Atmos. Chem. Phys. Discuss., 10, C1265–C1280, 2010
www.atmos-chem-phys-discuss.net/10/C1265/2010/

Atmospheric Chemistry and Physics Discussions

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

P. Zieger et al.

paul.zieger@psi.ch

Received and published: 6 April 2010

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

Comment: This paper summarizes 3 months of measurements of aerosol optical and physical properties at Zeppelin station in the Arctic. While Zeppelin has had long term measurements of light scattering and absorption at low RH, the unique finding reported here is the measurement of hygroscopic growth f(RH) measured by two nephelometers. Using size distribution measurements, an assumed composition and Mie code they do achieve closure on the dry scattering measurements. Based on the success of

this closure they derive estimates of g(RH) - the diameter change of the aerosol as a function of relative humidity. I like the discussion of the hysteresis index - I think that is quite clever and useful!
This paper is quite timely given the interest in the Arctic and the importance of aerosol optical properties to radiative forcing. I think the difficulty with this paper is in its introduction which should be expanded (described below) and the resulting confusion (to this reader) as the various parameterizations of hygroscopic growth are derived. Also, because these are the first f(RH) measurements in the Arctic I think it might be useful for the authors to include a table comparing the f(RH) results to other f(RH) results. There are a couple sentences at the end of section 5, but I think this should be expanded - there are a lot of f(RH) measurements out there. I don't think a lit review is necessary but putting the zeppelin f(RH) measurements in terms of polluted aerosol, dust and smoke as well as marine and free troposphere would be nice to see.

Reply: We think that our comparison to other literature values of f(RH) for different air mass types is sufficient for our purpose. An additional table will go beyond the scope of our paper. It will definitely be part in our planned publication, were we will summarize and compare all our WetNeph measurements performed at various European sites (Jungfraujoch, MaceHead and Spitsbergen) each being representative for certain aerosol types. We will also include resent measurements from Cabauw (The Netherlands) and Melpitz (Germany).

Comment: Below I list (in chronological order) a few technical comments which should be addressed as well as editorial (e.g., word-smithing) suggestions. Apologies for not separating out the technical and editorial - I did it this way and then read the instructions...

Comment: P3660 Line 9: change to: '...first time for aerosol particles present in summer and fall in the high Arctic.'

Reply: Sentence changed accordingly.

Comment: P3660 Line 15: Define dry RH e.g. RH $< 40\,\%$ (or whatever it was) in the dry neph

Reply: We added to that sentence: "...with a second nephelometer measuring at dry conditions with an average RH $< 10\,\%$ (DryNeph)."

Comment: P3660 Line 15 sentence: here and in all the rest of the paper -the humidified nephelometer would more appropriately be called the humidified nephelometer system. It's a commercially available nephelometer with a humidifier upstream of it.

Reply: This is an interesting suggestion, however, we would prefer to keep the name as is: The name has been used in the previous papers, it is short and simple. And it describes what it is: a nephelometer with a humidifier in front of it. In addition, we would like to mention that the nephelometer did need some important modifications as described in Fierz-Schmidhauser et al., 2010a): the original instrument would not have allowed for this application, due to its temperature increase within the instrument.

Comment: P3660 Line 27: Change 'found on average for g values of 1.61' to 'found, on average, g values to be 1.61'

Reply: Sentence changed accordingly.

Comment: General suggestion - starting in abstract and through paper - change 'g' to 'g(RH)' and consistently call the relative humidity induced scattering enhancement measured by the nephelometers f(RH). Doing so ties these two similar measurements together.

Reply: Yes, this is a good idea. We have changed it within the entire manuscript.

Comment: P3661 Lines 10-13: Split into two sentences and rewrite as follows: Continuous measurements of aerosol properties in the field, such as the wavelength dependent aerosol light scattering coefficient are often performed at dry conditions (RH<30-40% as recommended by WMO/GAW (2003)). These measurements at low RH can differ from the ambient conditions and thus may not be climatically relevant. [There are places where measurements at low RH are representative of ambient conditions.]

Reply: Sentence changed accordingly.

Comment: P3662 Lines 4-6: Rewrite as follows: Since no clear wavelength dependence of f(RH) was found during this study, lambda will be omitted for simplicity and the scattering enhancement factor will be written as f(RH).

Reply: Sentence changed accordingly: "Since no clear wavelengths dependence of $f(\mathrm{RH},\lambda)$ was found during this study, λ will be omitted for simplicity and the scattering enhancement factor will be written as $f(\mathrm{RH})$."

