Particulate matter measurement indoors: a review of metrics, sensors, needs and applications

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11 Abstract

12 Many populations spend ~90% of their time indoors, with household particulate matter being linked to 13 millions of premature deaths worldwide. Particulate matter is currently measured using particle mass, 14 particle number and particle size distribution metrics, with other metrics, such as particle surface area, 15 likely to be of increasing importance in the future. Particulate mass is measured using gravimetric methods, 16 tapered element oscillating microbalances and beta attenuation instruments and is best suited to use in 17 compliance monitoring, trend analysis and high spatial resolution measurements. Particle number 18 concentration is measured by Condensation Particle Counters, Optical Particle Counters and Diffusion 19 Chargers. Particle number measurements are best suited to source characterization, trend analysis and 20 ultrafine particle investigations. Particle size distributions are measured by gravimetric impactors, Scanning 21 Mobility Particle Sizers, Aerodynamic Particle Sizers and Fast Mobility Particle Sizers. Particle size distribution measurements are most useful in source characterization and particulate matter property investigations, 22 23 but most measurement options remain expensive and intrusive. However, we are on the cusp of a 24 revolution in indoor air quality monitoring and management. Low-cost sensors have potential to facilitate 25 personalized information about indoor air quality (IAQ), allowing citizens to reduce exposures to PM indoors 26 and to resolve potential dichotomies between promoting healthy IAO and energy efficient buildings. Indeed, 27 the low cost will put this simple technology in the hands of citizens who wish to monitor their own IAO in 28 the home or workplace, to inform lifestyle decisions. Low-cost sensor networks also look promising as the 29 solution to measuring spatial distributions of PM indoors, however, there are important sensor/data quality, 30 technological and ethical barriers to address with this technology. An improved understanding of 31 epidemiology is essential to identify which metrics correlate most with health effects, allowing indoor 32 specific PM standards to be developed and to inform the future of experimental applications.

33 1. Introduction

Poor indoor air quality (IAQ) has been estimated to be the 9th largest global burden of disease risk,¹ with 34 the World Health Organization (WHO) attributing >4.3 million premature deaths to household air pollution 35 36 in 2012, compared to 3.7 million deaths attributed to ambient (outdoor) air pollution.² The Institute for 37 Health Metrics and Evaluation attributed 2.57 million premature deaths to household air pollution in 2016,³ 38 with Asia, Africa, Europe and the Americas contributing 74%, 23%, 1% and 2% respectively.⁴ Particulate 39 matter (PM), the sum of all solid and liquid particles suspended in the air, is a major metric of IAQ. PM is 40 strongly associated with myocardial infarction, strokes, heart failure, asthma, chronic obstructive 41 pulmonary disease and lung cancer.⁵ PM <200nm in diameter has also been observed in the brain, and this 42 may be causally linked to neurodegenerative diseases, for example, Alzheimer's disease.⁶ It is well 43 established that modern populations on average spend >90% of their time indoors, and on a daily basis 44 indoor air contributes 19-76% of an individual's exposure to particles <100nm in aerodynamic diameter.^{7,8} 45 Brunekreef⁹ has summarized 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 behavior 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.

50 Atmospheric PM is generally measured using two main metrics; particle mass concentration (Pmass) and 51 particle number concentration (Pnum). Pmass is the mass of particles within a given volume (usually µg/cm³) 52 and Pnum is the number of particles within a given volume (particles/cm³). However, there are other 53 metrics that provide valuable information on the nature of atmospheric PM. Another important characteristic 54 of PM is the particle size distribution (Psd); this is the particle concentration (either mass or number) 55 measured over a range of different particle sizes. The chemical composition of particles can also be 56 measured, although chemical composition is not covered in detail by this paper. It is still largely unknown 57 how each of these metrics relate to health effects and therefore which metric is the best suited to measuring health risk. 58

59 The challenges associated with measuring PM in the indoor environment are distinctly different to those of 60 measuring PM in the outdoor environment. Indoor environments can have much more variable PM levels 61 than outdoors,¹⁰ in absolute terms and over small distances. PM levels can be much lower than outdoors 62 when there are no dominant indoor sources; however, in the presence of indoor sources, PM levels can 63 rapidly rise to several orders of magnitude greater than outdoor levels. The confined nature of indoor 64 environments allows PM to accumulate, but can also lower PM below ambient concentrations through 65 removal of particles during building shell penetration. With pressure placed on developing energy efficient 66 buildings with low heat losses and therefore low air exchange rates, accumulation of indoor generated PM pollution is likely to be an increasing problem.¹¹ This means that PM monitoring equipment used indoors 67 68 needs to be able to accurately measure PM over a wide range of concentrations. Indoor particle events are 69 frequently time and space specific; brief, intermittent and highly variable.⁸ This means that high spatial and 70 temporal resolution measurements are necessary to further understand the controls and influences on PM. 71 High temporal resolution measurements are common practice, with any "real time" measurement 72 instrument providing sufficient resolution in most cases; however, spatially varying measurements are less 73 common with campaigns rarely deploying multiple sensors within a single indoor environment.

74 There are many other practical issues associated with measurement of indoor PM. There are constraints of 75 size, noisiness and intrusiveness placed on equipment. For example, in an occupied school classroom it 76 would be inappropriate to deploy large noisy equipment. This is much less of an issue with outdoor 77 measurements. Some of the most severe indoor air pollution (IAP) is experienced in middle and low-income 78 homes in both rural and urban areas of less economically developing countries (LEDCs), often with high 79 levels of ambient pollution and use of coal or biomass stoves. Many of these locations lack access to 80 electricity, and hence, measurements must be made with either battery-operated devices or passive 81 samplers. The increasing focus on indoor PM has driven researchers to develop smaller, lighter, inexpensive 82 battery-operated sensors. However, developing sensors that are sufficiently sensitive when measuring over 83 a large range of concentrations and size ranges of PM is challenging.

A wide range of technologies and sensors has been 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, 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.

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

96 a. The sensor must operate within sensible ranges for the criteria listed above, whilst considering
97 their applications.

98 b. There must be evidence that the sensor has been used to measure PM within indoor environments,
99 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 more than one of the metrics listed. Indoor measurements are usually collected in the center of the environment, at 0.75-1.8m height representing the breathing zone, with a height of 1.5m recommended.¹²

104 Current developments in PM monitoring are primarily influenced by PM sensors that have traditionally been 105 used in occupational health and regulatory compliance monitoring, and developments in miniaturization & 106 wireless technology. Low-cost, portable sensors, capable of measuring high temporal resolution 107 concentrations have been at the forefront of meeting the rapid increase in public awareness and interest 108 in quantifying personal exposure to IAP. It is important to consider how these sensors may play a role in

measuring IAQ specifically.¹³ Many of the sensors now being 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 categorized by their measurement method.

Oscillating microbalance, beta attenuation, and low-pressure impaction measurement methods are not included within this review; they are less commonly used indoors and more practical alternatives are often available. Detailed descriptions of these methods are outlined by this international standard.¹⁵

118 2. Review of Metrics

119 In the atmosphere there are three broad particle modes; the fine/nuclei mode ($<0.1\mu$ m), the accumulation 120 mode (0.1-2.5 μ m) and the coarse mode (>2.5 μ m); in short, these are the consequence of physical 121 processes such as emission, nucleation, accumulation and scavenging. Pmass is usually measured as PM_{2.5} 122 or PM₁₀ and this is the mass of all particles with an aerodynamic diameter of $\leq 2.5 \mu m$ and $\leq 10 \mu m$, 123 respectively. PM₁ is also increasingly 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 PM₁₀ and PM₁ are arbitrarily 124 125 selected as size cut-offs out of convenience, whereas PM_{2.5} is selected more purposefully to include 126 accumulation and fine particles (which remain suspended for longer), but to exclude the coarse particles,

127 which are deposited more rapidly. These cut-offs are also indicative of historic progress; for example, the 128 UK Department of Environment, Food and Rural Affairs (DEFRA) introduced PM₁₀ and PM_{2.5} in 1997 and 129 2007, respectively, as sensing technologies and understanding of PM improved. Other measurement ranges 130 include total suspended particulate (TSP), a measurement of the mass of all particles present in the air, 131 and inhalable, thoracic and respirable size fractions, used in health-related sampling. The inhalable, thoracic 132 and respirable fractions represent particles that can enter the respiratory system through the mouth and nose (50% penetration efficiency (D_{50}) at 100 μ m), pass through the larynx and enter the bronchial region 133 134 of the lungs ($D_{50}=10\mu$ m) and enter the deepest part of the lungs, the ciliated alveoli ($D_{50}=4\mu$ m), respectively.¹⁷ It should be noted that these definitions were developed in a workplace exposure context. 135 136 The EPA¹⁸ provide a useful visualization of the sizes of PM_{2.5} and PM₁₀ and Nazaroff¹⁹ outlines the sources, 137 compositions and behaviors of different particle size fractions in indoor environments.

