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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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We thank Anonymous Referee #2 for his valuable comments and suggestions, which certainly helped us to improve this paper and make it more concise and better structured. In the following we give detailed answers and explanations to the issues raised.

Comment: Line 21-24 on page 3 Since the status of Schmidhauser et al. (2009) is “submitted”, the necessary technical details about the humidification nephelometer are needed. Did the authors modify the TSI nephelometer and use the humidifier control the relative humidity inside the nephelometer? Or the humidifier was set up in the upper stream of the nephelometer, then how was the relative humidity inside the neph-

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Interactive Discussion

Discussion Paper



Interactive
Comment

elometer measured? By the internal sensor of the TSI 3563 nephelometer or any other extra sensor were mounted? Since there could be large uncertainties regarding the RH control and especially inside the nephelometer because of the light source heating in the sampling chamber.

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 $\pm 0.3^{\circ}\text{C}$ and $\pm 1.5\%$ 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: Line 34-38 on page 3 The correction parameters given in Anderson and Ogren (1998) are particle size spectrum and chemical composition dependent. Since the nephelometer measured light scattering for total suspended particles, whether the no size cut parameters from Anderson and Ogren (1998) can be directly taken to correct the scattering coefficients measured at Jungfraujoch? Is the particle refractive index calculated with the AMS measure chemical compositions within the suitable range suggested by Anderson and Ogren (1998)?

Response: The real part of the refractive index calculated from AMS and aethalometer measurements varied between 1.48 and 1.56 and the imaginary part between 0 and 0.1. Anderson and Ogren (1998) did their calculations for refractive indices between 1.4 and 1.52 (real part) and 0 and 0.01 (imaginary part). The refractive indices of ammonium sulfate, ammonium nitrate and BC (see Table 1) are all above the values suggested by Anderson and Ogren (1998), so if a certain fraction of the chemical composition is composed of those compounds the resulting refractive index will be

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higher than the values of Anderson and Ogren (1998).

Comment: However, especially during the dust events, the particle size distributions may be very different than those at normal conditions and the forward scattering increase significantly. Due to the changes of particles size spectrum, particle shape and chemical compositions, similar truncation simulation as in Anderson and Ogren (1998) might be needed to find out the specific correction parameters for the Jungfraujoch dust cases.

Response: We have the opinion that a truncation simulation would go to far away from the topic of this paper. The focus of the paper should be on measurement of free tropospheric aerosol and not how to correct scattering coefficients. But we will change the sentence on page 20068, line 12 to: "... corrected for the truncation error according to Anderson and Ogren (1998) (no size cut)."

Comment: Line 23-25 on page 4 and line 13-19 on page 9 The authors should be careful to simply state that the 20% discrepancy between calculated dry/humid scattering and the measured ones were attributed to a systematic bias in the measured model input parameter. Because when the SMPS and OPC measured particle number size distributions were combined, a simple multiplication of the diameter by 1.12 (12%) were applied to the OPC data. Actually this cannot be described as a "slightly shift", because the connecting point was at 340 nm. Around this particle size (accumulation mode), particle number concentrations are normally high, and they are also within the most efficient light scattering size range at midwavelength. So a "slightly shift" at this size range, may cause notable changes in the simulated scattering coefficients. Uncertainty analyses are needed for both scattering simulation and also the for the calculated enhancement factor for scattering coefficients by taking into account uncertainties of all the input parameters in the Mie calculations.

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

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Interactive
Comment

$f(\text{RH}=85\%)$, and compared the values from shifted and non-shifted size distribution. Both dry and humid scattering coefficients increased by 20% by shifting the size distribution as mentioned in the manuscript. The shift led to an increase of the $f(\text{RH}=85\%)$ of 3%. With the shift, the calculated scattering coefficients are still lower than the measured scattering coefficients. If we assume an error of +2% in the RH the scattering coefficient at 85% RH with the shifted OPC is 27% and the $f(\text{RH}=85\%)$ 7% too low. If the error in the RH is -2%, the scattering coefficient at 85% with the shifted OPC is 14% too low and the $f(\text{RH}=85\%)$ is 12% too high. We did not include the uncertainty of the growth factor and the refractive index to this analysis, since we expect both to have a smaller influence than the size distribution and the RH. We will add a sentence on page 20070, line 4: “This shift increases the scattering coefficients on average by 20% and the $f(\text{RH}=85\%)$ by 3%.” and change the following sentence to: “A similar disagreement between SMPS and OPC was found in a different study. . .”.

