explored, for example, how factors like AP placement, number of APs, rate of air exchange and mixing may influence AP performance within a residential setting.

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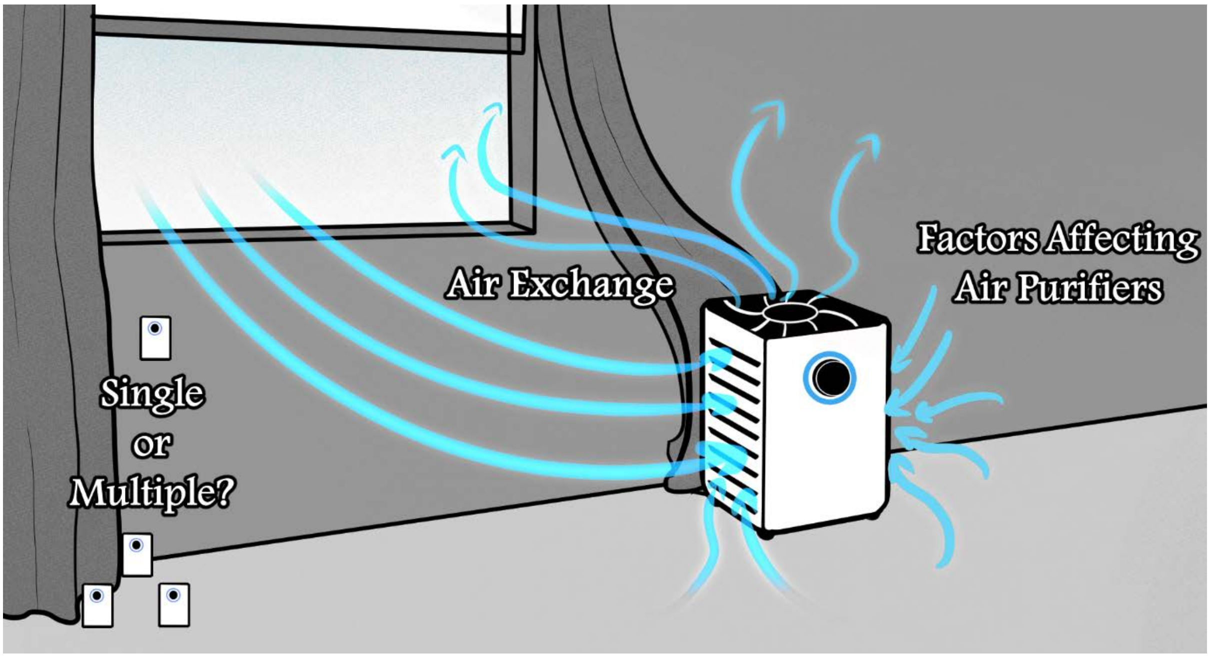
Chapter 4. Factors affecting real-world applications of HEPA purifiers in improving indoor air quality



Abstract

With modern populations spending ~90% of their time indoors, particulate matter (PM), a significant component of indoor air quality (IAQ), is of serious concern within indoor environments. High-efficiency particulate air (HEPA) filter technologies are commonly used to remove PM. Although their performance is well defined within a laboratory setting, many aspects of their real-world use remain poorly understood. This study investigated (i) the impact of air change rate on air purifier effectiveness, and how this influences energy-efficiency and other gaseous components of indoor air quality, and (ii) the relative effectiveness of operating single and multiple air purifiers within a multi-room residence. Measurements of air change and PM concentrations made in an Asian mega-city apartment, were used alongside air purifier performance data and external PM measurements to create a box model to simulate air purifier performance under different scenarios. Increasing air change rate inhibited the performance of air purifiers by acting as a source of outdoor PM into the indoor environment. Although sealing indoor environments is recommended to maximize the removal of PM, this permits the accumulation of gaseous components of IAQ and reduces energy efficiency. Use of multiple air purifiers in a multi-room residence reduces PM at a greater rate than use of a single more powerful air purifier.

Moreover, use of multiple air purifiers is more energy-efficient, although the maintenance and upfront costs are likely to be greater.

Introduction

It is well established that modern populations spend ~90% of their time indoors, with household air pollution being the seventh-largest global risk factor to disability-adjusted life years.[1,2] Particulate Matter (PM), the sum of all liquid and solid particles suspended in air, is a major determinant of indoor air quality (IAQ)[3] and is associated with a range of adverse health effects including myocardial infarction, stroke, heart failure, asthma, chronic obstructive pulmonary disease (COPD) and lung cancer.[4]

Rapid reductions of ambient (outdoor) PM concentrations are being observed in China. For example, a ~30% drop in population-weighted annual mean $PM_{2.5}$ concentrations was reported between 2013 and 2017.[5] However, PM is still of concern given that in 2017, 73% of 338 Chinese cities failed to meet China's national air quality standards for $PM_{2.5}$ and PM_{10}.[6] Moreover, 66-87% of total exposure to $PM_{2.5}$ of outdoor origin occurs within indoor environments. This exposure contributed up to three-quarters of total premature mortalities in urban China in 2015.[7]

Indoor PM originates from within the indoor environment or penetrates inwards from outdoors. Typical indoor sources of PM include cooking, smoking, cleaning and burning incense or candles.[8] When ambient PM concentrations are low, indoor sources are the main determinant of indoor PM concentration, and because of the confined nature of indoor environments, these concentrations can be raised to several orders of magnitude higher than ambient concentrations.[3] However, in the absence of indoor PM sources, indoor concentrations correlate strongly with outdoor concentrations.[9,10] Therefore, in Chinese mega-cities where ambient pollution is often severe, the impact of outdoor sources on indoor PM is of concern. Although PM is especially important within a health context, a holistic view of IAQ must also consider gaseous components, for example, carbon dioxide, VOCs, nitrogen dioxide, and sulfur dioxide.[11]

Air change rates (h^{-1}) are a measure of how many times the air within a room is replaced within an hour. Air change can be an important control on indoor PM; for example, when ambient concentrations are lower than indoor concentrations, air change can reduce indoor PM; conversely, when ambient concentrations are higher than indoor concentrations, air change can increase indoor concentrations. In more economically developed countries, reducing air change rates to improve the energy efficiency of buildings is becoming increasingly common. However, this has implications for IAQ, allowing some pollutants to accumulate to much higher concentrations.[3] Conversely, in areas with high ambient concentrations, such as Chinese mega-cities, reducing air change rates by sealing indoor environments can be beneficial to IAQ.[3]

High-efficiency particulate air (HEPA) type air purifiers, hereon referred to as APs, are valuable for reducing PM concentrations within indoor environments.[12] It is well established that the use of HEPA APs is associated with considerable reductions in PM concentration.[13–16] These reductions are associated with modest improvements in health outcomes.[17] Health improvements are most consistently observed within homes in Asian mega-cities, likely due to significant rates of ambient PM ingress, and therefore more significant reductions in PM.[15] Air purifiers are soon to become the fifth largest-selling home appliance in China, with sales of APs increasing from 112 million units in 2011 to 982 million in 2017.[18] With the burgeoning domestic use of air purifiers in China likely to further increase, understanding their real-world performance is essential.

Research into APs removal of PM under controlled conditions is extensive, and intervention studies are improving the understanding of APs effects on health.[17] However, recommendations on how APs should be operated in real-world conditions are not clearly defined.[12,17] Therefore, it is necessary to better understand how technical (e.g., runtimes, noise, maintenance, filter changes) and practical aspects (e.g., AP positioning, the quantity of APs, air change rates) of APs impact their effectiveness.

