

# 1 **Particulate matter measurement indoors: a review of metrics,** 2 **sensors, needs and applications**

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## 10 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  
22 measurements are most useful in source characterization and particulate matter property investigations,  
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 IAQ 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 IAQ 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

34 Poor indoor air quality (IAQ) has been estimated to be the 9<sup>th</sup> largest global burden of disease risk,<sup>1</sup> with  
35 the World Health Organization (WHO) attributing >4.3 million premature deaths to household air pollution  
36 in 2012, compared to 3.7 million deaths attributed to ambient (outdoor) air pollution.<sup>2</sup> The Institute for  
37 Health Metrics and Evaluation attributed 2.57 million premature deaths to household air pollution in 2016,<sup>3</sup>  
38 with Asia, Africa, Europe and the Americas contributing 74%, 23%, 1% and 2% respectively.<sup>4</sup> 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.<sup>5</sup> 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.<sup>6</sup> 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.<sup>7,8</sup>  
45 Brunekreef<sup>9</sup> has summarized the short and long term effects of PM on health.

46 Historically, the focus of measurement has been on outdoor PM. However, indoor PM is being increasingly  
47 identified as an area that requires more research. Understanding the sources, sinks and behavior of PM  
48 within indoor environments is important to accurately predict personal exposures and population health  
49 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  $\mu\text{g}/\text{cm}^3$ )  
52 and Pnum is the number of particles within a given volume (particles/ $\text{cm}^3$ ). 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  
58 health risk.

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,<sup>10</sup> 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  
67 pollution is likely to be an increasing problem.<sup>11</sup> This means that PM monitoring equipment used indoors  
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.<sup>8</sup> 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.

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

90 This paper critically reviews PM metrics, the sensors and measurement techniques associated with these  
91 metrics, and the sensors and techniques that are most appropriate for each experimental application. The  
92 future of IAQ and PM measurement, the challenges, solutions and anticipated shifts in focus are also  
93 discussed. This will aid future studies in selecting the most appropriate metrics, measurement techniques,  
94 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.

100 PM is currently measured using gravimetric, optical, oscillating microbalance, beta-attenuation and  
101 electrical current techniques. Some of the sensors can measure more than one of the metrics listed. Indoor  
102 measurements are usually collected in the center of the environment, at 0.75-1.8m height representing the  
103 breathing zone, with a height of 1.5m recommended.<sup>12</sup>

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

109 measuring IAQ specifically.<sup>13</sup> Many of the sensors now being used in indoor environments were initially  
 110 developed to measure vehicular emissions or industrial environments which tend to have high  
 111 concentrations of pollutants compared to indoor environments.<sup>14</sup> Therefore, it is important to establish  
 112 which role each sensor can most effectively play in IAQ measurement. Table 1 outlines the sensors  
 113 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         |

114 **Table 1.** A list of reviewed sensors and their abbreviations categorized by their measurement method.

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

## 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 $\mu$ m) and the coarse mode (>2.5 $\mu$ m); in short, these are the consequence of physical  
 121 processes such as emission, nucleation, accumulation and scavenging. Pmass is usually measured as PM<sub>2.5</sub>  
 122 or PM<sub>10</sub> 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. PM<sub>1</sub> is also increasingly used but is not yet widely implemented and there are discussions  
 124 currently on whether PM<sub>0.5</sub> or PM<sub>0.1</sub> should be introduced in the future.<sup>16</sup> PM<sub>10</sub> and PM<sub>1</sub> are arbitrarily  
 125 selected as size cut-offs out of convenience, whereas PM<sub>2.5</sub> 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 PM<sub>10</sub> and PM<sub>2.5</sub> 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  
133 nose (50% penetration efficiency (D<sub>50</sub>) at 100µm), pass through the larynx and enter the bronchial region  
134 of the lungs (D<sub>50</sub>=10µm) and enter the deepest part of the lungs, the ciliated alveoli (D<sub>50</sub>=4µm),  
135 respectively.<sup>17</sup> It should be noted that these definitions were developed in a workplace exposure context.  
136 The EPA<sup>18</sup> provide a useful visualization of the sizes of PM<sub>2.5</sub> and PM<sub>10</sub> and Nazaroff<sup>19</sup> 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 PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub>, 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<sub>2.5</sub> of 10µg/m<sup>3</sup> (annual mean) and 25µg/m<sup>3</sup> (24 hour mean)  
145 and for PM<sub>10</sub> of 20µg/m<sup>3</sup> (annual mean) and 50µg/m<sup>3</sup> (24 hour mean).<sup>20</sup> However, these standards are not  
146 indicative of where adverse health effects begin to occur (for PM<sub>2.5</sub>, this is currently understood to be just  
147 above background concentration at 3-5µg/m<sup>3</sup>).<sup>20</sup> Rather, they are the lowest levels at which "total,  
148 cardiopulmonary and lung cancer mortality have been shown to increase with more than 95% confidence  
149 in response to long-term exposure to PM<sub>2.5</sub>".<sup>20</sup> The WHO investigated whether it was necessary to introduce  
150 specific indoor Pmass standards.<sup>21</sup> 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

