

158 For consistency we demand $\langle PN \rangle = c_{PN}$, $\langle PS \rangle = c_{PS}$ and $\langle PMx \rangle = c_{PMx}$, which
 159 means that the expectation value of each metric is equal to the respective ambient particle
 160 concentration on a large scale. We can use Eq. (2) to express the expectation value of the number
 161 of particles counted $\langle N \rangle$ as stated in Eq. (6). While all three equations are valid, it makes sense
 162 to use only the equation with the corresponding ambient concentration of each metric for further
 163 evaluations respectively.

164

$$\langle N \rangle = c_{PN} V_{\text{Sample}}$$

$$\langle N \rangle = \frac{c_{PS}}{\langle S \rangle} V_{\text{Sample}}$$

165 $\langle N \rangle = \frac{c_{PMx}}{\langle \rho \rangle \langle V \rangle} V_{\text{Sample}} \tag{6}$

166

167 We further assume uncorrelated particle positions, leading to uniformly distributed particles in
 168 space. Therefore, the total number of particles in a given volume will be Poisson distributed
 169 (Badger, 1946; Hinds, 1999). Although the assumption that particle positions are uncorrelated is
 170 under debate (Damit et al. 2014; Larsen 2006) we draw the conclusion that the Poisson
 171 assumption is a valid starting point and neglect any possible correlations of particles positions
 172 such as for example a clustering of particles of same size or a complete avoidance of such
 173 clusters. In other words, we assume completely uniformly distributed particle positions.

174 According to Larsen 2006, assuming that particles are randomly distribute in a volume, the
175 Poisson distribution describes the number of particles within any subvolume.

176 Since we assume the number of particles within a sample is Poisson distributed, Eq. (7)
177 expresses the variance of N as the variance of the Poisson distribution is equal to its expectation
178 value.

179

$$180 \quad \text{Var}(N) = \langle N \rangle \quad (7)$$

181

182 ***Coefficient of Variation***

183 In order to compare the uncertainties of the metrics, we use the coefficient of variation, which
184 is the standard deviation divided by the expectation value and therefore shows the relative
185 variability. The standard deviation of PN, PS and PMx is given by the square root of the
186 respective variance as given in Eq. (3). This is the inherent variability of particle concentrations
187 on an arbitrary scale and is therefore independent of the type of PM sensor, e.g. whether the
188 sensor will analyse single particles and add them up, or take a cumulative measurement. The type
189 of sensor used will of course influence the metric of the measurement and vice versa. The
190 coefficient of variation of the measurement can be written as stated in Eq. (8) by using Eqs. (2-3)
191 and (7).

192

193
$$\sigma_{PN,rel} = \frac{\sqrt{\text{Var}(PN)}}{\langle PN \rangle} = \frac{1}{\sqrt{\langle N \rangle}}$$

$$\sigma_{PS,rel} = \frac{\sqrt{\text{Var}(PS)}}{\langle PS \rangle} = \frac{1}{\sqrt{\langle N \rangle}} \sqrt{1 + \frac{\text{Var}(S)}{\langle S \rangle^2}}$$

$$\sigma_{PMx,rel} = \frac{\sqrt{\text{Var}(PMx)}}{PMx}$$

$$= \frac{1}{\sqrt{\langle N \rangle}} \sqrt{1 + \frac{\text{Var}(V)}{\langle V \rangle^2} + \frac{\text{Var}(\rho)}{\langle \rho \rangle^2}} \quad (8)$$

194

195 In Eq. (8) we can clearly see that the coefficient of variation of each metric considered drops as
196 the sample size increases with a typical $\frac{1}{\sqrt{N}}$ behaviour. As stated in Eq. (6) $\langle N \rangle$ is directly
197 proportional to the sample volume V_{sample} and the mean ambient particle concentration in either
198 metric. Therefore, the variability decreases for increasing sample volumes and higher
199 concentrations which both leads to better statistics. Another contribution to the variability of the
200 measurement stems from the distribution of the particles surface for PS and the volume for PMx,
201 which are both directly related to the particles size distribution. In addition, the distribution of the
202 mass density affects the variability of mass related metrics PMx.

203 The formalism can easily be adapted to metrics like PM10, PM2.5 or PM1. The upper bound
204 of the metric influences the expectation value and variance of the volume of single particle's
205 $\langle V \rangle$ and $\text{Var}(V)$. For example, the variance of particle sizes of PM1 is typically lower than for

206 PM10 because the particles of interest expand over one more decade in diameter for PM10. Since
207 the volume is proportional to the diameter cubed, the variance in volume is significantly affected
208 by the particle sizes that are taken into account. Also the majority of particles is typically below 1
209 μm of diameter which means that the Poisson counting uncertainty is not greatly affected by
210 lowering the upper bound of the metric to 1 μm . For example Klejnowski et al. (2013) report that
211 during a measurement campaign in Zabrze in Poland, 99% of all particles by number
212 concentration are below 1 μm in diameter. Given a distribution $p(D)$, the variance for the metric
213 of interest can be calculated by a normalization of $p(D)$ within the size window of interest. The
214 probability outside this window is set to zero. Then again Eq. (4) and Eq. (5) can be used to
215 calculate expectation value and variance of single particle's volume respectively. Note that the
216 ambient concentration, which is used to calculate the expectation value of number of particles
217 $\langle N \rangle$, must therefore match the metric of interest.

218

219

220 **RESULTS AND DISCUSSION**

221 We begin our discussion with the uncertainty of particle number concentration measurements.

222 This uncertainty is straightforward to calculate and to estimate for a measurement as for a PN

223 measurement the number of particles counted has to be known obviously. Eq. (8) can be

224 evaluated which gives the uncertainty. The uncertainty of the measurement can be lowered by

225 prolonging the measurement until a relative uncertainty is reached that is acceptable for the user.