Within existing literature, air change is often considered a removal mechanism of PM.[19] However, this is the perspective of more developed countries, where indoor concentrations typically exceed ambient concentrations.[10] In megacities with high levels of ambient PM pollution, where air purifiers are most commonly used, ambient PM concentrations often exceed indoor concentrations, making air change a source of PM, rather than a sink.[20] Therefore, it is necessary to understand the effect of air change on AP performance and PM removal; to assess the advantages and disadvantages of decreasing air change in Chinese residences, and the implications of this on the gaseous components of IAQ.

Furthermore, it is unknown how effective a single AP is in reducing PM spatially throughout a residence, given the barriers to mixing presented by walls, doors, and furniture. Conversely, the benefits of having multiple APs deployed throughout a residence are also poorly defined.[12]

Therefore, this investigation aims to determine (i) how air change affects the efficiency of AP use and the implications of this on a holistic view of IAQ and energy-efficiency and (ii) how effective single and multiple APs scenarios are in reducing PM in a multi-room residence. This is investigated in a multi-room residence in Guangzhou China and is applied in a broader context using modelling.

Methodology

Description of the study location and approach

The investigations were conducted in a typical residential apartment located within the Guangzhou Institute of Geochemistry's Campus, in Guangzhou, China. The layout of the apartment is illustrated in Figure 1. The two-bedroom, third story, seventy-five square meter, the non-occupied apartment was lightly furnished, with furniture and flooring being mostly wooden, typical for a residential apartment in south China. The investigation area used within the experiments was forty-five square meters. The investigation area is a smaller segment of the whole apartment, because it needed to be of an appropriate size to be cleaned by the largest AP alone (less than fifty-three square meters).

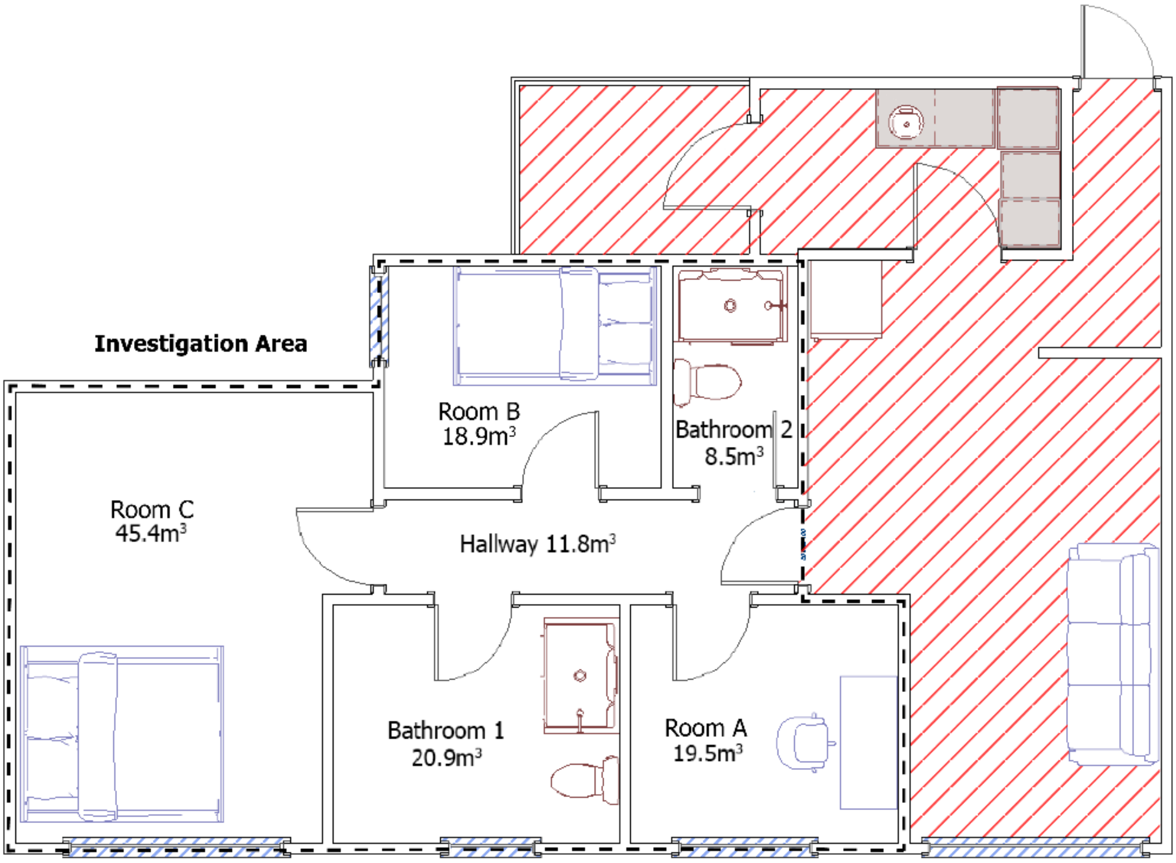


Figure 1. Floor plan of the investigation apartment; a two-bedroom, third story, seventy-five square meter apartment in Guangzhou, China. The investigation area (contained within the dotted line) marks the section of the apartment used within these experiments and the room height was 2.8 meters.

The investigation consisted of two experiments. Experiment 1 aimed to determine how air change rates affect the effectiveness of APs in removing PM from indoor air. This is designed to inform whether inhabitants should aim to reduce air changes in indoor environments when using an AP, but also considers the effect this will have on the gaseous components of IAQ. Experiment 1 uses a combination of measurement and modelling components.

Experiment 2 aimed to determine how effectively a single AP can clean an apartment of an appropriate size given barriers to mixing like walls, doors and furniture, and how this compared to operating multiple air purifiers throughout the apartment.

Instrumentation and Measurements

PM size-resolved number concentrations from 18-514nm were measured in the residence using a TSI SMPS (Classifier model 3082, CPC model 3775) with a full scan completed every minute. For both experiments, the SMPS was located in Room A and sampled air from three locations, namely, Room A, Room B and Room C. Sampling throughout the apartment was necessary to understand the spatial distributions of PM in the residence. The PM loss rates of the TIS conductive tubing taking samples from the rooms to the SMPS unit were accounted for by sampling air from the same room with each of the three lengths of hose, and then calculating the difference between measurements made by the longer hoses vs having no hose. During the experiments, the SMPS inlet hose was switched at 5-minute intervals, to sample from different locations throughout the apartment. An example time series, showing how it is divided into multiple rooms, is illustrated in Figure 2. AP performance was measured as total particle number decay rate, with increasing decay rate being associated with improved AP performance.

