

## ***Interactive comment on “Facility for generation of ambient-like model aerosols in the laboratory: application in the intercomparison of automated PM monitors with the reference gravimetric method” by Stefan Horender et al.***

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We would like to thank Reviewer 2 for the valuable feedback, discussion and comments that have helped us to improve the quality of the manuscript. Below please find the point-by-point response to the Reviewer's comments as well as the sections of the manuscript that were modified.

Reviewer 2: General comments: I have one major comment: The strategy of generating ambient-like particle mixtures in the lab is more convenient and reproducible than

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a field campaign, but seems to have the same main weakness for validation and calibration – it is difficult to understand which aerosol component(s) and properties (size, shape, composition, . . .) are responsible for discrepancies between instruments, and hence how to make improvements. For example, here the authors are puzzled why the DustTrak overestimates mass. Aside from the SOA-coated soot, the aerosol population in the flow tube is an external mixture. For the current application to particle mass sensors I think it would be easier and more conclusive to test with each particle type separately? But given the emphasis of the current paper, what role do the authors see for using ambient-like aerosol mixtures beyond proof of concept?

Answer: We believe that we should distinguish between i) instrument characterisation and ii) instrument calibration.

i) We agree with the Reviewer that experiments with single-component aerosols would be valuable for characterising certain aspects of the measuring instruments, such as the 50% cut-off at small particle sizes (150 nm - 300 nm) in the case of light-scattering detectors, which depends on material properties. Such experiments can easily be performed with the facility presented here by simply switching on the aerosol generator of interest (and keeping all other generators switched off).

ii) For calibrating PM monitors with respect to PM mass concentration, however, the use of single-component aerosols (e.g. dry mineral dust) would be unrealistic. It is known that the TEOM suffers from artefacts related to losses of (semi)volatile material. The light-scattering PM monitors suffer from artefacts due to hygroscopic growth of the particles and from biases related to the built-in algorithms which convert the measured particle size distributions into mass concentrations by assuming a size-dependent particle density function. Therefore, for the PM mass calibration to be meaningful, the calibration aerosol needs to comprise the right fraction of hygroscopic and volatile compounds, exhibit a realistic particle size distribution (accumulation and coarse mode) and a realistic size-dependent density. This can only be achieved by generating complex ambient-like aerosols in the laboratory. In the last three months, i.e. since the

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manuscript was posted online, we have already been able to go beyond "proof-of-concept". We have used complex aerosol mixtures to characterise a newly developed Black Carbon monitor with aerosols of different single scattering albedo, and calibrate a commercial PM low-cost sensor as well as two light-scattering PM monitors against the reference gravimetric method.

Reviewer 2: Title/abstract: Would the authors like to give a name/acronym to the facility? This may sound like a strange suggestion but could help with adoption and referencing in future!

Answer: We agree with the Reviewer's suggestion and we have changed the title to "Facility for production of ambient-like aerosols (PALMA) in the laboratory: application in the intercomparison of automated PM monitors with the reference gravimetric method". We have made a similar change in the "Abstract" and "Conclusions" of the manuscript.

Reviewer 2: Line 18 and line 115: please clarify photo-oxidation vs ozonolysis. What is the light wavelength in the Micro Smog Chamber? Which aging processes (ozonolysis, direct photolysis, OH oxidation etc. . .) occur under these conditions?

Answer: The UVC lamps in the MSC emit at  $\lambda=254$  nm and 185 nm. Since the relative humidity in the photo-oxidation reactor remains <5 % (see Section 2.1), the main ageing process is the ozonolysis of  $\alpha$ -pinene. The generation of OH radicals is negligible at such low RH. In the abstract, we have made the following clarification: "Model aerosols containing elemental carbon, secondary organic matter from the ozonolysis of  $\alpha$ -pinene...".

Reviewer 2: Line 117: How is alpha-pinene delivery controlled? Is the concentration monitored or can an estimate be provided?

Answer: We have added the following clarification in the text: "The concentration of  $\alpha$ -pinene at the inlet of the MSC was determined with a photoionization detector (PID

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PhoCheck TIGER, Ion Science Ltd, UK) after filtering out the particles. The concentration could be varied by adjusting the flow of air through the  $\alpha$ -pinene container (gas bubbler) and typically ranged between 60-70 ppm".