153 any specific, more stringent indoor Pmass standards. WHO outdoor Pmass guidelines are therefore  
154 assumed to apply to indoor environments.<sup>21</sup> This assumption makes sense for now, given our currently  
155 limited understanding of the nature of IAQ; however, in the future it is likely that specific indoor standards  
156 will be established, when the effect of composition and concentration on health are better understood. For  
157 example, if it is discovered that acute exposure to PM has more detrimental health outcomes than chronic  
158 exposure to PM, the standard might change from something based on the average to something that  
159 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  $\leq 100\text{nm}$  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.<sup>22</sup> Unlike  
165 Pmass, Pnum varies by several orders of magnitude - from  $<10^3$  particles/cm<sup>3</sup> in relatively clean  
166 environments to  $>10^6$  particles/cm<sup>3</sup> when there are dominant sources of indoor pollution.<sup>23</sup> In contrast to  
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  $<0.5\mu\text{m}$  size range than in the  $0.5\text{-}18\mu\text{m}$  size  
169 range.<sup>24</sup> 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,<sup>25</sup> 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  
173 important predictor of health impacts than Pmass.<sup>26,27</sup> This is because Pnum better represents the smaller  
174 particle size fractions, which penetrate further into the respiratory system, potentially causing more  
175 damage.<sup>28</sup>

## 176 Particle Size Distributions

177 Understanding the Psd in indoor environments is important for several reasons. The size of particles  
178 determines how far they can penetrate the respiratory tract. Smaller particles often have a higher toxicity

179 per unit mass, due to a larger surface area to mass ratio.<sup>29</sup> The Psd metric is commonly used in studies  
180 which attempt to identify the major PM sources within indoor environments, and to understand the  
181 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.<sup>30</sup> 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  
192 (LDSA)".<sup>30</sup> Measuring compliance or exposures using the Pmass and Pnum metrics can be of limited use,  
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 quantifying the sources generating  
195 the most reactive species of UFPs is suggested.<sup>30</sup>

## 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.<sup>31</sup> 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



204 assumption that particles are spherical whilst active surface concentration does not.<sup>32</sup> 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,  
208 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.<sup>33</sup> 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.<sup>34</sup> 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.  
221 This data can then be used to calculate the Pnum and Psd that the sampler was exposed to.<sup>33,35-37</sup> This  
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.

226 Passive gravimetric samplers are extremely small (1.5-5cm diameter) and light (1-5 grams) and require no  
227 maintenance. These samplers can be deployed for both long and short periods, however, this is subject to  
228 the ambient concentrations as the sensors can have too few or too many particles deposited for the

229 automated image analysis to be effective. If the samplers are operated in especially low PM environments,  
230 or for very short periods, this requires more scanning electron microscope (SEM) images to be analyzed to  
231 determine an accurate concentration. With gravimetric samplers the chemical composition of particles can  
232 be acquired using instrumental neutron activation analysis (INAA), particle-induced X-ray emissions (PIXE)  
233 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),<sup>15</sup> 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.<sup>15,38</sup> 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  
247 extinction (the sum of scattering and absorption) to determine the particle concentrations of an aerosol.<sup>38</sup>  
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.<sup>39</sup> 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 PM<sub>1</sub>, PM<sub>2.5</sub>, PM<sub>10</sub> 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 P<sub>mass</sub>

