

Dear Editor,

We would like to thank you for the time you spent on the manuscript and finding the reviewers. The answers to the review comments were submitted as supplement files on 14 May, 2014, and we have modified the manuscript according to the point-by-point responses to the reviewer comments (see the responses below, also posted online in the Interactive Discussion). We have also updated the reference list, and corrected a few small typos in the manuscript. We hope that you can consider our manuscript for publication in *Atmos. Chem. Phys.* and are looking forward to any comments you might have.

Best regards,

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Reviewer #1

We thank Reviewer #1 for his/her positive comments on our manuscript. We have accounted for the comments by Reviewer #2 in the revised manuscript (see our responses to Reviewer #2).

Reviewer #2

We thank Reviewer #2 for his/her constructive and detailed comments which we will account for in the revised manuscript. Our point-by-point responses to the issues raised by the reviewer are below.

Scientific questions:

Figure 2 suggests that you aren't really using the nephelometer measurements other than as a check on the size distribution measurements. Didn't Zieger et al (2010) already show that the calculated/measured scattering was in good agreement at the site so you could skip all the mention of the nephelometer measurements and calculated/measured nephelometer comparisons and just use the size distribution measurements to do what you've done. Similarly don't think the $f(RH)$ discussion provides anything really new.

There are two important differences between our results and those of Zieger et al. (2010). First, Zieger et al. (2010) analysed data only from the campaign in the summer and fall, while our scope is to look at the full seasonal cycle. Second, Zieger et al. (2010) had a simpler kappa-parameterization using the coarse mode fraction as input, while we had a different approach in obtaining kappa values. Since the chemical composition of the aerosols at Zeppelin vary considerably throughout the year, we needed to adopt an approach where we assumed an internally mixed aerosol with five different components and consequently evaluate this

approach with the nephelometer data. We also evaluated the dry measurements with the SU-nephelometer data that was not performed before in Zieger et al. (2010). Because of these additional analysis / considerations, we feel that presenting a summary of the model evaluation is warranted.

Alternatively, why do you need to do the size distribution+Mie theory part when you are measuring wet and dry scattering (and absorption) and can determine other parameters (asymmetry parameter, SSA) from the measurements? Doesn't the radiative transfer allow for input of vertical profiles of aerosol optical properties without the added complication of using size distribution to calculate the optical properties? If you have the measured optical properties why would you introduce additional error by calculating them from size distributions?

The reason for using the aerosol size distribution data as a basis for our calculations is linked to the data availability: the wet nephelometer data was restricted to the 90-day period in the summer and fall 2008, while we are interested in the seasonal behaviour of the optical properties during the full year. Furthermore, the 12-year data set of the aerosol size distributions also allowed us to evaluate the representativeness of 2008 as compared with other years. It would indeed be ideal to have wet optical properties available throughout the year, but unfortunately this was not the case. However, as shown by Figs. 4-7, we believe that we get a reasonable representation of the aerosol scattering and absorption using the combination of the size distribution and composition data.

P 7068

line 25 "A better understanding on the radiative" → "A better understanding of the radiative"

We will modify the revised manuscript accordingly.

P7069

Line 5-6 "aerosol particles, thus affecting ADRE" → "aerosol particles, and thus can affect ADRE"

We will modify the revised manuscript accordingly.

Line 26-27 "depending on the location and atmospheric conditions." → "depending on the location." Comment: Table 1 doesn't provide any information about atmospheric conditions. Really, I think that the point is that aerosol properties in different at places.

This is a good point. We will modify the revised manuscript accordingly ("atmospheric conditions" will be deleted).

Line 27-29 "Fierz-Schmidhauser et al. (2010c) measured the hygroscopic growth and optical properties of 100nm ammonium sulfate and sodium chloride particles in laboratory and found values of $f(85\%)$ ranging from 6 to 18." Comment: I personally would probably delete this sentence since citing a single lab study of non-ambient particles is not as relevant as the field studies listed in Table 1. Otherwise I would rearrange the sentence to contrast it with the ambient measurements mentioned in Table 1: "Even higher values ($f(85\%)$ ranging from 6 to 18) have been found for 100 nm ammonium sulfate and sodium chloride particles in the laboratory (Fierz-Schmidhauser et al., 2010c)."

We will modify the revised manuscript accordingly, i.e. delete the discussion of the Fierz-Schmidhauser et al., 2010c paper.

