The paper presents results of laboratory measurements of aerosol optical properties measured with a CAPS PMssa, a nephelometer and a filter-based absorption photometer, the TAP. Different types of BC particles were produced with a nebulizer and a burner. Also, purely scattering aerosols (ammonium sulfate) were nebulized. The data were used for calculating scattering and absorption, single-scattering albedo and Ångström exponents of extinction, scattering and absorption. The results show that scattering, absorption and extinction and their wavelength dependencies - except AAE - can be measured in the aerosol phase with the CAPS PMssa with a reasonably low uncertainty. This is very valuable because all filter-based instruments have artifacts in the absorption measurements. It is also valuable that different types of BC particles and size distributions were used in the experiments.

The paper is well written and I can recommend publishing it in AMT after some additions and answers to some questions that puzzle me.

Comments and questions

1) Compared to ambient measurements the concentrations were fairly high. The best results for the absorption coefficient with the CAPS PMssa were observed for absorption coefficients > 10 Mm-1 . Such levels are observed in very polluted environments, for instance in China and India. I think you could mention this and also refer to a new intercomparison where the CAPS PMssa was used in real background conditions, see Asmi et al. (2021) (ref below) and also compare your results with theirs.

Answer:

We will reference Asmi et al in the paper. The Pallas station is at clean artic air conditions, which results in low aerosol light absorption levels. That paper shows, that DM methods (EMS called there) are at the detection limit even after one hour of averaging. This result is in accordance with our results. We see some issues with Asmi at al. concerning inlet setup and truncation correction but in principle we can state that the CAPS Instrument performs well for Ext. Sca and SSA at rural conditions like previously stated by Onasch et al 2015, Massoli et al. 2010. Four our own measurements at low absorption coefficient conditions for AQ (5 Mm-1) we have calculated an error of the DM method of up to 2 Mm-1 (compared to TAP 0.2 Mm-1).
If no direct absorption measurement is available, the use of CAPS SSA gives you also a fair measurement of the absorption coefficient. The situation is different for the AAE calculation with a reported value of AAE=0.4 +- 1.7. Thus, our conclusion for rural sites is that AAE calculations based on DM method using CAPS must be taken with caution. We clarified this in the text.

2) Related to this, I am missing a table where you would show the extensive and intensive aerosol optical properties and the length of each experiment. Maybe in a supplement? Now the tables have various ratios and regression constants – which is important of course – but I think it would be useful to show also the range of absorption and scattering coefficients you have produced. Or if you don't want to make that supplement, at least you could add some lines to the overview table, Table 3: number of experiments, average length of experiments, averages and ranges of scattering and absorption coefficients.

Answer:

Table 3 shows the measured and calculated values for the pure substances. We add the information about the ranges for absorption/ extinction of the different experiments in the experimental section. Here the extinction coefficients are ranging from 15-2000 Mm^{-1} (all aerosol Types) (0-1000 Mm^{-1} for light absorption coefficient of mixed aerosol types). We have aimed for

extinction values ranging from 30 to 100 Mm^{-1} to stay at atmospheric relevant values. We will add information about the ranges of scattering and absorption in the tables 7,8 and 9.

3) The results in the scatter plots and the tables are based on experiment averages or ensemble averages and their ratios. As an example I take Table 5. I do not find anywhere information of how long data are collected for one experiment's average. In the table there are the numbers N=xx. I suppose xx the number of experiments, right? Please explain clearly both in the text and the table captions what N means.

Answer:

We will clarify on Page 13 in the Extensive Parameter Section that for each experiment (run) a different aerosol mixture was generated with different overall extinction levels. We used a 100 sec average (1 HZ sampling rate) per experiment (run), after the nephelometer reached a steady state. In Figures 3-5 100 second averages and standard deviations are plotted. Table 5 compiles EMS (DM) derived absorption coefficients. Here we report ensemble averages, where N denotes the number of experiments (run) used for the average.

4) Further on the same averaging question. So, the scatter plots are based on averages which I assume means using the average of each experiment. How would the results change, if you used shorter averaging times, from some seconds to minutes? Or was the aerosol production so stable that it would not matter, which averaging time was used? Usually it is assumed that the uncertainty due to noise decreases with one over the square root of averaging time. This then propagates to the uncertainties of the derived optical properties. For instance, I guess that in Fig. 6 the data points would fill in the grey shaded error bands if shorter averaging times were used. Discuss this a bit.

