Reply to the reviews of the manuscript “Impact of particle size, refractive index, and shape on the determination of the particle scattering coefficient – an optical closure study evaluating different nephelometer angular truncation and illumination corrections” by Teri et al.

We would like to thank Reviewer 2 for the thoughtful comments, which helped to improve the manuscript. In the following, the questions and comments raised by the reviewer are marked in blue. Our answers are given in black and also include a description of changes made to the manuscript.

Reviewer #2

This manuscript presents a comprehensive evaluation of different angular corrections for the Aurora 4000 nephelometer. The study combines specifically designed laboratory experiments with model simulations, with special focus on irregularly shaped coarse particles. The manuscript is very well written and structured and reads very easily.

General comments

Although the authors performed a very comprehensive study evaluating different angular corrections and using different input data, I feel like there are still some points that should have been addressed in this manuscript. The first one is the evaluation of the angular corrections for moderately absorbing aerosols. The SAE-derived correction is the most widely use angular correction and the recommended one in this article (for unknown aerosol type, which is the general case), however, it is highly uncertain for absorbing particles. What is then the recommendation for absorbing particles? Secondly, the laboratory experiments focused on different aerosol types (PSL, AS, dust,...) but I wondered why ambient aerosols were not explored (in particular urban aerosol with low single scattering albedo).

We agree that exploring absorbing aerosol would be interesting. However, the focus of our laboratory experiment was on irregularly shaped coarse mode aerosol. In order to include urban aerosol with low single scattering albedo, we would have to repeat the experiment with different aerosol types which is out of the scope of the present manuscript. We will include a recommendation in the manuscript that future studies should also explore absorbing aerosols.

Title: To what extent the results obtained in this study can be extrapolated to other nephelometers, like the TSI3563? I recommend the authors to include in the title that the study is limited to the Aurora 4000. Something like “Impact of particle size, refractive index, and shape on the determination of the particle scattering coefficient – an optical closure study evaluating different nephelometer angular truncation and illumination corrections for Aurora 4000 nephelometer”

In the laboratory experiment, we used the Aurora 4000 nephelometer. The evaluated angular corrections were calculated considering the angular sensitivity function of the Aurora 4000 nephelometer measured by Müller et al. (2012). The results of our analysis are consistent with literature for other nephelometers, e.g., Anderson and Ogren (1998), Bond et al. (2009), Massoli et al. (2009), Müller et al. (2011). We therefore expect the recommendations/decision tree given in Figure 8 to be also valid for other nephelometers such as for example the TSI 3563 nephelometer. There are some differences in the angular sensitivity function for different nephelometers, and therefore the value of the angular correction slightly differs. As a consequence, also the uncertainties given in Figure 8 may be slightly different for other nephelometers.

For multi-wavelength nephelometers with a known angular sensitivity function, the angular corrections based on the phase function (C_{phase}) and the Scattering Angström Exponent (C_{SAE}) can be derived. The angular correction based on the polar factor (C_{polar}) requires scattering measurements for different angular sectors currently performed only by the Aurora 4000 nephelometer. Therefore, the correction C_{polar} is limited to the Aurora 4000. Since the general conclusions also apply to other nephelometers, we decided to leave the title as it was.

Line 23: "... size distribution measured with optical particle spectrometers": not only optical spectrometers, some studies have been carried out using mobility and aerodynamic spectrometers. My suggestion is to remove "optical" to broaden the applicability.

The word "optical" was removed.

Line 59: The work by Sorribas et al. (2015, Q.J.R.Meteorol.Soc.141: 2700 – 2707, https://doi.org/10.1002/qj.2557) already explored the effect of particles with irregular shape in the Anderson and Ogren (1998) correction. They found that for fine mode particles, the angular correction underestimates the observed scattering for both spherical and spheroidal approximations by less than ~3%; while for particles within the coarse mode, the uncertainty for scattering was about 8%. I recommend the authors to read this manuscript carefully and take it into account both in the introduction (acknowledgement to previous work) and in the discussion of results.

We thank Referee #2 to point out the study by Sorribas et al. (2015) which is now included in the manuscript.

We modified line 59 as follows: "Meanwhile, Sorribas et al. (2015) estimated an uncertainty of about 13% for the Anderson and Ogren (1998) angular correction in case of coarse mode desert dust aerosol and recommended to re-evaluation of this angular correction for coarse mode aerosol.

This work aims to evaluate and compare ..."

In Table 2, we included the sentence: "Sorribas et al. (2015) recommend this correction to be re-evaluated for coarse mode aerosol."

