**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.

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Received and published: 10 January 2010

We thank Anonymous Referee #1 for all of his valuable suggestions and comments, which certainly helped us to improve this paper and make it more concise and clear. In the following we give detailed answers and explanations to the issues raised.

Comment: 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.

Response: It is written on page 20067, line 19, that the nephelometer measured scattering coefficients of total suspended matter (TSP). It is written on page 20071, line 2, that the Aerosol Mass Spectrometer (AMS) measured the size resolved aerosol chemical composition of non-refractory submicron aerosol particles. And we also wrote that the aethalometer has measured light absorption coefficients of TSP at seven wavelengths (page 20071, line 15). In the revised manuscript we will change the sentence on page 20068, line 12 to "... were corrected for the truncation error according to Anderson and Ogren (1998) (no size cut)."

Comment: 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)?

Response: In the revised manuscript we will add the following sentences on page 20068, line 2: "The RH in the nephelometer is measured with a combined temperature and capacitive RH sensor (Rotronic HygroClipS, accuracy +/-0.3°C and +/-1.5% RH, according to the manufacturer), which replaces the original RH sensor of the nephelometer. We further modified the nephelometer to achieve a temperature difference between inlet and sensing volume of the nephelometer of less than 1°C by placing an air-cooled infrared filter between the lamp and the sensing volume and by enlarging the outside of the nephelometer sensing volume with cooling fins, where four additional blowers surround the nephelometer and adjust its temperature close to room temperature (Fierz-Schmidhauser et al., 2009)."

Comment: 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.

Response: How we calculated the one free parameter a is explained on page 20069 with Eq. 4. For every measured f(RH) with related RH we calculated a value a, which we then used in Eq. 5 to get the recalculated f(RH) at the target RH. So for every f(RH) we have a value a. Before we finally used "our" empirical fit, we performed tests by fitting the data with the gamma function (used by Doherty et al., 2005). We found that with this fit-function we force values below the target RH to overestimate the f(RH=target) and force values above the target RH to underestimate the f(RH=target). If we reduce the RH range for fitting f(RH=85%) from 75-95% to 80-90% RH this will result in about 10% less data points (312 instead of 350) and the average f(RH=85%) changes by only 0.014. Since we would loose some valuable information of f(RH) (for example for the time of the SDE) we prefer to keep the RH range for fitting f(RH=85%) between 75 and 95% RH.

Comment: Do the authors use the inversion given by the TSI SMPS software to correct for charging efficiency? How is the aerosol neutralized?

Response: We corrected the charging efficiency with an own algorithm, which however does the same as the TSI SMPS software, with the only exception that our software does not force negative values to zero. We will change the sentence on page 20069 to: "This instrument consists of a Kr-85 source to neutralize the aerosol before the aerosol enters a differential mobility analyzer (DMA) followed by a condensation parti-cle counter (CPC, TSI Inc., model 3772)."

Comment: The merging of dma and OPC data is a bit simplistic considering the differ-ences 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].

Response: To investigate the error which results from the shift in diameter of the OPC, we calculated the mean dry scattering coefficient, scattering coefficient at 85% RH and f(RH=85%), and compared the obtained values using shifted and non-shifted average OPC size distributions. The calculated dry and humid scattering coefficients increased both by 20% by shifting the size distribution by 12% in diameter. Due to compensating effects the the size shift translates in an increase of only 3% in f(RH=85%). The result of this error analysis will be included in the revised version of the manuscript.

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?

Response: If we reduce the time of the humidogram of the 6./7. May, which lasts 17 hours to (for example) 9 hours the variability in the f(RH) stays. So we can conclude that this variability comes from a lower signal to noise ratio and not from a change in the air masses. The inorganic mass fraction changed between 0.52 and 0.46 within the 17 hours.

Comment: 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?

Response: We looked at mass fractions of the different compounds and not at absolute

concentrations, since a change in the absolute concentrations does not directly influence f(RH). The mass fraction of BC is not specially low or high on the days with a low f(RH), but the fraction of organic matter is high on May 6 and therefore the inorganic fraction is low on that day. The mass fraction of BC is never higher than 6%. Since the data confirms that the organic fraction/inorganic fraction (and not the BC fraction) is the main difference on the days with low and high f(RH) we would like to keep the sentence on page 7, line 45 as it is.

Comment: 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?