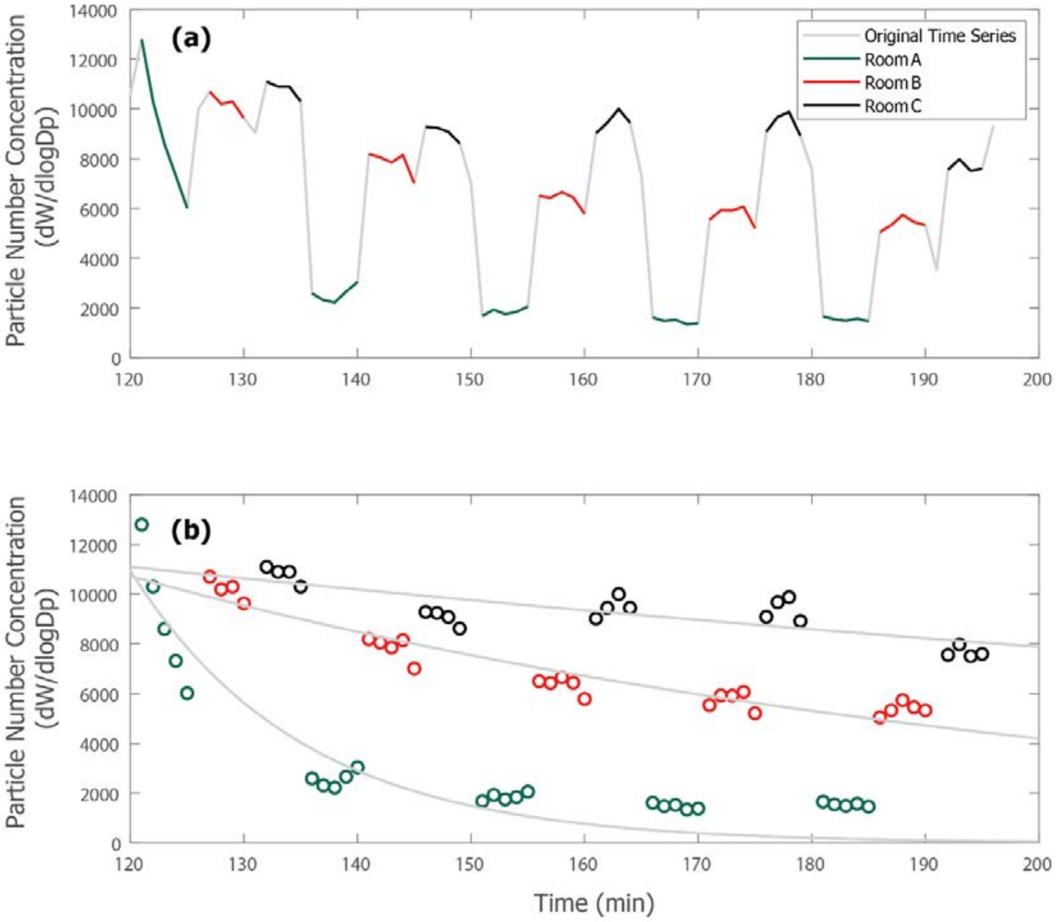


Figure 2. Measuring particulate matter decay across three rooms with a HEPA purifier active; by switching the inlet hose on the SMPS at five-minute intervals. **(a)** Shows an example time-series and how this varies for three rooms and **(b)** shows an example exponential fit from which a rate of decay can be calculated.

Three popular HEPA type air purifiers on the Chinese market were used for these experiments, these are referred to as AP large, AP medium and AP small and are described in detail in Lowther et al.

(2020)[12]. The HEPA filters had been used previously in Lowther et al. (2020)[12], but only for several hours under ambient conditions, so filter loading was minimal.

The measurements for Experiment 1 and 2 were conducted in May and June of 2019.

Experiment 1

The effect of air change on AP performance is largely unknown within the context of mega-cities with higher levels of ambient PM. Therefore, this investigation measures the effect of different air change conditions on the performance of three air purifiers within a real-world indoor environment. Modelling was conducted to give a better understanding of how changes to air change rate might be important when considering a holistic view of IAQ.

Three ventilation scenarios were selected to test in each room: sealed (windows, doors and air conditioning closed); air conditioning on, and windows open. These ventilation scenarios were selected as they were available for each of the measurement rooms. Air change rates were quantified for each of the three rooms, for each of the ventilation conditions by using the CO_2 decay method.[21] Briefly, CO_2 was released from portable canisters and was mixed using fans, then, CO_2 decay was measured using the NAQTS V2000.[22] Air change rates displayed within Table 1 were the average of three decays for each of the nine scenarios.

During these experiments, every room was ventilated until indoor PM concentrations matched ambient PM concentrations. Then, the desired ventilation condition was set up and finally, the AP was activated. Once particle concentrations had reached equilibrium, the air purifier was switched off, the room was ventilated, and the next repeat began. The point of equilibrium was determined between the competition of two processes, i.e. ventilation, the source of PM and the AP, the sink of PM.

Experiment 2

Although commercially available APs are given a recommended room size,[23] when considering the barriers to mixing which are present within most multi-room residences it is largely unknown to what degree they can reduce PM throughout the dwelling. Therefore, this study assessed the advantages and disadvantages of operating single and multiple APs within a multi-room residence.

For the single AP scenario, AP large was operated on its maximum fan speed and was placed in Room A. At maximum fan speed, this AP is rated to clean a 53m^2 area and was therefore suitable for use within the 45m^2 investigation area. For the multiple AP scenario, Room A, Room B and Room C contained the small, medium and large APs respectively, each running on their lowest fan speed. The Clean Air Delivery Rate (CADR) of the two scenarios was chosen to be roughly equivalent with the single AP and multiple AP scenarios outputting 316 and 301 ft^{3}min^{-1} respectively.[12] The multiple AP scenario is more energy-efficient, consuming 43.5 W compared to 69.7 W for the single AP scenario.[12]

Before the experiments, the investigation area was ventilated with air from outdoors until indoor PM concentrations matched outdoor concentrations. Next, the investigation area was sealed, with windows closed and air conditioning turned off, to minimize infiltration of outdoor PM. During these experiments, doors within the investigation area remained open, to facilitate mixing between rooms.

Model Development

The laboratory-determined performance of APs, room volumes and measured air change rates were supplemented with externally sourced data to develop a box model of how PM is affected by APs and air exchange. These values are displayed in Table 1. The model accounted for two processes: PM removal by the AP, and PM exchange with the outdoors. The model simulated the behaviour of an independent room and therefore did not account for air change between rooms. The box model is described by the following equations.

1. $M = C_{t-1} * \exp{(-CADR * \Delta t / V)}$ (PM removed by AP)
2. $C_t = M + (A - Ct) * \exp{(ach * \Delta t)}$ (PM exchanged with outdoors)

Where M = Model output, C_t = concentration at time t, C_{t-1} = concentration at previous time step t-1, V = Room Volume (ft^3), CADR = clean air delivery rate (ft^{3}min^{-1}), Δt = time step (min), A = Particle concentration of ambient air and ach = air exchange rate (min^{-1}).

A simpler model using source terms and ventilation rates (outlined in Table 1) was used to model gaseous components of IAQ, as seen in Figure 7 and 8.

Table 1. Model parameters and their respective sources.

