Author Reply to Anonymous Referee #1

We would like to thank Referee #1 for the useful comments, which help clarifying and improving our paper. Answers to specific issues of broader relevance are addressed below, while detailed suggestions for rephrasing sentences and correcting typos will be considered accordingly in the revision of the manuscript. We will also add a point-to-point reply to the revised paper.

GENERAL REMARKS

1. Comment on title: A more appropriate title might be "Intercomparison Study of the CAPS PMex (Cavity Attenuated Phase Shift Particle Light Extinction Monitor) with the combination of an Integrating Nephelometer and a Filter-based Light Absorption Photometer".

Reply: We discussed and decided to modify the title to: "Intercomparison of a Cavity Attenuated Phase Shift-Based Extinction (CAPS PM_{ex}) Monitor with an Integrating Nephelometer and a Filter-Based Absorption Monitor". This title is less specific and contains all relevant information.

2. Comment on aerosol mixtures: "Have you measured or performed any calculations to support your assumption that particles are not lost in the sample tubing between mixing chamber and the various instruments? This entire study is based on your assumption that each instrument is sampling the same aerosols. This is not always the case when instruments sample in parallel. Each line appears to have a different flow rate. The loss mechanisms will therefore be different in the tubing leading to each instrument, and size-dependent particle losses could affect the results."

Reply: We have not performed loss calculations during our studies but designed the sampling set-up in a way that the lengths of tubing to the instruments were as short as possible (in general < 1 m) and almost equal for the different instruments and the flow splits at bifurcations were as close to equal as possible. Furthermore, considered particle sizes ranged from 0.1 µm (lower limit set by optical activity of particles at visible light) to approx. 1 µm; see Fig. 10 of Petzold et al. (2012). For this range of particle sizes the following line loss estimates were performed based on Hinds (1999) and AEROCALC (P. Baron, 2001).

Pathlength adjustment studies: For the adjustment of the pathlength (see Fig. 1 of Petzold et al. (2012)), CAPS PMex and OPC were sampling both with approx. 1 lpm, and tube length downstream the bifurcation were approx. 1 m each. For the pathlength adjustment studies, particle loss effects can be neglected because they will be of equal magnitude for both instruments.

Aerosol mixture studies: All key instruments for the intercomparison were connected to the same branch downstream the bifurcation (45° angular split) which split the flows to the MAAP on one hand and to the other instruments on the other hand; see Fig. 2 of Petzold et al. (2012). The next flow split divided the NEPH flow (11 lpm) from the flow to the other instruments (in total, 3 lpm). We performed loss estimates starting with this flow split for the NEPH line as upper limit estimate. Line properties are line length 1 m, flow 11 lpm, velocity 2.5 m/s. For particles of 100 nm in diameter we found diffusion loss < 1%; inertia loss at bends of 45deg < 1%; losses by gravitational settling are negligible for sizes from 0.1 to 1 µm in diameter. Concluding from this loss estimate we argue that particle losses due to inertia and diffusional processes are of the order of 1% and can therefore be neglected in our data analysis. A paragraph on the line loss estimate will be added to the revised manuscript.

3. Comment on mixing chamber: "Is a 3 liter mixing chamber large enough given a total flow rate to all instruments in excess of 22 lpm? A larger mixing chamber damps out variability in the ambient or generated source aerosols."

Reply: We are comparing properties of a mixed aerosol after having passed a tube of approx. 3 m length with turbulent flow conditions (flow of 22 lpm, tube inner diameter of 0.95 cm). Hence, we assume well mixed conditions.

4. Comment on instrument temperature: "Unless the PSAP was a modified instrument (with internal T and p sensors), the authors are making the assumption that the pressure and temperature at the PSAP filter was quite similar to that measured inside the nephelometer. This is probably a reasonable assumption for the pressure, but the internal T of the nephelometer can be 4-5C above ambient because of the heating of the internal volume by the lamp."

Reply: The PSAP was a commercial instrument with no internal sensors so that NEPH sensors were used for temperature and pressure adjustment. Assuming heating of the NEPH by the lamp of 5K above the ambient level of approx. 300 K as suggested by the referee and keeping the pressure unchanged would result in an overestimation of σ_{ap} by 1.5%. Since we have no exact data on the PSAP internal temperature available, we decided to neglect this correction.

5. Comment on the wavelength adjustment of σ_{ap}: "The authors are assuming they know that the theoretical small particle limit of $1/\lambda$ wavelength dependence of absorption applies in these tests. That is a big assumption, even for the laboratory aerosols, and appears to be a 'quick and dirty' approach. The ambient aerosols could have a substantially different wavelength dependence of absorption, especially if significant amounts of organic aerosols were present (likely in that suburban setting). A more robust way of scaling the PSAP data to 630 nm would be to use log interpolation of the 530 and 660 nm absorption coefficients. This is an easy and defensible way to scale the PSAP absorption to 630 nm. At the very least a comparison should be performed on the wavelength-adjusted PSAP data from the log-interpolation and $1/\lambda$ methods to convince yourselves (and readers) that they are not significantly different."

