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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Effects of relative humidity on aerosol light scattering in the Arctic" *by* P. Zieger et al.

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We thank the second anonymous referee for all of his comments and suggestions. They helped in improving the paper in content. In the following we will respond to each comment in detail.

Comment: This paper presents new findings on the f(RH) quantity in a region with large uncertainty in modeling due perhaps to a lack of such measurements. The dual nephelometer method appears useful and would be beneficial if extended to other sites in the Arctic. The most surprising finding was that f(RH) stays mostly constant throughout the observation period despite changes in composition and size distribution.

My comments refer to questions I had in order of appearance:

Comment: [1] "Observations made at the Zeppelin station are in general less affected by local particle production occurring in the surf zone and assumed to represent boundary layer conditions" I do not follow the difference between the â€IJsurf zoneâ€$™ and the â€IJboundary layerâ€$™. To me they imply the same thing, although I suspect the authors mean differently. A simple clarification would make this sentence better, and perhaps explain why the local surf zone is not as important.
Reply: We have added a sentence for clarification: "Compared to the stations located at the airport and in the village, the altitude and the distance from the shoreline gives the advantage that the Zeppelin station is less susceptible to the surf and sea spray from breaking waves around the fjord area."

Comment: [2] Why is coarse mode fraction defined as $D_{\text{dry}} > 462$ nm? Modes have often have ambiguous definitions in the literature, and coarse is sometimes defined as particles greater than 1 micron, or even particles greater than 2.5 microns, and sometimes greater than 10 microns. Maybe a different term could be used, to avoid confusion with regulatory terms?
Reply: We agree. Previously we have defined the coarse in this way, because we wanted to use the SMPS in its full range, as this is the instrument with the better precision compared to the OPC. The coarse mode fraction is than the volume measured by the OPC divided by the total volume of SMPS+OPC. The wording "coarse mode fraction" might be a bit confusing for the reader and therefor we have changed it to "volume fraction of large particles" within the entire manuscript.

Comment: [3] How were soluble ions measured? I would expect ion chromatography to be used, but it was unclear what method was used.
Reply: Yes, ion chromatography was used. We have added a paragraph with the detailed analysis method:

"Prior to ion chromatography analysis, the Zefluor Teflon filters were soaked in Milli-Q water (10 ml) and subjected to ultrasonic agitation (30 min). The extracts were analyzed with respect to Ca$_2^+$, K$^+$, Mg^{2+}, Na$^+$, and NH$_4^+$ on a Dionex 120DX ion chromatograph, using a Dionex cation exchange CS12A column (4 mm x 250 mm), and a conductivity detector. The sample was eluted using sulfuric acid at a flow rate of 1 ml min^{-1}. Cl$^-$, NO$_3^-$, and SO$_4^{2-}$ were analyzed on a Dionex 120DX ion chromatograph, using a Dionex anion exchange AS9-SC column (4 mm x 250 mm), and a conductivity detector. The sample was eluted using carbonate at a flow rate of 2 ml min^{-1}"

Comment: [4] How is it possible to have losses in BOTH accumulation and coarse? Diffusion and Impaction are separate processes that should occur in different flow rates.
Reply: Yes, we agree, these are different processes. The sources of the losses are not fully understood. We have added a section about the inlet systems and also added a second hypothesis (in Sect. 2.5), that the differences might have been caused by different flow expositions of the two inlets and differences in the inlet characteristics themselves (ill-defined virtual impaction behavior, sedimentation losses for larger particles/hydrometeors, see comments of reviewer 1).

Comment: [5] Kelvin effect is small for large particles ... relevant to light scattering and absorption" The term â€IJsmall and largeâ€$™ are vague. Again, I am confused by size definitions. This statement would be better if distinct diameter values were stated?
Reply: We have modified the sentence: "This is justified in our case, because the Kelvin effect is small for large particles ($D > 100$ nm), which are relevant to light scattering and absorption."

Comment: [6] Could the assumption of fixed refractive index (without an imaginary part) actually be controlling the rather constant optical properties? If chemical composition changes, then so should index of refraction. Especially with the seasonal influence of black carbon to the Arctic, which would add imaginary absorption.

Reply: Yes. We have added several sentences that our assumptions are only valid for non absorbing aerosols, which could be made since we did not detect any local or long range transport of pollution during our period. In addition, we have added a sentence that shows the sensitivity to an imaginary part of the refractive index (see comments by reviewer 1).

Comment: [7] A HGF can be predicted with thermodynamic water uptake, as in the Ming and Russell papers that were cited. Which could be used as the g in your equation.
Reply: Yes, this could be made, although no full chemical analyses were available for our period (especially the organic compounds are missing). This makes the calculation difficult for us, because additional assumptions would have been necessary.