Answer:

We agree with the reviewer on this important point. We chose to average 100 seconds of the 1 HZ data per experiment based on reported Allan standard deviation results. The Allen Plots in Massoli et al., 2010 for CAPS extinction measurements (http://dx.doi.org/10.1080/02786821003716599**) show that there is a minimum detection limit close to 100 sec. For longer integration times, the measured variance starts to increase again due to baseline drift. Baseline drift is corrected in the CAPS by taking zeros at a user-defined frequency. In this study, we chose to measure zero baselines for all CAPS every 10-12 minutes, thus a 100 second average makes sense. A similar result is observed for the TAP/CLAP instrument shown in Ogren et al., 2017 Figure 7. In this study, Ogren observed the TAP/CLAP variance reached a minimum level of 10% for the aerosol loadings (comparable to our study) after 100 seconds. For rural, ambient measurements, Ogren et al recommend a longer averaging time than 100 seconds.**

Lines 68 – 73. There is discussion on AAE. It is written that AAE depends on chemical composition and that AAE > 1 is due to brown carbon or mineral dust. This is not the whole truth. It is easy to show with your Mie code and it has been shown in several papers that AAE also depends on the size distribution of the light absorbing particles and that both AAE>1 and AAE<1 values are observed even for pure BC particles. Here are just some references: Gyawali et al. (2009), Lack and Cappa (2010), Lack and Langridge (2013), Liu et al. (2018), Zhang et al. (2020) and Virkkula (20219. Actually, it is interesting that if you compare median diameters the AAE values in your Table 3 with Fig. 6 of Liu et al. (2018) they seem to be qualitatively in agreement

Answer:

We agree with the reviewer's comment. Our intent was to state that for the conditions we used in this study (i.e., solid absorbing particles externally mixed with solid non-absorbing particles), the AAE is expected to be near unity. We will make this clear in the text in on page 3 in the introduction section and will add your suggestions.

Line 112: " Because all instruments were connected to one central aerosol supply line." The sentence should continue, now there is a full stop.

Fixed. Thank you.

Line 136: "Data inversion for the nephelometer …". Is "inversion" really the correct term here? The scattering coefficients are simply multiplied with a correction factor that depends on SAE.

We changed "inversion" to "data correction".

Line 199: Size distributions were measured beforehand. Why not all the time? Any idea of the stability of the size distributions?

The size distributions of the aerosol production single particle types (i.e., absorbing, or non-absorbing) were characterized before the study and checked several times during the different experiments. Size distributions just relate to the MFC setup. The Size distribution is not expected to change during one experiment if the solution centration and flows do not change. (Liu, Aerosol generator of high stability, 1975)

References

Great, thank you for your efforts!

Asmi et al.: Absorption instruments inter-comparison campaign at the Arctic Pallas station, Atmos. Meas. Tech., 14, 5397–5413, 2021.

Gyawali et al.: In situ aerosol optics in Reno, NV, USA during and after the summer 2008 California wildfires and the influence of absorbing and nonabsorbing organic coatings on spectral light absorption, Atmos. Chem. Phys., 9, 8007–8015, 2009.

Lack and Cappa: Impact of brown and clear carbon on light absorption enhancement, single scatter albedo and absorption wavelength dependence of black carbon, Atmos.Chem. Phys., 10, 4207–4220, 2010.

Lack and Langridge: On the attribution of black and brown carbon light absorption using the Ångström exponent, Atmos. Chem. Phys., 13, 10535–10543, 2013.

Liu et al: The absorption Ångström exponent of black carbon: from numerical aspects, Atmos. Chem. Phys., 18, 6259–6273, 2018.

Zhang et al: The absorption Ångstrom exponent of black carbon with brown coatings: effects of aerosol microphysics and parameterization, Atmos. Chem. Phys., 20, 9701–9711, 2020.

Virkkula: Modeled source apportionment of black carbon particles coated with a light-scattering shell, Atmos. Meas. Tech., 14, 3707–3719, 2021