Comment: Line 48-49 on page 7 and line 32-38 on page 8 The changes of wavelength dependency in the time series of measured $f(\text{RH})$ is quite interesting. Do the authors think the universal correction parameters for the measure scattering coefficient might introduce some uncertainties into this wavelength dependency?

Response: The changes in wavelength dependency of the measured $f(\text{RH})$ goes in line with changes of the Ångström exponent and also with the hygroscopic growth factors. In the beginning of the measurement campaign and during the SDE, when the $f(\text{RH})$ at 450 nm is larger than the $f(\text{RH})$ at 700 nm, the Ångström exponent is between 0 and 1.5 (or even below 0 during the SDE), and the growth factors of the particles with a dry diameter of 265 nm are smaller than the growth factors of smaller particles. The Ångström exponent is between 1.5 and 2.5 when we observed that the $f(\text{RH})$ at 700 nm is larger than the $f(\text{RH})$ at 450 nm. During this time period the growth factors increase with increasing dry particle size. Because of these results from other measurements, we don't think that the correction parameters of the scattering coefficient introduce uncertainties into the wavelength dependency, but that it is a real phenomenon. We

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Interactive
Comment

will add a sentence on these findings on page 20076, line 2: "... in the latter case, which is confirmed by higher Ångström exponents between 1.9 and 2.5. "

Comment: Since Mie calculations were used to predict the $f(\text{RH})$, whether similar wavelength dependency were found in the calculated $f(\text{RH})$ as well when the input size distributions significantly changed during the dust events?

Response: Unfortunately we could not perform Mie calculations during the Saharan dust event to predict $f(\text{RH})$, since the H-TDMA was not running during that time. As we observed also a different wavelength dependency of $f(\text{RH})$ in the beginning (1-3 May) and the middle of the measurement campaign (11-14 May) we investigated the wavelength dependence of the predicted $f(\text{RH})$ for these two time periods. The predicted $f(\text{RH})$ at 700 nm is always larger than the predicted $f(\text{RH})$ at 450 nm, but the ratio between $f(\text{RH}, 700 \text{ nm})$ and $f(\text{RH}, 450 \text{ nm})$ is smaller for the beginning than for the middle of the measurement campaign. For the prediction of $f(\text{RH})$ a mean growth factor was used. As explained above the growth factors of the particles with a dry diameter of 265 nm are smaller than the growth factors of smaller particles in the beginning of the measurement campaign whereas in the middle on the measurement campaign the growth factors increased with increasing dry particle size. We conclude that in the beginning of the measurement campaign the larger particles had a smaller growth factor, resulting in a decrease of the measured $f(\text{RH}, 700 \text{ nm})/f(\text{RH}, 450 \text{ nm})$ ratio. This is not reproduced by the model prediction as there a diameter independent growth factor was used.

Comment: Line 48-50 on page 10 Do the authors have some clue why the Nessler's simple Ångström approach performs better even during dust events, what mechanism drove this results... ?

Response: The Ångström exponent is a measure of the average size of aerosol particles population. Since the Nessler approach uses this information it can infer the relative contributions of hygroscopic fine mode particles and non-hygroscopic coarse

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Interactive
Comment

mode dust particles. When predicting the $f(\text{RH})$ with the growth factor g from the AMS and aethalometer, only species with a size below one micrometer were taken into account and even more important no information on refractory material like dust is given, since it is not detected by the AMS. The prediction of $f(\text{RH})$ with g from the H-TDMA during the SDE could be expected to be better than with the AMS/aethalometer since the H-TDMA characterizes also non-hygroscopic dust aerosols. On the other hand it is limited to relatively small particles (with a dry diameter < 265 nm).

Comment: Figure 4 on page 22 If we take a look at specific RH range (e.g., 30-50%, 50-60%), the model (except Nessler's approach in Figure 4d) has the tendency to underestimate the $f(\text{RH})$ at certain RH range, especially at lower RH range. What is the significance of this linear correlation? The uncertainties of the measured and calculated $f(\text{RH})$ need to be taken into account when doing the linear regression.