Model Parameters		
Air change Parameters (h^{-1})		
Parameter	Value	Source
5th Percentile of annual infiltration rate of 294 Chinese residences	0.08	Hou 2019[24]
25th Percentile of annual infiltration rate of 294 Chinese residences	0.22	Hou 2019[24]
50th Percentile of annual infiltration rate of 294 Chinese residences	0.34	Hou 2019[24]
75th Percentile of annual infiltration rate of 294 Chinese residences	0.56	Hou 2019[24]
95th Percentile of annual infiltration rate of 294 Chinese residences	1.12	Hou 2019[24]
Room A (AC on)	0.55	Measured
Room A (Sealed)	0.30	Measured
Room A (Window Open)	1.64	Measured
Room B (AC on)	2.30	Measured
Room B (Sealed)	0.40	Measured
Room B (Window Open)	1.68	Measured
Room C (AC on)	2.22	Measured
Room C (Sealed)	3.04	Measured
Room C (Window Open + AC)	8.31	Measured
Room C (Window Open)	6.10	Measured
CO_2 Model Parameters		
Average Apartment Size	39 m^2	Chinese Bureau of Statistics 2019[25]
Model Bedroom Size	15 m^2	N/A
Model Kitchen and Living room Size	7.5 m^2	N/A
Breathed Volume	6 L min^{-1}	Carroll 2007[26]
CO_2 Concentration in Exhaled air	38000 ppm	CO_2 Meter[27]
Cooking Model Parameters		
PM Generation Rate	24.7×10^{10} #/s	Zhao 2018[28]
Total VOC (TVOC) Generation Rate	2.14 mg/m^3/min	Zhao 2014[29]
Other Parameters		
Air Purifier CADRs	Various	Lowther 2020[12]
Outdoor Background PM Concentration	1.0092×10^4 #/cm^3	Measured
Indoor Background PM Concentration	6.614×10^3 #/cm^3	Measured

Results and Discussion

Study location background information

The effect of air change on AP performance is a function of the PM concentrations in the ambient air penetrating inwards. Therefore, it is necessary to understand the likely concentrations and composition of PM in ambient air outside the apartment and its variability. In the presence of no dominant indoor sources, the average indoor concentration was 6900 #/cm^3 compared to 11500 #/cm^3 outdoors, (see Figure 3). Guangzhou ambient PM$_{2.5}$ in the wet season (when this campaign was conducted) was composed of secondary organic aerosol (21%), sulphate (18%), primary organic aerosol (14%), nitrate (10%), ammonium (8%), elemental carbon (5%) and unidentified fractions (25%).[30] Key sources of wet season Guangzhou PM$_{2.5}$ include; vehicular (37%), industrial (32%), power generation (12%), residential (7%) and indistinguishable emissions (12%).[30] However, these data were collected in 2013 and compositions may have changed since then. It is important to note that the majority of PM$_{2.5}$ is generated by vehicular sources, which predominantly generate particles <500 nm.[31]

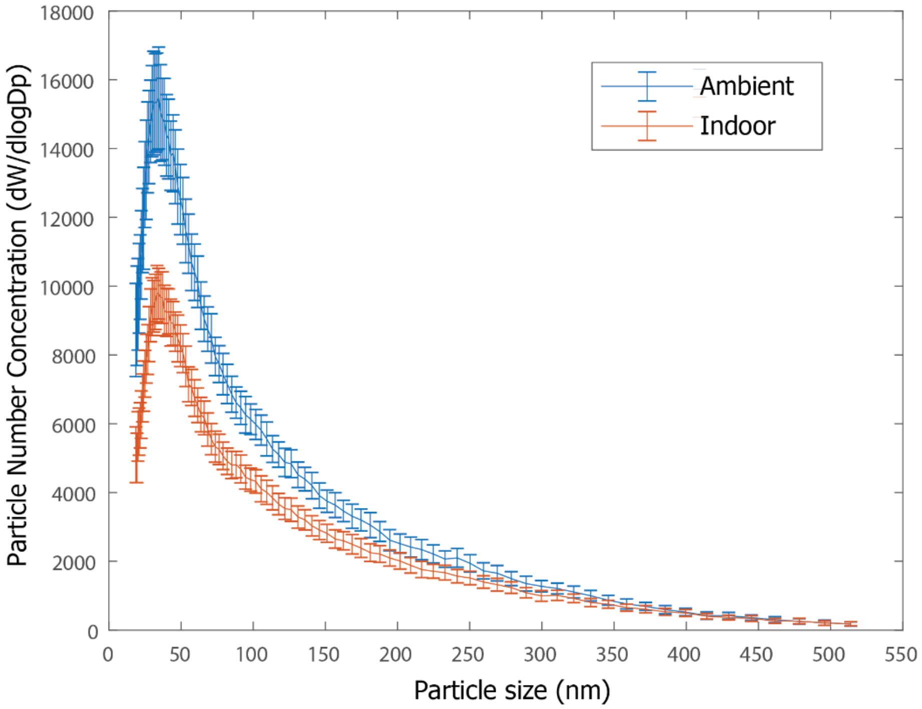


Figure 3. PM size distributions of ambient and indoor air from 18-514nm collected by a TSI SMPS, over 12 hours (2 hours per day), with the measurement between ambient and indoor switched every 20 minutes, on six days of the sampling campaign. dW/dlogDp units display particle size distributions normalised to one decade of particle size.

Experiment 1

Generally, increased air change rate is associated with decreasing particle decay rate and therefore decreased AP performance, as seen in Figure 4. For each of the three rooms, the performance of APs is least effective under the window open ventilation scenario. It should be noted that some of the measured air change rates appear unusual, particularly the greater air change in Room C when sealed than with the AC on and the greater air change in Room B with AC on than with the window open. These experiments were conducted in an urban megacity context where ambient PM often exceeds indoor PM, and hence increasing air change can act as an additional source of PM and reduce the effectiveness of air purification.

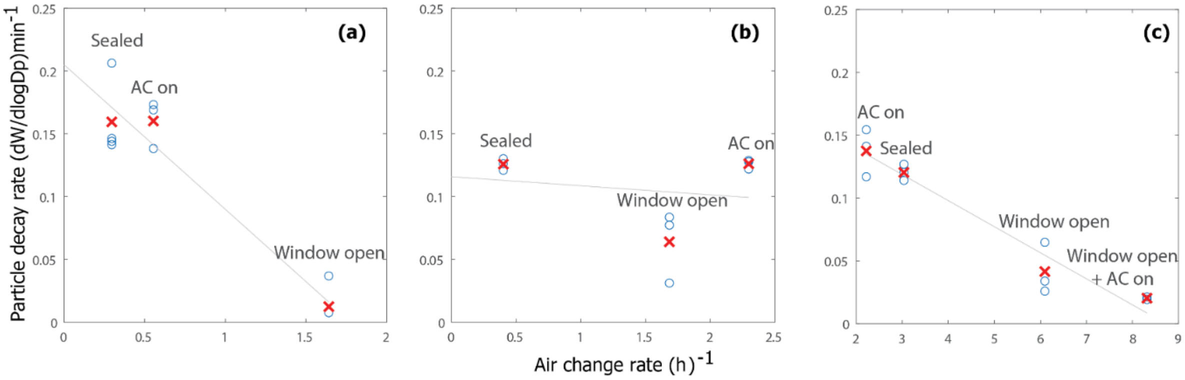


Figure 4. The measured relationships between air change and particle decay rates for three rooms under the different ventilation scenarios; **(a)** Room A, **(b)** Room B and **(c)** Room C. The blue circles and red cross represent the repeats and the mean respectively.

Particle decays when the AP was operated under the different ventilation conditions are shown in Figure 5. Both the modelled and measured outputs show that increasing air change rate decreases the rate of net particulate removal (and therefore the APs performance). In addition, the air change rate appears to determine the equilibrium particle number concentration achieved. Therefore, at lower air change rates, the AP performs more efficiently, and a lower equilibrium concentration is reached. The model appears consistent with what was measured and this would indicate that our understanding of processes within the indoor environment is reasonable.

