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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Reconciling aerosol light extinction measurements from spaceborne lidar observations and in-situ measurements in the Arctic" *by* M. Tesche et al.

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We thank Referee 2 for his/her comments. We have incorporated them into the revised manuscript. Please find our point-by-point answers below.

Overview:

The manuscript deals with a comparison of extinction coefficients at the Arctic which were derived in situ from nephelometer measurements with subsequent corrections at Zeppelin station and which were taken from remote sensing data (corrected backscatter data measured by CALIOP) in the wider vicinity of Zep-

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Interactive Discussion

Discussion Paper

pelin station. This is a very challenging task. The problem that the measurements were not taken at the same time and place was tackled by using trajectories in order to assure that at least the same air mass was probed. The authors applied a correction method regarding relative humidity which links the dry nephelometer measurements with the ambient CALIOP measurements.

It seems like the reviewer misunderstood the methodology we describe in the submitted manuscript to retrieve ambient extinction coefficients from the dry in-situ measurements. We did not humidify the dry nephelometer measurements to derive ambient extinction coefficients. The parameter was retrieved using the particle size distribution and Mie-scattering theory. Dry and wet nephelometer measurements were only used to validate the microphysical model that has been used to obtain scattering and extinction coefficients from the dry and humidified size distribution data.

However, we have now added the direct aerosol optical in-situ data to the comparison. We investigated if we can also use measurements of the dry nephelometer together with scattering enhancement factors to derive ambient extinction coefficients. This approach has the advantage of including the contribution of the coarse-mode fraction (that is not included in the size distribution measured with the DMPS) to the extinction coefficient. The scattering enhancement factor was obtained using median, minimum and maximum γ-values of 0.57, 0.35, and 0.85, respectively, according to Zieger et al. (2010). We restructured Section 2.2 accordingly to describe this procedure in Section 2.2.2. The results were added to Figures 1, 3, 4, and 5 as well as to Table 1. The description and discussion of these figures and the table has been revised accordingly. We now used the ambient extinction coefficients obtained from applying the scattering enhancement to dry nephelometer measurements for the comparison to CALIPSO findings presented in Figures 4 and 5.

Finding measurements which are suitable for comparisons required an extensive screening of the data. Besides this huge amount of work, the manuscript lacks an analysis of data quality, especially determination of measurement er-

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rors including error propagation through application of the correction method. Without tackling errors, it is difficult interpreting and understanding the results.

The main message of the manuscript is that one has to be very cautious when attempting to integrate spatially non-coinciding observations from different platforms and that the choices for data averaging have huge implications on the results. In particular, the highly averaged data that are often used for such a task can lead to coherence of the observations that is no longer present when comparisons are performed on a case-by-case basis. We refined the comparison methodology to take the influencing factors into account. Each of these factors has the potential to make any meaningful comparison impossible and to cause differences of orders of magnitude ("apples and oranges" comparisons).

The reviewer is correct that our initial approach of determining ambient extinction coefficients from DMPS measurements with the help of hygroscopicity and Mie modelling was lacking a proper representation of the error in the derived parameter. As suggested by other reviewers, we determined ambient extinction coefficients from the nephelometer measurements using scattering enhancement factors derived with γ-values measured at Zeppelin station by Zieger et al. (2010). This procedure is described in new Section 2.2.2 of the revised manuscript. We use a γ of 0.57 to describe the most likely conditions together with minimum and maximum values of 0.35 and 0.85, respectively. The latter are now used as an estimate of the error in the correction method in revised Figures 3 and 4.

Detailed comments

The line and page numbers are taken from the printed version not from the online display.

General

*** I have difficulties in understanding phrases such as "agreement of a factor of**

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1.85" (page 1), "agreement of a factor of ca. two" (page 4), "factor of five in agreement" (page 12). Does it mean that the data agree with each other or they disagree? I think, data can agree within their error bars and in case there is a factor of something it points towards disagreement.

We changed the respective formulation to emphasize if we see agreement or rather disagreement.

*** The whole issue of errors, error propagation, error bars is almost ignored. I am also missing comments about calibration of the nephelometer (how often, how old was the latest calibration, the data were reprocessed in order to account for a shift in the calibration constant between the day of calibration), about the detection limit.**

We now provide error bars for the ambient extinction coefficients obtained from the nephelometer measurements as derived from using different estimates in the γ-value for retrieving the scattering enhancement factor (see answer to general comments). The dry nephelometer underwent the usual quality assurance steps (regular CO2 and zero air calibration). The standard nephelometer truncation and illumination correction (Anderson and Ogren, 1998) has been applied as well. The nephelometer was also indirectly validated by using the measured size distribution and Mie theory (see Rastak et al, 2014).

*** It is not clear how the authors tackled the problem of the particle absorption which is not measured by the nephelometer. It seems from the comments on page 6 (lines 193-195) that particle absorption is only taken into account through the refractive index of particles omitting the issue of the mixing state (external/internal mixture).**

In the retrieval of the ambient extinction coefficient from the DMPS measurements, light absorption is accounted for through the refractive index used in the Mie-scattering calculations. In the now added retrieval of the ambient extinction coefficient from neph-

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elometer measurements, we accounted for light absorption with the help of PSAP measurements. The latter do not show increased light absorption for the cases considered in this study. The high single-scattering albedo of Arctic aerosols limits the effect of absorption to a negligible contribution to the ambient extinction coefficient (see answer to next comment).

The analyzed data contain biomass burning episodes? The approach of measuring dry aerosol particle scattering coefficient and calculating ambient aerosol extinction coefficients is valid for such episodes? Furthermore the time resolution for OC/EC ratio is one month (page 6). I doubt that monthly values can be reasonably used for single hourly measurements and correct for absorbing aerosols.