226 For the particle surface concentration the situation is already more complex as can be seen in Eq.

227 (8). There the expectation value and the variance of a single particle's surface, which is a function

228 of the particle size distribution, plays a role. Depending on the measurement technique, the size

229 distribution can be known or unknown. If there is no information on the particle size distribution,

230 the exact uncertainty cannot be determined. Since the situation for mass concentrations, such as

231 PM_x is similar and those metrics are the most common metrics for low-cost sensors, we focus the

232 discussion on mass metrics.

233 As shown in Eq (8) and Fig. 1 we can see that the coefficient of variation of the measured

234 PM_{2.5} value is strongly dependent on the size distribution of particles. For this evaluation, we

235 assume the particle diameter D to be lognormal distributed and set ρ to (1850 ± 140)

236 kg m^{-3} according to Hand and Kreidenweis (2002). For narrow particle size distributions, the

237 variability is dominated by the Poisson contribution and the contribution from the mass density

238 distribution. If we assume monodisperse particles the geometric standard deviation (GSD) is one
239 and $\text{Var}(V)$ is zero. In this case, we are left with the Poisson uncertainty and the small
240 contribution due to the mass density distribution. At broader particle size distributions with
241 higher GSD, the size distribution related contribution dominates. Note that the standard
242 deviations stemming from different sources do not add up but the variances do as stated in Eq. (3).
243 The total coefficient of variation for the investigated case for PM_{2.5} ranges from roughly 4 % to
244 over 30 % depending solely on the parameters of the size distribution. This major influence of the
245 underlying particle size distribution makes the estimation of confidence intervals for a real
246 measurement tricky. In a real case, users are forced to use the measured PM_x value as an estimate
247 for the ambient concentration. This is reasonable since the expectation value of the measurement
248 equals the ambient concentration when there are no systematic errors in the measurement. If the
249 measurement technique counts and sizes particles such as for example a light scattering device
250 does, then Eq.(8) can be used to estimate the uncertainty. In this case, a mean mass density for the
251 particles has to be assumed as well as a variance of the mass density. However, if the
252 measurement technique does not provide information on the particle number and the size
253 distribution, there is no estimate for the uncertainty of the measured PM_x value as the coefficient
254 of variation as stated in Eq. (8) cannot be evaluated. This is for example the case for gravimetric
255 measurements. Also the evaluation of Eq. (8) for a real measurement case will not lead to the

256 exact uncertainty as the information on the mean ambient concentration and the size distribution
257 is not exact for finite sample sizes. In this work, we have all information of the ambient situation
258 and use it in a forward calculation to find the variability of the particle concentrations. In a real
259 case, there is little to no information on the mean ambient particle concentration, the size
260 distribution and the mass density distribution of particles.

261 Mathematically speaking, the influence of the particle size distribution and the influence of the
262 mass density distribution are equal. Nevertheless, we can state that for cases, which are to be
263 expected in the ambient air, the size distribution of particles dominates. This is reasonable since
264 the particle-size distribution ranges over decades while the mass density distribution can safely be
265 assumed to extend over less than one decade. Therefore the term $\frac{\text{Var}(\rho)}{\langle \rho \rangle^2}$ is in general small
266 compared to $\frac{\text{Var}(V)}{\langle V \rangle^2}$.

267 Fig. 2 shows the uncertainty of PM1, PM2.5 and PM10 for lognormal distributed particles with
268 an exemplary geometric mean (GM) of 0.3 μm as a function of the geometric standard deviation
269 (GSD) of the size distribution. The ambient mass concentration is set to 25 $\mu\text{g m}^{-3}$ in either of the
270 metrics. The relative uncertainty in general rises as GSD and therefore the width of the particle
271 size distribution rises. At GSD values of approximately lower than 1.5, the relative uncertainties
272 of the metrics are equal as the metrics do not differ from each other as long as all particles are
273 below 1 μm in diameter. For higher GSD values, the size distribution relevant for each of the

274 metrics begin to differ, leading to a higher uncertainty of PM10 compared to PM2.5 and
275 compared to PM1. We can state that for the investigated parameters, the estimation of a mass
276 concentration based on a small sample yields results that are more robust the lower the upper
277 cutoff is. Since typically only a small number of particles has a diameter of greater than 2.5 μm
278 but their total volume is still of relevance, PM10 is among the most used mass metrics especially
279 prone to a relatively high uncertainty.

280 Fig. 3 shows the measurement uncertainty of PN, PS and PM2.5 as a function of the sample
281 volume. We assumed lognormal distributed particles with an exemplary GM of 0.3 μm and a
282 GSD of 1.7. The particle density ρ is set to $(1850 \pm 140) \text{ kg m}^{-3}$ according to Hand and
283 Kreidenweis (2002). We only took particles with diameters below 2.5 μm into account for this
284 evaluation. In general, the relative uncertainty drops as the sample volume increases. The
285 uncertainty of PM2.5 is the highest, followed by the uncertainty of the total particle surface PS.
286 The particle number concentration PN has the lowest uncertainty, which is immediately evident
287 in Eq. (8) as for the uncertainty of PMx, and PS more terms are added. In Appendix A we show
288 that for lognormal distributed spherical particles, the relative uncertainty of the particle surface
289 concentration is necessarily lower than the uncertainty of the volume concentration. The reason is
290 that the surface is proportional to the square of the diameter of the particle, whereas the volume is
291 proportional to the diameter cubed. This makes the mass metrics more prone to a relative high

292 uncertainty due to a small number of large particles. For the investigated parameters, a sample
293 volume of 1 ml yields a relative uncertainty of 6% for PN, 11% for PS, and 19% for PM2.5.
294 Especially for PM2.5, this might be a high uncertainty bearing in mind that it is only the inherent
295 uncertainty of the measurement where the sensor's imperfections are yet to be considered. For a
296 sample volume of one liter, the uncertainty of below 1% for each metric is most likely
297 overshadowed by the sensors imperfections in a realistic case, which is why this uncertainty has
298 never been observed using high-end laboratory equipment providing high flowrates and therefore
299 has not been discussed in literature yet.

