”Low hygroscopic scattering enhancement of boreal aerosol and the implications for a columnar optical closure study”: Reply to all review comments

We would like to thank all three reviewers for their detailed and constructive comments. They have greatly helped to improve the structure and content of our work. Our replies are given in blue colour below.

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Reply to reviewer #1 (A. Jefferson)

Review: Low hygroscopic scattering enhancement of boreal aerosol and the implications for a columnar optical closure study. Authors: P. Zeiger et al.

Please note that my original review was from a version of the paper with incorrect Greek symbols throughout the paper. I retract the original review, as many of my comments no longer apply. My sincere apology goes to the authors.

This paper looks at s study of aerosol scattering hygroscopic growth during the summer of 2013 in the boreal forest of Hyytiala, Finland. They compare the surface measurements for ambient extinction corrected for ambient RH to remote sensing of the AOD from a CIMEL sun photometer. A vertical profile of the aerosol extinction was obtained by scaling the surface extinction with aircraft vertical profiles of the total aerosol number concentration. The calculated extinction was remarkably lower than the reported Cimel AOD values. I recommend the paper for publication provided the authors make revisions and address the concerns outlined below.

2. The Field Site:

Can you specify the aerosol inlet heights for the system in the parking lot as well as the cottages in the forest? What were the inlet heights with respect to the forest canopy?

For clarification we have added to the manuscript:

“The inlets of the measurement containers were ~ 4 m above ground, while the inlets of the aerosol cottage were ~ 5 m above ground and ~ 10-15 m below the top of the canopy.”

3. Instrumental

The low RH values of the reference nephelometer will lower the scattering coefficients and may not be appropriate especially in a region with high organic and nitrates from forests and agricultural activity. Semi-volatile organics or nitrates will evaporate from the aerosol and greatly reduce the aerosol scattering coefficient. Even for a mostly salt aerosol the low RH values will result in a solid aerosol on the lower part of the hysteresis curve. This will obscure the fRH measurements, as optimally you'd like the fRH value to approach 1.0 around an RH of 40% when water content is minimal, but the aerosol is still on the upper branch of the hysteresis curve. Do you see much variation in the wet/dry scattering ratio with aerosol composition or size (Angstrom exponent) at the lower RH values of the humidifier scans? What was the RH inside the ACSM? The "a" parameter is the offset between the wet and reference scattering measurements in the lower RH region of the curve. It should become 1.0 around 40% RH, not 0% RH as the equation isn't valid at this low of an RH value. Offsets can result from aerosol loss in the humidifier, calibration offsets between the nephs, hysteresis and evaporation of low volatility compounds. For this experiment the values were close to1.0, but if they were otherwise it wouldn't necessarily be an indication of hysteresis.

The possible decrease of particle scattering due to evaporation of semi-volatile organics at low RH is difficult to prove. The optical closure study revealed that the in-situ measurements themselves were consistent (DMPS+APS compared to dry nephelometer). If there has been a "greatly reduced" aerosol scattering then it would have effected all in-situ measurements (size and chemical composition measurements) in the same manner. The organic mass dominates the aerosol composition and the scattering coefficient actually shows an increase with increasing organic mass fraction (see Figure 1). The effect of nitrates is probably small due to the low concentrations (see Tab. 2). Nevertheless, we can't fully rule it out so we have added to the revised manuscript (in the Sect. "Consistency of in-situ measurements: optical closure study"):

"Partitioning effects of semi-volatile organics (Donahue et al., 2006) or nitrate components (due to the low concentration to a lesser extent, see Tab. 2) that could have caused a potential decrease in the overall particle properties cannot be ruled out completely."

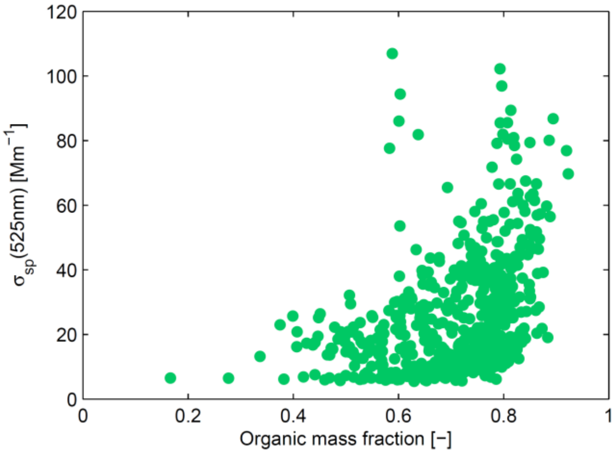

Figure 1: Particle light scattering coefficient at 525 nm vs. organic mass fraction.>