138 Particle Mass

139 Pmass is the most commonly used metric to measure PM; it is the easiest metric to measure and can be 140 measured accurately at a relatively low cost. Due to the characterization as PM₁₀, PM_{2.5} and PM₁, it is 141 convenient to draw comparisons between different indoor environments and measurements collected by 142 different equipment. Easy comparisons allow us to develop appropriate Pmass concentration standards. 143 Currently there are no universal standards for Pmass specifically for indoor environments. The WHO 144 recommends outdoor Pmass guidelines for $PM_{2.5}$ of $10\mu g/m^3$ (annual mean) and $25\mu g/m^3$ (24 hour mean) and for PM₁₀ of 20µg/m³ (annual mean) and 50µg/m³ (24 hour mean).²⁰ However, these standards are not 145 indicative of where adverse health effects begin to occur (for PM_{2.5}, this is currently understood to be just 146 above background concentration at 3-5µg/m³).²⁰ Rather, they are the lowest levels at which "total, 147 148 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 149 150 specific indoor Pmass standards.²¹ They concluded that because there was no significant difference 151 between the hazardous nature of PM in indoor and outdoor environments, and because indoor PM levels 152 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.

160 Particle Number

161 As measurement technologies have advanced and as focus moves towards the importance of ultrafine 162 particles (UFPs, particles \leq 100nm in aerodynamic diameter), Pnum is becoming an increasingly used metric 163 for measuring PM within indoor environments. Although the Pnum metric is being used increasingly in 164 outdoor and indoor PM studies, it was originally used to characterize vehicular exhaust emissions.²² Unlike 165 Pmass, Pnum varies by several orders of magnitude - from <10³ particles/cm³ in relatively clean environments to $>10^6$ particles/cm³ when there are dominant sources of indoor pollution.²³ In contrast to 166 167 Pmass, Pnum is mostly made up of smaller particles. For example, Pnum concentrations have been 168 measured to be two orders of magnitude greater within the $<0.5\mu$ m size range than in the $0.5-18\mu$ m size 169 range.²⁴ This means that when fine or UFPs are the focus of a study the Pnum metric is often employed. 170 Currently there are no standards for Pnum in indoor or outdoor environments,²⁵ likely due to the relatively 171 recent adoption of ambient Pnum measurements, the highly variable nature of the Pnum metric, and the 172 difficulty of comparing studies measuring Pnum. Epidemiologists have suggested that Pnum is a more important predictor of health impacts than Pmass.^{26,27} This is because Pnum better represents the smaller 173 174 particle size fractions, which penetrate further into the respiratory system, potentially causing more damage.28 175

176 Particle Size Distributions

Understanding the Psd in indoor environments is important for several reasons. The size of particlesdetermines 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.²⁹ 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.

182 The Future of Ultrafine Particle Metrics

183 There is currently much debate on what should be the flagship metric for the measurement of UFPs. The 184 metrics mainly considered are Pnum, surface area, "active" surface area, and particle reactivity (which 185 relates to chemical composition). There is general agreement that the metric should provide insight into 186 how PM interacts with the body through intake, uptake and transport. Therefore, there is some consensus 187 that a surface area-related metric should be introduced, since these better correlate with the biological and 188 toxicological activity of particles than either Pmass or Pnum.³⁰ However, there are concerns over whether 189 it is plausible to achieve accurate measurements with relatively simple equipment. It seems likely that the 190 UFP maximum size cut-off will be moved from 100nm to between 200-500nm and that diffusion chargers 191 may be the most effective way of measuring the proposed "active" or "lung deposited surface area (LDSA)".³⁰ Measuring compliance or exposures using the Pmass and Pnum metrics can be of limited use, 192 193 given that physiochemical properties of particles give rise to varying levels of toxicity and this toxicity is 194 also an important determinant of health effects. Instead, identifying and guantifying the sources generating 195 the most reactive species of UFPs is suggested.³⁰

196 The Future of Epidemiological Metrics

197 There is strong agreement that future metrics should provide more information on the health impacts of 198 particles than just physical properties. Particle length concentration and active or geometric surface 199 concentrations might be more indicative of a particle's effect on health than Pmass or Pnum.³¹ Whereas 200 Pnum takes no account of particle size, particle length concentration is Pnum multiplied by the diameter of 201 particles within a given size range. The geometric surface area concentration equals the particle number 202 concentration multiplied by the particle's diameter squared, within a certain size range. Although the 203 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.³² These may eventually

205 become more important metrics for PM in epidemiological studies.

206 3. Characterization of Measurement Approaches and Sensors

207 Here, principles of measurement techniques and sensors associated with each PM metric are briefly outlined,

and references to more detailed descriptions are provided.

209 Particle Mass

210 Pmass is measured using gravimetric and optical methods.

211 Gravimetric methods are based on weighing a filter sample before and after a sampling period and 212 calculating Pmass from the difference in weight. Filters have a collection substrate on which particles of all 213 sizes are deposited, unless there is a cyclone or impactor used to remove larger particles. Conventionally 214 this is active sampling, with a pump pulling air through the filter at a known flow rate, however, passive 215 samplers that do not require a pump have also been developed.³³ Passive samplers are much lighter, 216 smaller and less noisy than pump-operated active samplers, but must be deployed for longer times to be 217 effective and – depending on the design - can be influenced substantially by wind speed and particle size. 218 The University of North Carolina (UNC) passive sampler, has been shown to correlate well against active 219 samplers within indoor environments.³⁴ After a sample has been collected, it can be examined using an 220 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.^{33,35–37} This 221 222 method uses a scanning electron or optical microscope and automated image analysis. Unfortunately, 223 gravimetric sample analysis is labor intensive and cannot be used to measure UFPs (<0.1µm) which are 224 too small to be observed. There are also inherent errors in particle size and surface area measurements 225 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 analyzed to determine an accurate concentration. With gravimetric samplers the chemical composition of particles can be acquired using instrumental neutron activation analysis (INAA), particle-induced X-ray emissions (PIXE) or other processes, although this can be time-consuming, expensive and specialized.

234 Impactors can separate particles based on their inertia, and this allows for measurement of a mass-based 235 size distribution. The most commonly used type of Impactor is the Cascade Impactor, in which particles 236 flow through a series of sections (typically 3 to 15),¹⁵ each containing an impaction plate. In each section, 237 particles above a certain size cut-off are deposited onto the impaction plate. Between each stage the 238 diameter of orifices decreases, which leads to increased velocity of the aerosol and impaction of 239 progressively smaller particle sizes. The Cascade Impactor is made up of several stages followed by a final 240 filter, which collects any particles that were not deposited in any of the previous stages. Conventional 241 Cascade Impactors cannot size particles <400nm, however some Low-Pressure Cascade Impactors can size 242 particles from 30nm upwards, for example, the electrical low pressure impactor (ELPI) which can provide 243 size distributions in real-time.^{15,38} The collection substrates can be removed from the Cascade Impactor 244 and are processed in the same ways as standard gravimetric filters.

245 Optical methods are based on the interactions of particles with light. When light hits a particle, it is either 246 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.³⁸ 247 248 Scattering Laser Photometers measure the intensity of scattered light in one or more directions using a 249 photometer detector; the combined intensity of scattered light is directly proportional to the volume 250 concentration of the aerosol within the optical volume.³⁹ Photometers collect real time measurements with 251 a frequency of 1s, and measure particles from ~40-100nm upwards, however, measurement efficiencies 252 are significantly lower at smaller particle sizes. They are commonly used with impactors or cyclones to 253 measure PM1, PM2.5, PM10 or the respirable aerosol fraction and are extremely popular due to their small, 254 portable, robust and reliable nature. Photometers can measure accurately over a larger range of Pmass

concentrations (0.001-200 mg/m³), making them suitable for both clean and highly polluted environments.
 Examples of commercial photometers include the DustTrak, DataRAM 4 and UCB-PATS.

257 Gravimetric samplers are more accurate than optical methods since they measure PM directly rather than 258 indirectly. Therefore, at the start of campaigns, optical Pmass sensors are often co-located with gravimetric 259 samplers to be calibrated.⁴⁰ Optical measurements vary depending on the optical properties of particles, 260 and therefore do not provide absolute mass concentrations.⁴¹ However, there is a significant trade-off as 261 gravimetric filters are labor intensive and not real-time. When using gravimetric filters to measure Pmass, 262 the samples need to be dried and weighed and then chemical testing requires additional labor. In addition, 263 the process of drying the filters can remove the more volatile compounds, affecting the mass measurement. 264 Collecting data on Pnum, Psd, particle shape, particle surface area and particle structure requires use of a 265 scanning electron microscope, however, automation has made this process less labor intensive.

