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

Overview:

The study provides a comparison of ground-based in-situ measurements (Zeppelin Station) of aerosol light extinction to observations from the satellite CALIOP sensor.

The manuscript details the difficulties in quantitatively comparing satellite and

in-situ measurements, including discrepancies in space and time, uncertainty associated with aerosol humidification, and differences in actual measurement techniques. The authors use a complex approach in order to match appropriate CALIOP overpasses with in-situ data involving back trajectory analysis, CALIOP cloud-screening, and use a combination of humidified size distribution and chemical composition measurements to correct in-situ data to ambient humidity. The number of matching cases is extremely low (57 of a total of 2018 CALIOP overpasses), and a comparison of extinction coefficients yields agreement only within a factor of 10 (plus/minus a factor of 5). Additionally, the overpasses closest to the ground-site showed the worst correlation. Since the focus of the manuscript seems to be more about the process of linking the two measurements, rather than on the fairly uncertain results, it is suggested that more work is required to explore which steps are most important and if simplified methods could produce similar results.

The paper is indeed about the process of linking the different observations. We feel that this topic requires some attention as integrated observation systems and multi-platform synergies are considered as the next step in improving our understanding of atmospheric processes. Hence, reconciling such observations will pose a major challenge for the future.

From the reviewer comments we realized that we rushed into describing the detailed comparison approach in Section 3 without proper motivation for that course of action. Consequently, this and two other reviewers are interested in a quantification of the effects of the steps we apply during the comparison procedure. We actually considered the influence of the individual steps when we refined the comparison procedure from the simple closest approach method to what is described in Section 3. The simple comparison could not be used to reconcile the measurements at Zeppelin with CALIOP observations. While the number of comparison cases was much larger than the 57 cases we present in the manuscript, the difference in the extinction coefficients was in the

range of three to four orders of magnitude. The increased complexity of the comparison approach decreased the difference in the compared values and led to physically meaningful situations for comparison. To make it easier for the reader to understand the rationale of our approach we added a paragraph to Section 3 that describes the background of why we believe that simplified comparison scenarios, i.e. missing any of the used steps, will lead to physically meaningless results in environments as considered in this study (with low aerosol load, high cloud cover, strong influence of relative humidity).

We started our investigation by applying the closest approach method to link CALIPSO observations in the region of interest to coincident dry in-situ measurements at Zeppelin station. While this course of action led to a high number of matches, it did not enable reasonable case-by-case reconciliation of in-situ and remote-sensing data. Differences in the compared aerosol optical properties ranged between two and three orders of magnitude. Perpetual refinement of the comparison procedure as described below showed that the failure in reconciling the different observations in the initial comparison is due to:

1. *Physically meaningless comparison scenarios in which no connection can be established between the locations of the ground site and the satellite track during heterogeneous aerosol conditions*
2. *The inclusion of apparently unrealistic signal spikes into the CALIOP extinction coefficient in case of fixed or inappropriately selected along-track averaging intervals*
3. *Humidification effects*
4. *The temporal delay in the observations*

The first two points make reasonable comparisons impossible. The latter two can still introduce uncertainties of up to 100%.

Major comments:

I fully appreciate the amount of work that went into this study and the detailed approach was thorough and well-presented, involving humidity correction, spatial scale matching with back-trajectories, and careful cloud-screening. The uncertainties of this process, coupled with the uncertainties associated with CALIOP measurements in the clean Arctic somewhat expectedly lead to non-ideal comparisons between the two measurements. Still, it is unclear how this study does anything more than point out these uncertainties in the form of Figure 4, including what visually looks like a lack of correlation at all.

We show that taking the uncertainties into account can lead to physically meaningful comparison cases. Highly averaged data on the other hand are likely to show agreement for the wrong reason. Consequently, the outcome of any study that attempts to reconcile CALIOP measurements with ground-based observations strongly depends on the comparison approach and data treatment. We see our study also as a critical assessment of the many issues involved with such endeavors.

Results indicating that increased overpass proximity to the ground site leads to decreased accuracy only suggest that the method was fundamentally unnecessary.