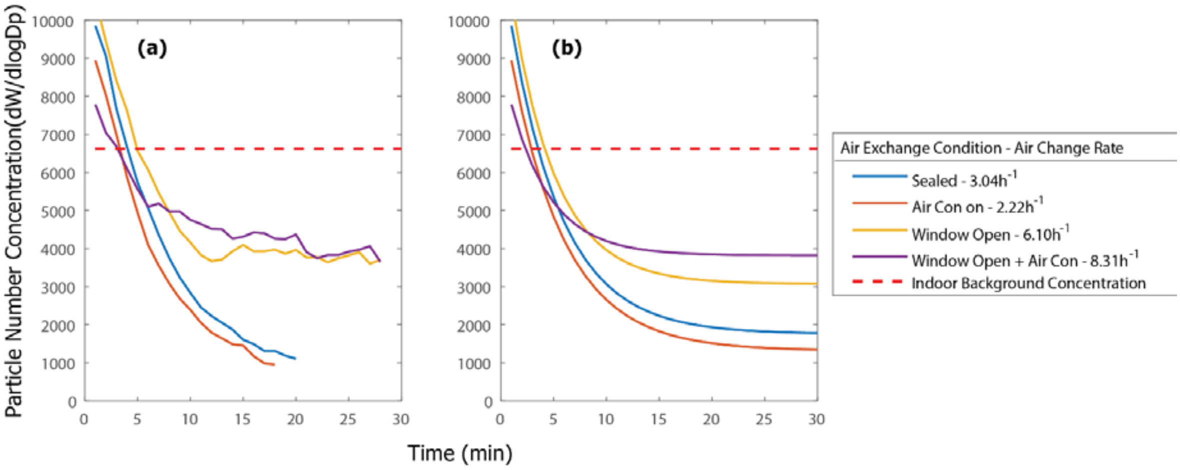


Figure 5. (a) Measured and **(b)** modelled total particle number decays when an air purifier is active in Room C under four ventilation conditions.

Figure 6 shows how the decay of PM due to AP use changes under different ventilation conditions when indoor PM concentrations are initially greater than outdoor concentrations (i.e. when a dominant indoor source of PM is present). Figure 6 demonstrates that when indoor PM concentrations exceed ambient PM concentrations, as is the case initially, then air change acts as a sink of PM, removing PM from the indoor environment. Conversely, when indoor PM concentration is less than ambient PM concentration, air change inhibits the performance of the air purifier, leading to decreased particle decay rates and increased equilibrium PM concentration, as for the measurements shown in Figure 6. Looking at total PM exposure (area under the curve) for each of the ventilation scenarios in Figure 6, it seems that when PM is of concern, and when there are limited indoor sources of PM, sealing the environment is the best strategy for reducing exposure. Although sealing the environment initially removes PM at a slower rate (when ambient PM>indoor PM), this effect is small compared to the benefits of increased removal rate and decreased equilibrium concentration when indoor PM>ambient PM. If indoor and ambient PM concentrations could be quantified in real-time, as may be possible in a future smart home,[32] ventilation could be automatically controlled to maximise the removal efficiency of PM. However, until then it is likely best to seal the environment and allow the AP to remove PM with limited inhibition.

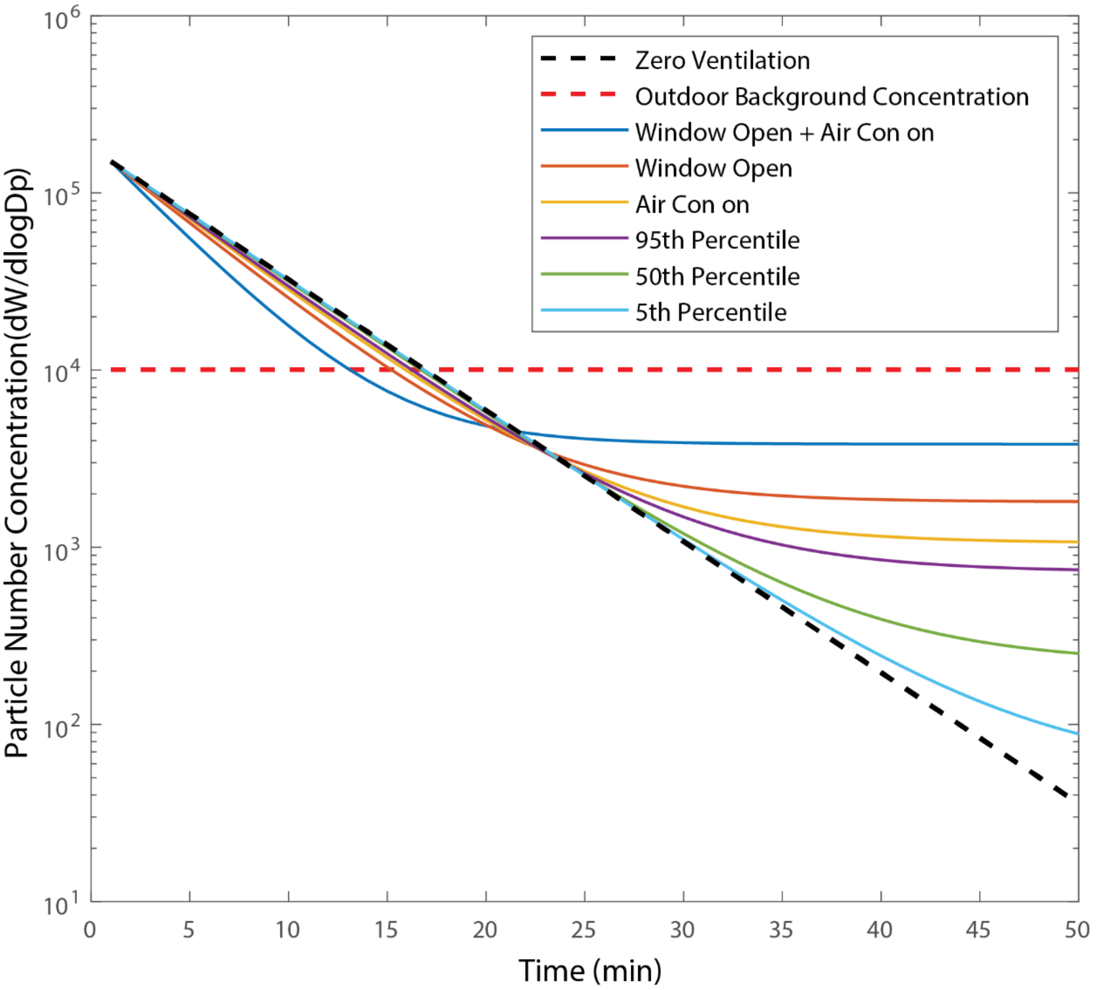


Figure 6. Modelled total particle number decays when the Large AP is active on medium fan speed in an average-sized Chinese bedroom under five ventilation conditions. The initial concentration is significantly greater than the ambient background to simulate a potent indoor PM source. The window open and air-con on, air change rates used were averages for measurements across the three rooms. The percentiles are of annual infiltration rate of 294 Chinese residences as in Hou et al, 2019.[24]

When PM is of primary concern, and there are limited sources of indoor PM, sealing the environment and allowing the AP to reduce PM with minimal inhibition seems like the most logical strategy. However, this may not be coherent with a holistic view of IAQ. Therefore, it is also necessary to consider the effect of this sealing on the gaseous components of IAQ.

Figure 7 considers the impact of sealing on the accumulation of CO_2 from breathing within a 'sealed' bedroom overnight. It demonstrates that by the end of the night, concentrations can be 4-10 times greater than the background. It is thought that CO_2 concentrations greater than ~1000 ppm can affect concentration and comfort,[33] with the 8-hour time-weighted-average exposure limit value to CO_2 being

5000 ppm.[34] Within an average-sized Chinese bedroom with average air change, and a single occupant, CO_2 concentrations can reach ~2000 ppm overnight.