Comment: P3662 Lines 6-7: Omit the sentence Dry means a RH lower $30-40\,\%$ inside nephelometer - you've defined dry in the first sentence of the introduction.
Reply: Sentence deleted.

Comment: P3662 Line 8: Change to: 'Modeled and measured enhancement factors have been described...'

Reply: Sentence changed accordingly.

Comment: P3662 Line 11: Change 'or free tropospheric' to 'and free tropospheric'

Reply: Sentence changed accordingly.

Comment: General comment on the introduction - it would be quite helpful to the reader if you spent a little time discussing the various parameterizations of aerosol hygroscopicity and what the advantages/disadvantages of each are. Later in the paper you present g(RH), gamma and kappa as well as f(RH) without much background so it kind of blurs together into calculations that all show different values for clean and polluted air. Remember that people reading this may not be as familiar with the different parameterizations of aerosol hygroscopicity.

Reply: For clarification we have modified the description of g(RH) and κ from Sect. 3 (P3667) and moved the entire paragraph into the introduction (before the definition of f(RH)). "The growth of an aerosol particle due to water uptake is described by the hygroscopic diameter growth factor g(RH) which is defined as the particle diameter D_{wet} at a certain RH divided by its dry diameter D_{dry}:

$$g(RH) = \frac{D_{wet}(RH)}{D_{dry}} \tag{1}$$

The RH dependence of g(RH) can be parameterized in a good approximation by a one parameter equation, proposed e.g. by Petters and Kreidenweis (2007):

$$g(a_w) = (1 + \kappa \frac{a_w}{1 - a_w})^{\frac{1}{3}} \tag{2}$$

Here, a_w is the water activity, which can be replaced by the relative humidity RH, if the Kelvin effect is omitted. 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. The coefficient κ is a simple measure of the particle's hygroscopicity and captures all solute properties."

Comment: P3662 Line 16: '...humidified nephelometer system...'

Reply: See comment above.

Comment: P3662 Line 22-23: change to: 'Low RH aerosol light scattering measurements...' [no need to say long term because you say they've been made since May 2001]

Reply: Sentence changed accordingly.

Comment: Write a separate section on the two inlets. Give flow rates, heights, information about sizecut or not, what instruments are hooked to each, then discuss the instruments in more detail. This would also be a fine place to note that the humidity of the air brought into the lab is low because of the temperature difference between inside and outside.

Reply: We have added the following paragraphs at the end of the experimental section:

"**2.6 Inlet systems**

The WetNeph, aethalometer, OPC, and SMPS were all connected to one inlet which had no specific aerosol diameter size cut. The inlet consisted of a vertical pipe which sampled in about 2 m height on the roof of the station (pipe diameter approx. 5 cm, covered on top by a precipitation shelter). The instruments were located in the room directly below the inlet about 1 – 1.5 m away from the inlet entering the laboratory. The total flow was approx. 25 l min^{-1} (WetNeph: 16.6 l min^{-1}, SMPS: 0.3 l min^{-1}, OPC: 1.2 l min^{-1}, aethalometer: 8 l min^{-1}).

The inlets to the routine aerosol instrumentations run by the Stockholm University (DryNeph, CPC, DMPS, and PSAP) do not have an aerosol size characteristic cut off. The inlets consist of a 10 cm diameter carrier shaft with 0.25 inch stainless steel tubing to support the different instruments. The dry nephelometer has its own 0.25 inch inlet with a flow of approx. 6 l min^{-1}. The PSAP, CPC, and DMPS share another 0.25 inch inlet with a total flow of ca 5.5 l min^{-1}. The shaft and tubings are covered by a precipitation shelter approx. 25 cm high and are approx. 25 cm in diameter. The low flow rates of around 5-10 l min^{-1} will, for the most time, prevent hydrometeors and large aerosol particles to enter the sampling system.

During transport to the instruments, the ambient air sample will accommodate to room temperature, which is typically much higher than the outside temperature. This often makes the air dryer than 10 % RH."

Comment: P3663 Lines 7++: Change the section entitled 'humidified nephelemeter' to 'nephelometers'. discuss the dry neph measurements and then discuss the humidified neph system. What is the flow rate through the dry neph?

Reply: We have changed the section name to "Humidified and dry nephelometer" and described the WetNeph first, as it is our main instrument and then the DryNeph as our reference instrument. The flow rate of the DryNeph was 6 l min^{-1} (described now in the separate inlet section, see comment above).

Comment: P3664 Line 13: Is there a model/manufacturer for the SMPS (just to be consistent with your descriptions of the rest of the instruments)

Reply: It is a custom-built SMPS without a model number. Since the CPC was manufactured by TSI, we have added: "(CPC, TSI Inc., Model 3772)".