Page 7070

Line 8-9 “contributed in a continuous reduction” → “contributing to a continuous reduction”

We will modify the revised manuscript accordingly.

Line 13 “both by e.g. altered” → “both by altered”

We will modify the revised manuscript accordingly.

Line 20-21 “campaignwise data” → “campaign data”

We will modify the revised manuscript accordingly.

Page 7072

Line 3 “water and produce saturated aqueous solutions” → “water, thus becoming saturated aqueous solution droplets”

We will modify the revised manuscript accordingly.

Line 16-19 need to define ‘T’ in equation 1.

We will modify the revised manuscript accordingly (T will be defined in the text).

Page 7073

Description of Mie model should clarify whether it is for homogeneously mixed spheres or shell/cores (I know it’s described later, but that’s something that should be mentioned here.)

We will modify the revised manuscript accordingly, i.e. add “assuming aerosol particles as homogeneously mixed spheres”.

Description of input to the radiative transfer model should also refer to section 4.1.3 for information on how the vertical profiles of aerosol properties were determined. (Currently in section 3.3 you just explain how you come up with values for the aerosol properties using Mie code, but not how you get the aerosol profiles.

In fact in the description of radiative model (p. 7073, line 21) we already do refer to section 4.1.3 for information on how the vertical profiles were determined. We assume this is a misunderstanding.

Page 7074

Line 9 I think you need a space: “calculations(see” → “calculations (see”

We will modify the revised manuscript accordingly.

Line 9 “8.00 to 12.00 a.m.” 12 am is defined as midnight. Do you mean 12 pm (noon)? Also, should clarify that it’s local time

We will modify the revised manuscript accordingly (change 12 am to 12 pm (noon) and add local time in the text).

Line 12 “10.00 to 12.00 a.m”, see comment above.

We will modify the revised manuscript accordingly.

Line 14 “sufficient sunlight” clarify, how many hours is ‘sufficient’

The chosen months correspond to months with typical daytime solar radiation exceeding 10 Wm⁻², which is approximately five times more than the typical variation of the instrument zero point. In the text we add “These are the months with daytime solar radiation higher than a threshold of 10 Wm⁻² (see sect. 4.1.4 for more details).” and in Sect. 4.1.4 we add “This value is chosen to be approximately five times more than the typical variation from the instrument zero point.”

Page 7075

Line 2 “The RH measurements” → “The surface ambient RH measurements”

We will modify the revised manuscript accordingly.

Line 13 “Twelve Whatman” → “Whatman ” Comment: 12 sheets were used because there was 1 sheet/month? I don’t think you need to include the 12. If you want to clarify the number of samples you could change the previous sentence “on a monthly basis from 1 September 2007 to 9 September 2008” → “on a monthly basis from 1 September 2007 to 9 September 2008 (for a total of 12 filter samples)”

We will modify the revised manuscript accordingly (remove “twelve” from the text).

Line 25 “MWS-OS was” → “MWS-OC was”

We will modify the revised manuscript accordingly.

Page 7076

Line 4-5 “determind” → “determined”

We will modify the revised manuscript accordingly.

Page 7077

Line 19 “This lead to” → “This leads to”

We will modify the revised manuscript accordingly.

Page 7078

Line 16-17 “do not represent the size-dependent chemical composition at the Zeppelin site, but rather an average bulk composition” Comment: In section 4.1.2 you’ve already noted that you are not dealing with a size dependent chemical composition, so maybe add something here reminding readers of that?

We think there is a misunderstanding here: In this section we are introducing the HTDMA measurements (i.e. they also represent a bulk composition) whereas in section 4.1.2 we explained the chemical composition that we used in our calculations.

Line 22-24 Comment: have you compared the asymmetry parameter and single scattering albedo from the Mie model output with the calculated values from the measurements? You can calculate an asymmetry parameter from the nephelometer and SSA from the nephelometer + aethalometer.

We only compared the scattering coefficients from calculations with the measurements, but given the relatively good agreement we feel that a more detailed comparison is beyond the scope of this paper and left for a potential future study (see also responses to the first comments).

Page 7079

Section 4.2.3 – I think you should mention $f(RH)$ in this paragraph just to clarify in the reader's mind the difference between GF (with a TDMA) and $f(RH)$ (with a nephelometer).