Because of differences in the chosen methods to measure the size distribution and to estimate the uncertainty of the angular corrections, it is not straightforward to compare the results of the present work with those of Sorribas et al. (2015).

Line 105: What is the uncertainty in the scattering angles?

We are unsure what reviewer #2 means since the angles reported in line 105 are the angular shutter positions α and not the scattering angles θ.

Concerning the uncertainty in the scattering angles: the light detector is always orthogonal to the light source, and the intensity function of the light source has a maximum at θ=90°. Therefore, the nephelometer is always aligned, and there is no bias in the scattering angle.

Concerning the uncertainty of the angular shutter positions α: the uncertainty of the scatter shutter position is unknown. We rely on the calibration procedure and the manufacturer's nominal values. During the laboratory experiment, we performed several calibration checks with CO_2. The Rayleigh scattering signal measured for α=90° was half of the total signal measured for α=0°. Thus, the angular shutter position was correct, and the instrument was well-calibrated.

Another uncertainty is given by the angular sensitivity function including the truncation angles, the non-Lambertian illumination and the non-perfectly sharp separation of the shutter (see Sec.3.1). The angular sensitivity function measurements for Aurora 4000 (parameters given in Table 1) were repeated several times and the repeatability was high (T. Müller, personal communication). Therefore, we assume the uncertainty for the given unit to be low. As mentioned in line 624 of the manuscript, we cannot exclude small differences between different units of the same model.

Line 315: I have several concerns about the use of an optical spectrometer instead of using an aerodynamic spectrometer for this type of study. I am aware that there is no consensus on a standard instrument for measuring the coarse aerosol size distribution.

However, since the focus of the study is to use the size distribution to retrieve aerosol optical properties I feel that it would be more appropriate to use an instrument that measures the size independently of the optical properties of the particles. Said that, and being aware of the amount of work behind these experiments, I recommend to include more information on the performance of the OPS: First of all, a discussion on the uncertainties in the size distribution obtained with the

OPS for aerosol particles other than PSL. The instrument is calibrated with PSL, but how accurate is the size distribution for particles with different refractive index and shape (like dust)? How this would affect the results obtained? This is briefly discuss in lines 555-567, but this should be further elaborated. Using an APS instead of OPS would have been more optimal for this study.

We agree that there is no consensus on a standard instrument for measuring the coarse aerosol size distribution. Optical particle counters are widely used to measure the coarse mode size distribution during aircraft field experiments (e.g., Weinzierl et al., 2009, 2011, 2017., Spanu et al., 2020, Brock et al., 2021). The lower size detection limit of the APS is relatively high, e.g., the TSI APS Model 3321 can detect particles with aerodynamic diameters starting from 500 nm. Using a combination of OPCs during our experiment allowed us to measure the aerosol size distribution down to 60 nm thereby also covering the optically important size range between 60-500 nm.

We added at line 79: „The TSI OPS 3330 is an optical particle spectrometer, a class of instruments widely used to measure the aerosol size distributions during aircraft field experiments (e.g., Weinzierl et al., 2017, Spanu et al., 2020, Brock et al., 2021).“

Following the suggestion of the reviewer, we included the following section to the supplementary information to discuss the performance of the OPS.

„S3 Sensitivity of the TSI OPS 3330 to particle refractive index and shape

An optical particle sizer, such as the TSI OPS 3330, measures the particle scattering cross sections and sorts particles into different size bins based on their detected scattering cross section. For the TSI OPS 3330, the manufacturer provides nominal diameter values for the bin boundaries for the calibration material, i.e., PSL particles with refractive index m = 1.59 + i 0.00 and spherical shape. The particle size distribution obtained with such a measurement is a PSL-equivalent size distribution. Any deviation from the refractive index or shape of the calibration material will increase the uncertainty of the derived particle size distribution.

Figure S3 shows the relationship between the TSI OPS 3330 scattering cross section and the particle volume equivalent diameter for the calibration material (in black) and for different refractive indices in the range of mineral dust particles, both in the case of spherical particles and in the case of irregularly shaped particles.