It is not clear to us what the reviewer suggests with this comment. We state that overpasses closest to the ground site (increased overpass proximity) generally present the worst comparison cases (decreased accuracy). This means that the presented method was in fact fundamentally necessary (the exact opposite of the reviewer's comment). The closest approach method leads to physically meaningless comparisons ("apples and oranges") for the conditions met in the Arctic. Our complex procedure on the other hand establishes a link between the different observations and reduces the effect of

atmospheric variability as best as possible. While this reduces the number of comparison cases, it increases the overall quality of the comparisons. We think that the closest approach method's advantage of having a large amount of comparison cases does not outweigh its implicit drawback of including physically meaningless comparisons.

If accuracy to a factor of 10 is the best possible result, and if presenting this approach is the real result of the manuscript, then I believe a sensitivity study is necessary to assess each step in the process.

When we started our investigation, we intended to perform a systematic comparison of aerosol extinction coefficients as obtained from in-situ and lidar measurements. From this we would have evaluated the representativeness of measurements at Zeppelin for the Arctic and gained additional value to the CALIPSO observations and vice versa. Along the way we realized that such comparisons (even for a high overpass rate as in the Arctic) require significant efforts to ensure acceptable and usable data quality. As the Arctic is one of the hot spots of current research on aerosols and climate change, we believe that it is in the interest of the scientific community to see that reconciling aerosol properties from different platforms is not straightforward. This is of particular importance for data users that don't necessarily have a strong background in (the limitations of) the different measurement techniques.

A factor of ten was the worst agreement we found from our investigations. Most comparison cases were actually within a factor of two. This is orders of magnitude better than what could be obtained when doing the simple closest approach comparison. As suggested by the reviewers, we investigated the potential of using extinction coefficients from humidifying the dry nephelometer measurements with the help of reasonable scattering enhancement factors. This course of action leads to improved agreements as is stated in the revised manuscript.

For example, what does the comparison look like prior to each step in the analysis process? Examples of steps that could be simplified and evaluated for the

effect on accuracy and uncertainty of the final in-situ/CALIOP comparison are (but shouldn't be limited to):

As stated in the beginning of our answer to the reviewer's comments, we started with a closest approach comparison which could not even give us the same order of magnitude of the different observations. Considerable refinement to was required to come up with comparison scenarios that are likely to yield physically meaningful results rather than dealing with an apple-and-oranges situation.

1.) Can a constant humidification factor be used instead of necessitating continuous size distributions and chemical composition data?

For the revised manuscript, we calculated ambient extinction coefficients from the scattering and absorption coefficients measured with the dry nephelometer and PSAP, respectively. For that we used mean, minimum, and maximum scattering enhancement factors obtained by assuming γ-values of 0.57, 0.35, and 0.85, respectively, according to Zieger et al. (2010).

We restructured Section 2 to account for the added information. The procedure of humidifying dry nephelometer measurements is described in new Section 2.2.2 (Nephelometer + PSAP + scattering enhancement factor) as:

The DMPS measurements used in the previous section only cover particles up to a diameter of 790 nm and provide no information on the concentration of larger particles. These coarse-mode particles can have a huge effect on the overall aerosol optical properties as they are much more efficient scatterers of light compared to smaller ones. Hence, missing even low concentrations of coarse particles can cause an underestimation of the aerosol scattering and extinction coefficients by as much as 50% (Zieger et al., 2010, 2013). In addition, it is more straightforward to determine ambient extinction coefficients directly from the nephelometer measurements if the scattering enhancement factor is known or can be estimated within a reasonable range of values.

Therefore, ambient extinction coefficients were also calculated using the dry absorption and scattering coefficients measured with the PSAP and nephelometer, respectively, together with scattering enhancement factors that represent the median, minimum, and maximum effect of hygroscopic growth on light scattering. Values of γ = 0.57, 0.35, and 0.85, respectively, were used to obtain the scattering enhancement factor as $f(RH) = (1 - RH)^{-\gamma}$ (Zieger et al., 2010).