We will add also the definition of $f(RH)$ to Sect. 4.2.3 for clarity.

Page 7080

Line 3-5 “The large deviation for the June sample is probably due to the fact that during the period with high sea salt concentrations, the high volume sampler was not operating.” Comment: need to clarify – what does the hi-vol not working have to do with high sea salt concentrations?

The largest deviation can be seen in June, because in June there was a period that the high volume sampler was not operating (it was out of order) and at the same time there was a high sea salt concentration. So in the HTDMA measurements we missed high GF values related to the high sea salt concentration. Therefore, the measured GF values are lower than what were calculated in the model for June. We will clarify this in the revised manuscript by changing the paragraph to “The large deviation for the June sample is probably due to the fact that the high-volume sampler was out of order during part of June. This period coincided with high sea salt concentrations, causing apparent difference between the average predicted and measured GFs.”

Line 12 “of 80–120nm the filter samples” → “of 80–120nm, while the filter samples”

We will modify the revised manuscript accordingly.

Line 14-15 “Previous studies on the seasonal trends of chemical composition at several monitoring sites in” Comment: do you need to specify ‘coastal’ monitoring sites here? (Summit in Greenland probably doesn't get much sea salt)

This is a good point. We will specify in the revised manuscript that the previous studies that we refer to are those with marine influence. We add “with marine influence” to page 7080, line 15.

Page 7081

Line 4 “the modelled coefficients” → “the modelled scattering coefficients”

We will modify the revised manuscript accordingly.

Line 18-21 “The main reason for this difference is probably related to somewhat different dry references used in the studies (see Zieger et al., 2010 for details). The different dry reference values can be partly due to the different operating conditions, and partly due to e.g. different inlet structures and resulting losses.” Comment – this seems vague – are you talking about assumed dry RH? Assumed dry scattering? Other? You didn't discuss inlets/losses at all here – did those change between Zieger and your paper or is there another paper describing those changes that should be cited?

We will add “The measured size distribution and dry nephelometer data were taken from instruments connected to the SU (Stockholm University) inlet (without a size cut), while Zieger et al. (2010) performed their measurements on their own total inlet. However, their scattering coefficient and number size distribution measurements were approximately 25% higher compared to the SU inlet (see Zieger et al, 2010 for more details).” to the text for more clarity.

Line 23-25 “enhancement factors and size distribution together with an assumption on dry refractive index to retrieve the apparent hygroscopic growth factor.” → “ $f(\text{RH})$ and size distributions together with an assumption about dry refractive index to retrieve the apparent hygroscopic growth factors.”

We will modify the revised manuscript accordingly.

Page 7083

Line 3-4 “Seasonal variations of aerosol number size distributions, scattering coefficients and enhancement factors $f(\text{RH})$ are presented in Fig. 9.” → “Seasonal variations of measured aerosol number size distributions, and modelled scattering coefficients and enhancement factors $f(\text{RH})$ are presented in Fig. 9.”

We will modify the revised manuscript accordingly.

Line 22 “The enhancement factor $f(\text{RH})$ ” → “The enhancement factor $f(\text{RH}=\text{ambient})$ ”

We will modify the revised manuscript accordingly.

Page 7085

Line 15 “RH values about 50%” → “RH values of about 50%” Comment: This profile RH in Figure 11a is a lot lower than the RH values depicted in Figure 8a. Is there an RH profile that is more representative?

Indeed, the RH humidity in the example case shown in Fig. 11 is close to the lower end of typical values in April and consequently the same is true for the predicted scattering enhancement. To clarify this we add “It is worth noting that the surface-level RH on this example day is somewhat towards the lower end of typical values observed in April (see Fig. 8a), so the expected difference between Dry and Wet cases is larger on days with higher RHs.” To the text.

Line 18-21 “In contrast, a significant difference between the scattering coefficients calculated for the dry and wet cases is predicted, especially below 2 km (about 50%), where both RH and the aerosol particle concentrations are high.” Comment: I am surprised at the amount of scattering enhancement ($f(\text{RH})$) at RH 50%. It seems high compared to lots of other studies on hygroscopic growth, even at clean marine sites which should be similar to the high hygroscopicity sea salt aerosol measured at Ny-Ålesund. I think this is worthy of more comment/discussion (or cite other Ny-Ålesund $f(\text{RH})$ papers if they discuss this).