300 In Fig. 4 and Fig. 5 we show the relative uncertainty for PM2.5 as a function of the total mass
301 that is sampled. As the uncertainty depends on the total number of particles that are sampled,
302 which is proportional to the sample volume times the ambient mass concentration, the total mass
303 sampled by the sensor is a reasonable parameter. In Fig. 4 the uncertainty of PM2.5 is shown for
304 lognormal distributed particles with a GM of 0.3 μm and various GSD values. The particle
305 density ρ is set to $(1850 \pm 140) \text{ kg m}^{-3}$ according to Hand and Kreidenweis (2002). In Fig. 5 the
306 same is shown for a fixed GSD value of 1.5 and various values of GM. The relative uncertainty
307 declines when higher total masses are sampled which can be either due to a high sample volume,
308 a high ambient particle mass concentration, or both. In general, the uncertainty rises for higher
309 GSD values as the size distribution becomes broader and therefore the variance of the particles

310 volume becomes higher. The uncertainty is also dependent on the geometric mean of the size
311 distribution. The lower GM the more particles are necessary to have a fixed mass concentration.

312 Fig. 6 shows the relative uncertainty of PM1, PM2.5 and PM10 as a function of the total mass
313 that is sampled at an exemplary lognormal particle size distribution with a GM of 0.3 μm and a
314 GSD of 1.7. The particle density ρ is set to $(1850 \pm 140) \text{ kg m}^{-3}$ according to Hand and
315 Kreidenweis (2002). PM1 has the lowest cutoff and therefore the lowest relative uncertainty at
316 equal masses that are sampled, whereas PM10 has the highest of the three.

317 As a rule of thumb, we propose that for PM2.5 and PM10 a minimum total mass of 100 μg
318 should be sampled, which yields a measurement uncertainty for PM2.5 of around 10% as can be
319 seen in Fig. 4 and Fig. 6. This assumes lognormal distributed particles with a geometric mean of
320 0.3 μm and a GSD of 1.7 and a particle density ρ of $(1850 \pm 140) \text{ kg m}^{-3}$. We consider this a
321 realistic worst-case scenario. At a particle mass concentration PM2.5 of $25 \mu\text{g m}^{-3}$ this would
322 mean that approximately 4 ml of volume have to be sampled, at $10 \mu\text{g m}^{-3}$ 10 ml would be
323 necessary to stay below 10% measurement uncertainty. For PM1 we propose a minimum of 30 μg
324 to be sampled in order to have a relative uncertainty of below 10%. We would like to emphasize
325 that this is not valid for arbitrary particle size distributions and mass density distributions. In
326 order to evaluate the exact uncertainty Eq. (8) has to be utilized.

327 For particle number measurements, the rule of thumb we propose is to count at least 100
328 particles, which yields a relative uncertainty of 10%. Keeping in mind that imperfections of the
329 measurement system itself come on top of this inherent uncertainty, 10% inherent uncertainty
330 may well be low compared to the uncertainties coming from technological boundaries.

331

332 **CONCLUSIONS**

333

334 We have established a model for the local variability of particulate matter concentrations on
335 arbitrary small scales and evaluated the model for various parameters. The model covers the
336 uncertainty of the particle number concentration, the surface concentration as well as mass
337 concentration such as PM1, PM2.5, and PM10. The variability is due to the small number of
338 particles within a small sample, which should represent a rather complex distribution of particle
339 size and mass density. While we suppose the variability on small scales has no influence on
340 human's health or other measurable consequences, it does have a major impact on the validity of
341 miniaturized PM sensors measurements. We find that even a sensor without any imperfections is
342 hardly able to assign a sound particle concentration to the ambient if the sample volume is on the
343 order of milliliters. Depending on the metric used, the uncertainty depends on the distribution of
344 the influencing parameters. The overall uncertainty depends on the total particle concentration
345 and the sample volume for particle number concentration, additionally there is a dependence on

346 the size distribution for particle surface concentration as well as the mass density distribution of
347 particles for mass metrics such as PM1, PM2.5 and PM10. Except for the sample volume, those
348 might be unknown during and after measurement with low-cost sensors and low sample volumes,
349 which makes the estimation of the uncertainty of the measurement hardly possible. The total
350 uncertainty is on the order of ten percent for realistic cases and a sample volume of a few
351 milliliters.

352 We have shown that for the same ambient situation the relative uncertainty of the particle
353 number is lower compared to surface concentration, which again is lower than the relative
354 uncertainty of the mass concentration. A small sample volume is therefore better suited to
355 estimate number concentration or surface concentration than it is for estimating ambient mass
356 concentrations. We have shown that when a mass metric is used, typically the estimate for PM1
357 has a lower uncertainty than the estimate for PM2.5. PM10 typically has the highest relative
358 uncertainty as for PM10 the particle sizes that are taken into account expand to the highest
359 diameters out of these metrics.