The statistical analysis of the ambient extinction coefficients derived from humidification of the nephelometer measurements were included to revised Figures 1 which is now discussed in new Section 2.2.3 (Dry versus ambient optical properties):

The box plots in Fig. 1 visualize the importance of transforming dry optical properties to ambient conditions. About 75% of the hourly aerosol scattering coefficients at 550 nm measured with the dry nephelometer at Zeppelin station in 2008 are smaller than 5 Mm^{-1}. Humidity correction to ambient extinction coefficients increases the median value for 2008 from 2 to 7 – 10 Mm^{-1}. The differences found in the median values of the ambient extinction coefficients derived according to the two methods described in Sects. 2.2.1 and 2.2.2 is likely to be the effect of coarse-mode particles that are not captured by the DMPS. These particles may contribute to about 20% – 30% of the total extinction coefficient at Zeppelin station (Zieger et al., 2010). The geometric mean has a much lower standard deviation than the arithmetic mean and is similar to the arithmetic median value. Independent of the retrieval method, the ambient extinction coefficient is on average a factor of three to five larger than the dry one when resolved according to different seasons. The Arctic haze period in spring shows the highest median values of the ambient extinction coefficient (17 – 22 Mm^{-1}) followed by winter (8 – 12 Mm^{-1}). Summer and fall are associated with very low median values (3 – 4 and 4 – 6 Mm^{-1}, respectively). Summer is the slightly cleaner season and a larger variation is observed during fall. This is in agreement with previous observations at Zeppelin station (Ström et al., 2003; Zieger et al., 2010; Tunved et al., 2013).

In the following, we use the ambient extinction coefficients derived from the humidified

nephelometer measurements. This is because the lower and upper estimate in the γ-value for the determination of the scattering enhancement provides as with an error estimate that is more reliable than what can be obtained using the model approach described in Sect. 2.2.1.

We also revised Figures 3, 4, and 5 as well as Table 1 and their respective captions and discussions according to the new values of the ambient extinction coefficient obtained from the humidification of the nephelometer measurements.

2.) Can back trajectories be avoided by using the overpass point closest to the ground site?

We believe that trajectories should be considered to guarantee that comparisons are physically meaningful. Even for homogeneous aerosol conditions one should use trajectories to ensure that the closest approach method is a valid simplification and applicable with minor impact on the comparison result. It is actually a message of this study that closest approach is not a suitable choice for the complex aerosol/cloud situation in the Arctic as it leads to physically meaningless solution, i.e. an error of 100%. This is addressed in the discussion of Figure 4.

3.) What are the results if a less rigorous could-screening process is applied?

We performed a signal screening rather than a classic cloud screening. Instead of excluding all CALIOP aerosol profiles that are flagged as cloud-containing, we regarded the aerosol extinction coefficient in our height range of interest of 250 to 750 m above sea level. CALIOP extinction coefficient is not automatically of bad quality if high clouds are present in the lidar profile. Considering the actual extinction coefficients allowed us to assess if these are in a realistic range of values. This investigation showed that CALIOP aerosol profiles – even when flagged as cloud-free – often show signal spikes that are clearly unrealistic, i.e. single height bins with values that are an order of magnitude larger than the adjacent bins. These spikes are an artifact of the low signal-to-noise ratio of the observations (“garbage in, garbage out”), and thus, should not be

considered in the comparison. In our case of heterogeneous aerosol/cloud conditions with low signal-to-noise ratios, less rigorous signal screening will lead to physically meaningless comparisons, and hence, an error of 100%.

The beneficial result of a less-rigorous point-matching process is more comparison points and better statistics.

As stated earlier, we believe in quality over quantity. There is no benefit in 'better statistics' if these are skewed towards meaningless comparisons.

The step-by-step evaluation will also be useful for readers without such comprehensive in-situ measurements, and help to justify the benefits of the process.

We added the results of using ambient extinction coefficients derived from dry nephelometer measurements and reasonable assumptions in γ-values for determination of the scattering enhancement factor to the paper. This is a way to simplify the comparison from the side of the in-situ measurements.

Additionally, I would suggest presenting a few case studies that highlight good/bad correlations that may shed light on the underlying issues with the method.

This is already included in the manuscript. We present individual cases in Fig. 3 and discuss why only half of the overpasses in the chosen time window can be used for comparison.