We do not agree concerning the statement that the *f*(RH) should be 1 at 40%. It is known that especially organic dominated particle already take up water at RH below 40%. This has been observed at several sites before (see e.g. Fig. 1 in Zieger et al., 2014, for the Melpitz site or Fig. 6 in Zieger et al., 2010, for the Arctic at Ny-Ålesund). The scattering enhancement factors can be easily underestimated (approx. by 10-15%) if they are normalized to 1 at 40% RH. This is why we first correct for relative differences of our nephelometers (at dry conditions) and then calculate the *f*(RH)-values. The parameter *a* in Eq. 2 is then ideally around 1 when the lower branch of the hysteresis branch of the measured data is fitted. For the upper branch it can be different and larger to 1 for example when inorganic sea spray particles dominate the aerosol distribution. In general, one would expect even at 40% RH that if the particles still contain water that their *f*(RH)>1. The losses of the humidifier/drier system are a separate issue and were experimentally evaluated by Fierz-Schmidhauser et al. (2010) and showed to be below 2.5 - 5%.

In opposite to other sites, we have not observed clear deliquescence behaviour at the intermediate RH range in Hyytiälä (as already stated in the manuscript) due to the dominance of organics. The variation is only slightly higher at low RH (e.g. at low RH=30-40% the standard deviation (STD) of *f*(RH) varies for 450nm on average between 0.06-0.09, while varies between 0.05 and 0.06 (mean) at high RH=80-90%). However, the statistics is also influenced by more measurement points at elevated RH.

The RH of the ACSM was on average 30%. We have added this information to the manuscript (see comment & sentence below; page 18).

Page 3334 The term "successfully compared" is ambiguous. How close did the instruments compare, across what size range or Angstrom exponents and with what type of aerosol? Can you specify that they came with a certain percent of the other using a certain aerosol type of a particular size and growth factor? You don't need to be elaborate, but give more information, otherwise "successful" is open to interpretation.
We agree that the statement is not sufficient as it is. The comparison of the WetNeph to the ACS will be shown in a separate instrumental manuscript of the ACS which is currently in preparation. Both instruments showed a good comparison for the periods when they were both operated in parallel. As a preview, the figure below (Fig. 2) shows the comparison of the fit-parameter gamma of the scattering enhancement as determined from Eq. 2. The instrumental paper for the ACS is still in preparation. We have modified the sentence and added the degree of agreement for the 525 nm wavelength as an example:

"The WetNeph showed a good agreement to a novel commercially available humidified nephelometer system (aerosol conditioning system (ACS1000) by Ecotech Pty Ltd.) for certain periods of the campaign. At 85% RH the median f(RH) agreed within 6% for 525 nm (M. Laborde, pers. com., April 2015)."

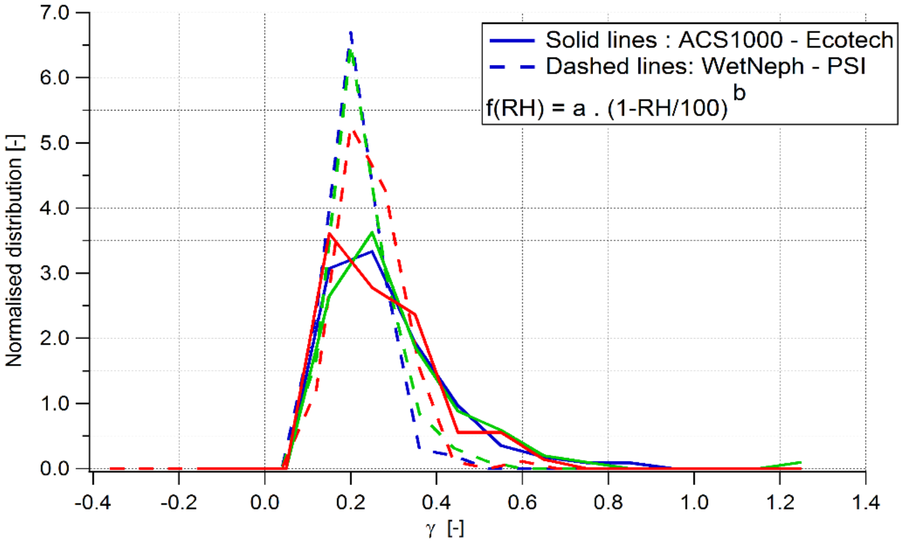

Figure 2: Probability density function (PDF) of the fit-parameter γ (see Eq. 2) describing the f(RH) determined from the WetNeph (instrument used here) and from the new commercially available instrument ACS (Aerosol Conditioning System, Ecotech; Laborde et al., 2015, in. prep.).

4. Trajectories

The back trajectory calculations seem long enough to allow for substantial particle loss, dispersion, cloud processing and multiple sources that could obscure the interpretation and disproportionally

favor an aerosol source region that had a minor contribution. Would you get different results with 5-day back trajectories or does the removal parameter in equation 4 aptly account for this?
Yes, the removal factor in Eq. 4 accounts in a simplified way for the removal of aerosols (assuming a half-lifetime of one week). Other removal mechanisms (e.g. due to precipitation) were not taken into account. As stated in the manuscript this part of the analysis is based on very simplified assumption, as are the trajectories based on reanalysis data and model calculations with attributed uncertainties. Nevertheless they allow us to give further insights on the origin and properties of the aerosol encountered at Hyytiälä.