360 Miniaturized sensors with small footprints can overcome these limitations only by allowing
361 relatively long measurement times or providing high flowrates. Both of these measures are in
362 contradiction to miniaturization since both increase energy consumption, and a high flowrate
363 typically comes together with a large footprint. On top of these inherent limitations, there are the

364 sensors imperfections be accounted for. As a rule of thumb for PM_{2.5} and PM₁₀, we propose a
365 minimum total mass of 100 pg to be sampled in order to have around 10% relative uncertainty for
366 the investigated cases we consider realistic for ambient measurements. This equals typically 4 ml
367 of sample volume at an ambient mass concentration of 25 µg m⁻³. For PM₁ we propose a
368 minimum total mass of 30 pg to be sampled to have a relative uncertainty of around 10% for the
369 assumed situations. The miniaturization of particle sensors might hit a boundary at sample
370 volumes of around 1 ml due to the inherent statistical variability of particle concentrations on
371 such small volumes. In the opinion of the authors a sample volume of less than 1 ml may at
372 reasonable ambient particle concentrations be insufficient to provide sound results at all. This is
373 especially severe for mass metrics such as PM₁, PM_{2.5} and PM₁₀.

376 **ACKNOWLEDGMENTS**

377
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421

List of Figure Captions

422

423 **Fig. 1.** Coefficient of variation as a function of the geometric standard deviation of the particle
424 sizes for PM2.5. GSD corresponds to the width of the distribution. The calculation is done at an
425 exemplary geometric mean of 300 nm. For the calculation, only particles with diameters lower
426 than 2.5 μm are taken into account in order to show the uncertainty for PM2.5. The calculation is
427 shown for a constant ambient PM2.5 concentration of 25 $\mu\text{g m}^{-3}$, a particle mass density $\langle \rho \rangle$
428 of 1850 kg m^{-3} with a variance $\text{Var}(\rho)$ of $(140 \text{ kg m}^{-3})^2$, and a sample volume of 1 ml. Although
429 the ambient PM2.5 concentration is held constant and therefore the total mass that is sampled is
430 also constant, the total uncertainty ranges over tens of percent. For realistic cases, the uncertainty
431 stemming from the particle size distribution dominates. The Poisson contribution also rises as
432 GSD rises since the number of particles at a constant ambient concentration gets lower as the
433 variance of the size distribution rises.

434

435 **Fig. 2.** Coefficient of variation as a function of the geometric standard deviation of the particle
436 sizes for PM1, PM2.5 and PM10. GSD corresponds to the width of the distribution. The
437 calculation is done at an exemplary geometric mean of 0.3 μm . The calculation is shown for a
438 ambient particle mass concentration of 25 $\mu\text{g m}^{-3}$ in either metric, a particle mass density $\langle \rho \rangle$
439 of 1850 kg m^{-3} with a variance $\text{Var}(\rho)$ of $(140 \text{ kg m}^{-3})^2$, and a sample volume of 1 ml. Although

440 the ambient particle mass concentration is held constant in each metric and therefore the total
441 mass that is sampled is also constant, the total uncertainty ranges over tens of percent and is a
442 function of the upper cutoff of particle sizes relevant. We can clearly see that at a GSD of lower
443 than about 1.5, the uncertainties are almost equal as there are almost no particles that have a
444 diameter above 1 μm which makes the metrics output equal. At higher GSD, the metrics begin to
445 differ and so do the uncertainties. The lower the upper cutoff of the metric is, the lower is $\text{Var}(V)$
446 of the relevant size window. We can see that for equal distributions and similar mass
447 concentrations, the relative uncertainties of PM1, PM2.5 and PM10 differ although all of them
448 take into account mass.

449
450 **Fig. 3.** The plot shows the measurement uncertainty as a function of the sample volume. The
451 particle size distribution is assumed lognormal with an exemplary GM of 0.3 μm and a GSD of
452 1.7. The ambient PM2.5 concentration is set to $25 \mu\text{g m}^{-3}$ and the particle density ρ to $(1850 \pm$
453 $140) \text{ kg m}^{-3}$ according to Hand and Kreidenweis (2002). σ_{rel} drops as the sample volume
454 increases for each of the metrics. Particles with diameters higher than 2.5 μm are neglected for
455 each metric. The relative uncertainty of PN is the lowest, followed by PS and PM2.5 having the
456 highest uncertainty.

457

458 **Fig. 4.** The plot shows the relative uncertainty of PM_{2.5} as a function of the total mass that is
459 sampled. A lognormal particle size distribution with an exemplary GM of 0.3 μm is assumed
460 whereas results for various GSD values are shown. Assuming a PM_{2.5} concentration of 25 $\mu\text{g m}^{-3}$
461 a volume of 4 ml is typically necessary in order to sample a total mass of 100 μg . Depending on
462 the GSD of the particle size distribution, the relative uncertainty of PM_{2.5} can differ significantly.

463
464 **Fig. 5.** The plot shows the relative uncertainty of PM_{2.5} as a function of the total mass that is
465 sampled. A lognormal particle size distribution with a GSD of 1.5 is assumed. The results for
466 various GM values are shown. Depending on the GM, the relative uncertainty of PM_{2.5} can
467 differ significantly. This is reasonable since at lower GM values, a greater number of particles is
468 necessary to have the same mass concentration as for higher GM values. The greater number of
469 particles lowers the relative uncertainty.