Reviewer 2: Line 155: The tube is marked as 2.1 m in Fig 1. Please clarify.

Answer: The length of the tube is indeed 2.1 m and therefore Fig. 1 is correct. We have corrected, however, the first sentence of Section 2.2 as follows: "The homogenizer is a 2.1-m-long custom-made stainless steel tube with an inner diameter of 16.4 cm". Thank you to the Reviewer for pointing out this inconsistency between Figure 1 and the text in Section 2.2.

Reviewer 2: Line 212: Please define "coverage factor".

Answer: Thank you to the Reviewer for pointing this out to us. The sentence now reads: "By calculating the standard deviation of all 28 measured data points, the spatial inhomogeneity of the aerosol in terms of number concentration was found to be 1.3 % for coverage factor  $k=1$  (i.e. 68 % confidence level) or 2.6 % for  $k=2$  (i.e. 95 % confidence level)."

Reviewer 2: Line 313: 10-20% "other material" is attributed to contamination or mineral coatings in Fig 5. This seems concerningly high given the controlled aerosol production. What purities of ammonium sulfate/nitrate and water were used in the atomiser? Are the collection substrates or other aspects of aerosol chemical analysis potential contamination sources? If the mineral dust is analysed separately, how much "other material" is identified?

Answer: We believe that the major source of impurities is contamination of the flow tube homogeniser with NaCl from previous experiments (aerosol spatial homogeneity studies, Figure 3). Although we made sure to flush the tube with clean air for several hours before each experiment, it seems there were still NaCl particles deposited in the aerosol inlet. By cleaning meticulously the aerosol inlet with wet wipes, it is pos-

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sible to keep the mass fraction of "other material" well below 10%. We have added a clarification in Table 2.

Reviewer 2: Line 326-382: I suggest this discussion and Fig 6 be re-ordered to start with the currently labelled aerosol 2 and 3. The discussion around aerosol 1 is confused by technical problems with the DustTrak and soot source.

Answer: The Reviewer is right that the experiments related to Aerosol 1 are complicated by the fact that i) the soot concentration decreased during the PM sampling duration and ii) the DustTrak failed to measure correctly. However, we think that there is merit in presenting the experiments in the same order in which they were performed. In this way, the readers can follow the progress achieved in the course of the experiments: in the case of Aerosol 2 and 3 both technical issues were rectified. If we were to rearrange the order in which the experiments are presented and discussed, the readers would be left with the impression that the aerosol generation cannot be stabilised, which is not true. The experiments with Aerosols 2 and 3 (as well as subsequent experiments we have performed since then) prove that the aerosol generation can remain stable over several hours.

If the Reviewer agrees, we would suggest to leave the discussion (and Figure 6) as is.

Reviewer 2: In general the composition of the three model aerosols seems similar and it's unclear if we are meant to focus on any differences? The main differences seem to be RH and temperature.

Answer: Model aerosols 1,2 and 3 differ in temperature, RH, mass concentration and chemical composition. It is true that the difference in chemical composition is not so pronounced: The % mass fractions of the different chemical constituents was varied in the range  $\approx 30\text{--}40\%$  OM,  $\approx 5\text{--}15\%$  EC,  $\approx 7\text{--}15\%$  nitrate,  $\approx 5\text{--}15\%$  sulphate and  $\approx 10\text{--}20\%$  mineral dust. This is in accordance with past studies on European continental aerosols. See, for instance, the pie charts with the chemical composition of urban, suburban and rural PM10 aerosols in Switzerland (page 7, in german):

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[https://www.empa.ch/documents/56101/246436/chem\\_char\\_pm10/d7e07ec3-0442-4749-b34f-3d26c9687038](https://www.empa.ch/documents/56101/246436/chem_char_pm10/d7e07ec3-0442-4749-b34f-3d26c9687038)

We could easily increase the % mass fraction of any of the chemical components (OC, OM, inorganic ions, mineral dust) up to at least 80 %. However, such aerosols would not be representative of typical ambient aerosols any more. To summarise: By generating model aerosols 1, 2 and 3 we wanted to showcase the potential of the facility to produce "customised" aerosols which differ in T, RH, mass concentration and chemical composition. The focus of the study was on European continental aerosols.

