

Review of 'Effects of relative humidity on aerosol light scattering in the Arctic' by Zieger et al

Overview:

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.

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....

P3660

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

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

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.

Line 27: change 'found on average for g values of 1.61' to 'found, on average, g values to be 1.61'

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.

Introduction

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.]

P3662

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

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.

Line 8: change to: 'Modeled and measured enhancement factors *have been* described...'

Line 11: change 'or free tropospheric' to 'and free tropospheric'

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 $g(RH)$, γ and κ 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.

Experimental

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

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]

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.

P3663

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

P3664

Line13: Is there a model/manufacture for the SMPS (just to be consistent with your descriptions of the rest of the instruments)

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

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.

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 used to estimate aerosol equivalent black carbon (EBC)...'

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.

Line 25: What inlet is the PSAP on?

P3666

Line 14 combine this section with the wet nephelometer section as mentioned above?

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

Line 20: change '...the DMPS *also measured* 27% less...

P3667

Line 1: Change 'legitimate' to 'suggest'

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.

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

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.

P3668

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

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

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.

P3672

Line 1, rewrite: '...albedo w_0 is *also* very high *during* these seasalt events...'

Line 8: change to: The aerosol filter analyses were only available for *part of the* three month period...

Lines 19-20: 'Humidograms...were determined as daily median values of $f(\text{RH})$...' 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.

P3673

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

Parameterization of $f(\text{RH})$

Line 21: Equation 8 still has λ in it though you say you will simplify and not use that earlier in paper.

P3676

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

P3677

Line 25: change sentence to 'The index of refraction was assumed to be that of (NH₄)₂SO₄ for the entire period...' Note – it's not really complex because the absorbing part of the RI is zero

P3678

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

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.

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$$b_{ap}(\lambda_{550}) = b_{ap}(\lambda_{520}) * (520/550)^{\text{angst}(\lambda_{520}, \lambda_{590})}$$

$$\text{where } \text{angst}(\lambda_{520}, \lambda_{590}) = -\log(b_{ap}(\lambda_{520})/b_{ap}(\lambda_{590}))/\log(520/590)$$

I'm not sure if the symbols will work when I submit the review but if they do it would be:

$$\sigma_{ap}(\lambda_{550}) = \sigma_{ap}(\lambda_{520}) * (\lambda_{520}/\lambda_{550})^{\tilde{a}} \quad \text{where } \tilde{a} = -\log(\sigma_{ap}(\lambda_{520})/\sigma_{ap}(\lambda_{590}))/\log(\lambda_{520}/\lambda_{590})$$

Line 18: change negligence to insignificance

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.

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.)

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)

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

Figure 5 – add plot of ambient RH at zeppelin.