5.1 Influence of water uptake on the aerosol The lack of correlation of fRH to the aerosol single scatter albedo and scatter Angstrom is unusual, as most sites even with a high organic fraction will exhibit this behavior. Can you explain the finding? Does the SSA vary with the organic and inorganic fractions of the aerosol the same as fRH? How does the Angstrom vary with the sulfate and OC mass fractions?
The Ångström exponent of the scattering coefficient (if determined by a fit across the three nephelometer wavelengths) did not show any clear correlation with *f*(RH). However, we repeated the analysis calculating the Ångström exponent using pairs of wavelengths / scattering coefficients and also comparing all major in-situ parameter against each other. The result is seen in Figure 3 (below), were the squared correlation coefficient of all major in-situ parameters being compared is shown. It can be seen that the Ångström exponent (using a fit) and the single scattering albedo do not significantly correlate with *f*(RH). However, if the Ångström exponent is calculated without a fit just by using the scattering coefficient at 450 nm and 525 nm a negative correlation (R^2=0.45) can be observed if compared to *f*(85%,450nm). The red channel might have influenced the determination of the Ångström exponent through a fit due to lower signal strength and more noise. Still, the correlation is not as significant as for the organic/inorganic mass fraction. One reason for this can be the small variation of the Ångström exponent and thus small variation in size throughout the campaign. We have added a table with the statistics on the measured optical and chemical parameters (as suggested by reviewer #3). Figure 3 also shows that there is no correlation between the scattering Ångström exponent, the SSA and any chemical mass fraction (maybe a small negative one with EC, as one might expect).

The small inverse correlation (R^2=0.45) between Ångström coefficient and *f*(RH=85%) is an effect of particle size and hygroscopicity. This is also revealed when looking at the trajectory analysis (see Figure 4 below). The high Ångström exponents (smaller particles) were observed when the air originated from the East with increased organic contribution. However, the effect is not obviously seen in all in-situ measurements (e.g. the mean size parameter Volume/Surface does not correlate with any chemical mass fraction). We have added this information to the revised manuscript and modified the last paragraph of Sect. 5.1:

"The measured scattering enhancement factors have been compared to further in-situ measured aerosol and meteorological parameters. No clear and significant dependency was found if compared to the single scattering albedo, aerosol size distribution parameters (total number concentration and mean size), wind direction or wind speed. An exception was the small inverse *correlation (R^2 =0.45) that was found for the scattering Ångström exponent (only when using the 450 and 525 nm scattering coefficients) and the total particle surface area. This can probably be explained by the fact that an increased concentration of mainly smaller particles (increased Ångström exponent) were also composed of more organic components (lower hygroscopicity), which overall caused a decreased f(RH). This is also seen in the trajectory analysis, which revealed that air masses from the East showed generally a higher Ångström exponent similar to the organic mass fraction (see Fig. 5c)."*