255 concentrations (0.001-200 mg/m<sup>3</sup>), making them suitable for both clean and highly polluted environments.  
256 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.<sup>40</sup> Optical measurements vary depending on the optical properties of particles,  
260 and therefore do not provide absolute mass concentrations.<sup>41</sup> 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  
273 particles can be sized based on the amplitude of the current generated.<sup>38</sup> For example, the Alphasense OPC  
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  
291 reliable nature,<sup>42</sup> however, they have a relatively low sampling frequency (several seconds) due to zones  
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  
295 temporal resolution of measurements.<sup>43</sup> In a Mixing CPC, a cold aerosol flow is mixed with a warm saturated  
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.<sup>42</sup> 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  
302 capture data at 3s.<sup>44</sup> However, realistically, scans are unlikely to be reduced below 30s. Currently, Full Flow  
303 CPCs and Mixing CPCs have temporal resolutions of 0.25-3s and 16-100ms respectively.<sup>42</sup> 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

306 and saturator can change the  $D_{50}$  of a CPC. Handheld CPCs can make counting errors when multiple particles  
307 are located together in the optical detection region, and this is a common occurrence for  
308 concentrations  $>250000$  particles/cm<sup>3</sup>. CPCs need to be maintained level, to prevent the working fluid  
309 entering the optical circuitry and this makes them difficult to use for personal exposure monitoring.<sup>45</sup>

310 The Diffusion Size Classifier (DiSC) can estimate  $P_{num}$ , 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;  $I_{diffusion}$  and  $I_{filter}$ . The relationship between these generated currents can be used to  
317 calculate the  $P_{num}$  and average particle diameter.<sup>46,47</sup> 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  
322 charge which leads to overcounting.<sup>45</sup> When compared to the SMPS and CPC the mean particle size and  
323  $P_{num}$  were within  $\pm 30\%$  and  $\pm 50\%$  of reference values, respectively.<sup>48</sup> They identified that the presence  
324 of particles  $>400$ nm 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  
327 mandated to have  $P_{num}$  sensors by the 1<sup>st</sup> January 2021.<sup>49</sup> This large demand will drive the market to  
328 produce low-cost ( $< \$5000$ ), reliable and easy to use  $P_{num}$  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  
335 mobility.<sup>39</sup> The size selected by the DMA is determined by the magnitude of the voltage applied.  
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  
340 selects particles of a given size, and the CPC counts these particles. The SMPS is the most precise  
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 0.5-20 $\mu$ m  
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 200 $\mu$ m; 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

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

### 358 Particle Surface Area

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

361 Diffusion Chargers use corona discharge to create unipolar ions that diffuse onto the active surface of  
362 particles, and an electrometer then measures the charge that is transferred from the ions to the particle.  
363 This charge can be related to the active surface concentration, which is a fraction of the geometric surface  
364 area. This is a similar measurement premise to the DiSC. The nanoparticle surface area monitor (NSAM)  
365 measures particles between 20-400nm using the principle of unipolar diffusion charging.<sup>50</sup> It can measure  
366 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 Category (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/m³             | 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<       | <1x10⁴-1x10⁶ particles/cm³ | 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<       | <1x10⁴-1x10⁶ particles/cm³ | 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        | <1x10⁴ particles/cm³       | 1-2                                   | Lower cost and more portable than conventional CPCs.  | Unable to measure the smallest particles (<0.3µm).   |
|                            | DISC                 | Yes (1s)                    | High        | 10-700nm        | <5x10²-1x10⁶ particles/cm³ | 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      | 1-1x10⁷ particles/cm³      | 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/cm³         | 3-4                                   | High Temporal resolution, and can be used to supplement 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 less accurate particle number concentrations than the CPC used in the SMPS |
| Particle Surface Area      | NSAM                 | Yes (1s)                    | High        | 10-1000nm       | <10000 µm²/cm³             | 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

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

#### 372 Compliance Measurement, Temporal Trends and Source Apportionment

373 Compliance measurements are commonly collected when it is necessary to understand how the severity of  
374 pollution relates to national and international standards and to historical measurements, usually making  
375 P<sub>mass</sub> the focal metric. Many of these studies use real-time measurements and therefore often contain  
376 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).<sup>51</sup> This study demonstrates that long-term temporal variations can be measured  
381 using gravimetric samplers deployed and collected over several seasons.<sup>51</sup> They were also able to apportion  
382 sources using chemical analysis, with particular sources having distinctive chemical compositions.<sup>51</sup> Passive  
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 $\mu$ m, 10-2.5 $\mu$ m, 2.5-1 $\mu$ m and <1 $\mu$ m in 8 different indoor locations in France.<sup>52</sup> 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 P<sub>mass</sub> 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.<sup>53</sup>