Comment: [8] Why 24-hour e-folding lifetime? What size has this decay?
Reply: Within 24 hours we have an exponential decay in particle number particle concentration. Since we are using simplified assumption, we assume the same time decay as an average for all particle sizes and donâ€$™t consider explicit washout.

Comment: [9] "No clear long range transport of pollution to the measurement site has been observed within our measurement period?" How do you know this?
Reply: Yes, this statement should better be at the end of the paragraph, after the aethalometer, filter and FLEXPART results were presented. We have deleted that sentence at the beginning of paragraph and modified the second part:
"... The observations at Zeppelin indicated no clear long range transport of pollution to the measurement site, as can also be confirmed by transport simulations. The FLEXPART backward simulations showed that the air masses reaching the Zeppelin station mainly originated from the Arctic region surrounding Svalbard and the North Atlantic

Ocean (see Fig. 5). Hence, the measurement period was characterized by maritime and rather clean air masses."

Comment: [10] Could the last 3rd of the campaign be influenced by sea ice? This is when sea salt is higher if coming over Arctic. I recommend considering sea ice as sea salt aerosol source.
Reply: Yes. Sea ice can be a potential source for sea salt. We have implemented sea ice sources in the simulation according to the description of Yang et al. (2008) and references therein. Our general result has not changed; only for a few days during last third campaign (around 3/09, 34/09 and 12/10) higher sea salt contribution from sea ice covered areas was estimated. This points again to the result of the trajectory analysis that most of the dominant surface contacts were regions expected to be free of sea ice during that time of year (Fig. 5).
We have modified Sect. 4.2:

"Sea salt sources from open water were calculated following the parametrization given by Gong (2005) based on simulated 10-m wind speeds. Wind speeds were taken from ECMWF analysis and $+3$ h forecast fields and were available every 3 h with a 1 by 1 horizontal resolution. Sea salt in 3 different size ranges was considered: 0.01–10 µm, 0.1–10 µm, and 1–10 µm. Emissions from ice covered areas were considered seperately according to the parameterization by Yang et al. (2008) and references therein. The fraction of open sea water was obtained from daily sea ice analysis (http://cersat.ifremer.fr/data/discovery/by_parameter/sea_ice/psi_ssmi).

The sea salt aerosol number concentrations at the receptor site were calculated by summation of the products of residence times and sea salt over all grid boxes and for each simulated time, t:

$$S_N(t) = \sum_l \sum_{i,j} \exp\left(\frac{-T_l}{\tau_{\mathrm{ss}}}\right) \tau_{i,j,l} \frac{F_{N_{i,j}}}{V_{i,j}}\,, \tag{1}$$

where T_l is the time before arrival, τ_{ss} the life-time of the sea salt tracer, τ the residence times in units of s, F_N the sea salt source (from open water plus ice sheets) in units of N$/$(m^2 s) and V the grid box volume of the lowest FLEXPART output grid (100 m level top). The summation runs over all horizontal grid boxes i, j and along the integration time l."

Comment: My biggest comment is the feasibility of extrapolating the findings to the whole Arctic for the whole annual cycle. Especially when no long range transport was noticed, and when sea ice probably covered some of the ocean. Have you considered what might happen to the constant f(RH) if biomass burning, long range pollution/dust and sea ice influence reached the station? This is not a pan-Arctic study, and these findings might be different in Alaska, Siberia or Iceland, where chemical influences vary by region.
Reply: The influence of bio-mass burning aerosol will most probably reduce the f(RH), since enhanced organic and BC will lower the hygroscopic growth of the particle. Arctic haze mainly occurring in spring may also exhibit different f(RH). Thus these findings might be similar only for other sites in the Arctic region if the chemical composition is similar. We have stated clearly that this study is only representative for aerosol particles found in summer and fall at Zeppelin and it is difficult to generalize our results to the entire Arctic region. Therefore it is highly desirable to perform further measurements during other periods and other places in the Arctic. We have added a sentence in the conclusions: "In any case it is desirable to perform further measurements during other periods and other places in the Arctic."

References:

Yang, X., Pyle, J. A., and Cox, R. A., Sea salt aerosol production and bromine release: Role of snow on sea ice, Geophys. Res. Lett., 35, L16815, doi:10.1029/2008GL034536, 2008.

Gong, S. L.: A parameterization of sea salt aerosol source function for sub- and supermicron particles, Global Biogeochem. Cy., 17, 1097, 10.1029/2003GB002079, 2003.

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