266 Particle Number

267 Pnum is measured using optical and current methods. Optical Particle Counters (OPCs) work similarly to a 268 scattering laser photometer with a diode laser shining on the optical volume with the scattered flash being 269 measured by a photodetector; unlike photometers, only one particle is illuminated at once. The 270 photodetector converts the flash of light into an electrical current, and as each electrical current 271 corresponds to a different particle, the number of particles can be counted. The size of the particle is 272 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.³⁸ For example, the Alphasense OPC 273 274 can count particles with a diameter of 0.35-<40µm and place them into 24 size categories, with a sampling 275 frequency of 1s upwards. Other OPCs include the TSI Optical Particle Sizer 3330 and GRIMM OPC. OPCs 276 are light, portable, rugged and quiet, however their main disadvantage is their inability to count particles 277 <300nm. Particles smaller than this cannot be counted by the optics, hence are grown to a size where they 278 can be counted in the CPC (described below). Particles <300nm in size make a substantial contribution to 279 total Pnum concentrations.

280 Condensation Particle Counters (CPCs) share similar operating principles to OPCs but can count particles of 281 much smaller sizes. CPCs condense a solvent (typically butanol, isopropyl alcohol, or more recently water) 282 onto the surface of particles to grow them to a size where they can be counted. Once grown, the particles 283 pass through the focal point of the laser beam and are individually counted. Vapor around the particles 284 needs to reach a certain degree of super-saturation before condensation occurs, and the magnitude of this 285 super-saturation determines the minimum countable size of the CPC. There are two types of CPCs; Full 286 Flow CPCs (sometimes called Continuous Flow Laminar CPCs) and Mixing CPCs (sometimes called Fast 287 CPCs). In Full Flow CPCs the aerosol is drawn through a conditioner where it is saturated with vapor and 288 brought to thermal equilibrium. The aerosol then passes into a cooler growth tube where the liquid is 289 condensed onto the surface of particles. Full Flow CPCs require higher regulation of temperature control 290 than other CPCs. In aerosol research the Full Flow CPC is the most commonly used due to its robust and reliable nature,⁴² however, they have a relatively low sampling frequency (several seconds) due to zones 291 292 of recirculation and time needed to establish super-saturation.

293 Many of the atmospheric processes measured by CPCs are rapid and therefore there is a need to develop 294 a CPC capable of higher frequency sampling. Mixing CPCs were developed in the 1980s to increase the temporal resolution of measurements.⁴³ In a Mixing CPC, a cold aerosol flow is mixed with a warm saturated 295 296 gas flow; this dilutes the aerosol flow, allowing the CPC to cope with the dynamic range of indoor Pnum. 297 When this was designed it was able to achieve mixing times as fast as 0.6s, 10 times faster than any 298 commercially available full flow CPC.⁴² Therefore, Mixing CPCs are sometimes referred to as Fast CPCs. One 299 of the main uses of a Mixing CPC is as part of a Scanning Mobility Particle Sizer (SMPS) (described below), 300 which measures particle size distributions. SMPS's commonly have a scan time of 2-4 minutes and this is 301 mainly limited by the slow temporal resolution of the CPC. Under certain conditions a "Fast CPC" could capture data at 3s.⁴⁴ However, realistically, scans are unlikely to be reduced below 30s. Currently, Full Flow 302 303 CPCs and Mixing CPCs have temporal resolutions of 0.25-3s and 16-100ms respectively.⁴² The detection 304 limit, or "cut-off", of a CPC is described by its 50% detection efficiency diameter (d_{50}); the size at which 305 <50% of particles passing through the CPC are counted. Changing the temperatures within the condenser

and saturator can change the D₅₀ 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/cm³. CPCs need to be maintained level, to prevent the working fluid entering the optical circuitry and this makes them difficult to use for personal exposure monitoring.⁴⁵

310 The Diffusion Size Classifier (DiSC) can estimate Pnum, the average particle diameter in the size range of 311 10-700nm and LDSA. In the DiSC the aerosol is charged in a unipolar diffusion charger and then passes 312 through two electrometer stages. The DiSC is one type of Diffusion Charger, primed to be important in the 313 future of UFP measurement due to its ability to measure surface-area related metrics. The first stage or 314 "diffusion stage" consists of a stack of stainless-steel screens connected to sensitive electrometers; the 315 second stage is a HEPA filter connected to an electrometer. Deposition of particles in each of these areas 316 generates a current; Idiffusion and Ifilter. The relationship between these generated currents can be used to 317 calculate the Pnum and average particle diameter.^{46,47} The DiSC is small, portable and battery operated, 318 and this makes it highly suitable for field measurements. The DiSC performs very well for its size and cost 319 but is significantly less accurate than the larger and more expensive CPC and SMPS. Although the accuracy 320 is generally good, the DiSC can perform poorly under certain conditions; for example, measuring 321 monodisperse aerosols of specific compositions and sizes. This is because larger particles can carry more charge which leads to overcounting.⁴⁵ When compared to the SMPS and CPC the mean particle size and 322 323 Pnum were within ±30% and ±50% of reference values, respectively.⁴⁸ They identified that the presence 324 of particles >400nm drastically bias the mean particle size measurement. An example of this piece of 325 equipment is the TESTO DiSCmini. Traditionally, DiSCs are cheaper than CPCs; however new periodic 326 technical inspection regulations for vehicle emissions are being introduced in Germany, with 35,000 garages mandated to have Pnum sensors by the 1st January 2021.49 This large demand will drive the market to 327 328 produce low-cost (<\$5000), reliable and easy to use Pnum measurement devices. The likely result will be 329 that DiSCs and CPCs will be forced to become competitive on price, which will likely increase their 330 widespread use in other sectors, for example, IAQ measurement.

331 Particle Size Distribution

332 Psd can be obtained from SMPS, Fast Mobility Particle Sizer (FMPS), OPCs, APSs and passive air samplers. 333 In Differential Mobility Analyzers (DMA) particles are given a unipolar corona charge and are passed through 334 an electric mobility analyzer; from here particles of a given diameter are selected based on their electrical mobility.³⁹ The size selected by the DMA is determined by the magnitude of the voltage applied. 335 336 Exponentially increasing the voltage allows scanning through a particle diameter size range in several 337 minutes. The same concept underpins operation of the SMPS. An Electrostatic Classifier is a particle 338 neutralizer combined with a DMA; it can generate and subsequently size select particles. A SMPS is 339 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 340 341 instrument for measuring particle size distributions. The FMPS differs from the SMPS by using an 342 Electrostatic Classifier and multiple low noise electrometers to measure Pnum and Psd in the 5-600nm 343 range. The benefit of this over the SMPS is a 1s sampling frequency, making it more suitable for measuring 344 rapid aerosol processes. However, the SMPS measures the very smallest particles with a higher accuracy 345 than the FMPS. Examples of this equipment include TSI's SMPS 3938 and FMPS 3091. The SMPS with a 346 range of 1-1000nm is often paired with an Aerodynamic Particle Sizer (APS) with a range of 0.5-20µm 347 when Pnum and Psd of particles >1000nm are also of interest.

348 The APS uses the inertia of particles to determine their size; firstly, the aerosol flow is constricted through 349 a nozzle and this accelerates the aerosol. The velocity of the particle can then be related to the particle's 350 surface area and mass and therefore aerodynamic diameter. The aerodynamic diameter is determined 351 assuming spherical particles and uniform density. Secondly, particles then pass through two laser beams 352 separated by 200µm; as the particle passes through each beam, light is scattered onto a photodetector. 353 The time difference detected between the two pulses of scattered light can be used to determine the 354 velocity and therefore aerodynamic diameter. The magnitude of the electrical current generated by the 355 scattered light also provides a secondary estimate of the particle size. APSs are commonly operated

alongside a SMPS but can be used exclusively when particles <500nm are not of interest. For example,
 APS measurements are suitable for source characterization of resuspended material.

358 Particle Surface Area

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

Diffusion Chargers use corona discharge to create unipolar ions that diffuse onto the active surface of particles, and 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, which is a fraction of the 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.⁵⁰ It can measure lung-deposited particle surface area concentrations, based on lung particle deposition models.

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

	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/m³<	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/m³	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

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

369 4. Experimental Applications

This section outlines the experimental applications of the previously described technologies, and aims to provide insight into the practicalities of deploying these indoors.

372 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 and international standards and to 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.

377 One cheap and robust method for testing compliance is by using gravimetric aerosol samplers; however, 378 these are not real time and therefore cannot be used to identify short-term temporal variation. Passive 379 aerosol samplers have been used in primary schools, with polycarbonate and quartz filters deployed at child 380 breathing height (1.2m).⁵¹ This study demonstrates that long-term temporal variations can be measured using gravimetric samplers deployed and collected over several seasons.⁵¹ They were also able to apportion 381 sources using chemical analysis, with particular sources having distinctive chemical compositions.⁵¹ Passive 382 383 samplers are smaller and quieter than larger active samplers and so are appropriate for use in schools, 384 many workplaces and homes.