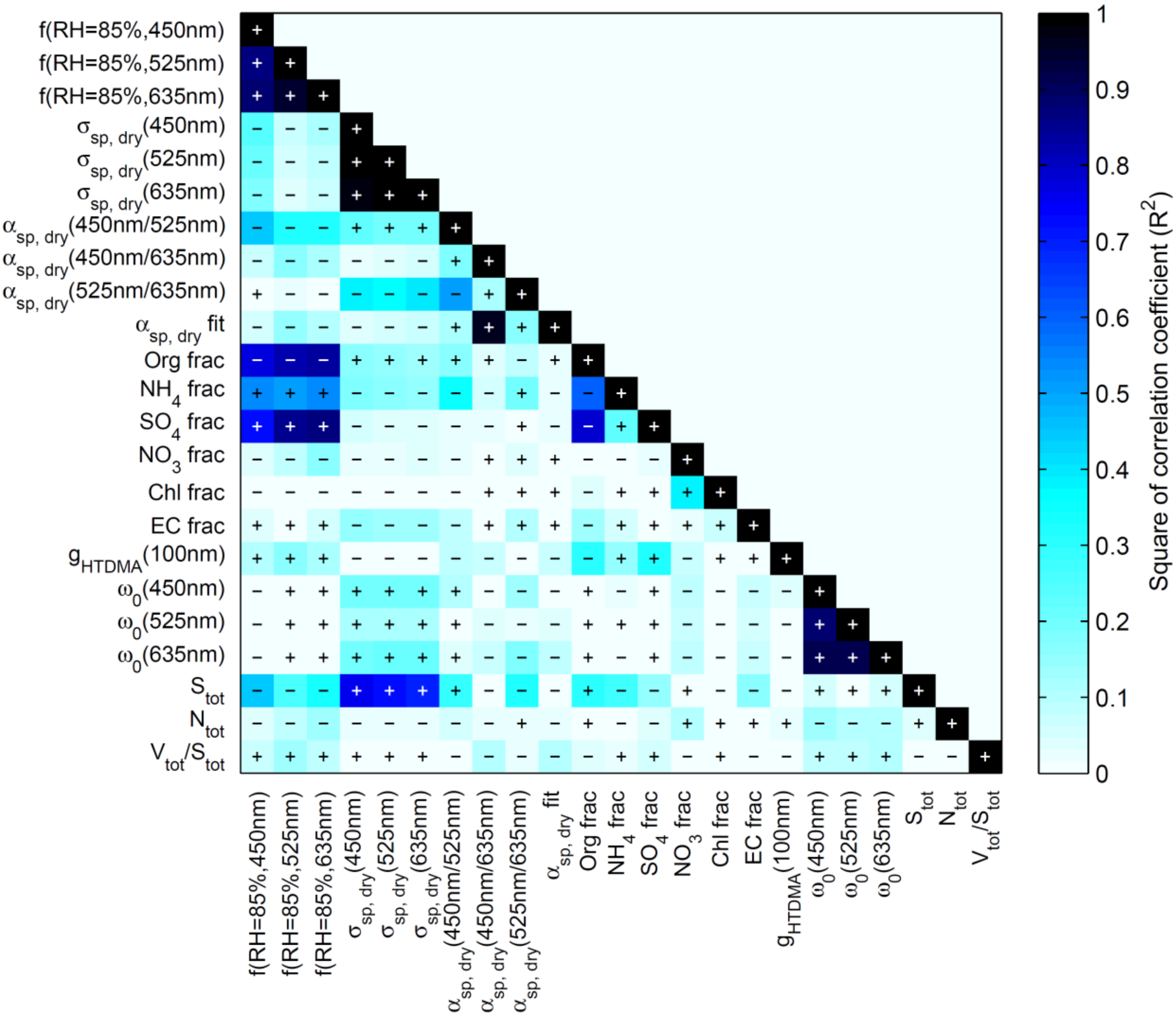

Figure 3: Correlation plot of different aerosol parameters measured at ground level. The colour code states the degree of the squared correlation coefficient, while the sign indicates a negative or positive correlation.

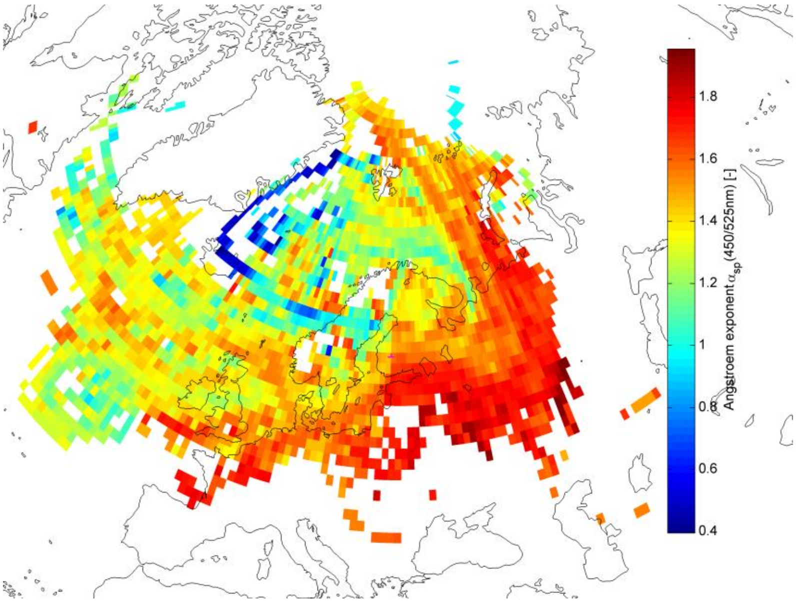

Figure 4: Result of the trajectory analysis (see Sect. 4 and 5.1. and Fig.5) but for scattering Ångström exponent using the scattering coefficients of the 450 and 525 nm wavelengths.

5.2 Instead of using the entire CPC count to scale the extinction coefficient can you use only the counts greater than 50 nm that are more optically active? The altitude dependent extinction may be skewed by the small particles with a low scattering efficiency.