We checked the data of PSAP measurements at Zeppelin station for periods with an increased contribution of absorbing aerosols. We found a mean single-scattering albedo (SSA) of 0.985 ± 0.014 (median of 0.989). The cases we consider in our study did not show SSAs that are lower that these values. The behavior of the scattering enhancement factor with relative humidity dominates the error of the ambient extinction coefficient. Uncertainties in the treatment of humidity effects outweigh the influence of absorption to total light extinction in the Arctic. Hence, we conclude that the contribution of biomass-burning aerosols is negligible for observations at Zeppelin.

*** The authors investigated data for the whole year of 2008. The humidity correction was based on a model taking into account certain chemical components of aerosol particles. This model was evaluated using measurements done between July and October 2008 (see reference Rastak et al., 2014). Is this model for humidity correction suitable for measurements outside the evaluation period? How much did the chemical composition changed over the entire year? Seeing Fig. 3 in Rastak et al. 2014 manuscript, it seems that EC is much less during the evaluation period compared to the months December-May/June.**

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We used the chemical data for the respective day (sulphate and sea salt which dominate hygroscopicity) or month (OC/EC which dominates absorption) together with individual size distribution measurements. From the chemical data we derive the kappa values required in the humidification model for the considered measurement days. The increase of the extinction coefficient due to hygroscopic growth is much larger than the contribution of absorption. Hence, even larger errors in the absorption coefficient (as a result of coarse resolution in the OC/EC data) are negligible in comparison to the uncertainties that can be introduced by improper description of hygroscopic growth. However, the chemical data that are required for the latter are available with daily resolution.

The model performed well for the evaluation period when using a kappa value based on the chemical information representative for this period. We don't see why it should not be suitable for another part of the year when the underlying particle chemistry (i.e. hygroscopicity) is adapted accordingly.

To assess the validity of our approach we now also present wet extinction coefficients as obtained by applying a constant scattering enhancement factor to the dry nephelometer measurements. The procedure is described in new Section 2.2.2. The results are presented in revised Figures 1, 3, 4, and 5 and in Table 1. The discussion has been revised according to the new findings.

Specific comments

Line 73: "above seal level" Typo; sea instead of seal

corrected

Line 191: "are water-soluble and insoluble organics, sulfate, sea salt, and black carbon." For avoiding misunderstandings, "sulfate" should be changed to "ammonium sulfate" (see Rastak et al., 2014)

changed

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Lines 301-302: Which kind of meteorological data were used (GDAS, ReAanalysis. . .)?

We ran the HYSPLIT model with meteorological data from the Global Data Assimilation System (GDAS) archive. The last paragraph of Section 2.4 was extended to now state:

Meteorological parameters from the Global Data Assimilation System (GDAS) are provided along the trajectories and used in this study to estimate RH at the location of the CALIPSO overpass.

Line 388: "the CALIPSO observation is in poor agreement with the result of the in-situ measurement" I would not say it is poor agreement. It seems more like a disagreement.

We changed the wording from "poor agreement" to "disagreement".

Line 389-390: "This emphasized that using a closest approach for comparison of ground-based measurements and CALIPSO observations might not always be the best choice." I disagree with this conclusion. The closest distance approach is related to the idea of spatial homogeneity/inhomogeneity (or representativeness) of a quantity of interest, whereas the approach of this manuscript is related to the idea of probing the same airmass. In the end it is important whether it can be expected that the quantities could be compared or not.

The only thing that is important is whether a comparison of a certain quantity is physically meaningful or not. The closest approach method relies on temporal and spatial homogeneity of a quantity. This condition is often fulfilled if column-integrated properties are considered. This manuscript deals with vertically resolved observations of aerosol optical properties, and thus, an additional level of complexity. The refinement of probing the same air-mass enhances the chance of performing a physically meaningful comparison. It also improves the chances of obtaining quantities that can actually be compared, especially when individual overpasses are considered. None of this is

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implied in the closest approach method which is furthermore often applied to highly averaged data, i.e. mean values for a certain time period are compared to mean values over a certain area during that period.

Lines 417-419: "It was found that the most characteristic outliers in Figs. 4 and 5a occur for cases that were identified predominantly as polluted dust, polluted continental, and dust in the CALIPSO retrieval. These aerosol types are rather uncommon at 78N and suggest misclassification in the CALIPSO retrieval." Misclassification is a possible cause for explaining the outliers. However it is possible that classification was correct, pointing to an important contribution of aerosol absorption which seemed to be not well taken into account by the correction scheme for the nephelometer? Please note that even uncommon aerosol types could easily be present in single measurements.

The reviewer is correct to point out that single cases could easily represent exotic conditions rather than an erroneous measurement. However, it is more likely that these aerosol types are the result of misclassification – especially when the low signal-to-noise ratio of CALIOP observations in the Arctic during summer is considered. The CALIPSO aerosol model can chose from a wider pool of lidar ratios (aerosol types), if measurements are performed over land and open water (i.e. to the west of Svalbard) rather than over snow and ice. This leads to a larger variation in the selected aerosol type for westerly flow. We added the following statement to the discussion of Figure 5:

On the other hand, the CALIOP aerosol classification scheme can choose from a larger pool of lidar ratios for observations over ocean and land compared to those over snow and ice (Omar et al., 2009).

Misclassification can occur as a result of signal noise, improper cloud screening, or due to surface effects. Given the structure of the CALIPSO aerosol classification scheme described in Omar et al. (2009), CALIOP observations in the Svalbard region during background conditions (weakly depolarizing and integrated attenuated backscatter co-

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efficient not exceeding the threshold value of 0.0015 at 532 nm) should be classified as clean continental (over land and snow/ice) and clean marine (over ocean).

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 5687, 2014.