385 Cascade Impactors can be used upstream of traditional gravimetric samplers when the mass-based particle 386 distribution is of interest. For example, cascade impactors were used to simultaneously sample particles 387 of >10 μ m, 10-2.5 μ m, 2.5-1 μ m and <1 μ m in 8 different indoor locations in France.⁵² This included private 388 residences, a school and a restaurant across a range of urban, suburban and rural settings. The particles 389 were chemically analyzed to determine the concentrations of 16 US-EPA priority Polycyclic Aromatic 390 Hydrocarbons (PAHs). Gravimetric samplers are also a relatively unobtrusive method of sampling, for 391 example, size segregated Pmass was measured in a well-ventilated primary school gym during PE lessons, 392 using personal cascade impactors to minimize intrusiveness and health and safety risks.⁵³

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 centers, with spatial and temporal distributions of LDSA being assessed.⁵⁴

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 PM₁₀ in Hong Kong schools to assess compliance against Hong Kong's Air Quality Objectives, with increased levels of PM attributed to sources such as traffic and construction.⁵⁵

When measuring compliance, it is important to consider whether a device is suitable for the levels of PM being measured; 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.⁵⁶ The photometers were converted from smoke detectors and were developed specifically to measure in high pollution environments, with a lower detection limit of 50µg/m³. However, this caused difficulties with 49% of PM_{2.5} measurements falling at or below 50µg/m³.

408 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, air pollution generated within households can have significant influence on ambient air pollution concentrations, for example, in Los Angeles, consumer volatile chemical products are the largest source of ambient VOCs.⁵⁷ 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 in Pittsburgh.⁵⁸ The Personal Exposure Monitors (gravimetric filters) 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; they compared the indoor/outdoor ratios of 27 chemical species.

425 The SMPS is often the equipment of choice in investigations aiming to understand indoor penetration and 426 deposition behavior. This is because these processes are determined by particle size, which can be 427 measured by the SMPS. For example a SMPS and APS were used to measure both indoor and outdoor Psd 428 simultaneously by alternating between indoor and outdoor air inputs using a specially designed sampling 429 manifold.⁶⁰ A similar study was conducted in modern offices.²⁴ In this investigation they were able to 430 determine differential infiltration and deposition as a function of size. Indoor and outdoor Pnum and Psd 431 were measured simultaneously in a school using two SMPS units; they were able to identify the main 432 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.

439 Our current understanding of the relationship between indoor and outdoor PM is summarized by Chen⁶³.

440 Source Characterization

Source Characterization 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.

444 In a chamber setup fine and UFP emissions were measured from 13 particle sources using an OPC and 445 CPC.⁶⁴ A total-capture dilution tunnel system was 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 446 447 and APS in the real world, namely in 15 homes in Brisbane, Australia.⁶⁶ Elevated Pnum was linked to 21 448 other indoor activities using occupant logs and these events were measured using a CPC and Dustrak. 449 Controlled environments allow for more accurate and repeatable measurement of particle generation, but 450 real-world measurement can provide a more realistic basis for investigating processes; for example, 451 dispersion, ageing and deposition of PM.

452 OPCs are ideally suited to resuspension studies because they negate the OPCs biggest weakness, the 453 inability to measure particles <300nm. This is because the process of resuspension by human activities contributes most to the coarse particle fraction $(>1\mu m)$.⁶⁶ OPCs are commonly used when the UFP fraction 454 455 is not of interest or to supplement the measurements made by other equipment. OPC measurements can 456 add Pnum and Psd data to data collected by CPCs and photometers. Alternatively, at additional cost, the 457 OPC could be paired with a diffusion size classifier to measure a much wider range of 10nm-20µm. Of 13 458 studies measuring resuspended particles and how they varied in size between different human activities, 8 459 utilized OPCs and 3 utilized 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 (0.3-20µm) were used to characterize the emissions from seven 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 a FMPS to measure Pnum and
Psd at 1s resolution. The nanoparticle emissions of burning incense were characterized 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.

469 High Resolution Spatial and Temporal Measurements

470 It is becoming increasingly important to understand the lateral PM variations within rooms and buildings, 471 to link sources, ventilation and purification systems. Therefore, there is a need for low-cost, portable real-472 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 However these investigations are 473 474 currently still largely testing the premise, it remains to be seen whether these instruments are sufficiently 475 accurate and sensitive. 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, 476 India to understand the spatiotemporal resolution of PM generated by cookstoves.⁴¹ In lab tests the sensors 477 478 agree well with reference grade equipment, however, these sensors become saturated at 4-5mg/m³ 479 compared to the 20 mg/m³ of the SidePak, making them less suitable for high pollution environments.⁷²

480 5. Discussion

481 Whether it is important to measure multiple metrics in an investigation depends upon the study design, 482 purpose and focus is. Using a variety of instruments, which, measure the same or different metrics will 483 improve confidence in results and understanding of the nature of indoor PM. For example, in photometer-484 based studies photometers are regularly co-located with reference grade gravimetric samplers to ensure 485 the data collected is of suitable accuracy or to calibrate the photometer if necessary. With Pnum and Psd 486 measurement, it is important to consider whether it is possible to expand the particle diameter range 487 measured by using a number of different pieces of equipment. For example, supplementing the SMPS (2.5-488 1000nm) or FMPS (5-560nm) with use of an APS (0.5-20µm) or OPC (0.3-20µm). Combining use of high 489 cost, less portable and more accurate equipment with low-cost, highly portable equipment can also improve 490 spatial mapping results. This is important, because air pollution is not homogeneous throughout a single 491 room or building. Given the disparity between the concentrations of PM indoors in LEDCs, and higher 492 economically developed countries (HEDCs) (responsible for ~97% and 3% of premature deaths due to 493 household air pollution in 2016),³ it is essential to be aware of the range limitations of sensors. For example, 494 many low-cost photometers are able to measure effectively in high concentration environments, but are 495 too insensitive to use at low concentrations.⁷² Conversely, mixing CPCs and photometers are extremely 496 versatile in measuring a wide dynamic range, with sensitivity at low concentrations and with inundation 497 only at very high concentrations. This is due to the mixing CPCs dilution of the aerosol flow and the 498 photometers historical use for occupational health monitoring in high pollution environments, (e.g. in saw 499 and flour mills).

500 Challenges of PM metrics

501 A challenge of the Pnum/Psd/Surface area metrics are the difficulty in comparing measured values across 502 literature. Different investigations often operate equipment with differing size ranges or operate the same 503 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 504 505 between 6-15nm made reliable comparisons difficult and therefore only two out of the eight studies 506 included were directly comparable. Having no standardized size range is one of the reasons why universal 507 standards have not yet been developed. Different types of Pnum and Psd sensor have highly variable size 508 ranges depending on the method being implemented; this adds further complexity to the task. Without 509 regulation, expecting original equipment manufacturers (OEMs) to comply to standardized size ranges is 510 difficult as they will want freedom to change and increase their size ranges as technology improves. On the 511 other hand, OEMs like regulation-backed markets because it mandates the need for their products. It will 512 be important to allow OEMs to contribute to discussions around standardization. Being unable to directly 513 compare to other literature or universal standards, investigations instead must rely on internal comparisons, 514 for example, the levels were x times greater than background levels or Site A was x relative to Site B. 515 Although there are standards issued for the Pmass metric, very few are specific to indoor environments 516 and this is a problem because the variability in concentration, chemical composition and Psd can all be

517 significantly different to outdoor environments. This is because indoor air is made up of contributions from 518 both indoor and outdoor sources; this means that although they share many physico-chemical properties, 519 they are often far from homogenous.⁸ Future campaigns should aim to deploy high grade sensors to 520 measure the physio-chemical properties of IAP under a variety of scenarios; one such campaign is 521 HOMEchem.⁷³ HOMEchem is a collaborative field investigation aiming to determine how everyday activities 522 effect the emissions, interactions and removal processes of particles and trace gases in indoor 523 environments.⁷⁴ Once the nature of IAQ is better understood it can be compared to outdoor air quality in 524 order to make informed decisions on whether health standards should differ between them.

525 Standardizing the field of PM measurement

526 Given the difficulty in making comparable measurements, there is a need for the community to come 527 together and to work towards standardized measurements. Accordingly, there are two types of guidelines 528 that need to be defined: 1) standardized size ranges or D_{50s} for individual measurement techniques e.g. 529 CPCs, OPCs, DiSCs and NSAMs, and 2) regulatory guidelines for what are allowable concentrations of PM 530 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 1.

Old paradigm New paradigm Set interim guidelines Limited ability to (based on an incomplete set useful guidelines understanding) Increased standardized **Reform** existing measurement guidelines Limited No guidelines understanding set Cyclic Issue of PM Breaking the Measurement cycle Standardization More intercomparable Regular review of guidelines through specialist workshops data and conferences No standardized Limited intercomparable measurements Improved understanding data ง and real-world pressure tested guidelines

Figure 1. 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 distributions as opposed to a mechanistic relationship to health effects. As the "new paradigm" cycle continues and epidemiological understanding improves, we could start to implement the more useful health related guidelines.