During the preparation of the manuscript, we have tested the particle surface area measured by the SMPS as a scaling factor and the results looked similar. However, we prefer to use the total CPC as scaling factor, because it covers all particles including those above 270 nm, which are important for light scattering (see scattering size distribution in Fig. 12b in the original manuscript). We have added below (Figure 5) the result of the columnar closure study (the relative differences to the observed AOD by the Sun photometer) if the particle surface is being used instead of the total number concentration for the scaling factor. Although small differences occur for certain days, the overall picture and the magnitude of underestimation of the in-situ AOD stays unchanged. Thus we prefer to keep the total number concentration as scaling factor. For clarification, we have added to the revised manuscript (Sect. “Extrapolation to the atmospheric column using aircraft measurements”):

“It should be noted here that the following results are in a similar range if the particle surface is being used as scaling factor c(h) in Eq. 7 and 8, however that factor would omit optically active particles above the upper size limit of the airborne SMPS.”

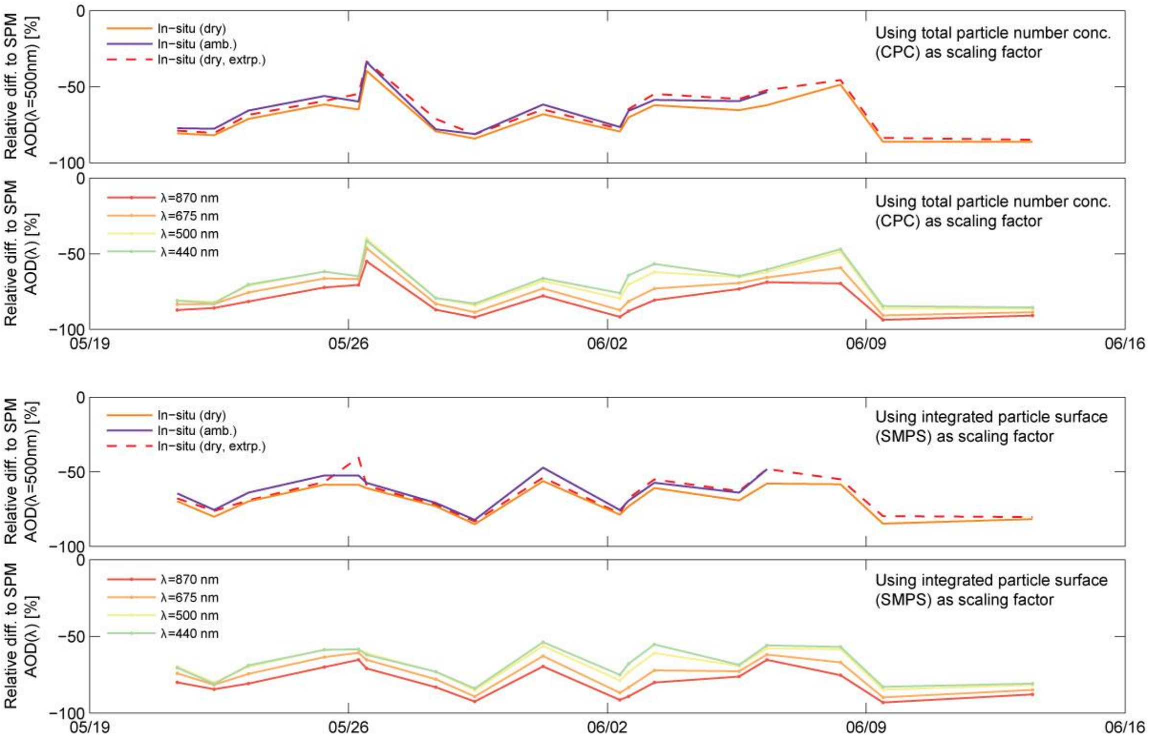

Figure 5: Relative differences of the in-situ AOD compared to the measured one from the Sun photometer. The upper two panels show the result when using the total number concentration as scaling factor, the lower to panels show the same relative differences if the particle’s surface of the airborne SMPS (with D<270nm) would be taken as scaling factor. This figure will replace Fig. 9 from the original manuscript (only the upper panels which are added to Fig. 8 in the revised manuscript).

In addition, we have plotted in Figure 6 the scaling factor *c(h)* using the integrated surface (from the SMPS for particles < 270 nm) vs. the scaling factor using total number concentration of the CPC (all particles, as which is finally being used). As one can see, the differences are not systematic and no large differences can be observed. Thus it is more consistent to use the total CPC to calculate the scaling factor since it captures all particles.