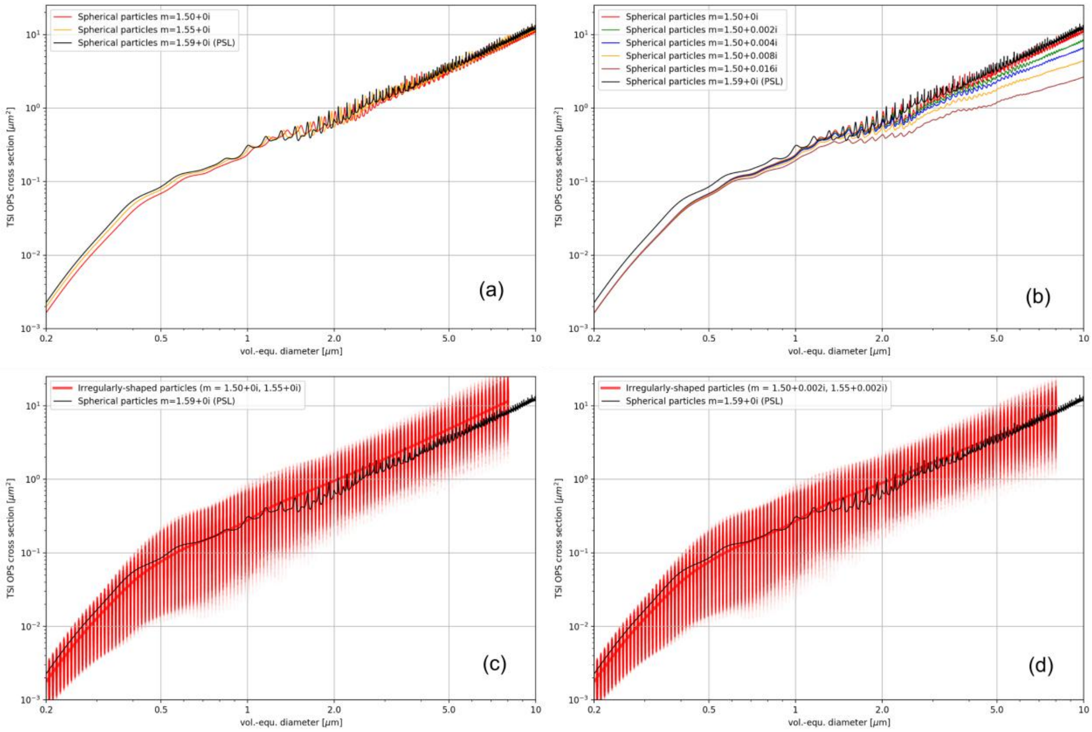

Figure S3. Scattering cross section modeled for the TSI OPS 3330. The black line in all four panels is the TSI OPS 3330 scattering cross section for PSL particles, i.e., the calibration material (m=1.59 + i 0.00 and spherical shape). In addition, each panel shows the TSI OPS 3330 scattering cross section for particles with different refractive indices or shapes relevant for mineral dust particles: (a) non-absorbing spherical particles with different real part of the refractive index (n = 1.50, 1.55); (b) spherical particles with different imaginary part of the refractive index (k = 0.000, 0.002, 0.004, 0.008, 0.016); (c) non-absorbing and (d) slightly absorbing irregularly shaped particles. In panels (c) and (d), the red dots correspond to 18000 orientations of six different mineral dust shapes (Gasteiger et al. 2011) and the red line is the average.

The TSI OPS 3330 scattering cross section for coarse mode particles (d > 1 µm) is not very sensitive to the real part of the refractive index between m = 1.50 + i 0.00 and m = 1.59 + i 0.00 (Fig.S3a). Thus, the uncertainty of the PSL equivalent particle size distribution for non-absorbing spherical particles with mineral dust-like refractive index is negligible. The TSI OPS 3330 scattering cross section for coarse mode particles is more sensitive to the imaginary part, and the PSL-equivalent diameters underestimate the diameter of absorbing particles (Fig.S3b). For example, a moderately absorbing spherical particle with imaginary part k = 0.004 and a diameter of about 5 µm would have the same scattering cross section as a PSL particle with about 4 µm diameter, and therefore, the OPS would size it with a PSL-equivalent diameter of about 4 µm.

If particles have an irregular shape, the relationship between TSI OPS 3330 scattering cross section and particle volume equivalent diameter is more complex as also the shape and the orientation of the particle plays a role. Figure S3c and S3d show the TSI OPS 3330 scattering cross section for 6 irregular shapes typical for mineral dust (Gasteiger et al., 2011) in 18000 orientations (red dots). The average is indicated by the red line. Coarse non-absorbing irregular particles have, on average, a slightly increased scattering cross section with respect to spherical particles with the same volume equivalent diameter (Fig.S3c). Therefore, the particle size is slightly overestimated by a TSI OPS 3330 calibrated with PSL particles. In case of slightly absorbing particles (e.g., with imaginary part ~ 0.002) the TSI OPS 3330 calibrated with PSL has on average a better performance (Fig.S3d). Therefore, the effect of irregular shape might partly compensate the effect of the imaginary part of the refractive index.