Minor comments:

Page-line

5689-13. remove "among either"

"among other" has been omitted in the sentence

5695-4. Observations from the summer were not used for comparison because

of difficulties by CALIOP. If scattering enhancement factors were only derived from July-October, were they used at all in the analysis? If not, it may make sense to remove them.

In the submitted manuscript, we used the measurements with the humidified nephelometer and scattering enhancement factors derived with this instrument to validate the performance of the humidification model that gives us ambient extinction coefficients. Scattering enhancement factors were not used directly in the analysis presented in the submitted manuscript.

As suggested by the reviewer, we added a new part to the revised manuscript in which we investigate if we can use dry nephelometer measurements and a constant scattering enhancement factors to obtain similar results. The scattering enhancement is parameterized with the help of the γ-value which depends on aerosol chemical composition. The used γ-values refer to the median, minimum, and maximum values derived by Zieger et al. (2010). This is described in new Sect. 2.2.2. As seasonal changes in the chemical composition of the aerosols at Zeppelin station are not dramatic (see Fig. 3 of Rastak et al., 2014), we are confident that using this range of γ-values will lead to reasonable ambient extinction coefficients.

5689-15. The four 'issues' you present are certainly pertinent to the study and provide a good review of the difficulties associated with remote/in-situ comparisons. I would suggest providing examples for each, e.g., specifically reference lidar and radiometer techniques under 2.

We thought of providing examples for each point presented in the introduction when we were working on the manuscript. We decided against it to keep these points more general. As these points are universally applicable, we wanted to prevent readers from feeling ignored if we wouldn't address their particular instrument of interest.

5695-9. What variability in the enhancement factor was observed? A factor 3 is very large compared to mid-latitude, continental sampling.

Zieger et al. (2013) present measurements of the scattering enhancement factor at different European sites. Highest f(RH)-values were indeed found in the Arctic at Ny-Ålesund with average values of 3.24 ± 0.63 at RH = 85%. For maritime air masses at Cabauw values could also reach 3 or higher at 85% RH. Even in the mid-latitudes, high values of 2.77 ± 0.37 were measured, e.g. at Melpitz, Germany, and explained by the high inorganic content of the aerosol.

Was the humidified nephelometer system verified with known substances like ammonium sulfate and nitrate?

The wet nephelometer has indeed been characterized and verified with known hygroscopic substances (ammonium sulphate and sodium chloride), as is described in detail by in Fierz-Schmidhauser et al. (2010). See also recommendations given in Zieger et al. (2013).

Fierz-Schmidhauser, R., Zieger, P., Wehrle, G., Jefferson, A., Ogren, J. A., Baltensperger, U., and Weingartner, E.: Measurement of relative humidity dependent light scattering of aerosols, Atmos. Meas. Tech., 3, 39-50, doi:10.5194/amt-3-39-2010, 2010.

Was there any evidence of biomass burning aerosols being transported to the site, which would likely reduce the enhancement factor significantly?

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. Hence, we conclude that the contribution of biomass-burning aerosols is negligible for observations at Zeppelin.

5695-20. How is bimodal (externally mixed) aerosol treated in this scheme? How often were distributions simple and mono-modal? Where changes in the chemical composition consistent with variability in the scattering enhancement factor?

Individual size distributions were monomodal during most of the time. The aerosol was treated as internally mixed with a sectional distribution over 20 size bins. The variability of the enhancement factor was found to be consistent with changes in chemical composition. Further details can be found in Rastal et al. (2014).

5696-1. The average contribution is minor, but were any biomass burning episodes observed which would result in anomalous comparison data points?

As stated in the answer to a previous comment, we are certain that biomass-burning aerosol did not affect the comparison cases conserved in this study. We added the following text to the new Sect. 2.2.1 to elaborate on the issue of absorbing particles:

Note that absorption contributes less than 75% to the ambient dry extinction coefficient of Arctic aerosols (Eleftheriadis et al., 2009; Zieger et al., 2010). This is in agreement with the PSAP measurements at Zeppelin. The effect of light absorption decreases even further when ambient extinction coefficients are considered.

5697-3. The effective radius calculation seems superfluous and seemingly was not used in the analysis. I would suggest removal.

