Atmos. Chem. Phys. Discuss., 9, C6104–C6108, 2009
www.atmos-chem-phys-discuss.net/9/C6104/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measured and predicted aerosol light scattering enhancement factors at the high alpine site Jungfraujoch" *by* R. Fierz-Schmidhauser et al.

Anonymous Referee #1

Received and published: 21 October 2009

The paper presents interesting aerosol observations. Part of the value of this paper is that these important measurements related to climate change, and the Jungfraujoch site is a globally important site for such measurements. The paper does a lot with the dataset and provides some interesting results. The paper is well-written generally and clearly presents the ideas and conclusions. A few shortcomings need to be addressed before this is published which it should be ultimately.

The Anderson and Ogren model gives corrections for 1 micron or no size cut for their non-ideality corrections. The authors should state if any size cut was used on the nephelometer measurements as well as the other supporting measurements. If they

use size cuts on any of the measurements, these should be given in the appropriate sections.

The authors should state in their measurement protocol how they measure RH as this is a significant uncertainty with f(RH) measurements. This is particularly true with the light scattering instrument used which results in a significant heating of the sample and thus influences sample humidity. Do the measurements rely on the internal RH sensor of the TSI nephelometer? Is there any additional verification (e.g. chilled mirror hygrometer+dry bulb temperature)?

For their empirical fit (Equation 4), How did the authors find the one free parameter, a? Is the same value used for all cases? A dust aerosol may have a much different fit parameter than a sulfate aerosol. More detail here is needed.

I suggest the authors make an estimate the uncertainty this introduces. Numerous empirical fit models have been made to f(RH) data. One such uses an exponential fit using the single free parameter 'gamma' [e.g. Doherty et al, 2005]. Using an alternate fit like this would yield a measure of uncertainty in extrapolating the measurements to a different RH.

Along the same lines, the variability in RH for their measurement is a wide range (75<RH<95%), over which light scattering is very sensitive to this variation. I would characterize this as more than a 'slight variation'. This variability must also be considered in the uncertainty estimation.

Do the authors use the inversion given by the TSI SMPS software to correct for charging efficiency? How is the aerosol neutralized? The merging of dma and OPC data is a bit simplistic considering the differences in the techniques. A uniform shift of diameter does not address the difference in measurement technique where OPC measured data is sensitive to particle shape and refractive index variability. These cannot be merged completely realistically without consideration of these different measurement techniques [e.g. Hand and Kreidenweis, 2002].

ACPD

9, C6104–C6108, 2009

Interactive Comment

The time frame of the humidograms is quite long, 4 to 17 hours. Based on their measurements, the authors should comment on the potential for changes in air masses and aerosol chemical composition over these time frames. Could this impact the higher variability seen on the 17 hour measurement on May 6th? On P.7, line 45: "By passing over populated areas the air probably picked up more organic matter which results in a decrease of f(RH)." I would change this to carbonaceous material as elemental carbon would also have low hygroscopicity. Does the aethalometer confirm or deny higher BC concentrations during this period?

On p. 9, line 14: "The scattering coefficients were calculated for each measurement point of the humidified nephelometer. The calculated dry and humid scattering coefficients were ∼20% below the measured ones, which we attribute to a systematic bias in the measured model input parameters." Are the authors better able to substantiate this claim?

The assumptions used for the Mie modeling appear to be sound and appropriate. However, use of the AMS data particularly for prediction of growth factors and then f(RH) is uncertain. The collection efficiencies for the various constituents range from less than 50% to near 100%, dependent on which species as well as dependent on aerosol acidity, RH, and other factors. How can the authors claim across the board collection efficiency of 100%? These can't be ascertained without parallel conventional measurements of chemical composition, e.g. filter-based sampling. And how do you determine the molecular form of these compounds? What about dust species that the AMS will not see but which clearly affect the overall f(RH)? It's not surprising that the AMS doesn't well predict the f(RH). The AMS is generally sensitive to particles < 1um aerodynamic diameter whereas the other measurements are done presuming no size cut. These populations can be quite different, e.g. Saharan dust events. The calculation of refractive index from the AMS is likely a smaller uncertainty since the real part of the refractive index is similar for the species assumed. Can the authors state that the dust contribution (unidentifiable by the AMS) is negligible outside of the Saharan episode. I

ACPD
9, C6104–C6108, 2009

do recommend the AMS calculation of f(RH) be removed. At the minimum some more detailed information including justifications and caveats of using a limited composition dataset is needed to justify AMS predictions of f(RH).

The sensitivity analysis is a bit hard to comprehend at first, but I think I get it now. The reader comes away with the notion that size distribution does not matter for f(RH) at all. The authors should clearly make the assertion that the relatively constant size distribution at JFJ results in only a small variability in f(RH). Much more important at this site is the influence of changes in aerosol chemical composition. Size distribution would certainly influence f(RH) if the size distribution of the dust aerosol was used as the input in the sensitivity analysis.

On p.11, line 27: Since the interest is not only on the two RH ranges shown in Fig. 6, we display box plots of b and w for different RH bins in Fig. 7. Here we present a subset of the whole dataset where humidograms were measured (totally 51 hours of measurements). Each of the eight bins comprises a 10% range between 15% and 95% where N represents the number of 10-minute data. The limits of the boxes show the 25th and 75th percentiles, whereas the whiskers denote the 10th and 90th percentiles. The circles represent the mean values, whereas the horizontal lines in the boxes display the median values. The backscatter fraction b decreases with increasing RH from about 0.13 at 20±5% RH to about 0.09 at 90±5% RH. The decrease is not perfectly monotonous, but we assume that this is mainly due to the low number of points measured at 50% RH. w shows the opposite behavior above 50% RH, it increases with increasing RH. The observed deviation at 50% RH is again caused by poor statistics which is based on 12 data points."

I recommend the text description of the figure "Each of the. . . .the median values" be deleted since the caption more appropriately describes the figure details. Is the perturbation at 50% caused by a single outlier (b∼0.25 and w∼0.7)? The difference between the mean and median suggests such in which case its removal is merited. On P.3, near line 23: "The humidification system consists of a humidifier to rise the RH of the

ACPD

9, C6104–C6108, 2009

aerosol up to 95% RH, followed by a dryer, which dries to aerosol to the desired RH (Schmidhauser et al., 2009)." I suggest this be state that the humidifier 'raises the humidity of the aerosol to RH = 95%'.

On P.3, line 39 "Dividing σbsp by σsp results in the backscatter fraction b, which is the percentage of radiation that is scattered by an angle smaller than 90°." Don't you mean to say the fraction scattered between 90 and 180 degrees?

On P.3, line 18 "No drying of the air is needed to achieve this low RH, since the temperature difference between the ambient atmosphere and the laboratory is typically more than 25°C." This is not a negligible heating. Do the authors expect any losses of species other than water (e.g. organics, nitrate)?

On p.8, Line 22: "At the JFJ extensive aerosol properties undergo diurnal variations, strongest seen in spring and summer (Baltensperger et al., 1997, Lugauer et al., 1998, Weingartner et al., 1999)." Suggest 'most strongly observed'

Doherty, S. J., P. K. Quinn, A. Jefferson, C. M. Carrico, T. L. Anderson, and D. Hegg (2005), A comparison and summary of aerosol optical properties as observed in situ from aircraft, ship, and land during ACE-Asia, J. Geophys. Res., 110, D04201, doi:10.1029/2004JD004964.

Hand, J. L., and S. M. Kreidenweis (2002), A new method for retrieving particle refractive index and effective density from aerosol size distribution data, Aerosol Sci. Technol., 36 (10), 1012-1026.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 20063, 2009.