The scattering enhancement (about 1.5) that we observe is in line with the results of e.g. Zieger et al. (2010). We will point this out in the revised manuscript. Without any more detailed references it is difficult to know which studies the reviewer is referring to. We add “The magnitude of the enhancement is comparable to typical values reported by e.g. Zieger et al. (2010).” to the text.

Page 7086

Line 1 “(Bohren and Huffman., 1983),” → “(Bohren and Huffman, 1983),”

We will modify the revised manuscript accordingly.

Line 18-19 “while after the haze period there are a large number of smaller particles” → “while after the haze period the size distribution shifts to primarily smaller particles”

We will modify the revised manuscript accordingly.

Line 25 “which expectedly peak during” → “which peak during”

We will modify the revised manuscript accordingly.

Page 7087

Line 6 “vertical distribution of the aerosol particles and their chemical composition” Comment: there’s also uncertainty in the aerosol size distribution with height – it seems likely that the size distribution would look different within and above the boundary layer.

This is a good point, we will clarify this in the revised manuscript. The new text will be “...due to lack of information about the exact vertical distribution of the aerosol particle concentration, their size distribution and chemical composition.”

Line 14-15 “The dry size become more significant compared to RH when calculating the ADRE instead of the surface layer scattering coefficients” → “The relative importance of RH and dry particle size are reversed for ADRE and surface layer scattering coefficients.” Comment: I think my suggested change is what you were trying to say?

This is a good suggestion. We will follow it in the revised manuscript.

Line 23-24 “Figure 13 also demonstrates the importance of knowing the surface albedo for accurate predictions of ADRE, particularly during the early spring months.” Comment: You should expand on this – the importance of surface albedo is almost as large as the dry size for some months so give a range of the effect similar to what you did for RH and dry size. I am assuming the surface albedo at Ny-Ålesund changes because of snow melting and exposing bare ground? You could even include a statement saying something along the lines of: transitional months from snow cover to (rock and vice versa) might have the most uncertainty and that if this corresponds to the haze season (does it?) then things are even more uncertain.

This is a good suggestion and we will modify the revised manuscript accordingly by clarifying the driving processes behind the surface albedo changes. We add “when surface albedo is higher due to the snow covered surface. Surface albedo at Ny-Ålesund changes because of snow melting and exposing to bare ground. During the transitional months, from snow cover to rock and vice versa we have high uncertainty in ADRE.” To the end of the paragraph.

Tables/Figures

Table 1 – Sheridan measurements were made in Oklahoma (delete ‘South Kansas’) Add the results from this current Ny-Alesund study to the table either at the top or the bottom of the table. If Zieger et al (2013) repeats results from Fierz-Schmidhauser (2010ab) for Mace Head and JFJ, then maybe don’t repeat here – it’s like double counting. I’m assuming that the mineral dust $f(\text{RH})$ mentioned for Zieger was for JFJ and the Arctic aerosol was for Ny-Alesund, then just list those two sites for that paper?

These are good suggestions and we will follow them in the revised manuscript.

Table 2 Aren't some of the chemical components only known on a monthly basis (e.g., as noted in Figure 3)?

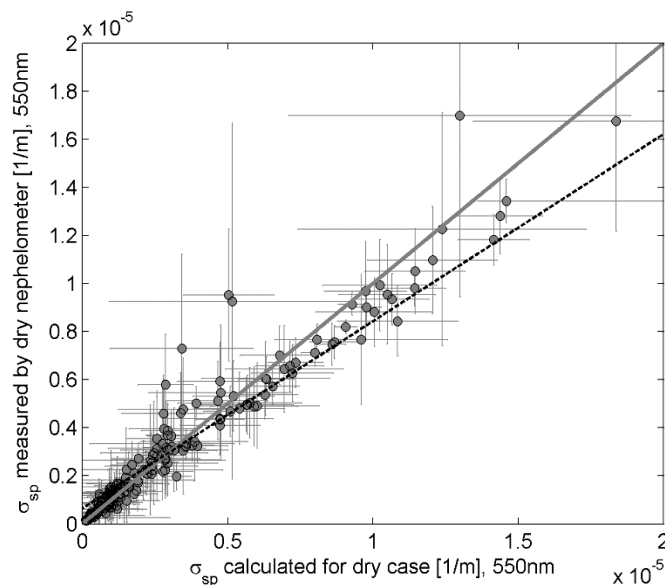
The mass fraction of organics is measured on monthly basis, but as the mass fraction of sulphate and sea salt are changing in daily basis the volume fraction used in Mie model changes on a daily basis (because of the different densities of the inorganic components). For more clarification we add "Please note that some of the individual chemical components are available on a monthly basis only (see text for details)." To the caption of Table 2.