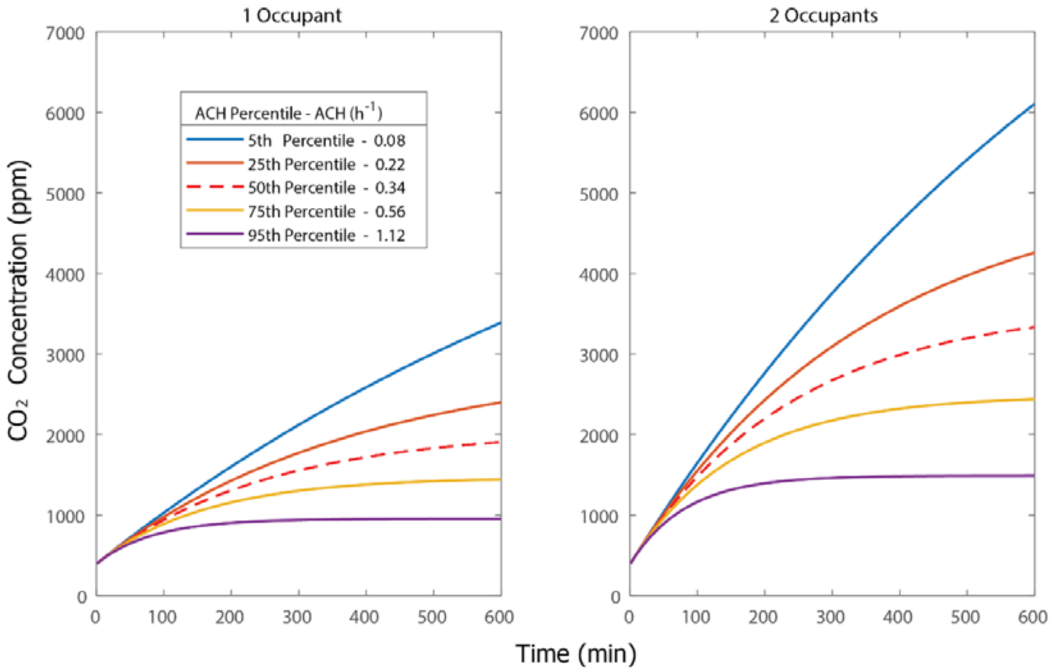


Figure 7. Carbon dioxide accumulation from one and two occupants breathing in an average-sized Chinese bedroom over ten hours, for different air change conditions. The percentiles are of annual infiltration rate of 294 Chinese residences as in Hou et al, 2019.[24]

Figure 8 shows how different ventilation conditions will influence the removal of PM and total volatile organic compounds (TVOCs) after a cooking event. Regardless of the ventilation condition, PM is reduced to an acceptable level within ~50 minutes. However, the ventilation condition does define the equilibrium concentration reached, as outlined in Figure 4 and 5. TVOCs, a gaseous component of IAQ, which are therefore not removed by the AP are not reduced to acceptable levels within the 3-hour duration, regardless of the ventilation strategy. This demonstrates that although ‘sealing’ indoor environments can be beneficial for the removal of PM, this will have clear implications on the accumulation and decay of gaseous components of IAQ.

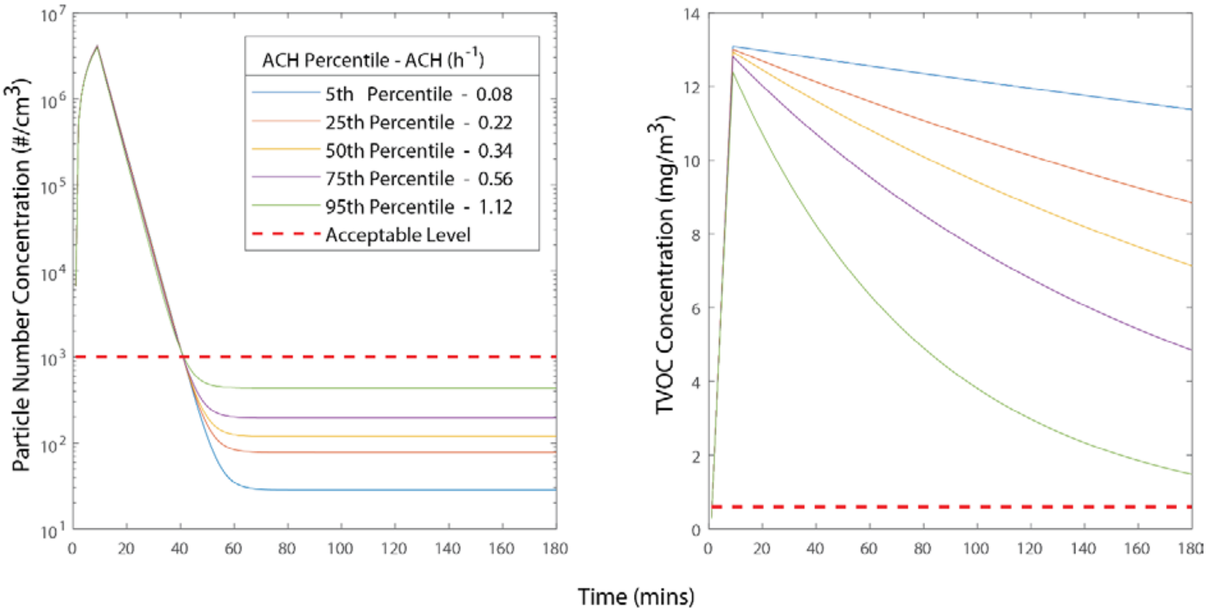


Figure 8. Particle number concentration (#/cm^3) and TVOC concentration (mg/m^3) over time for simulation of a ten-minute Chinese style stir-frying event in a 21m^3 room; after the ten-minutes of cooking small AP is turned on at maximum fan speed. This is simulated for different air change conditions. The acceptable TVOC concentration is 0.6 mg/m^3.[35] The acceptable level of PNC is set as 10^3 #/cm^3, defined as "relatively clean" by Lowther et al[3] and Bo et al[36].

Experiment 2

Table 2 compares the effectiveness of single and multiple AP usage within a two-bedroom residence.

Table 2. Particle decay rates and percentage particle reductions over two hours for the three measurement rooms for the single AP and multiple AP scenarios.

	Single AP scenario				Multiple AP scenario			
	Decay rate min^{-1}				Decay rate min^{-1}			
	Repeat number				Repeat number			
	1	2	3	Mean	1	2	3	Mean
Room A	0.0272	0.0155	0.0291	0.0239	0.0311	0.0264	0.0248	0.0274
Room B	0.0108	0.0113	0.0143	0.0121	0.0318	0.0335	0.0297	0.0317
Room C	0.0045	0.0118	0.0046	0.0069	0.0219	0.0164	0.0187	0.0190
	Percentage reduction per hour				Percentage reduction per hour			
	Repeat number				Repeat number			
	1	2	3	Mean	1	2	3	Mean
Room A	80.4	60.5	82.5	74.5	84.5	79.5	77.4	80.4
Room B	47.8	49.2	57.7	51.6	85.2	86.6	83.2	85.0
Room C	23.5	50.6	24.0	32.7	73.2	62.6	67.4	67.7

Under the single AP scenario, Room A was cleaned most efficiently, as this was where the AP was located. The reduction in PM decreased for Room B and Room C as the distance away from the AP increased. For the multiple AP scenario, where one AP was located in each room and was operated on the lowest fan speed, ~70-80% reductions in PM can be seen consistently across the three rooms. Therefore, not only is it more energy-efficient to run multiple APs on lower modes than it is to run a larger AP on maximum fan speed, it is also more efficient in removing PM. It should be noted that, although a single AP was able to reduce PM across the investigation area, this was with all the doors open, which often will not be the case in an occupied residence, especially whilst sleeping, when AP use is arguably most important. Under a scenario where all the doors were closed, the improvements from a multiple AP scenario would likely be significantly greater than from a single AP scenario. Even within typical Chinese residences which are comparatively small, a single AP is likely insufficient to clean the area it is rated for, given the barriers to mixing that are present within residences, i.e. walls, doors and furniture. Therefore, a better strategy would be to have APs located in any room where extended periods are spent. This would also be beneficial for energy consumption, being more efficient than running a single AP on greater fan speed. However, the upfront cost of multiple APs will be greater.