541 Development of these guidelines are beneficial to a variety of stakeholders. Firstly, PM researchers and 542 epidemiologists will have more inter-comparable measurements, which will improve understanding on their 543 respective fields. Regulators will be better informed, allowing for development of more robust guidelines. 544 This will expand the usefulness of Pnum, Psd and surface area metrics for regulatory purposes, and this in 545 turn will benefit equipment manufacturers by increasing demand for their products. Some other broader 546 challenges associated with aerosol science community are addressed by Sorensen.⁷⁵

547

548 The relationship between IAQ and energy efficiency

549 In recent years improvements to the energy efficiency of buildings has reduced air exchange rates between 550 indoors and outdoors, to improve energy efficiency through reducing heat losses. However, this has created 551 indoor environments where pollution can become "trapped" and accumulate to much higher levels than before.¹⁰ A balance is needed between improving energy efficiency, reducing operation costs and improving 552 553 IAQ.⁷⁶ However, in some environments such as in the mega-cities of many LEDCs, reducing the exchange 554 between indoor and outdoor air may improve the IAQ, especially in indoor environments without dominant 555 sources of IAP. Improved energy efficiency may motivate increased research on IAQ, how it relates to 556 outdoor air quality and the nature of indoor air pollutants ageing indoors. It is worth noting that indoor air 557 chemistry will have similar complexity to ambient atmospheric chemistry. Some of the most important guestions relating to IAQ and "green buildings" are addressed by Steinemann.⁷⁷ 558

559 Low-cost sensors

560 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.^{13,78} Static and sparse air pollution 561 562 monitoring stations are not sufficient to fully understand the behavior of pollution in urban areas. Existing 563 networks could be supplemented with a low-cost high-density network. The same concept applies indoors, 564 but on a smaller scale; by taking a single measurement in a room or building the assumption is that the air 565 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 566 not be fully indicative of individual exposure given the "personal cloud" effect,⁷⁹ therefore, in the future 567 low-cost sensors will also be needed for personal exposure measurements. Switching from wired to 568 569 wireless data transmission has been shown to reduce initial investment and annual operation cost by 3x and 5x, respectively, in the US.^{75,80} Wireless data transmission also allows for unattended large-scale 570 571 monitoring. Wireless data transmission is especially important for indoor data collection because minimizing 572 intrusiveness is important. Intrusion into residences and workplaces can be disruptive, and being able to 573 minimize intrusion will increase the participation in projects. Furthermore, in unoccupied environments,

574 intrusions can influence indoor PM through resuspension. Some of the issues currently associated with the 575 future of low-cost sensor networks are the consistency and durability of sensors, the reliability of data and 576 the high costs associated with data processing. Moreover, most low-cost sensors have a short lifetime of 6 577 months to a few years, which will need to be improved for these sensors to be viable in the future. The 578 costs of calibration, servicing (replacing batteries), integrating the sensor into a device and data processing 579 often exceeds the cost of the sensors themselves; these costs need to be reduced. Sensor performance 580 will need to be tested under a wide range of environmental conditions before they are ready to be deployed 581 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.⁸⁰ Moreover, 582 583 with gas sensors, the focus is purely on concentration, while with PM it is important to understand Pnum, 584 Pmass, Psd and chemical composition. This makes PM a lot more difficult to effectively quantify than 585 ambient gas concentrations. Another shortcoming of low-cost devices is their signal to noise ratio, making 586 them much less useful in low pollution environments. This can be negated to some extent by recording for 587 longer periods and averaging the results, or by co-locating several sensors and taking an average. Digital 588 filters are commonly applied to the data from these sensors to remove high-frequency noise.⁷¹

589 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 LEDCs due to their nature of being easily deployed and operated and having low power requirements. Moreover, with PM concentrations usually higher in LEDCs, 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 and a large database is generated, several issues need to be considered. It is important that the data collected is easily accessible to interested stakeholders, and that the database is not held by a private company that might misuse it for financial gain.⁸⁰ It is 600 necessary to consider who would own such a network and database; public bodies, research bodies, 601 commercial entities or citizens. Minimizing the need for data processing and analysis through automation 602 could make air pollution data from portable sensors more accessible to the public. If the data can be 603 interpreted into an easily accessible form, then citizen uptake will increase. Fortunately, low-cost sensors 604 require very little knowledge to be deployed and require less technical maintenance than more complicated 605 equipment, which may also increase citizen uptake. Low-cost sensors could also provide immediate value, 606 by warning occupants of high pollution levels and offering suggestions to mitigate these, for example, 607 opening the window or automatically turning on an air purifier. It is also worth considering whether it is 608 essential for these low-cost sensors to perform well at low-concentrations; if their primary purpose is to 609 inform users in real time of dangerously high levels. People may be more inclined to remediate IAQ 610 problems based on personalized measurements of IAQ, rather than city wide pollution measurements and 611 warning systems.

612 Ethical concerns of real-time measurement

613 Real time measurement within residences also raises various ethical concerns. For example, IAQ data can 614 be used to infer activity, such as when the house is occupied or unoccupied, whether residents are awake 615 or asleep and whether activities such as cooking, or bathing are occurring. Therefore, personal data is 616 inadvertently collected alongside the IAQ data. This needs to be considered when viewing the data in real-617 time and ensuring anonymity when viewing data afterwards. A further ethical concern occurs if dangerously 618 high pollutant levels are observed in real time. What level of responsibility and duty of care should residents 619 expect? Should looking at results in real-time be avoided completely, even though this is an important part 620 of maintaining sensors and troubleshooting?

621 6. 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 remain useful due to its long history of use in compliance testing; however, future epidemiological studies may conclude that Pmass has less correlation to human 626 health effects than other metrics. Pmass is likely to continue its evolution from PM₁₀ and PM_{2.5} down to PM₁ 627 or PM_{0.5} as technology improves. The dominant contribution to Pmass comes from larger particles 628 (>100nm), whereas Pnum is largely influenced by the smallest particles (<100nm). The Pnum metric is 629 becoming increasingly useful as focus is shifting towards measurement of UFPs. However, with Pnum it is 630 very difficult to make comparisons to previous studies and it seems unlikely that any air quality standards 631 will be developed soon. The Pnum metric is considered a better determinant of health effects than Pmass,²⁶ 632 but Pmass sensors are being reduced in size at a much faster rate than Pnum sensors. This might make 633 Pmass sensors the preferable choice for the low-cost sensor networks of the future. Although Pnum sensors 634 such as OPCs are being reduced in size, they cannot measure the very smallest particle sizes. The Psd 635 metric can allow for observations that cannot be determined from Pmass or Pnum alone and will become 636 increasingly implemented as sensors improve. Psd will likely remain as a "research grade" measurement, 637 solely for understanding the nature of PM and applying that to developing better metrics and sensors for 638 compliance monitoring. There is consensus that future UFP metrics should provide insight into how PM 639 interacts with the body through intake, uptake and transport. Therefore, many researchers are favoring a 640 surface area related metric, which correlates better with toxicological or biological activity than either Pmass 641 or Pnum.³⁰ NSAMs are likely to become an increasingly important measurement option, as interest in 642 measuring surface area related metrics increases.

643 Many countries and organizations are having to decide whether to solely legislate using the historical and 644 conventional Pmass metric or to adopt other metrics. However, this is an extremely difficult task given the 645 current lack of understanding of PM and its associated health effects, even though PM is one of the best 646 understood forms of air pollution, there is still much that needs to be understood. Several essential epidemiological questions still need to be answered.⁸¹ Firstly, which properties, or combination of properties, 647 648 are the most important determinants of potency; for example, size, surface area, or chemical composition. 649 Secondly, are chronic background pollution levels or acute high-level pollution events a greater determinant 650 of health effects. This will largely inform what type of standard should be enforced; for example, daily 651 average or "should not exceed x for longer than y duration". It will also inform the needs of regulatory

652 compliance measurements, for example, if high level pollution events are deemed the most important 653 determinant of health effects, then having PM monitors which are sensitive to low concentrations is far less 654 important. Until these questions are answered, we should continue to experiment using a wide range of 655 metrics to better understand the nature of PM.

656 With low-cost real-time sensors at the forefront of meeting rapidly increasing public interest, and with 657 citizen science projects becoming more frequent, personalized information about IAQ is likely to become increasing available.^{13,82} Eventually this could lead to real-time IAQ warning systems in homes; allowing 658 residents to more easily mitigate IAQ issues.⁸² This level of technology would also allow us to resolve many 659 660 of the issues related to IAQ and energy efficiency. As real-time low-cost sensors become increasingly 661 common, high spatial distribution measurements which were previously difficult are likely to become more 662 common. However, it is unlikely that the current generation of low-cost PM sensors will be able to detect 663 subtle variations in the indoor environment. But this will likely change as sensors are periodically improved. 664 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.⁸¹ A better understanding of epidemiology 665 666 will inform the future of compliance measurement and source characterization in indoor environments.

667 Many premature deaths worldwide are attributed to indoor PM. Several essential metrics and measurements 668 techniques are available that practitioners and scientists can use to better understand and reduce indoor 669 PM. In the future it is essential to better understand what PM properties most strongly effect health and to 670 channel this information into the development of improved metrics, measurement techniques, legislative 671 standards and new experimental applications.