Furthermore, the variability of the scattering cross section of irregularly shaped particles of fixed size might lead to additional effects on higher orders of the size, e.g., particle area or volume, so that the agreement of the average shown in Fig.S3d, does not necessarily result in good performance for higher orders using the same imaginary part. For any non linear function of the size separate investigations are needed.“

To estimate how the uncertainty due to different refractive indices and shapes affects the angular correction $C_{phase,OPS}$, we performed the simulated closure experiment described in the manuscript in section 5.

For coarse mode aerosols, the results are reported in the first column of Fig.7 of our manuscript and are consistent with the TSI OPS 3330 scattering cross section of Fig.S3. The angular correction $C_{phase,OPS}$ has an uncertainty of a few percent for non-absorbing spherical particles (Fig.7 first column, second row). When the imaginary part of the refractive index increases, the angular correction $C_{phase,OPS}$ underestimates the true angular correction. For spherical particles and mineral dust-like refractive indices the underestimation can be up to 22% for k=0.016 (Fig. 7 first column, fifth row). For mineral dust-like irregularly shaped particles, $C_{phase,OPS}$ overestimates the true angular correction by up to 8% in case of non-absorbing particles and underestimates it by up to 17% for absorbing particles (Fig.7 first column, last row). This comparison shows that also for $C_{phase,OPS}$ the effect of the particle absorption is partly compensated by the effect of the irregular shape.

We modified lines 555-567 of the manuscript as follows:

„The angular correction $C_{phase,OPS}$ is a rarely used method in literature (e.g.,Di Biagio et al.2019) as it requires simultaneous particle size distribution measurements and also knowledge about refractive index and particle shape. For the mineral dust measurements, the performance of the angular correction $C_{phase,OPS}$ is surprisingly good (Fig.3.h) even though no refractive index or shape is considered in the size distribution retrieval. This result might lead one to the conclusion that there is no need to take into account the refractive index or shape of the measured aerosol for the calculation of the angular correction $C_{phase,OPS}$. The optical particle spectrometer is an optical cross-section selector rather than a particle sizer. The use of the refractive index of the calibration material might lead to compensations of the effects of refractive index and shapes on the measured particle size distribution (see supplementary information in Sec.S3 and Fig.S3) and thus on the calculated angular correction. Results of our simulated closure experiment (Fig.7, last row) suggest that the good performance of the angular correction $C_{phase,OPS}$ for mineral dust measurements (Fig.3h) might be partly due to a compensation of the non-spherical shape effect by the small imaginary part of the refractive index. In the case of mineral dust measurements, the particle size distribution by an OPS, obtained with the manufacturer-provided PSL-equivalent nominal diameters of the bin boundaries, leads to an angular correction which is on average correct (within ± 0.3 %}). However, $C_{phase,OPS}$ has a strong dependence on the imaginary part of the refractive index; it overestimates the true angular correction by up to 8% for non-absorbing particles and underestimates it by up to 17% for absorbing particles (Fig.7, last row, approach a).„

The loss correction applied to both OPS, is size-dependent? Different losses are expected for particles of different sizes...

Yes, the applied particle loss corrections are size-dependent.

We changed line 323: “A size-dependent correction factor was estimated and applied to the UHSAS and the OPS 3330 measurements.”

Line 526: remove recently, it has been 10 years since its publication...

The word “recently” was removed.

Fig8: I found this figure very informative and interesting for the scientific community. However, in general for atmospheric measurements, the aerosol type is not known (aerosols are a mixture of different components), so the recommendation is to use the SAE derived correction despite its large uncertainty (which is the most widely used correction scheme). Also, no recommendation are given for environments with high load of absorbing particles like urban sites. Extending the study to moderately absorbing mixtures will significantly enhance the scope of this manuscript.

We thank Referee #2 for this comment. Since we focused our study on coarse mode aerosol with irregular shapes, we did not perform measurements for absorbing aerosol. Therefore, we cannot give recommendations for environments with a high load of absorbing particles.

We agree with Referee #2 on the importance of extending the study to absorbing particles, both in the case of urban sites and moderately absorbing mixtures, such as mineral dust and black carbon mixtures.

We include the following sentence at the end of the manuscript:

“By improving the understanding of the nephelometer angular correction for irregularly shaped coarse particles – in particular mineral dust - this study reduces uncertainties in observations of aerosol scattering properties. Future studies with the aim of further reducing the uncertainty of scattering measurements should focus on environments with a high load of absorbing particles like urban sites or moderately absorbing mixtures such as mixtures of mineral dust and black carbon.”

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