Response: We do not really understand what anonymous referee #2 means with this comment. The model is (if at all) rather overestimating the $f(\text{RH})$ at low RH and not underestimating. The horizontal structures (in red and yellow = at low RH) come from dividing the scattering coefficients measured by two different instruments which leads to some noise in the data.

Comment: Line 24 on page 3 “which dries to aerosol to the desired RH...” should be “which dries aerosol to the desired RH: : :”.

Response: We will change this sentence to: “. . . which dries the aerosol to the desired RH. . .”

Comment: Figure 1 on page 19 It is hard to distinguish the light green and grey color in Figure 1a. And why not to add a time series of AMS data in to Figure 1 too, for example, time series of sulfate, BC and OC (or their percentage in particle mass).

Response: We added to Figure 1 the time series of the mass fractions of OC, BC, sulfate, nitrate and ammonium. Therefore we had to change the figure caption to “Time

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series of the scattering coefficient at 550 nm wavelength (a), measured $f(\text{RH})$ recalculated to $\text{RH}=85\%$ of three distinct wavelengths (b) and mass fraction of the chemical composition measured by the AMS and the aethalometer (c). SDE indicates the time period, when a Saharan dust event was present. The scattering coefficients shown in light green are below 10^{-6} m^{-1} , those shown in grey are below $5 \times 10^{-6} \text{ m}^{-1}$. “ We changed the first sentence of section 3.1.1 (page 20074, line 16) to: “Figure 1 shows an overview of the measured scattering coefficient, the light scattering enhancement factor $f(\text{RH})$ at 85% RH and the submicron mass fraction of the aerosol chemical composition. “ And we changed the sentence on page 20075, line 17 to: “On these days we measured inorganic mass fractions higher than 0.57 (see Figure 1c), largest particle mean. . .”

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

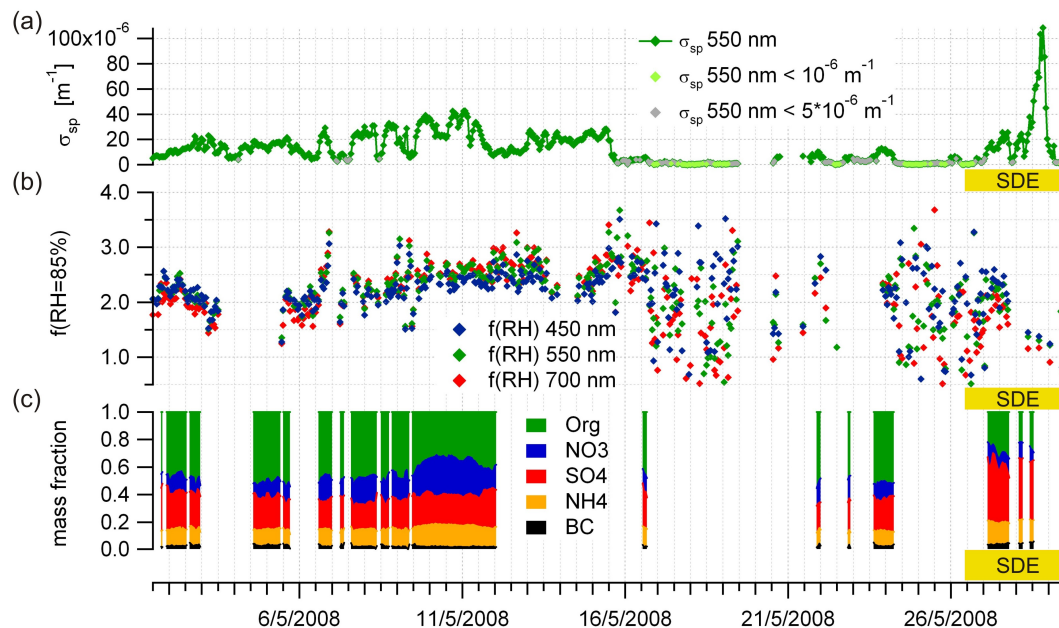
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Interactive Discussion

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