672 Words: 9400 + (2 Tables + 1 Figure) = 10000

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- 676 References:

- 678 (1) Forouzanfar, M. H.; Alexander, L.; Bachman, V. F.; Biryukov, S.; Brauer, M. Global, Regional, and
 679 National Comparative Risk Assessment of 79 Behavioural, Environmental and Occupational, and
 680 Metabolic Risks or Clusters of Risks in 188 Countries, 1990-2013: A Systematic Analysis for the
 681 Global Burden of Disease Study 2013. *The Lancet.* 2015, pp 2287–2323.
 682 https://doi.org/10.1016/S0140-6736(15)00128-2.
- World Health Organization. Burden of Disease from Household Air Pollution for 2012. *World Heal. Organ.* 2014, *35* (February), 17. https://doi.org/10.1016/S0140-6736(12)61766-8.Smith.
- (3) Institute for Health Metrics and Evaluation. Global Burden of Disease (GBDx Results Tool)
 http://ghdx.healthdata.org/gbd-results-tool (accessed Jul 17, 2018).
- 687 (4) Roser, M.; Ritchie, H. Indoor Air Pollution https://ourworldindata.org/indoor-air-pollution (accessed Jun 17, 2018).
- (5) Lim, S. S.; Vos, T.; Flaxman, A. D.; Danaei, G.; Shibuya, K. A Comparative Risk Assessment of
 Burden of Disease and Injury Attributable to 67 Risk Factors and Risk Factor Clusters in 21
 Regions, 1990-2010: A Systematic Analysis for the Global Burden of Disease Study 2010. *Lancet*2012, 380 (9859), 2224–2260. https://doi.org/10.1016/S0140-6736(12)61766-8.
- (6) Maher, B. A.; Ahmed, I. A. M.; Karloukovski, V.; MacLaren, D. A.; Foulds, P. G.; Allsop, D.; Mann,
 (6) D. M. A.; Torres-Jardón, R.; Calderon-Garciduenas, L. Magnetite Pollution Nanoparticles in the
 (6) Human Brain. *Proc. Natl. Acad. Sci.* **2016**, *113* (39), 10797–10801.
 (7) https://doi.org/10.1073/pnas.1605941113.
- Klepeis, N. E.; Nelson, W. C.; Ott, W. R.; Robinson, J. P.; Tsang, A. M.; Switzer, P.; Behar, J. V.;
 Hern, S. C.; Engelmann, W. H. The National Human Activity Pattern Survey (NHAPS): A Resource
 for Assessing Exposure to Environmental Pollutants. *J. Expo. Anal. Environ. Epidemiol.* 2001, *11*(3), 231–252. https://doi.org/10.1038/sj.jea.7500165.
- Morawska, L.; Afshari, A.; Bae, G. N.; Buonanno, G.; Chao, C. Y. H.; Hänninen, O.; Hofmann, W.;
 Isaxon, C.; Jayaratne, E. R.; Pasanen, P.; et al. Indoor Aerosols: From Personal Exposure to Risk
 Assessment. *Indoor Air.* December 1, 2013, pp 462–487. https://doi.org/10.1111/ina.12044.
- 704 (9) Brunekreef, B.; Holgate, S. T. Air Pollution and Health. *Lancet*. 2002.
 705 https://doi.org/10.1016/S0140-6736(02)11274-8.
- 706
 (10)
 Adgate, J.; Ramachandran, G.; Pratt, G. Spatial and Temporal Variability in Outdoor, Indoor, and

 707
 Personal PM 2.5 Exposure. *Atmos. ...* **2002**, *36* (20), 3255–3265. https://doi.org/10.1016/S1352

 708
 2310(02)00326-6.
- Seppänen, O. Ventilation Strategies for Good Indoor Air Quality and Energy Efficiency. In *International Journal of Ventilation*; 2008; Vol. 6, pp 297–306.
 https://doi.org/10.1080/14733315.2008.11683785.
- (12) ISO. ISO 16000-1 Indoor Air -- Part 1: General Aspects of Sampling Strategy. *International Organization for Standardization*. 2014.
- (13) Snyder, E. G.; Watkins, T. H.; Solomon, P. A.; Thoma, E. D.; Williams, R. W.; Hagler, G. S. W.;

- Shelow, D.; Hindin, D. A.; Kilaru, V. J.; Preuss, P. W. The Changing Paradigm of Air Pollution
 Monitoring. *Environ. Sci. Technol.* **2013**. https://doi.org/10.1021/es4022602.
- Kumar, P.; Skouloudis, A. N.; Bell, M.; Viana, M.; Carotta, M. C.; Biskos, G.; Morawska, L. Real-Time Sensors for Indoor Air Monitoring and Challenges Ahead in Deploying Them to Urban Buildings. *Science of the Total Environment*. 2016, pp 150–159. https://doi.org/10.1016/j.scitotenv.2016.04.032.
- (15) ISO. ISO 16000-34 Indoor Air Part 34: Strategies for the Measurement of Airborne Particles.
 International Organization for Standardization. 2018.
- Thongyen, T.; Hata, M.; Toriba, A.; Ikeda, T.; Koyama, H.; Otani, Y.; Furuuchi, M. Development
 of PM0.1 Personal Sampler for Evaluation of Personal Exposure to Aerosol Nanoparticles. *Aerosol Air Qual. Res.* 2015. https://doi.org/10.4209/aaqr.2014.05.0102.
- (17) British Standard. Workplace Atmospheres: Size Fraction Definitions for Measurement of Airborne
 Particles in the Workplace. *Br. Stand.* **1993**, *BS EN 481*:
- (18) US EPA. Particulate Matter (PM) Basics https://www.epa.gov/pm-pollution/particulate-matter-pm basics.
- 730 (19) Nazaroff, W. W. Indoor Particle Dynamics. *Indoor Air, Supplement*. 2004.
 731 https://doi.org/10.1111/j.1600-0668.2004.00286.x.
- WHO. WHO Air Quality Guidelines for Particulate Matter, Ozone, Nitrogen Dioxide and Sulfur
 Dioxide: Global Update 2005: Summary of Risk Assessment. *Geneva World Heal. Organ.* 2006, 1–
 https://doi.org/10.1016/0004-6981(88)90109-6.
- WHO Regional Office for Europe. Guidelines for Indoor Air Quality. *WHO Guidel.* 2010, *9*, 454.
 https://doi.org/10.1186/2041-1480-2-S2-I1.
- Andersson, J.; Giechaskiel, B.; Munoz-Bueno, R.; Sandbach, E.; Dilara, P. Particle Measurement
 Programme (PMP) Light-Duty Inter-Laboratory Correlation Exercise (ILCE_LD) Final Report. *Inst. Environ. Sustain.* 2007, *EUR 22775*, 2034–2042.
- 740 (23) Bo, M.; Salizzoni, P.; Clerico, M.; Buccolieri, R. Assessment of Indoor-Outdoor Particulate Matter
 741 Air Pollution: A Review. *Atmosphere*. 2017. https://doi.org/10.3390/atmos8080136.
- (24) Chatoutsidou, S. E.; Ondráček, J.; Tesar, O.; Tørseth, K.; Ždímal, V.; Lazaridis, M. Indoor/Outdoor
 Particulate Matter Number and Mass Concentration in Modern Offices. *Build. Environ.* 2015, *92*,
 462–474. https://doi.org/10.1016/j.buildenv.2015.05.023.
- 745 (25) DEFRA. Ultrafine Particles (UFP) in the UK; 2018.
- Penttinen, P.; Timonen, K. L.; Tiittanen, P.; Mirme, A.; Ruuskanen, J.; Pekkanen, J. Ultrafine
 Particles in Urban Air and Respiratory Health among Adult Asthmatics. *Eur Respir J* 2001, *17*,
 428–435. https://doi.org/10.1183/09031936.01.17304280.
- (27) Stephenson, D.; Seshadri, G.; Veranth, J. M. Workplace Exposure to Submicron Particle Mass and Number Concentrations from Manual Arc Welding of Carbon Steel. *Am. Ind. Hyg. Assoc. J.* 2003, 64 (4), 516–521. https://doi.org/10.1080/15428110308984848.
- Kim, K.-H.; Kabir, E.; Kabir, S. A Review on the Human Health Impact of Airborne Particulate
 Matter. *Environ. Int.* 2015, *74*, 136–143. https://doi.org/10.1016/J.ENVINT.2014.10.005.
- (29) Harrison, R. M.; Yin, J. Particulate Matter in the Atmosphere: Which Particle Properties Are Important for Its Effects on Health? *Sci. Total Environ.* 2000, *249* (1–3), 85–101. https://doi.org/10.1016/S0048-9697(99)00513-6.