470
471 **Fig. 6.** The plot shows the relative uncertainty of PM₁, PM_{2.5} and PM₁₀ as a function of the
472 total mass that is sampled. For each of the metrics the same lognormal particle size distribution
473 with a GM of 0.3 μm and a GSD of 1.7 is assumed. The relative uncertainties of PM₁, PM_{2.5} and
474 PM₁₀ differ due to the upper cutoff in particle size that is considered. At a total sampled mass of

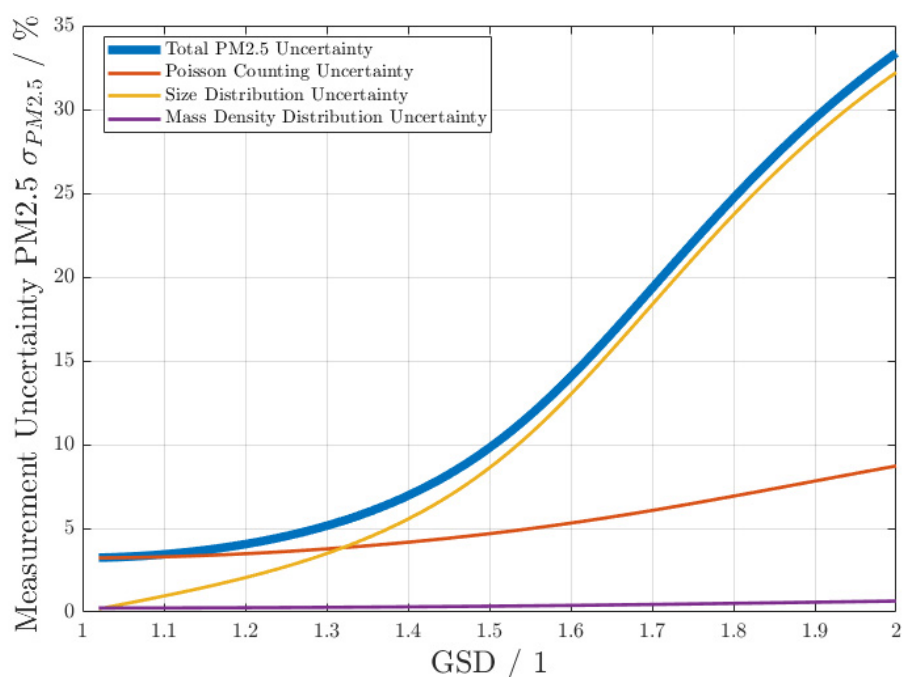
475 100 pg, the uncertainty of PM1 is 5%, for PM2.5 it is 10% and for PM10 is at about 11%. For

476 PM1 30 pg are sufficient for the assumed parameters to have a relative uncertainty below 10%.

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479

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481 sizes for PM2.5. GSD corresponds to the width of the distribution. The calculation is done at an

482 exemplary geometric mean of 300 nm. For the calculation, only particles with diameters lower

483 than 2.5 μm are taken into account in order to show the uncertainty for PM2.5. The calculation is

484 shown for a constant ambient PM2.5 concentration of 25 $\mu\text{g m}^{-3}$, a particle mass density ρ

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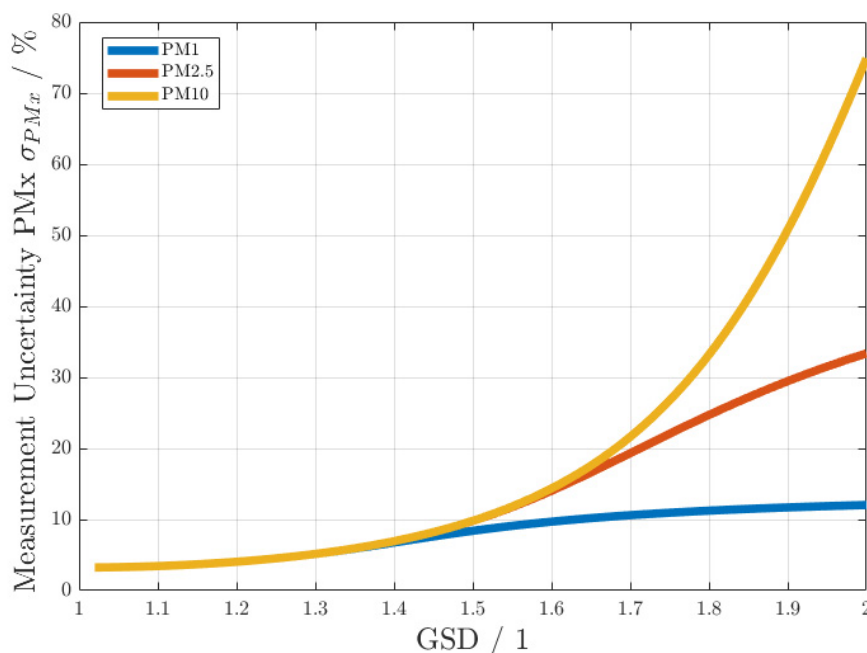
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488 stemming from the particle size distribution dominates. The Poisson contribution also rises as

489 GSD rises since the number of particles at a constant ambient concentration gets lower as the

490 variance of the size distribution rises.



491

492 **Fig. 2.** Coefficient of variation as a function of the geometric standard deviation of the particle

493 sizes for PM1, PM2.5 and PM10. GSD corresponds to the width of the distribution. The

494 calculation is done at an exemplary geometric mean of $0.3 \mu\text{m}$. The calculation is shown for a

495 ambient particle mass concentration of $25 \mu\text{g m}^{-3}$ in either metric, a particle mass density $\langle \rho \rangle$