Conclusions

Using a combination of measurement and modelling, this investigation aimed to determine the effect of air change on AP effectiveness and the benefits of using single or multiple APs within a multi-room residence.

Firstly, when ambient PM concentrations are greater than indoor PM concentrations, increasing air change rate inhibited the effectiveness of AP use, by acting as a source of PM into the indoor environment. The rate of air change determined the equilibrium concentration reached in the indoor environment, once the sink of PM (AP) and source of PM (air change) became balanced. Conversely, when indoor PM concentrations are greater than ambient PM concentrations, i.e. when there are strong sources of PM indoors, then air change can act as a sink of PM. However, this sink is negligible compared to the sink of the AP. Therefore, when PM is of major concern within the indoor environment, and when there are minimal indoor sources of PM, it seems logical to seal the environment as much as is possible, to minimize the inhibition of the AP. However, this allows for the accumulation of gaseous components of IAQ, which are not removed by the HEPA filter. Additionally, in contrast to many developing countries, where sealing of buildings is becoming increasingly prevalent as a method to improve energy efficiency,[3] in China, increasing natural ventilation is suggested to be beneficial to improving energy efficiency.[38] Therefore, there are many conflicting factors when suggesting an optimal air change rate. Future work could determine to what extent a "smart home", where concentrations of various pollutants can be measured in real-time, and where air change can be adjusted accordingly, can reduce exposures to PM.

Secondly, the use of multiple APs within a multi-room residence can reduce PM concentrations more than the use of a single AP on higher fan speed. Therefore, in a multi-room residence, it is beneficial to have APs located in any room where significant time is spent. Although the upfront and maintenance cost of APs is greater, utilizing multiple APs is also more energy-efficient than operating a single more powerful AP on higher fan speed. Future work could determine where an AP should be placed in a room relative to furniture to maximise its performance.

This investigation used measured and modelled data to determine that increasing air change inhibits the performance of APs. Although, sealing is beneficial in reducing exposure to PM, this conflicts with concentrations of gaseous components of IAQ and improving energy efficiency in Chinese buildings. Using multiple APs in a multi-room residence can reduce exposure to PM and have a lower energy consumption when compared to the use of a single AP.

Further work should investigate holistic AQ solutions, where internet of things (IoT) sensors make changes to purification and ventilation in real-time, to optimize IAQ.

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Chapter 5. Conclusions

PM measurement uses three key metrics, Pmass, Pnum and Psize. Pmass is a robust, low-cost measurement, and is easy to compare and benchmark against other Pmass measurements. The Pmass measurements of PM_{10} and $PM_{2.5}$ represent the most established and recognized PM measurements currently. Although Pmass provides a useful indication of the abundance of PM, it is thought that the total mass of suspended particles may not be as useful an indicator of health effects as some other metrics. Pnum, the number of suspended particles in air, is suggested to be more important in a health context, as it is disproportionately weighted by fine and ultrafine particles, which are thought to have a greater impact on health. Therefore, with Pnum measurement becoming increasingly technologically accessible, this is likely to become more prevalent in the future. However, Pnum measurements are disadvantaged by the difficulty in making reliable comparisons between them. Psize measurements, although extremely useful, in the short term (10-20 years) are likely to remain used largely only in a research capacity, as technology remains expensive and inaccessible. In the future, metrics that better link PM to health effects, such as surface area, particle length concentrations or compositional measurements, are likely to become more important, as better measurement technologies are developed.

Under controlled conditions, HEPA type air purifiers are effective at removing PM from indoor air. HEPA type air purifiers were found to be effective at removing ultrafine particulate matter (<100nm diameter), and coarser particles (>500nm), and were found to be least effective at an intermediate size (200-300nm) where the processes that removed the smallest particles (diffusion) and largest particles (interception, inertial impaction and sieving processes) were least effective. This is an important finding, as particles of this size can penetrate building shells effectively, reside airborne for extended durations, can enter the circulatory system, and can be found in human organs.

Air purifiers can work effectively in residences, but several factors affect their performance. Firstly, in environments where ambient PM exceeds indoor PM, ventilation will act as a net source of PM into the indoor environment. As ventilation rate increases, the performance of air purifiers is reduced. In this scenario, minimizing ventilation will improve the performance of the air purifier, reducing total exposure to PM. However, this may lead to the unintended consequence of the accumulation of gaseous IAPs e.g. CO_2, NO_2 and VOCs. Secondly, a single air purifier, although rated to clean an area comparable to a Chinese residence, will not be able to effectively mix air throughout the residence and therefore, will not be suitable to clean the whole flat area. However, using multiple air purifiers can clean a residence much more effectively, whilst having a lower energy consumption. So with the understanding that air purifiers are only most effective in reducing PM within the room they are located, if only a single air purifier is available, this should be located in the room where the residents spend the most time, to maximize the reduction in total exposure to PM.

This thesis outlines the complexity of measuring PM, with different metrics and sensors available, and the complicated relationship between the properties of PM and health impacts. It defines a need to better understand the relationship between PM properties and health endpoints, as this will determine the most important metrics, and which particle types require focus, whether it be in reducing the sources or developing effective filtration tools. It identifies HEPA as a useful real-world tool in reducing PM but recognises a size bin (200-300nm) which is removed least effectively and is important within an IAQ and health context. Although APs are effective in theory, this work identifies that there are several factors that influence their performance, and it is important to raise awareness of this, as small changes may lead to significant reductions in total exposure. Finally, it describes the characteristics necessary for a holistic IAQ monitor, a tool that will be of increasing importance in the coming decades.

Reflections on the IAQ field

The conflict between IAQ and energy efficiency is likely to increase as governments are pressured to achieve climate targets. However, awareness of IAQ is also increasing and this is reflected in some building codes, with the number of air changes mandated to improve occupant comfort and indoor environmental quality. Striking a balance between these two important environmental issues is difficult.

The metrics most commonly used to measure PM may see a shift as measurement technologies and focuses change. Pmass is likely to remain useful as a tool for comparing PM concentrations against historical measurements or differing environments. However, this may be considered less important as other characteristics of PM (surface area, size, composition etc.) are likely to be considered more indicative of potential health impacts than mass measurements. As particle number, surface area and compositional measurements improve and become more widespread, this will, in turn, improve our currently limited epidemiological understanding of PM exposure. It is suspected that some particle types are more harmful to health than others, i.e. combustion particles more so than pollen particles, and understanding these relationships is essential, as it will allow for the direct targeting of particularly potent and harmful sources. As all particles may not be equal in their health impact, targeting several important sources may reduce health outcomes much greater than the reduction in total Pmass might suggest.