Comment: P3664 Line 22: Aerosol size distribution section- change '(like the SMPS at dry conditions)' to '(also at dry conditions)'

Reply: Sentence changed accordingly.

Comment: P3665 Lines 19-20: 'Due to the high SSA of the measured aerosol at zeppelin station, R was set to be unity...' This makes it sound like you are using the aethalometer data to determine the correction factor for the aethalometer. Please clarify.

Reply: Sentenced changed: "R was set to be unity, since the contribution of absorption to total light extinction is small (Weingartner et al., 2003), similar to the measurements at the JFJ (Fierz-Schmidhauser et al., 2010b)."

Comment: P3665 Line 22: The aethalometer does not measure black carbon! As you say above it measures light attenuation which can be used to calculate light absorption. Need to make some assumptions to come up with a black carbon concentration. I would suggest changing the sentence to: 'The 880-nm channel is sued to estimate aerosol equivalent black carbon (EBC)...'

Reply: We have changed the sentence: "The 880 nm channel is used to estimate aerosol equivalent black carbon (BC) concentrations, with the manufacturer's calibration." We would like to stick with the abbreviation BC, because it is more known and easier for the reader to follow in the later parts of the paper.

Comment: P3665 Lines 24-25: When you compare the PSAP and aethalometer what are you comparing? Aethalometer EBC at 880nm with PSAP light absorption at 565nm? please clarify.

Reply: Sentenced changed to: "A comparison of the aethalometer $\sigma_{ap}(\lambda = 565$ nm) (calculated using Eq. 4) with $\sigma_{ap}(\lambda = 565$ nm) measured by a soot absorption photometer (PSAP) running in parallel showed a good agreement with approx. 10 % difference and a high correlation (R^2=0.86). Note that Eq. 11 was used to interpolate the aethalometer values to the PSAP wavelength of $\lambda = 565$ nm."

Comment: P3665 Line 25: What inlet is the PSAP on?

Reply: Same inlet as the DMPS and CPC (see Sect. 2.6 Inlet systems).

Comment: P3666 Line 14: Combine this section with the wet nephelometer section as mentioned above?

Reply: We would prefer keeping this section here, since we also refer to size distribution measurements and mention acronyms which were described in a separate section before.

Comment: P3666 Line 15: Change to: 'A comparison of both nephelometers at low RH ($< 40\,\%$) conditions...'

Reply: Sentence changed accordingly.

Comment: P3666 Line 20: Change '...the DMPS also measured 27 $\%$ less...

Reply: Sentence changed accordingly.

Comment: P3667 Line 1: Change 'legitimate' to 'suggest'

Reply: Sentence changed accordingly.

Comment: Two general comment about neph comparison section:
(1) I'm assuming that you did do closure on the dry scattering with the measured size distribution using mie code. I think it would be good to state that and this would be one reasonable place to do so (the other place would be the size distribution section). I'm not sure which makes more sense.
(2) discuss particle losses (or lack thereof) in humidity system at high RH either here or in neph section.

Reply: Yes, good comment. We mention now that such a Mie closure has been done. The main argument is that both separate size distribution measurements (SMPS+OPC and DMPS) showed the same difference in number concentration. We have also added a sentence with an assumption where the losses were coming from. The following sentences were added:

"Since both DMPS and DryNeph had separate inlet lines but a joint precipitation shelter within the SU inlet system the differences might also be explained by different flow exposition to the SU inlet and/or differences in the inlet characteristics themselves (ill-defined virtual impaction behavior and sedimentation losses for larger particles/hydrometeors)."

"The differences between DMPS and DryNeph may be due to the fact that they sampled air from two different inlet lines."
"The measured size distribution (SMPS and OPC) and the measured scattering coefficients (WetNeph, when measuring at low RH) were found to agree well within a performed closure study using Mie theory (see Sect. 5.5)."
"Particle losses in the humidifier and dryer were characterized in a laboratory study for particle diameters 100-300 nm and found to be less than 5 $\%$ (Fierz-Schmidhauser et al., 2010a)." (Sentence added to the nephelometer section).

Comment: P3667 Line 18-19: '...Kelvin effect is small for large particles...' define large particles (D$>$X μm).

Reply: We added: "... ($D > 100$ nm) ..."

Comment: Comment on model description: While the Zeppelin aerosol is primarily scattering, it can have an absorbing component when sampling polluted air. However, you set the imaginary part of the refractive index to zero. I think it would be appropriate to acknowledge this and suggest what the effect (uncertainty) of ignoring the absorbing aerosol is in your Mie calculations.