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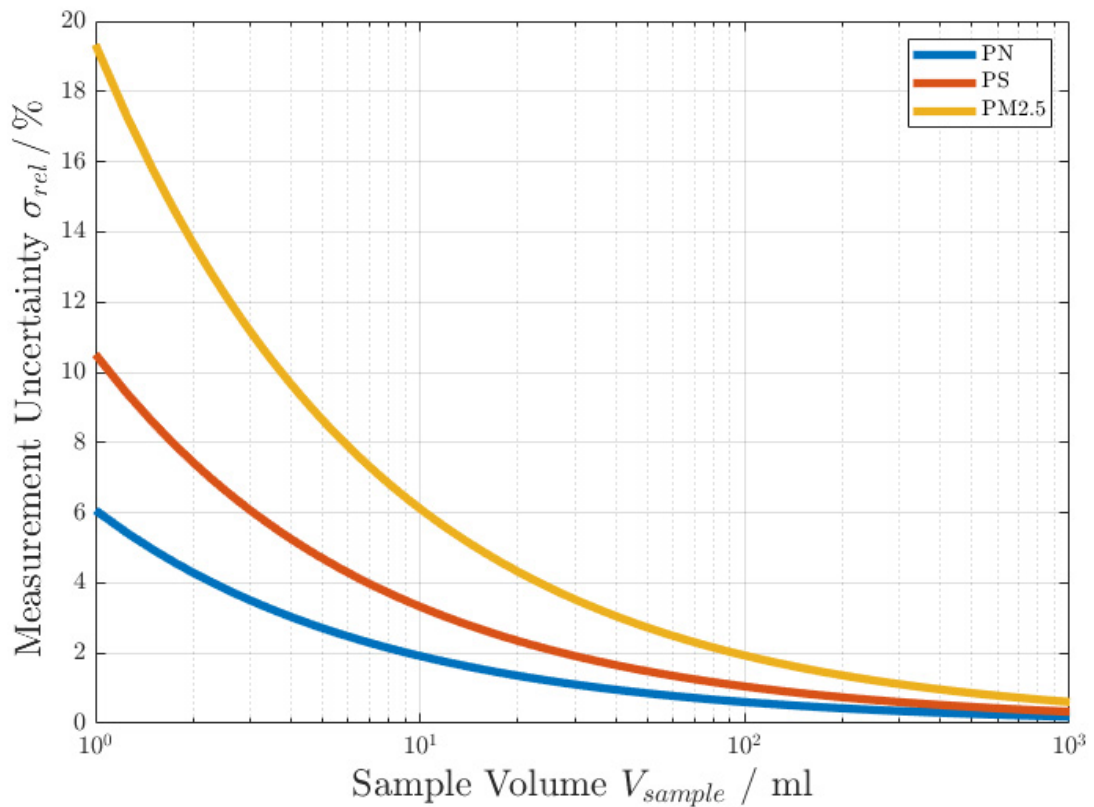
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504 concentrations, the relative uncertainties of PM1, PM2.5 and PM10 differ although all of them
505 take into account mass.

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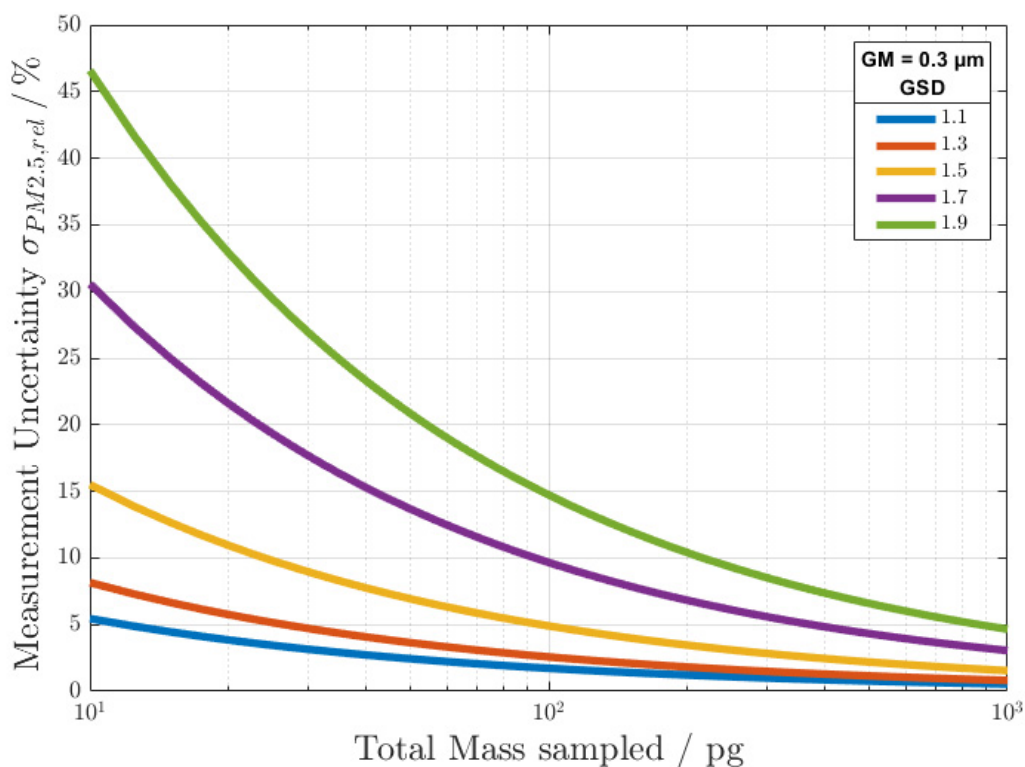
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Fig. 3. The plot shows the measurement uncertainty as a function of the sample volume. The particle size distribution is assumed lognormal with an exemplary GM of $0.3 \mu\text{m}$ and a GSD of 1.7. The ambient PM2.5 concentration is set to $25 \mu\text{g m}^{-3}$ and the particle density ρ to $(1850 \pm 140) \text{kg m}^{-3}$ according to Hand and Kreidenweis (2002). σ_{rel} drops as the sample volume increases for each of the metrics. Particles with diameters higher than $2.5 \mu\text{m}$ are neglected for each metric. The relative uncertainty of PN is the lowest, followed by PS and PM2.5 having the highest uncertainty.