Low-cost sensors are becoming increasingly viable as a tool to collect real-world data. In controlled conditions, sensors can measure with high precision and accuracy, however, these sensors often drift and are sensitive to temperature and humidity interferences. Currently, this means that low-cost sensors ideally need to be in a temperature-controlled housing and calibrated regularly to work effectively. With low-cost sensors, mathematical tools like rolling averages and adjusting data based on temperature are important to yield useful data.

Low-cost sensors allow for high spatial density measurements, which can be used to understand the distribution, movement and behaviour of IAPs. Currently, spatial models of air pollution are used to understand the spatial distribution of air pollutants across cities, to identify which areas perform worst, and where improvements are needed. Low-cost sensors may be an alternative to this method, with high distributions of sensors being able to measure at regular intervals across a city, with interpolation made between points. If low-cost sensor accuracy improves, this may be a higher cost, but also a more reliable and detailed air quality mapping tool. Alternatively, these may be used more effectively in tandem, with sensors used to feed models and test the outputs.

With low-cost sensors becoming increasingly cheap and available, more low-cost sensing bundles are coming to market, Earthsense Zephyr, AQ mesh, Dyson backpack and NAQTS V2000 etc. However, as the sensing components are becoming smaller and lower cost, it is no longer the cost of sensors that determines the overall cost of development or efficiency. Instead, the development of data adjustment algorithms and temperature/humidity adjustments, which will determine the overall effectiveness of a sensor. Put succinctly, much of the low-cost sensing field is becoming more of a software battle, as the hardware is usually third-party, low-cost and accessible to all.

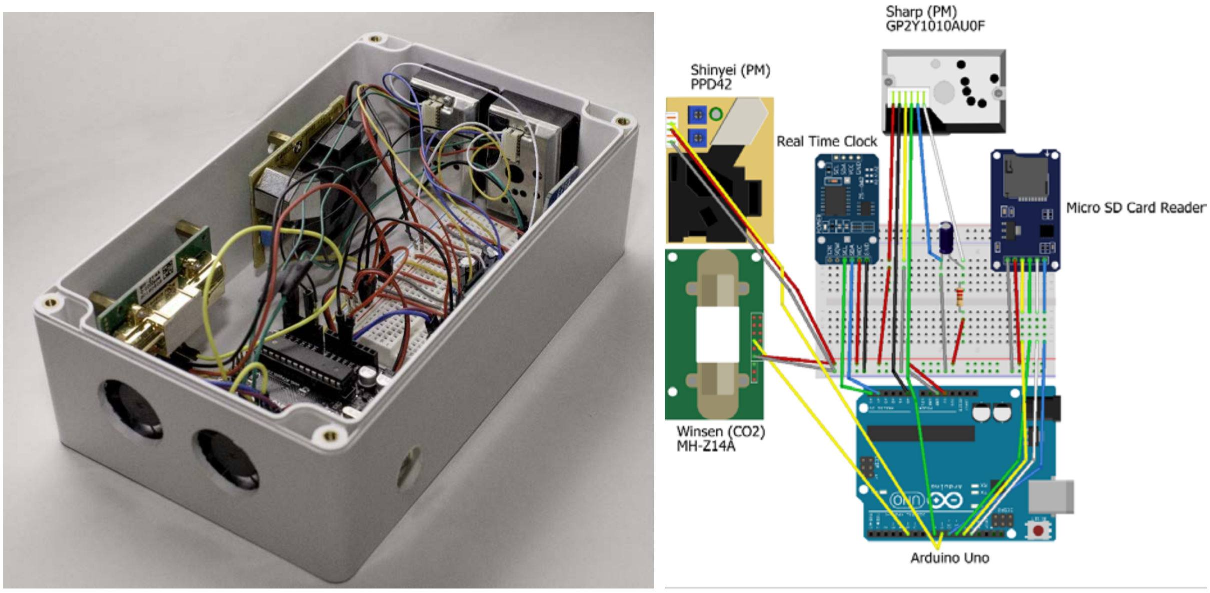

Figure 3. A low-cost sensor developed by the author during the PhD process.

It appears likely that IAQ low-cost sensors will be increasingly seen in future “smart homes”, with the emergence of home products with in-built IAQ sensors (e.g. air purifiers, heaters and humidifiers) and a general increase in the public awareness of air quality. In the next 30 years, most homes will likely have some “internet of things” IAQ sensors, and this will be connected with the ability to change ventilation rates (open or close windows, change extractor fan speed, etc) and control appliances (air purifiers, heaters and humidifiers etc) (Blanco-Novoa *et al.*, 2018). Each home may have an internal (IAQ) and external (ambient) air quality sensor, and this will define how ventilation is controlled, to maximise air quality. With smart homes already able to measure energy efficiency, this will likely be a known variable, with homes able to optimise for improved IAQ, energy efficiency or a balance between the two.

With smaller and lower cost sensors becoming increasingly available, there is also likely to be an increase in the collection of personal exposure data, where people are carrying sensors on their person all day. This wealth of personal AQ exposure data may be important in informing health research and setting appropriate guidelines for recommended exposure levels.

Indoor air quality is likely to be considered increasingly important in China, as ambient levels are consistently dropping over time, making IAQ more important in comparison. As the biggest sources of ambient pollution are targeted first in a “low hanging fruit” fashion, in China, residential sources of air pollution are likely to become more important in defining ambient air quality. By this point, certain cooking or indoor combustion methods may be restricted to universally improve indoor and ambient air quality in China.

Recommendations for future research

It is poorly understood how individuals operate air purifiers in real-world residential environments, how this affects their performance, and how much this reduces total exposure to PM. Further research should aim to determine how individuals are using air purifiers, and develop guidance on how to use them most effectively, to maximise reductions in personal exposures to PM. Although HEPA type air purifiers are effective in removing PM, further research may enable the development of filters with higher efficiencies in the 200-300nm size range. Additionally, further chemical filter types are being researched to facilitate the removal of some gaseous components of IAQ, for example, formaldehyde.

With ADMS Urban spatial modelling of air pollutants across cities becoming increasingly accurate, and with IAQ in many mega-cities being driven largely by ambient air quality, it may be useful to estimate in which areas residents are exposed to the greatest degree. For countries with information on building types, ages and energy-efficiency classifications (information that may allow for rough estimations of air change rates), it may be possible to estimate IAQ levels on a city scale. However, this would only be an estimate of baseline IAQ (without indoor sources considered) but may indicate where residents in a city may be exposed to the greatest degree.

With PM thought to be of high health relevance, and with various properties of PM being able to be measured with increasing accuracy, it is necessary to research the properties of PM, and how they may affect health. This could inform which measures of PM are most important and focus the conversation away from mass measurements like PM2.5 and PM10 (which may be less indicative of health outcomes), and towards alternative metrics of particle number, surface area or composition.

With the technology now available for air quality adaptive smart homes, something that is likely to join the current “internet of things” ecosystem, it is necessary to understand how beneficial this technology may be in reducing exposure to IAPs. Firstly, experimenting with indoor and ambient air quality sensors in the home, and adapting ventilation based on the ratio of indoor to ambient air quality, and secondly, controlling air purifier systems remotely based on real-time air quality measurements may determine how effectively adaptable systems can reduce exposures to pollutants. In the future, these techniques may be indispensable in improving indoor air quality, energy efficiency, health and wellbeing.

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