393 NSAMs are used to account for UFP exposure by measuring LDSA. When measuring UFP exposure and  
394 dosing, LDSA is often chosen because it is likely a better indicator of health effects than other metrics. For  
395 example, exposures were measured in four elderly care centers, with spatial and temporal distributions of  
396 LDSA being assessed.<sup>54</sup>

397 Major sources are typically identified through chemical composition, but sources can also be apportioned  
398 through temporal measurements, although this is less accurate, and speculative. For example, apportioning  
399 increases in PM during rush hour traffic periods to vehicular emissions. A DustTrak was used to measure  
400 PM<sub>10</sub> in Hong Kong schools to assess compliance against Hong Kong's Air Quality Objectives, with increased  
401 levels of PM attributed to sources such as traffic and construction.<sup>55</sup>

402 When measuring compliance, it is important to consider whether a device is suitable for the levels of PM  
403 being measured; some sensors cannot detect low levels of PM whilst others cannot detect high levels of  
404 PM. For example, photometers were used to measure P<sub>mass</sub> in highly polluted, densely populated, low  
405 income housing in urban Dhaka, Bangladesh.<sup>56</sup> The photometers were converted from smoke detectors  
406 and were developed specifically to measure in high pollution environments, with a lower detection limit of  
407 50µg/m<sup>3</sup>. However, this caused difficulties with 49% of PM<sub>2.5</sub> measurements falling at or below 50µg/m<sup>3</sup>.

#### 408 Indoor vs Outdoor Comparisons

409 Indoor vs Outdoor studies are usually focused on understanding the contributions of outdoor air to IAQ,  
410 the penetration rates of particulates and how ventilation and building design may improve or worsen IAQ.  
411 In addition, air pollution generated within households can have significant influence on ambient air pollution  
412 concentrations, for example, in Los Angeles, consumer volatile chemical products are the largest source of  
413 ambient VOCs.<sup>57</sup> These studies are not limited to use of any specific metric, in fact, each metric adds  
414 different value and should be selected based on the specifics of the investigation.

415 For example, the influence of outdoor air pollution and smoking on indoor PM<sub>2.5</sub> and Black Carbon levels  
416 was quantified in 21 industrial community homes in Pittsburgh.<sup>58</sup> The Personal Exposure Monitors  
417 (gravimetric filters) needed to be replaced every 3 days to prevent particle overload on the impaction plate

418 or perturbation of the size cut-off inlet. The samplers were placed in the main activity room away from the  
419 windows, and heating and combustion sources to try to ensure the samples were representative of the  
420 whole room. Gravimetric samplers were chosen because industrial particle emissions are mainly composed  
421 of larger sized particles, contributing more to particle mass.

422 Indoor, outdoor and personal exposure samples were collected for 6 Beijing residences whilst operating air  
423 purifiers.<sup>59</sup> Gravimetric sampling was chosen because the researchers wanted to chemically quantify various  
424 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 P<sub>sd</sub>  
428 simultaneously by alternating between indoor and outdoor air inputs using a specially designed sampling  
429 manifold.<sup>60</sup> A similar study was conducted in modern offices.<sup>24</sup> In this investigation they were able to  
430 determine differential infiltration and deposition as a function of size. Indoor and outdoor P<sub>num</sub> and P<sub>sd</sub>  
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.<sup>61</sup>

433 Another study used an OPC, CPC and gravimetric samplers simultaneously; the OPCs size distribution was  
434 used to supplement the absolute P<sub>num</sub> measurements of the water-based CPC.<sup>62</sup> With the water-based  
435 CPC also having a maximum size cut-off of 3µm, the OPC was able to extend this range up to 10µm. This  
436 study aimed to understand indoor and outdoor source contributions to indoor air over different size ranges,  
437 so the OPC was important to measure the size distributions. OPCs provides a more affordable and portable  
438 measurement of P<sub>sd</sub> 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 Chen<sup>63</sup>.