- (30) Baldauf, R. W.; Devlin, R. B.; Gehr, P.; Giannelli, R.; Hassett-Sipple, B.; Jung, H.; Martini, G.;
 McDonald, J.; Sacks, J. D.; Walker, K. Ultrafine Particle Metrics and Research Considerations:
 Review of the 2015 UFP Workshop. In *International Journal of Environmental Research and Public Health*; 2016; Vol. 13. https://doi.org/10.3390/ijerph13111054.
- Ruckerl, R.; Schneider, A.; Hampel, R.; Breitner, S.; Cyrys, J.; Kraus, U.; Gu, J.; Soentgen, J.;
 Koenig, W.; Peters, A. Association of Novel Metrics of Particulate Matter with Vascular Markers of
 Inflammation and Coagulation in Susceptible Populations –Results from a Panel Study. *Environ. Res.* 2016, *150*, 337–347.
- (32) Heitbrink, W. A.; Evans, D. E.; Ku, B. K.; Maynard, A. D.; Slavin, T. J.; Peters, T. M. Relationships among Particle Number, Surface Area, and Respirable Mass Concentrations in Automotive Engine Manufacturing. *J. Occup. Environ. Hyg.* **2008**, *6* (1), 19–31.
 https://doi.org/10.1080/15459620802530096.
- (33) Wagner, J.; Leith, D. Passive Aerosol Sampler. Part I: Principle of Operation. *Aerosol Sci. Technol.* **2001**, *34* (2), 186–192. https://doi.org/10.1080/027868201300034808.
- (34) Wagner, J.; Macher, J. M. Comparison of a Passive Aerosol Sampler to Size-Selective Pump Samplers in Indoor Environments. *Am. Ind. Hyg. Assoc. J.* 2003, *64* (5), 630–639. https://doi.org/10.1080/15428110308984856.
- (35) Wagner, J.; Leith, D. Passive Aerosol Sampler. Part II: Wind Tunnel Experiments. *Aerosol Sci. Technol.* 2001, *34* (2), 193–201. https://doi.org/10.1080/027868201300034826.
- (36) Leith, D.; Sommerlatt, D.; Boundy, M. G. Passive Sampler for PM10-2.5 Aerosol. *J. Air Waste Manag. Assoc.* **2007**, *57*(3), 332–336. https://doi.org/10.1080/10473289.2007.10465336.
- Whitehead, T.; Leith, D. Passive Aerosol Sampler for Particle Concentrations and Size
 Distributions. *J. Environ. Monit.* 2008, *10* (3), 331. https://doi.org/10.1039/b714225k.
- Amaral, S. S.; de Carvalho, J. A.; Costa, M. A. M.; Pinheiro, C. An Overview of Particulate Matter
 Measurement Instruments. *Atmosphere*. 2015, pp 1327–1345.
 https://doi.org/10.3390/atmos6091327.
- (39) Giechaskiel, B.; Maricq, M.; Ntziachristos, L.; Dardiotis, C.; Wang, X.; Axmann, H.; Bergmann, A.;
 Schindler, W. Review of Motor Vehicle Particulate Emissions Sampling and Measurement: From
 Smoke and Filter Mass to Particle Number. *Journal of Aerosol Science*. 2014, pp 48–86.
 https://doi.org/10.1016/j.jaerosci.2013.09.003.
- (40) Wang, Z.; Calderón, L.; Patton, A. P.; Sorensen Allacci, M. A.; Senick, J.; Wener, R.; Andrews, C.
 J.; Mainelis, G. Comparison of Real-Time Instruments and Gravimetric Method When Measuring
 Particulate Matter in a Residential Building. *J. Air Waste Manag. Assoc.* 2016, *66* (11), 1109–
 1120. https://doi.org/10.1080/10962247.2016.1201022.
- Patel, S.; Li, J.; Pandey, A.; Pervez, S.; Chakrabarty, R. K.; Biswas, P. Spatio-Temporal
 Measurement of Indoor Particulate Matter Concentrations Using a Wireless Network of Low-Cost
 Sensors in Households Using Solid Fuels. *Environ. Res.* 2017, *152*, 59–65.
 https://doi.org/10.1016/j.envres.2016.10.001.
- (42) Wehner, B.; Siebert, H.; Hermann, M.; Ditas, F.; Wiedensohler, A. Characterisation of a New Fast
 CPC and Its Application for Atmospheric Particle Measurements. *Atmos. Meas. Tech.* 2011, *4* (5),
 823–833. https://doi.org/10.5194/amt-4-823-2011.
- (43) Okuyama, K.; Kousaka, Y.; Motouchi, T. Condensational Growth of Ultrafine Aerosol Particles in a
 New Particle Size Magnifier. *Aerosol Sci. Technol.* **1984**, *3* (4), 353–366.
 https://doi.org/10.1080/02786828408959024.

- Wang, J.; McNeill, V. F.; Collins, D. R.; Flagan, R. C. Fast Mixing Condensation Nucleus Counter:
 Application to Rapid Scanning Differential Mobility Analyzer Measurements. *Aerosol Sci. Technol.* 2002, *36* (6), 678–689. https://doi.org/10.1080/02786820290038366.
- 804 (45) Mills, J. B.; Park, J. H.; Peters, T. M. Comparison of the Discmini Aerosol Monitor to a Handheld
 805 Condensation Particle Counter and a Scanning Mobility Particle Sizer for Submicrometer Sodium
 806 Chloride and Metal Aerosols. *J. Occup. Environ. Hyg.* 2013, *10* (5), 250–258.
 807 https://doi.org/10.1080/15459624.2013.769077.
- 808 (46) Fierz, M.; Burtscher, H.; Steigmeier, P.; Kasper, M. Field Measurement of Particle Size and Number 809 Concentration with the Diffusion Size Classifier (DiSC). *SAE* **2008**.
- (47) Fierz, M.; Houle, C.; Steigmeier, P.; Burtscher, H. Design, Calibration, and Field Performance of a
 Miniature Diffusion Size Classifier. *Aerosol Sci. Technol.* 2011, *45* (1), 1–10.
 https://doi.org/10.1080/02786826.2010.516283.
- 813 (48) Todea, A. M.; Beckmann, S.; Kaminski, H.; Bard, D.; Bau, S.; Clavaguera, S.; Dahmann, D.; Dozol,
 814 H.; Dziurowitz, N.; Elihn, K.; et al. Inter-Comparison of Personal Monitors for Nanoparticles
 815 Exposure at Workplaces and in the Environment. *Sci. Total Environ.* 2017, *605–606*, 929–945.
 816 https://doi.org/10.1016/j.scitotenv.2017.06.041.
- 817 (49) Booker, D. Performance and Operational Demands for Low-Cost Particle Number Measurements
 818 for Periodic Technical Inspection. In *Cambridge Particle Meeting*, 2018.
- (50) Asbach, C.; Fissan, H.; Stahlmecke, B.; Kuhlbusch, T. A. J.; Pui, D. Y. H. Conceptual Limitations
 and Extensions of Lung-Deposited Nanoparticle Surface Area Monitor (NSAM). *J. Nanoparticle Res.*2009. https://doi.org/10.1007/s11051-008-9479-8.
- (51) Canha, N.; Almeida, S. M.; Freitas, M. D. C.; Trancoso, M.; Sousa, A.; Mouro, F.; Wolterbeek, H.
 T. Particulate Matter Analysis in Indoor Environments of Urban and Rural Primary Schools Using
 Passive Sampling Methodology. *Atmos. Environ.* **2014**, *83*, 21–34.
 https://doi.org/10.1016/j.atmosenv.2013.10.061.
- Liaud, C.; Dintzer, T.; Tschamber, V.; Trouve, G.; Le Calvé, S. Particle-Bound PAHs Quantification
 Using a 3-Stages Cascade Impactor in French Indoor Environments. *Environ. Pollut.* 2014, *195*,
 64–72. https://doi.org/10.1016/j.envpol.2014.08.007.
- 829 (53) Braniš, M.; Šafránek, J.; Hytychová, A. Exposure of Children to Airborne Particulate Matter of
 830 Different Size Fractions during Indoor Physical Education at School. *Build. Environ.* 2009, *44* (6),
 831 1246–1252. https://doi.org/10.1016/j.buildenv.2008.09.010.
- Almeida-Silva, M.; Almeida, S. M.; Gomes, J. F.; Albuquerque, P. C.; Wolterbeek, H. T.
 Determination of Airborne Nanoparticles in Elderly Care Centers. *J. Toxicol. Environ. Heal. Part A Curr. Issues* 2014. https://doi.org/10.1080/15287394.2014.910157.
- kee, S. .; Chang, M. Indoor and Outdoor Air Quality Investigation at Schools in Hong Kong.
 Chemosphere **2000**, *41* (1), 109–113. https://doi.org/10.1016/S0045-6535(99)00396-3.
- (56) Gurley, E. S.; Salje, H.; Homaira, N.; Ram, P. K.; Haque, R.; Petri, W. A.; Bresee, J.; Moss, W. J.;
 Luby, S. P.; Breysse, P.; et al. Seasonal Concentrations and Determinants of Indoor Particulate
 Matter in a Low-Income Community in Dhaka, Bangladesh. *Environ. Res.* 2013, *121*, 11–16.
 https://doi.org/10.1016/j.envres.2012.10.004.
- 841 (57) McDonald, B. C.; De Gouw, J. A.; Gilman, J. B.; Jathar, S. H.; Akherati, A.; Cappa, C. D.; Jimenez,
 842 J. L.; Lee-Taylor, J.; Hayes, P. L.; McKeen, S. A.; et al. Volatile Chemical Products Emerging as
 843 Largest Petrochemical Source of Urban Organic Emissions. *Science (80-.).* 2018.
 844 https://doi.org/10.1126/science.aaq0524.