518

519 **Fig. 4.** The plot shows the relative uncertainty of PM2.5 as a function of the total mass that is

520 sampled. A lognormal particle size distribution with an exemplary GM of 0.3 μm is assumed

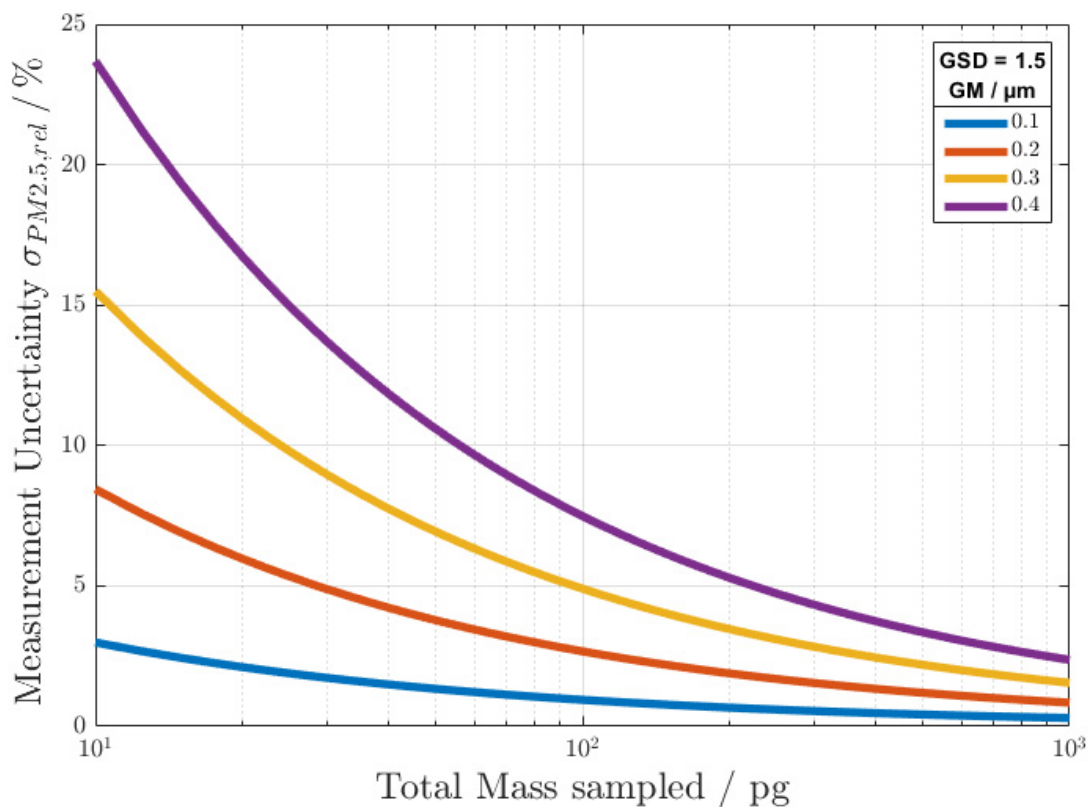
521 whereas results for various GSD values are shown. Assuming a PM2.5 concentration of 25 $\mu\text{g m}^{-3}$

522 a volume of 4 ml is typically necessary in order to sample a total mass of 100 pg. Depending on

523 the GSD of the particle size distribution, the relative uncertainty of PM2.5 can differ significantly.

524

525



526

527 **Fig. 5.** The plot shows the relative uncertainty of PM_{2.5} as a function of the total mass that is

528 sampled. A lognormal particle size distribution with a GSD of 1.5 is assumed. The results for

529 various GM values are shown. Depending on the GM, the relative uncertainty of PM_{2.5} can

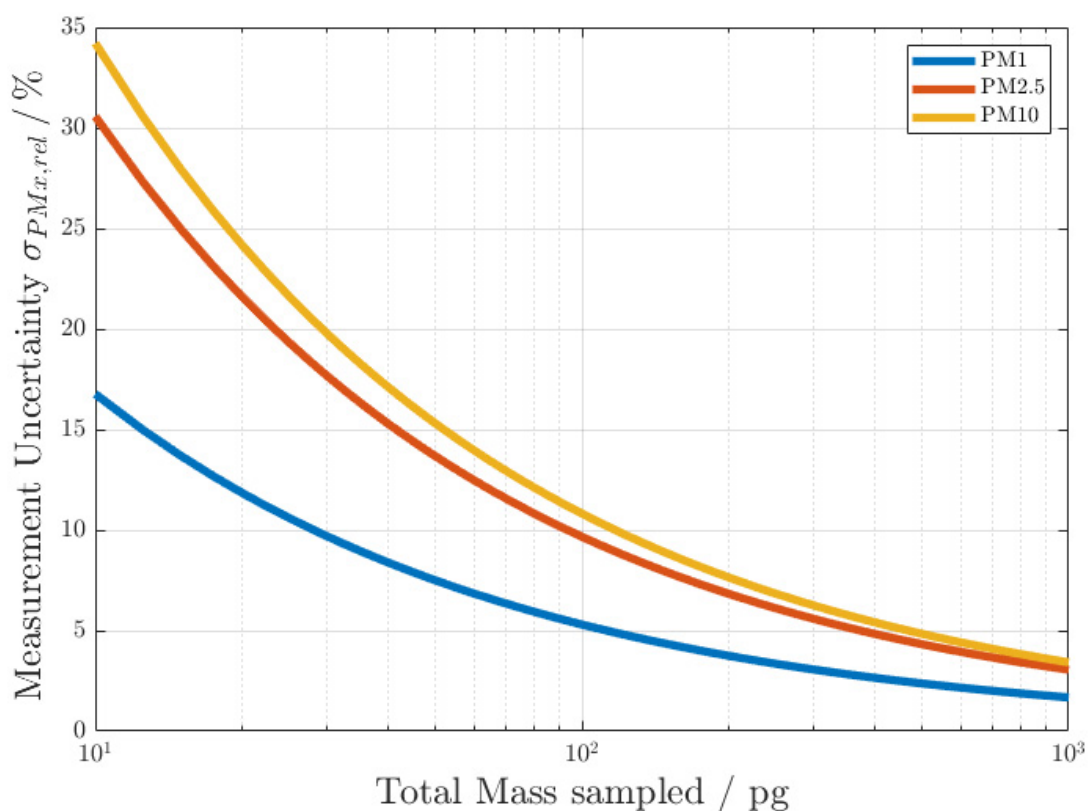
530 differ significantly. This is reasonable since at lower GM values, a greater number of particles is