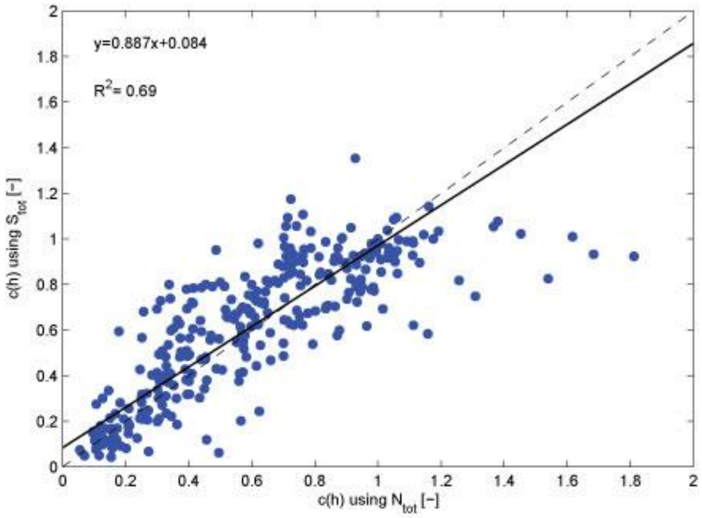

Figure 6: Scatterplot of the scaling factor c(h) (see Eq. 7) if the integrated surface of the SMPS is being taken vs. c(h) if calculated taking the total number concentration of the CPS as input.

Is there a measure of the boundary layer mixing height? Were flights conducted when the mixed layer was at its highest. i.e. 1-3PM LT?

The flights were conducted at all different times of the day. Besides the particle number concentration, no other PBL height measure was available for the campaign period at Hyytiälä.

Reply to reviewer #2

Using a WetNeph (measurement campaign in Mai-August 2013) the authors first calculate the scattering enhancement at Hyytiälä (Finland) and compare it with the one found at other stations over Europe. A clear correlation was found between the scattering enhancement and the organic mass fraction. Combining in-situ surface measurements with particle number concentration and humidity profiles measured by an aircraft, an estimation of the AOD was calculated at several wavelengths and compared to sun photometer measurements. The inconsistencies of this columnar optical closure study are then discussed in the light of an optical closure study for in-situ instrumentation and of the difference between in-situ and aircraft measured size distributions. Finally the contribution of high altitude aerosol concentration to AOD is estimated from aerosol backscatter coefficient profile from a lidar.

The paper is well written and scientifically very clear and the figures are self-explaining. It should be published after a major revision:

Main comments:

Results: 5.1, figure 4: you only plot the data of 2 over 6 sites. Taking into account all the sites, is the correlation between f(RH) and the organic mass fraction unique ? if yes is it possible to give a general rule f(organic mass fraction)?

Unfortunately, Hyytiälä and Melpitz (Germany) were the only two campaigns with concurrent and highly time-resolved AMS or ACSM measurements. At Jungfraujoch (Fierz-Schmidhauser et al., 2010) an AMS was also deployed but only for parts of the campaign with little variation and very low concentrations. This is also the reason why we did not show any scatter plots or correlations at that time in Fierz-Schmidhauser et al. (2010).

A general rule of *f*(RH) as a function of organics will only be valid for sites with similar coarse mode composition or a negligible coarse mode to the overall scattering coefficient in general. As we have shown in Zieger et al. (2013) in a general sensitivity study and in Zieger et al. (2014) specifically for the Melpitz site (continental aerosol), the coarse mode can significantly influence the *f*(RH) by having a decreasing (e.g. mineral dust) or an increasing (sea spray) effect. Nevertheless we have added a

linear regression to the joint Melpitz and Hyytiälä datasets of the γ-value which can be used to calculate f(RH) at any given RH. The Melpitz values were linearly interpolated to 525 nm and 635 nm (the original measurements were done with a TSI nephelometer at 450, 550, and 700 nm). It should be mentioned that we also tested other regression functions (like polynomial) but if looking at the three channels it turns out that the linear regression is the more trustworthy one. In the submitted manuscript we originally used the robust fit (a Matlab function that uses iteratively reweighted least squares and a bisquares weighting function and there is less sensitive to outliers), however, if x- and y-axis are switched and the inverse of slope and intercept slightly differentiate from the first fit. Therefore, we changed to a weighted bivariate fit which is the more reasonable one to use (here we used the STD of the averaged values as weights) and is based by the routine described by York et al. (2004). This procedure allows to derive an error of slope and intercept which we also state in the revised manuscript. We have removed the regression coefficients for the components that did not show any linear behaviour (NO_3, Cl, EC) since they are physically meaningless.
To improve the structure and readability we discuss the comparison to the chemical mass fraction and the proposed parameterization now in two separate subsections (Sect. 5.2. "*Comparison to the chemical composition measurements*" and Sect 5.3. "*A simplified parametrization for f(RH)"*). As reviewer # 3 also suggested, we compare our results to previous parameterizations. For the revised Sect. 5.2, 5.3 and revised Fig. 4 we like to refer to our answers to reviewer # 3 below (page 19-22) and the revised manuscript.