Reviewer 2: Line 355: The role of hygroscopicity and aerosol liquid water in influencing the performance of particle mass sensors (e.g. Crilley et al., 2018; Di Antonio et al, 2018) should be mentioned. The evaporation of water in the TEOM due to heating may be at least as important as nitrate/organic matter. Liquid water is a potential source of difference for all the online methods (70% RH) when comparing to the gravimetric reference (47.5% RH) especially if hygroscopic components effloresce on the filter. In future, the water content of the aerosol could be estimated from composition, RH and temperature using a thermodynamic model such as E-AIM (<http://www.aim.env.uea.ac.uk/aim/aim.php>).

Answer: Thank you to the Reviewer for the valuable comments. We have modified the sentence referring to TEOM artefacts as follows: "Since the aerosol stream sampled by the TEOM 1405 is heated to 30 °C, a fraction of the (semi)volatile components (e.g. nitrate, secondary organic aerosol and water) evolves into the gas phase and is therefore not collected on the filter". We have also modified the following section on light-scattering PM monitors: "Deviations may occur if the built-in functions differ substantially from the real density function of the aerosol. Hygroscopic growth of aerosol particles can also lead to considerable measurement artefacts especially when low-cost PM sensors are used (Di Antonio et al., 2018; Crilley et al., 2018)". In the future, we will try to estimate the water content by using a thermodynamic model such as E-AIM (<http://www.aim.env.uea.ac.uk/aim/aim.php>).

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Reviewer 2: Line 369: Capitalise “in”.

Answer: Thank you to the Reviewer for spotting this typo. The sentence now begins with a capital letter “In...”.

Reviewer 2: Line 390: The data in the paper focuses on 20-40  $\mu\text{g}/\text{m}^3$ . It’s hard to conclude from this that the stable operating range is “few to 500  $\mu\text{g}/\text{m}^3$ ”. Please include some additional evidence or modify this statement.

Answer: The Reviewer is right that the experiments presented in the manuscript were performed in the mass concentration range 20 - 40  $\mu\text{g}/\text{m}^3$ . The reason for this is that the EU target values (annual mean values) for PM<sub>2.5</sub> and PM<sub>10</sub> are 25  $\mu\text{g}/\text{m}^3$  and 40  $\mu\text{g}/\text{m}^3$ , respectively, and we wanted to focus on mass concentration ranges which are relevant for the EU member states. We can, however, generate up to about 80  $\mu\text{g}/\text{m}^3$  of fresh soot (depending on particle size), 150  $\mu\text{g}/\text{m}^3$  of aged soot, >500  $\mu\text{g}/\text{m}^3$  of inorganic salts and >500  $\mu\text{g}/\text{m}^3$  of mineral dust in the flow tube homogeniser (i.e. after dilution). Below please find a figure that presents PM<sub>10</sub> mass concentration measured by a low-cost optical sensor. The PM sensor reports an average concentration of about 650  $\mu\text{g}/\text{m}^3$ . The reference concentration was  $925 \pm 60 \mu\text{g}/\text{m}^3$ . This measurement shows that the facility can also produce higher particle loadings in a stable manner.

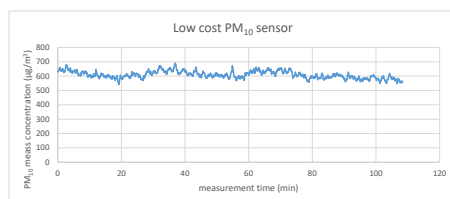
Reviewer 2: Figure 6: Please use different marker shapes as well as colours for the different instruments.

Answer: We thank the Reviewer for this suggestion. We have modified Fig. 6 so that each data set is marked with a different symbol (circles for TEOM, squares for the DustTrak and triangles for the Fidas Frog). The datasets were already marked in different colours but they might be hard to distinguish if the manuscript is printed on grayscale.

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**Fig. 1.** Calibration of low-cost optical sensor at PM<sub>10</sub> mass concentrations larger than 500  $\mu\text{g}/\text{m}^3$ .

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