## 440 Source Characterization

441 Source Characterization investigations aim to quantify the particle generation of various PM sources within  
442 the indoor environment. This can be studied in a real-world, laboratory or chamber environment,  
443 predominantly using P<sub>mass</sub>, P<sub>num</sub> and P<sub>sd</sub> 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.<sup>64</sup> A total-capture dilution tunnel system was used to investigate the P<sub>num</sub> and P<sub>sd</sub> emissions for 11  
446 household cook stove-fuel systems using a SMPS.<sup>65</sup> Conversely, cooking was characterized using a SMPS  
447 and APS in the real world, namely in 15 homes in Brisbane, Australia.<sup>66</sup> Elevated P<sub>num</sub> 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  
454 contributes most to the coarse particle fraction (>1µm).<sup>66</sup> OPCs are commonly used when the UFP fraction  
455 is not of interest or to supplement the measurements made by other equipment. OPC measurements can  
456 add P<sub>num</sub> and P<sub>sd</sub> 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.<sup>67</sup>

460 When high time resolution data is important, a FMPS may be used rather than a SMPS. For example, a  
461 FMPS (5.6-560nm) and OPC (0.3-20µm) were used to characterize the emissions from seven wood burning  
462 fireplaces in German homes.<sup>68</sup> Although these fireplaces largely have airtight seals, the combustion  
463 chamber needs to be opened regularly to put more wood in and this led to increased P<sub>num</sub> concentrations  
464 within the room. As the chamber only remains open for a few seconds, a shorter time resolution is required

465 than the >1-minute resolution of the SMPS. Therefore, the researchers used a FMPS to measure Pnum and  
466 Psd at 1s resolution. The nanoparticle emissions of burning incense were characterized using a FMPS within  
467 a chamber.<sup>69</sup> High time resolution data was important to understand periods of rapid change, for example  
468 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  
473 of developed low-cost sensors working on the light scattering principle.<sup>70</sup> However these investigations are  
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  
476 determine lateral variations in PM.<sup>71</sup> Similarly, these sensors have been deployed in households in Raipur,  
477 India to understand the spatiotemporal resolution of PM generated by cookstoves.<sup>41</sup> In lab tests the sensors  
478 agree well with reference grade equipment, however, these sensors become saturated at 4-5mg/m<sup>3</sup>  
479 compared to the 20 mg/m<sup>3</sup> of the SidePak, making them less suitable for high pollution environments.<sup>72</sup>

## 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),<sup>3</sup> 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.<sup>72</sup> 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

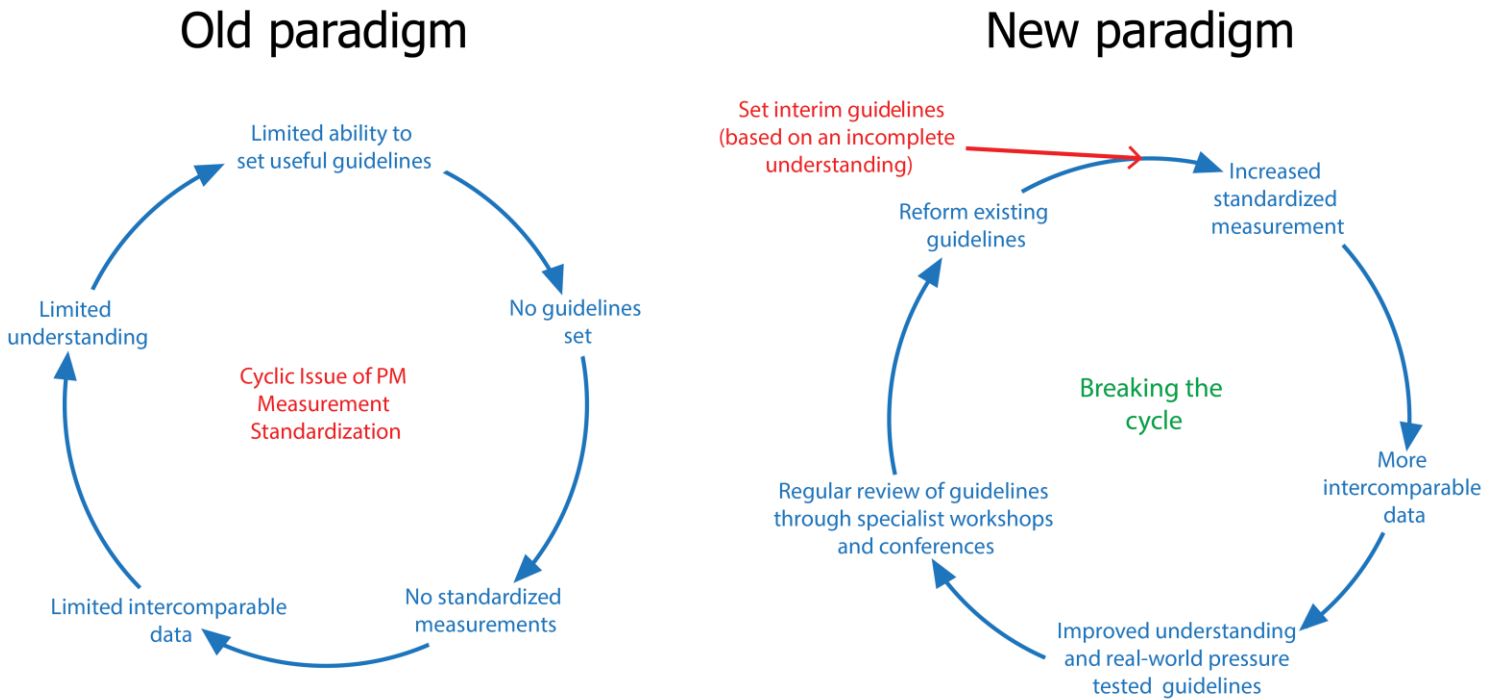
501 A challenge of the Pnum/Psd/Surface area metrics are the difficulty in comparing measured values across  
502 literature. Different investigations often operate equipment with differing size ranges or operate the same  
503 piece of equipment at different size ranges. Unfortunately, comparing measurements of different size  
504 ranges is not meaningful.<sup>8</sup> In this review paper, even the difference in minimum diameter measurement  
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.<sup>8</sup> 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.<sup>73</sup> 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.<sup>74</sup> 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.