Reply: Yes. As mentioned later, the refractive index of ammonium sulfate works fine for our period. For clarification we have added a sentence: "Neglecting the imaginary part of the refractive index is only possible if no strong absorbing aerosol is found, which is the case for our study."
Concerning the uncertainty we have added a sentence in Sect. 5.3: "As mentioned

above, neglecting the imaginary part is only possible because no strong absorbing aerosol was found during our period. The slope s between calculated and measured scattering coefficient (see regression line in Fig. 11) would decrease accordingly if an imaginary part was included in the refractive index (e.g. with m=1.53+0.001i → s=0.91, with m=1.53+0.01i → s=0.82 or with m=1.53+0.1i → s=0.56)."

Comment: P3668 Lines 17-19: Delete 'In this study...' through '...(NH4)2SO4 (see above)' as they are repetitive.

Reply: Sentences deleted.

Comment: P3668 Line 23++: 'Independent measurements of hygroscopic growth...makes it impossible to directly calculate or predict f(RH)...' in the literature, f(RH) is often referred to as hygroscopic growth and since you measure f(RH) here this sentence is confusing. I think what you mean to say is that since g(RH) was not measured. Please clarify.

Reply: We have modified the sentence for clarification: "Independent measurements of hygroscopic diameter growth factors (e.g. through measurements of g(RH) by a hygroscopic tandem differential mobility analyzer) were not available for this study, which makes it impossible to directly calculate or predict the scattering enhancement f(RH) (as done e.g. by Fierz-Schmidhauser et al., 2010b, for the Jungfraujoch)."

Comment: P3669 Lines 9-10: I'm confused about your use of the truncation correction. Earlier you say that you corrected the nephelometer data using the Anderson and Ogren correction and now you say you are only doing calculations for 7-170 degrees to avoid the truncation correction error. Please clarify.

Reply: The nephelometer delivers measured scattering coefficients for the angles 7-170. It is more precise to compare the calculated values directly to the measured

ones, if the calculation is directly done for 7-170. The Anderson and Ogren correction is based on a comparison of nephelometer measurements to model calculations to retrieve the entire scattering coefficient for 0-180 (via the measured Ångström exponent) and therefore brings additional uncertainties.
For clarification we have added a sentence: "By doing so, the calculated values can be directly compared to the direct measurements."

Comment: P3672 Line 1, rewrite: '...albedo wo is also very high during these seasalt events...'

Reply: Sentence changed accordingly.

Comment: P3672 Line 8: Change to: The aerosol filter analyses were only available for part of the three month period...

Reply: Sentence changed accordingly.

Comment: P3672 Lines 19-20: 'Humidograms...were determined as daily median values of f(RH)...' how much variability was there over the day? I know at other sites can see lots of variability in a day ± 0.5 even without much obvious change in airmass.

Reply: The variability can be seen in Fig. 7 (d), where the daily median values of $f(\text{RH} = 85\%)$ and the standard deviation as error bars can be seen. For most of the days the variability in terms of standard deviation is quite moderate (± 0.5), but for few days the variability is quite large ($> \pm$ 1) which is mostly due to instrument noise at low aerosol loadings and not air mass changes.

Comment: P3673 Lines 10-15: Using your parameterizations of f(RH) or fit equations in cited papers can you adjust one of the f(RH) values so they are all at the same RH for better comparison?

Reply: Yes, we can transform our mean values (mean for $84\,\% < RH < 86\,\%$) to different RH using Eq. 8. To allow a direct comparison, we have added in the manuscript (end of first part of the result section): "Using Eq. 8, our mean values can be transformed to the RH values used in the previous mentioned studies: f(RH $= 82\,\%$)=2.89 and $f(RH = 80\,\%) = 2.71$."

Comment: P3673 Parameterization of f(RH) Line 21: Equation 8 still has lambda in it though you say you will simplify and not use that earlier in paper.

Reply: We have deleted λ from the equation.

Comment: P3676 Line 5: change to '...tracer analysis also showed a possible...'

Reply: Sentence changed accordingly.

Comment: P3677 Line 25: change sentence to 'The index of refraction was assumed to be that of (NH4)2SO for the entire period...' Note - it's not really complex because the absorbing part of the RI is zero

Reply: Sentence changed accordingly.

Comment: P3678 Line 2: change sentence to 'The rest of the measurement period could be dominated...'

Reply: Sentence changed accordingly.

Comment: P3679 Line 2: Equation 11 is not clear and you can easily explain what beta is in terms of your measurements. I imagine what you did was something along the lines of the following if you were adjusting absorption to 550 nm using the 520 and 590 nm absorption measurements.