Reply: Following the recommendations of referee #1 the data analysis was slightly modified. First, we used the method of log-interpolation to adjust σ_{ap} from 660 nm to 630 nm and compared the values to the formerly applied method using the $1/\lambda$ - relationship. For the ambient aerosol data set, the absorption coefficient is underestimated by <2% on average when applying the $1/\lambda$ - relationship; see Fig. 1 for details. For the laboratory test aerosols deviations between both methods are < ±1%. Although the deviations between the recommended approach of logarithmic interpolation and the applied simpler method using the $1/\lambda$ - relationship were small, we changed the data analysis scheme as follows:
The values for σ_{sp} and σ_{ap} were determined for the wavelength 467 nm and 660 nm as described in the manuscript. Then the value for σ_{ep} (NEPH + PSAP) was adjusted to a wavelength of 630 nm by using the measured extinction Ångström exponent for the wavelength pair 467 nm/660 nm. This is a robust and sound approach. Additionally the value for σ_{ap} was adjusted to 630 nm by logarithmic interpolation. The new scheme will be shown in the revised manuscript as modified Fig. 3 (see Fig. 2 of the reply) and described accordingly. As the changes of the absolute values are small, the new data analysis scheme has no impact on the drawn conclusions.

6. Comment on pathlength adjustment: "Is this pathlength adjustment the same for all CAPS PMex instruments, or do you have to go through the process of 'calibrating' each one using PSL spheres for its own unique pathlength (I am asking this question because of the authors' association with the commercial production and manufacture of this instrument)? It is also important for readers that reference this paper to know whether they will also have to apply a correction factor of 1.05 to their CAPS extinction data or whether the firmware can take care of that in future models."

Reply: The pathlength adjustment was conducted for a single instrument. However, side-by-side run data as shown in Fig. 7b provide evidence that the pathlength misalignment from prototype instruments is similar for all prototypes. Finally all CAPS PMex instruments are now delivered with the 5% pathlength adjustment already included and users don't have to apply this adjustment again. We will make a clear statement on this issue in the revised manuscript.

7. Comment on relative humidity effects: "If the NEPH internal RH sensor was reading in excess of 80% at times inside the warm NEPH instrument, it is at least possible that there was condensation in some of the cooler sampling lines. High-RH air entering the PSAP would have likely caused the PSAP filter-based absorption data to be noisy, and possibly inaccurate, during these periods. If there was in fact condensation in the sampling lines, the amount of aerosols getting through these individual lines is unknown. Can the authors discount the possibility of condensation during the high-RH episodes?"

Reply: As for the laboratory test aerosol, the ambient aerosol was transported in a single line from the PM10 inlet to the bifurcation close to the instruments. In the potential case of condensation in the sampling lines this would affect all instruments in the same way. An impact on the instrument intercomparison can thus be rated as of minor relevance, although we cannot exclude it entirely. We will include a sentence on this issue.

8. Comment on disagreement between methods: "There are a number of possible explanations for this small level of disagreement.' What about organic aerosols? There are papers in the literature that suggest organic particles may influence the absorption measurement of the PSAP (and other filter-based instruments). See, for example, Lack et al. 2008 and Cappa et al., 2008, both in AS&T. This should at least be mentioned as a possibility for the disagreement on ambient aerosols, although this study (and some others) does not appear to support the Lack et al. and Cappa et al. results of a large positive bias in the PSAP measurement of ambient light absorption."

Reply: The observed disagreement can indeed be explained by an enhanced absorption measurement potentially caused by organic coatings on the filter. Since this phenomenon occurred during conditions with high organic loading we will mention this potential source for an overestimation of light extinction from a measurement artifact in the absorption measurement together with the recommended references (Cappa et al., 2008; Lack et al., 2008).

9. Comment on Table 3: "The CAPS/NEPH-PSAP ratio for AS aerosols appears to be related to aerosol amount. At the higher extinction levels the ratio is lower, while the lower extinction levels show the largest ratios. What can you say about this?"

Reply: We have no interpretation of this effect. For the pure AS runs the observed ratios between scattering and extinction are of the order of 1.01. So an erroneous compensation of light scattering by the PSAP can be excluded.

Figures:

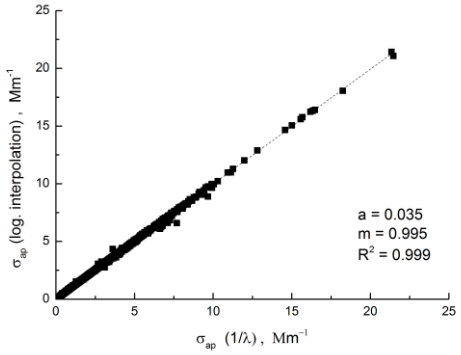

Figure 1. Comparison of wavelength-adjusted σ_{ap} data by using $1/\lambda$ (x-axis) and log. interpolation (y-axis).

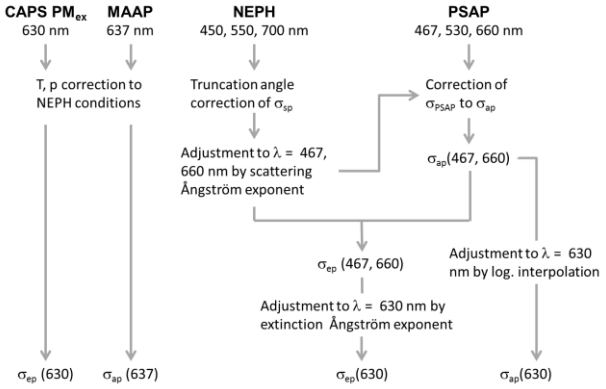

Figure 2. Revised schematic of the data inversion procedures for the optical instruments.

References

Cappa, C. D., Lack, D. A., Burkholder, J. B., and Ravishankara, A. R.: Bias in filter-based aerosol light absorption measurements due to organic aerosol loading: Evidence from laboratory measurements, Aerosol Science and Technology, 42, 1022-1032, doi: 10.1080/02786820802389285, 2008.

Hinds, W. C.: Aerosol Technology: Properties, Behaviour and Measurement of Airborne Particles., John Wiley & Sons, Inc., New York, 483 pp., 1999.

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