Results: 5.2: The AOD measured by the Sun photometer is a measurement of the whole atmospheric column beginning at the instrument altitude (18 m agl) above the canopy of the forest (§ 3.7). The calculated AOD begins at ground (It is not clear in your paper what is the beginning point for that integration) and end at the maximal altitude reached by the aircraft. If not done, you can probably begin the integration of equation (5) at 18 m with the extinction coefficient measured on the 17 m mast in order to have the same beginning point. I do not think that this will change your results that much, but the integration will at least begin above the canopy. This will however change part of discussion about the particle losses problem in the first § of 6.2.
The in-situ measurements were performed below the canopy at ground and not on top of the tower were the Sun photometer was located. We have added another sentence for clarification on the inlet heights (see comment below). Concerning the integration, the lowest level for averaging the CPC measurements of the aircraft was 200-400 m a.s.l. (and not above ground!), if taken into account that Hyytiälä is approx. 180 m a.s.l. and the canopy/tower about 20 m a.g.l., we do not miss a layer between Sun photometer and the layers to calculate the in-situ AOD. We have added "a.s.l." to the corresponding statements. In addition we added a sentence to Sect. 5.4:

"To calculate the in-situ AOD the atmosphere above was separated into 200 m wide levels in which the CPC measurements were averaged to determine c(h) for each layer starting at 200 m a.s.l. (close to the top of the canopy and location of the Sun photometer)."

Secondly, you never estimate the part of the AOD difference that is due to the different end points h1 of the integral (Eq. 5). Referring to fig. 8b and 15b, it seems obvious that the aerosol load over 3000 m is not always negligible. Would it be possible to fully integrate the calculated AOD on the whole atmospheric column by fitting a decrease of the extinction coefficient as a function of altitude? This would be a valuable approach at least for the first time period (21-28 May).
In the original work, we discussed the issue of aerosols above the maximum flight altitude by using the lidar measurements from Kuopio (Sect. 6.3 and Fig. 15). Here, we showed the percentage of the integrated AOD coming from layers above 3 km. It was shown and discussed that approximately 15-25 % of the AOD came from the elevated layers before the Saharan dust event (SDE) and up to almost

80 % during the SDE. This was done for the 1064 nm channel of the lidar which is most sensitive to coarse mode particles (the other lidar channels did not work appropriately during the entire campaign). As the reviewer suggested and as an additional sensitivity study, we have added to all profiles an assumed exponential decrease and integrated up to 7000 m (see Figure 7 below).

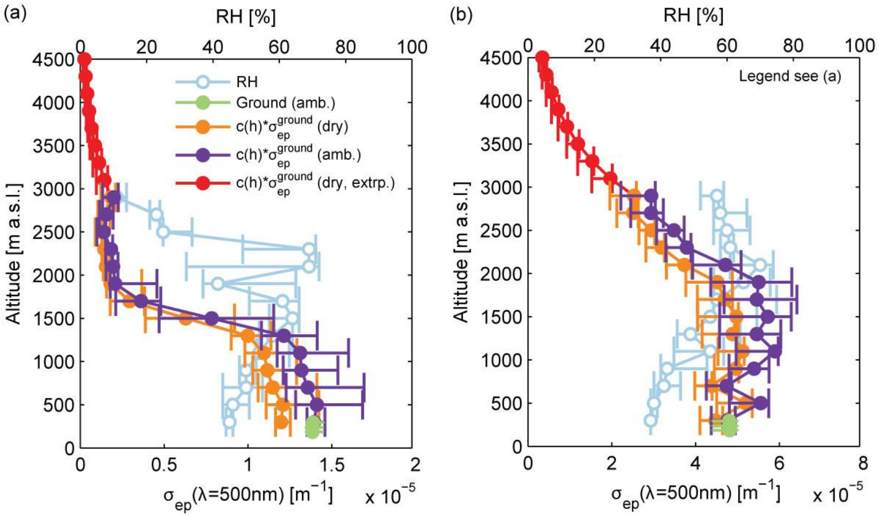

Figure 7: Modified Fig. 8 with the assumed exponential decreasing profile (red bullets). The profile shape has slightly changed since we now used 1-sec CPC data (before 2 min values) and show now everything related to sea level (a.s.l.), see comment below.

The AOD with the extended exponential decreasing profile has been only calculated for the dry case since no RH measurements are available above the maximum flight altitude of the aircraft. The overall effect is also small since the majority of particles are within the profiling range of the aircraft (< 3 km) for the period without influence of long-range transported mineral dust. This effect and contribution to the AOD is discussed using the lidar profiles later on. We have added this AOD values also to Fig. 8 in the revised manuscript as a time series (see Figure 8 below). To describe and discuss the result on the exponential assumed decreasing profiles we have added to Sect. 5.4 (after the description of the different approaches):

"To test the influence of the layer above the maximum flight altitude an exponential decrease of the total number concentration was assumed (with $c(h) = c(h_i)\exp(-0.25h)$ above the maximum flight altitude, where $c(h_i)$ is the scaling factor of the last height bin and h the altitude up to 7 km). This is a reasonable assumption only for cases without clear elevated layers, which was most likely only given for the first half of the airborne observation period (see Sect. 6.3). The in-situ AOD is only calculated for the dry case since no RH measurements are available above the maximum flight altitude."