531 However, setting these guidelines is not an easy task (especially No. 2), and therefore, a model for  
532 developing improved guidelines is illustrated in Figure 1.



534 **Figure 1.** The cyclic issue of PM Measurement standardization and breaking the cycle.

535 Initially, due to our limited understanding of the epidemiology of Pnum and surface area concentrations,  
 536 regulatory guidelines are difficult to relate to health effects, as is the case for existing Pmass guidelines.  
 537 Instead, good, satisfactory, poor and extremely poor standards, could be based on the frequency of  
 538 measurement, i.e. based on measurement frequency distributions as opposed to a mechanistic relationship  
 539 to health effects. As the "new paradigm" cycle continues and epidemiological understanding improves, we  
 540 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.<sup>75</sup>

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  
552 before.<sup>10</sup> A balance is needed between improving energy efficiency, reducing operation costs and improving  
553 IAQ.<sup>76</sup> 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  
558 questions relating to IAQ and “green buildings” are addressed by Steinemann.<sup>77</sup>

## 559 Low-cost sensors

560 Low-cost sensors are becoming increasingly important in the modern world and are essential to improving  
561 the spatial and temporal resolution of urban air pollution measurements.<sup>13,78</sup> Static and sparse air pollution  
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  
566 estimations.<sup>71</sup> It is worth noting however, that even high resolution spatio-temporal measurements may  
567 not be fully indicative of individual exposure given the “personal cloud” effect,<sup>79</sup> therefore, in the future  
568 low-cost sensors will also be needed for personal exposure measurements. Switching from wired to  
569 wireless data transmission has been shown to reduce initial investment and annual operation cost by 3x  
570 and 5x, respectively, in the US.<sup>75,80</sup> Wireless data transmission also allows for unattended large-scale  
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  
582 because it is difficult to develop particle monitors that are very sensitive to small concentrations.<sup>80</sup> Moreover,  
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.<sup>71</sup>

### 589 Applications of and Considerations for Low-cost sensors

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

597 If low-cost sensor networks are introduced nationally and a large database is generated, several issues  
598 need to be considered. It is important that the data collected is easily accessible to interested stakeholders,  
599 and that the database is not held by a private company that might misuse it for financial gain.<sup>80</sup> 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

622 Pmass is the most widely used metric of PM currently with measurements being robust, reliable, and easily  
623 compared to well defined standards and a long-standing literature. However, specific indoor health related  
624 standards are still to be developed. Pmass will remain useful due to its long history of use in compliance  
625 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 PM<sub>10</sub> and PM<sub>2.5</sub> down to PM<sub>1</sub>  
627 or PM<sub>0.5</sub> 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,<sup>26</sup>  
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.<sup>30</sup> 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  
647 epidemiological questions still need to be answered.<sup>81</sup> Firstly, which properties, or combination of properties,  
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  
658 increasing available.<sup>13,82</sup> Eventually this could lead to real-time IAQ warning systems in homes; allowing  
659 residents to more easily mitigate IAQ issues.<sup>82</sup> This level of technology would also allow us to resolve many  
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  
665 health, and which metrics are best able to capture this interaction.<sup>81</sup> A better understanding of epidemiology  
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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