Response: The ratio between calculated and measured scattering coefficients for the three wavelengths is between 0.8 and 0.82 with correlation coefficients (R^2) above 0.95, so the ratio is about the same for the whole measurement campaign. The scattering coefficient at dry conditions depends on the aerosol chemical composition (via the refractive index), on the shape of the aerosol size distribution (or mean size) and on the aerosol concentration. The influence of the refractive index is not large enough to account for the encountered discrepancy of 20%. It is speculated that this discrepancy is manly caused by a small systematic error in the measurement of the size distribution (i.e., either in the determination of the diameter or number concentration, or a combination of both).

Comment: 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.

Response: In general the collection efficiency (CE) of the AMS is unimportant for the purposes of this study which primarily rely on the mass fraction of species and not absolute concentration. However, the CE of 1 was determined based on intercomparisons of the AMS with both SMPS measurements and nephelometer measurements, a CE of 1 was determined. This particular instrument has been deployed at several other locations and intercomparisons with other instruments including other AMS instruments consistently report a CE of 1 for ambient aerosol.

Comment: 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 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).

Response: To determine the molecular form of the compounds we used a simplified ion pairing scheme as described by Gysel et al., (2007). The anonymous reviewer #1 is pointing to a weak point of our analysis: that first the AMS only detects sub micrometer aerosol (and the nephelometer TSP) and second that the AMS does not detect refractory material like dust. This is an important point because Cozic et al. (2008) and Henning et al. (2003) have indeed shown that there is always a non negligible mineral dust contribution at JFJ. Luckily, the real part of the refractive index of mineral dust is 1.56 (Seinfeld and Pandis, 1998), which is very similar to the ones of ammonium nitrate

and ammonium sulfate, which are two of the three most abundant chemical species at Jungfraujoch. In addition, the choice of the dry refractive index is not a critical issue in the Mie modeling, since the refractive index at high RH will be anyway dominated by the one of water. We are aware that using the AMS measurements to predict f(RH) have limitations in correctness due to the problems that the reviewer mentioned. Nevertheless, we think that the prediction of f(RH) with the AMS is interesting and valuable for the community since AMS measurements are more widely used than measurement of the hygroscopic growth factors.

Comment: 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.

Response: We will add a sentence to this topic on page 20080, after line 22: "If the time period of the SDE would have been included in this sensitivity analysis the conclusion could be different, because during the SDE the shape of the size distribution varied more, than during the rest of the measured time period."

Comment: 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.

Response: We agree on this suggestion and the sentences from "Each of the eight bins . . ." to ". . .the median values" (page 20082, line 14-18) will be deleted in the manuscript.

Comment: 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.

Response: It is true that a single outlier increases the mean value of b at 50% RH. If this outlier would be removed the mean would be 0.125 instead of 0.134. It is also a single outlier which decreases the mean of w at 50% RH. If this outlier is removed the mean value of w increases from 0.904 to 0.925. The w at 70% for example also has a single outlier, which decreases the mean w. Should we remove this value too? We decided to present b(RH) and w(RH) as box-plots since in this way the reader sees that certain average values are strongly influenced by single outliers. In this way we do not need to remove values rather randomly. For this reason we prefer to keep all outliers.

Comment: On P.3, near line 23: "The humidification system consists of a humidifier to rise the RH of the 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%'.

Response: We agree and will change the sentence to: "The humidification system consists of a humidifier that raises the RH of the aerosol up to RH = 95%, followed by a dryer, which dries to aerosol to the desired RH (Fierz-Schmidhauser et al., 2009)."

Comment: On P.3, line 39 "Dividing ïĄşbsp by ïĄşsp 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?

Response: We already noted that this sentence is very misleading and we will change it to: "Dividing sbsp by ssp results in the backscatter fraction b, which is the percentage of radiation that is scattered back at angles between 90° and 180°."

Comment: 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)?

Response: Nessler et al. (2003) showed that at Jungfraujoch the dry total number concentration is often considerably smaller (on average 28%) than the ambient total number concentration. They assume that this is due to evaporation of volatile material at the higher temperature of the dry measurements. But they could prove that the particle losses mainly concern small particles with dry diameters smaller than 100 nm. Since the contribution of light scattering from aerosol particles with D < 100 nm is negligible, we can neglect here the particle losses due to evaporation. We will add s sentence to the revised manuscript on page 20067, line 24: "This temperature increase can lead to evaporation of volatile aerosol particles, but mainly in the diameter size range below 100 nm (Nessler et al., 2003)."

Comment: 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'

Response: We will change the sentence to "At the JFJ extensive aerosol properties undergo diurnal variations, most strongly observed in spring and summer."

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