Table 3 in the table notes, wavelength should be one word. Figure 3 In caption MWL-OC → MWS-OC.

We will modify the revised manuscript accordingly.

Figure 5a – does this really need to be on log scale? I feel like log scale hides things sometimes. How does it look with linear axes?

Due to the large variation in the values of scattering coefficients, we would prefer keeping the figures in logarithmic scale in the revised manuscript. However, we attach the corresponding Figures 5a and 6a presented in linear scale for the reviewer's information below. As can be seen, this way of presenting the data suggests no significant changes to our conclusions.



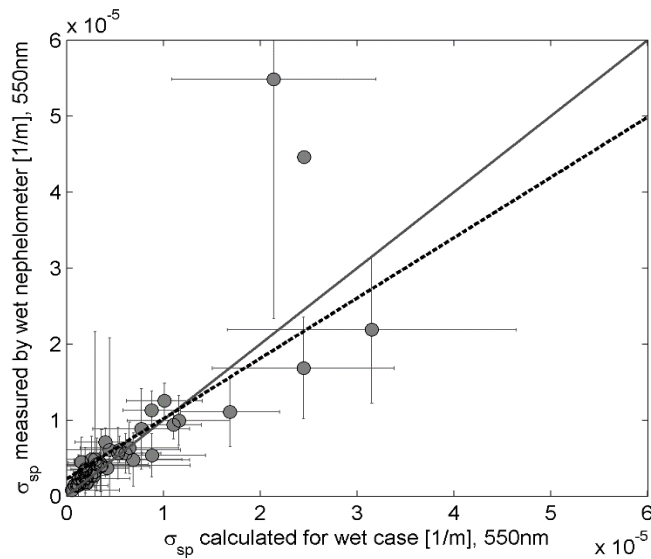


Figure 6a – are these for ambient RH or a set RH (e.g., RH=85%)?

Ambient RH, we will indicate this in the caption in the revised manuscript.

Figure 6b – if you split the histogram by season do you see a shift in the curves?

Unfortunately since there is no wet nephelometer data available for the whole year, we cannot present a seasonal comparison. However, for the four different months of the campaign are shown separately in the figure below. The positive values for August show model overestimation compared with the nephelometer and negative values for July, September and October show model underestimation compared with the nephelometer (we have 17 days of measurements for July and 12 days for October).

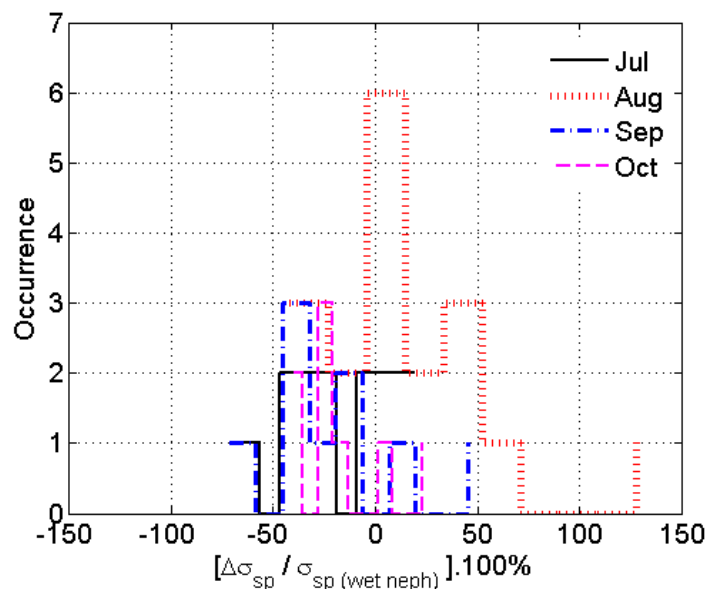


Figure 13 – In caption “(new cases)” is confusing – perhaps change caption to “The sensitivity of the ratio between the calculated ADRE and the ADRE (wet base case) to” or something like

We will modify the revised manuscript accordingly. We delete “new cases” from the caption.