I'm not sure if the symbols will work when I submit the review but if they do it would be:
$\sigma_{ap}(\lambda_{550})$ = $\sigma_{ap}(\lambda_{520})$*($\lambda_{520}$/$\lambda_{550}$)
where = -log($\sigma_{ap}(\lambda_{520})$/$\sigma_{ap}(\lambda_{590})$)/log($\lambda_{520}$/$\lambda_{590}$)

Reply: Yes, one can use two discrete wavelengths to calculate the absorption coefficients at the wavelengths of the nephelometer, but it is more precise to use all available aethalometer channels. We have used an empirical fit. Therefore we would like to leave Eq. 11 as it is, since it is the most general form of the Ångström law. To clarify it a bit more we have added a sentence. "Equation (11) was fitted to the averaged spectra of the aethalometer measurement (using all available channels) to retrieve α and β, which then allowed to calculate the absorption coefficient at the individual nephelometer wavelength."

Comment: P3679 Line 18: change negligence to insignificance

Reply: Sentence changed accordingly.

Comment: P3679 Line 22-23: where get assumed neph uncertainty? See for example neph and PSAP uncertainty calculations in Clarke et al 2002. 10% seems very low uncertainty for aethalometer. Can you justify it? Clarke, A. D., et al., INDOEX aerosol: A comparison and summary of chemical, microphysical, and optical properties observed from land, ship, and aircraft, J. Geophys. Res., 107(D19), 8033, doi:10.1029/2001JD000572, 2002.

Reply: The initially assumed errors were quite optimistic. We have repeated the error calculations with a 10 % nephelometer uncertainty (Anderson et al. 1996) and a 20 % aethalometer uncertainty (M. Collaud, MeteoSwiss, Payerne, personal communication) and updated Fig. 10. The errors bars increase accordingly. The sentenced was changed accordingly: "...assuming a 10 % uncertainty of the nephelometer (Anderson et al., 1996), a 20 % uncertainty of the aethalometer (M. Collaud, MeteoSwiss, Payerne, personal communication) and..."

Comment: P3680 Lines 7-8: rewrite: '...there are cases where using SSA_{dry} can cause opposite signs in the radiative forcing than if SSA_{wet} was used (possible citation: Randles, C.A., L.M. Russell, and V. Ramaswamy, Geophysical Research Letters, 31, doi: 10.1029/2004GL020628, 2004.)

Reply: Sentence changed accordingly.

Comment: P3680 Lines 10-12: There are two things that would show the need to account for hygroscopic growth for the Zeppelin aerosol: (1) high values of f(RH) which you show are possible in your time series and humidograms (figs 4&5) and (2) high ambient values of RH. You should add an additional plot to fig 5 showing the time series of ambient RH (or at least state some statistics about what the summer/fall RH values are at Zeppelin)

Reply: The ambient RH (daily mean plus standard deviation) is already depicted in Fig. 10 (a), where we show the influence of f(RH) on the single scattering albedo. This is the best place for presenting the ambient RH, since we show in the panel below the actual ambient f(RH), and we would prefer leaving it here. Nevertheless we added a sentence about the statistics and refer to Fig. 10 again: "The ambient RH during our investigated time period is characterized by high values (RH mean: 89.0 $\%$, 10th percentile: 70.3 $\%$, 90th percentile: 99.3 $\%$, see Fig. 10(a)), which in addition shows the need to account for hygroscopic growth."

Comment: Figure 1 - the two mie code boxes say 7-170 degrees, but you said you corrected the neph values for truncation...please clarify

Reply: For the retrieval, the calculation was done in the defined scattering angles of the nephelometer ($7 - 170$) in order to directly compare the measured values (without applying a correction scheme to them). See comment above.

Comment: Figure 5 - add plot of ambient RH at zeppelin.

Reply: The ambient RH is given in Fig. 10 together with the actual ambient f(RH). In addition we have added a sentence about the RH statistics in the manuscript. See comment above.

References:

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, 2010a.

Fierz-Schmidhauser, R., Zieger, P., Gysel, M., Kammermann, L., DeCarlo, P. F., Baltensperger, U., and Weingartner, E.: Measured and predicted aerosol light scattering enhancement factors at the high alpine site Jungfraujoch, Atmos. Chem. Phys., 10, 2319–2333, 2010b.

Weingartner, E., Saathoff, H., Schnaiter, M., Streit, N., Bitnar, B., and Baltensperger, U.: Absorption of light by soot particles: determination of the absorption coefficient by means of aethalometers, J. Aerosol Sci., 34, 1445–1465, 2003.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 3659, 2010.