531 necessary to have the same mass concentration as for higher GM values. The greater number of

532 particles lowers the relative uncertainty.

533

534



535

536 **Fig. 6.** The plot shows the relative uncertainty of PM1, PM2.5 and PM10 as a function of the

537 total mass that is sampled. For each of the metrics the same lognormal particle size distribution

538 with a GM of 0.3 μm and a GSD of 1.7 is assumed. The relative uncertainties of PM1, PM2.5 and

539 PM10 differ due to the upper cutoff in particle size that is considered. At a total sampled mass of

540 100 pg, the uncertainty of PM1 is 5%, for PM2.5 it is 10% and for PM10 is at about 11%. For

541 PM1 30 pg are sufficient for the assumed parameters to have a relative uncertainty below 10%.

542

543

544 Appendix

545

546 **A**

547

548 We can compare the relative standard deviations of the metrics we investigated as stated in Eq.
549 (8) in terms of magnitude. It is obvious that for the same conditions the particle number
550 concentration has the lowest relative uncertainty since for PS and PMx additional terms add to
551 the total uncertainty.

552 The comparison of surface concentration and mass concentration is more complex.

553 We can state that $\sigma_{PS,rel}$ is proportional to $\frac{Var(S)}{\langle S \rangle^2}$ while $\sigma_{PMx,rel}$ is proportional to $\frac{Var(V)}{\langle V \rangle^2}$. If

554 $\frac{Var(V)}{\langle V \rangle^2}$ is always greater than $\frac{Var(S)}{\langle S \rangle^2}$, $\sigma_{PMx,rel}$ is necessarily greater than $\sigma_{PS,rel}$. In terms of the
555 size distribution $p(D)$ we can interpret the expectation value of the volume of a single particle as
556 the second moment of the distribution times a constant by assuming spherical particles, as can be
557 seen in Eq (9). The variance is proportional to the sixth moment of the distribution as shown in
558 Eq. (10).

559

560
$$\langle V \rangle = \frac{1}{6} \pi \int_0^\infty p(D) D^3 dD \quad (9)$$

561
$$Var(V) = \langle V^2 \rangle - \langle V \rangle^2 = \left(\frac{1}{6} \pi \right)^2 \int_0^\infty p(D) D^6 dD - \left(\frac{1}{6} \pi \int_0^\infty p(D) D^3 dD \right)^2$$

562 (10)

563 In a similar way, we can state that the expectation value of the surface of a single particle is
564 proportional to the second moment of the distribution, while the variance is proportional to the
565 fourth moment.

566 If we assume a lognormal distribution of the particle diameter, we can use Eq. (11) to calculate
567 the n-th moment of the distribution.

568

569
$$E(X^n) = e^{n\mu + n^2 \sigma^2 / 2} \quad (11)$$

570

571 We can use Eq. (12) to postulate Eq. (13) .

572
$$\frac{Var(A)}{\langle A \rangle^2} = \frac{\langle A^2 \rangle - \langle A \rangle^2}{\langle A \rangle^2} = \frac{\langle A^2 \rangle}{\langle A \rangle^2} - 1 \quad (12)$$

573 $\frac{\langle V^2 \rangle}{\langle V \rangle^2} > \frac{\langle S^2 \rangle}{\langle S \rangle^2}$ (13)

574 If we can show that Eq.(13) is valid for any lognormal distribution, then the relative
 575 uncertainty of the volume and therefore the mass concentration is greater than the uncertainty of
 576 the surface concentration for any lognormal distribution. We use Eq. (11) to proof Eq. (13).
 577

$$\frac{e^{6\mu+36\sigma^2/2}}{e^{6\mu+18\sigma^2/2}} \geq \frac{e^{4\mu+16\sigma^2/2}}{e^{4\mu+8\sigma^2/2}}$$

578

$$e^{9\sigma^2} \geq e^{4\sigma^2}$$

579

$$e^{5\sigma^2} \geq 1$$

580 Since $\sigma^2 \geq 0$ we have shown that the relative variability of the surface concentration is
 581 necessarily higher than the variability of the mass distribution for spherical particles from a
 582 lognormal distribution. Only for cases where $\text{Var}(\rho) = 0$ and σ of the log-normal distribution
 583 approaches zero, leading to a monodisperse particle size distribution, the variability of the surface
 584 concentration and the variability of the mass concentration are equal to each other and are equal
 585 to the relative variability of the particle number concentration which can easily be seen in Eq. (8).

586 Therefore, we can state Eq. (14) which is valid for spherical lognormal distributed particles.

587

588 $\sigma_{PMx,rel} \geq \sigma_{PS,rel} \geq \sigma_{PN,rel}$ (14)

589

590