Drying the particles could move them into a size range at which they are no longer efficient light scatterers. Consequently, not accounting for humidity effects will make it impossible to reconcile dry in situ measurements with ambient remote sensing observations. This is one of the major problems of studies like the one presented here. The effective radius is a parameter that is of enormous relevance for optical measurements and is commonly used in the remote-sensing community. Hence, this paragraph is not superfluous at all as we use it to illustrate the problem described above. Nevertheless, we now omit the equation to reduce the paragraph to:

The humidification of the particle number size distribution obtained with the DMPS leads to an increase of the particle effective (surface-weighted) radius from 0.14 ± 0.02 *to* $0.23 \pm 0.04\,\mu$*m (yearly average, not shown). This moves the aerosol from an optically*

ineffective state to a size range in which they are more efficient in interacting with visible light. Contributions of particles larger than the maximum DMPS size bin would lead to an overall increase in the effective radius, and thus, further improve the light scattering efficiency of the probed aerosol.

5701-1. It is hard to believe that distances this large are applicable in most environments. Can you comment on this result, based on your work?

In a previous study (Tesche et al., 2013), we used the trajectory approach (i.e. linking CALIPSO ground tracks to a fixed ground site with the help of forward and backward trajectories) for a validation of CALIOP profiles (particle backscatter coefficient at 532 and 1064 nm, particle extinction coefficient at 532 nm, and particle depolarization ratio at 532 nm) with ground-based lidar measurements at Cape Verde. We could show that successful comparisons of profiles of aerosol optical properties can be performed for distances of as much as 500 km. We reduced the maximum distance for comparison to 300 km in the present study to account for the more heterogeneous meteorology in the Arctic compared to the tropical north Atlantic. It is the advantage of the trajectory approach that overpasses at larger distance compared to the closest approach method can be included in a comparison study. Similar results have been found by studies of Anderson et al. (2003) and Kovacs (2006), which we refer to in the beginning of Section 3 (Comparison Approach) together with Tesche et al. (2013).

5705-13. The dependence on wind direction is weak and only really depends on a few datapoints at high extinction. I would suggest an analysis more quantitative than point-coloring for this figure. Wind-rose plot?

The color coding in this figure refers to the lidar ratio used in the retrieval of the CALIOP extinction coefficient. It shows that the largest absolute differences in extinction coefficients occur for cases with westerly flow and unusual lidar ratios (aerosol type). As the west of Svalbard is ice-free, it is possible for the CALIOP aerosol classification scheme to select from a larger pool of lidar ratios. A respective statement has been added 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)

Figure 2. panel a, the colors for the labels ('no features' etc.) are difficult to distinguish.

Sorry for that. We increased the size of the labels to improve readability.

Figure 2. panel b, is 'cloudfree' and 'aerosol only' the same data? If so, please use consistent labels. Likewise for 'cloudy' and 'clouds and aerosols'.

We harmonized the labeling and the colors used in Figure 2: cloud-free was changed to aerosol only and cloudy was changed to clouds + aerosols. We also shortened the figure caption to:

Histograms of the monthly abundance of (a) CALIOP level 2 5-km aerosol profiles and (b) 60-m height-bins with aerosol observations as detected during 2018 CALIPSO overpasses in the region of interest during 2008. The color coding refers to the observed occurrence of atmospheric features (aerosols and/or clouds).

Figure 3. The triangles at the top are difficult to discern, please increase size. Since the colors are the same as other symbols in the figure, it is confusing to interpret. Consider using different shapes?

The colored triangles were replaced by different black symbols. The corresponding part of the caption was changed to:

Symbols and corresponding numbers mark CALIPSO overpasses that could be connected to the ground site for the considered time period: only aerosol features (triangles), aerosol and cloud features (diamond), and no or only cloud features (circles).

Figure 4. Is there any linear correlation between variables? Can a regression line

w/confidence limits be added to provide some statistical basis of the correlation? An average CALIOP/in-situ factor of 1.85 and is noted in the text, can these be shown in Figure 4? Are geometric means more appropriate for log/log plotting like this?

We could not find a linear correlation between the variables and refrain from adding a correlation line with a squared correlation coefficient of 0.16. Note that Fig. 4 now shows the results of humidifying the dry nephelometer measurements and that the discussion of this figure has been revised accordingly.

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