- Tunno, B. J.; Shields, K. N.; Cambal, L.; Tripathy, S.; Holguin, F.; Lioy, P.; Clougherty, J. E. Indoor
 Air Sampling for Fine Particulate Matter and Black Carbon in Industrial Communities in Pittsburgh. *Sci. Total Environ.* 2015, *536*, 108–115. https://doi.org/10.1016/j.scitotenv.2015.06.117.
- 848 (59) Zhan, Y.; Johnson, K.; Norris, C.; Shafer, M.; Bergin, M.; Zhang, Y.; Zhang, J.; Schauer, J. The
 849 Influence of Air Cleaners on Indoor Particulate Matter Components and Oxidative Potential in
 850 Residential Households in Beijing. *Sci. Total Environ.* **2018**, *626*, 507–518.
- (60) Long, C. M.; Suh, H. H.; Catalano, P. J.; Koutrakis, P. Using Time- and Size-Resolved Particulate
 Data to Quantify Indoor Penetration and Deposition Behavior. *Environ. Sci. Technol.* 2001, *35*(10), 2089–2099. https://doi.org/10.1021/es001477d.
- (61) Guo, H.; Morawska, L.; He, C.; Zhang, Y. L.; Ayoko, G.; Cao, M. Characterization of Particle
 Number Concentrations and PM2.5 in a School: Influence of Outdoor Air Pollution on Indoor Air. *Environ. Sci. Pollut. Res.* **2010**, *17*(6), 1268–1278. https://doi.org/10.1007/s11356-010-0306-2.
- (62) Chan, W. R.; Sidheswaran, M.; Sullivan, D. P.; Cohn, S.; Fisk, W. J. Cooking-Related PM2.5and
 Acrolein Measured in Grocery Stores and Comparison with Other Retail Types. *Indoor Air* 2016,
 26 (3), 489–500. https://doi.org/10.1111/ina.12218.
- (63) Chen, C.; Zhao, B. Review of Relationship between Indoor and Outdoor Particles: I/O Ratio,
 Infiltration Factor and Penetration Factor. *Atmospheric Environment*. January 2011, pp 275–288.
 https://doi.org/10.1016/j.atmosenv.2010.09.048.
- (64) Afshari, A.; Matson, U.; Ekberg, L. E. Characterization of Indoor Sources of Fine and Ultrafine
 Particles: A Study Conducted in a Full-Scale Chamber. *Indoor Air* 2005, *15* (2), 141–150.
 https://doi.org/10.1111/j.1600-0668.2005.00332.x.
- 866 (65) Shen, G.; Gaddam, C. K.; Ebersviller, S. M.; Vander Wal, R. L.; Williams, C.; Faircloth, J. W.;
 867 Jetter, J. J.; Hays, M. D. A Laboratory Comparison of Emission Factors, Number Size Distributions,
 868 and Morphology of Ultrafine Particles from 11 Different Household Cookstove-Fuel Systems.
 869 *Environ. Sci. Technol.* 2017, *51* (11), 6522–6532. https://doi.org/10.1021/acs.est.6b05928.
- (66) He, C.; Morawska, L.; Hitchins, J.; Gilbert, D. Contribution from Indoor Sources to Particle Number
 and Mass Concentrations in Residential Houses. *Atmos. Environ.* 2004, *38* (21), 3405–3415.
 https://doi.org/10.1016/j.atmosenv.2004.03.027.
- Qian, J.; Peccia, J.; Ferro, A. R. Walking-Induced Particle Resuspension in Indoor Environments.
 Atmospheric Environment. 2014, pp 464–481. https://doi.org/10.1016/j.atmosenv.2014.02.035.
- 875 (68) Salthammer, T.; Schripp, T.; Wientzek, S.; Wensing, M. Impact of Operating Wood-Burning
 876 Fireplace Ovens on Indoor Air Quality. *Chemosphere* **2014**, *103*, 205–211.
 877 https://doi.org/10.1016/j.chemosphere.2013.11.067.
- 878 (69) See, S. W.; Balasubramanian, R.; Man Joshi, U. Physical Characteristics of Nanoparticles Emitted
 879 from Incense Smoke. *Sci. Technol. Adv. Mater.* 2007, *8* (1–2), 25–32.
 880 https://doi.org/10.1016/j.stam.2006.11.016.
- (70) Jovašević-Stojanović, M.; Bartonova, A.; Topalović, D.; Lazović, I.; Pokrić, B.; Ristovski, Z. On the
 Use of Small and Cheaper Sensors and Devices for Indicative Citizen-Based Monitoring of
 Respirable Particulate Matter. *Environ. Pollut.* **2015**, *206*, 696–704.
 https://doi.org/10.1016/j.envpol.2015.08.035.
- Li, J.; Li, H.; Ma, Y.; Wang, Y.; Abokifa, A. A.; Lu, C.; Biswas, P. Spatiotemporal Distribution of Indoor Particulate Matter Concentration with a Low-Cost Sensor Network. *Build. Environ.* 2018, 127, 138–147. https://doi.org/10.1016/j.buildenv.2017.11.001.
- 888 (72) Wang, Y.; Li, J.; Jing, H.; Zhang, Q.; Jiang, J.; Biswas, P. Laboratory Evaluation and Calibration of

- 889
 Three Low-Cost Particle Sensors for Particulate Matter Measurement. *Aerosol Sci. Technol.* 2015,

 890
 49 (11), 1063–1077. https://doi.org/10.1080/02786826.2015.1100710.
- Konstantiation (73)
 Konstantiation (73)<
- 893 (74) Farmer, D. K.; Vance, M. E.; Abbatt, J. P. D.; Abeleira, A.; Alves, M. R.; Arata, C.; Boedicker, E.;
 894 Bourne, S.; Cardoso-Saldaña, F.; Corsi, R.; et al. Overview of HOMEChem: House Observations of
 895 Microbial and Environmental Chemistry. *Environ. Sci. Process. Impacts* 2019.
 896 https://doi.org/10.1039/c9em00228f.
- 897 (75) Sorensen, C. M.; Flagan, R. C.; Baltensperger, U.; Pui, D. Y. H. Grand Challenges for Aerosol
 898 Science and Technology. *Aerosol Sci. Technol.* **2019**, *53* (7), 731–734.
 899 https://doi.org/10.1080/02786826.2019.1611333.
- 900 (76) Persily, A. K.; Emmerich, S. J. Indoor Air Quality in Sustainable, Energy Efficient Buildings. In
 901 *HVAC and R Research*; 2012. https://doi.org/10.1080/10789669.2011.592106.
- 902 (77) Steinemann, A.; Wargocki, P.; Rismanchi, B. Ten Questions Concerning Green Buildings and 903 Indoor Air Quality. *Build. Environ.* **2017**. https://doi.org/10.1016/j.buildenv.2016.11.010.
- 904 (78) Mead, M. I.; Popoola, O. a M.; Stewart, G. B.; Landshoff, P.; Calleja, M.; Hayes, M.; Baldovi, J. J.;
 905 McLeod, M. W.; Hodgson, T. F.; Dicks, J.; et al. The Use of Electrochemical Sensors for Monitoring
 906 Urban Air Quality in Low-Cost, High-Density Networks. *Atmos. Environ.* 2013, *70*, 186–203.
 907 https://doi.org/10.1016/j.atmosenv.2012.11.060.
- 908 (79) Rodes, C. E.; Kamens, R. M.; Wiener, R. W. The Significance and Characteristics of the Personal
 909 Activity Cloud on Exposure Assessment Measurements for Indoor Contaminants. *Indoor Air* 1991.
 910 https://doi.org/10.1111/j.1600-0668.1991.03-12.x.
- (80) Kumar, P.; Morawska, L.; Martani, C.; Biskos, G.; Neophytou, M.; Di Sabatino, S.; Bell, M.;
 Norford, L.; Britter, R. The Rise of Low-Cost Sensing for Managing Air Pollution in Cities. *Environment International.* 2015. https://doi.org/10.1016/j.envint.2014.11.019.
- (81) West, J. J.; Cohen, A.; Dentener, F.; Brunekreef, B.; Zhu, T.; Armstrong, B.; Bell, M. L.; Brauer,
 M.; Carmichael, G.; Costa, D. L.; et al. What We Breathe Impacts Our Health: Improving
 Understanding of the Link between Air Pollution and Health. In *Environmental Science and Technology*, 2016. https://doi.org/10.1021/acs.est.5b03827.
- 918 (82) Schieweck, A.; Uhde, E.; Salthammer, T.; Salthammer, L. C.; Morawska, L.; Mazaheri, M.; Kumar,
 919 P. Smart Homes and the Control of Indoor Air Quality. *Renewable and Sustainable Energy*920 *Reviews.* 2018. https://doi.org/10.1016/j.rser.2018.05.057.
- 921