Further down in the discussion we added:

"Figure 8 also reveals that the addition of an assumed exponential decreasing profile above approx. 3 km only marginally leads to an increase of the in-situ derived dry AOD. This points towards the fact that most of the particles were captured by the aircraft profiling, if the assumption of an exponential decrease in particle number concentration is valid. However, this assumption is most likely not valid for the second half of the aircraft profiling period. As can be seen in Fig. 8b, the AOD increases in the beginning of June due to long-rang transport of mineral dust in elevated layers (see Sect. 6.3)."

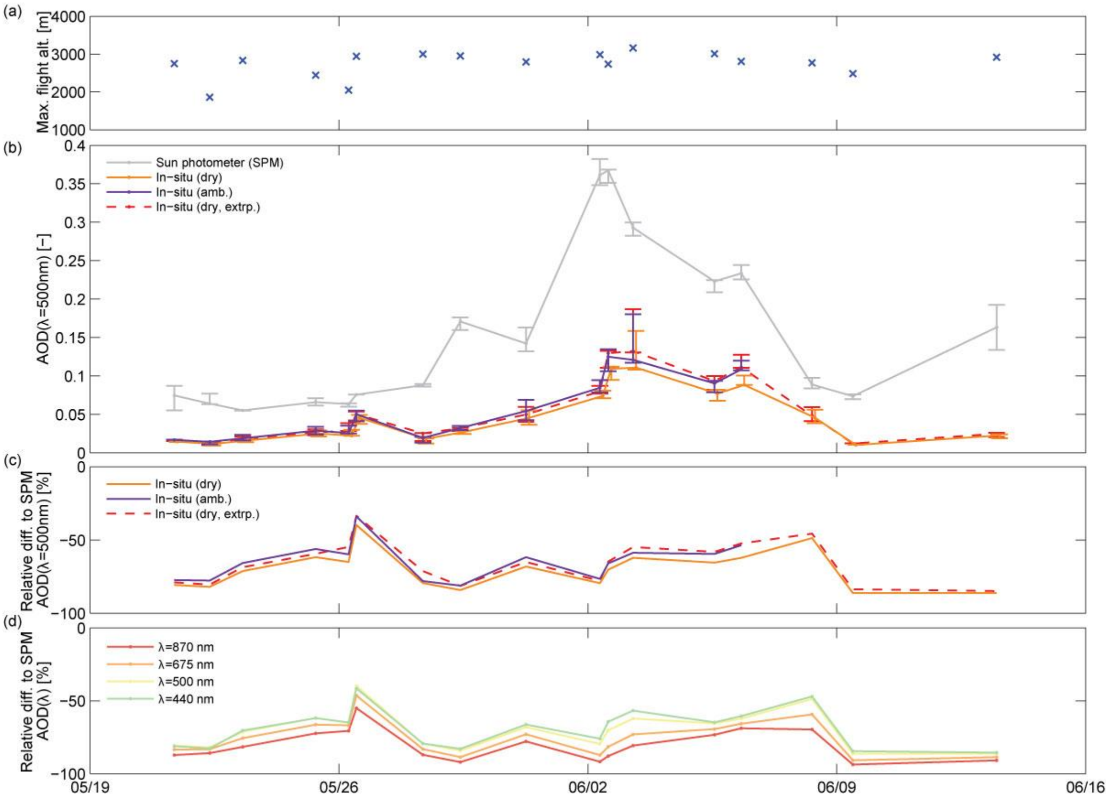

Figure 8: (Modified Fig. 9 from the original manuscript) We have added the line for the AOD (dry case) if an exponential decrease of the aerosol concentration is assumed above the maximum flight altitude (see red dashed curve in panel b). In addition, we added the relative differences for the three in-situ cases (panel c) and for the dry case but for four different SPM wavelengths in panel d (see comments below).

Results: Figure 10: it seems to me that the data taken to calculate Fig 10 are not mentioned. Since the measurements from the 30 May to the 3 June are clearly influenced by SDE at high altitude, I hope that this figure is produced with only the first time period (21-28 May). If it is not the case, the discussion concerning the coarse particle losses has probably to be changed.

I suppose that you use the extinction Angström coefficient to interpolate to all the CIMEL wavelengths. Due to the restricted wavelength range of the Nephelometer, what is the uncertainty in the interpolation up to 1600 nm ? Could this have an effect on the discussion 6.2 ? It seems from Fig. 11 that the measurements at the greatest wavelength are more dispersed than at 450 nm. This is also probably the case for the AE-31. Are therefore the uncertainties of the nephelometer and the Aethalometer similar at all the wavelengths ? If not, what is the effect of these uncertainties on Fig. 10 and on the discussion about coarse particle losses ?

In the original manuscript, Fig. 10 was calculated by using all points of the calculated in-situ and measured AOD’s (incl. the periods with probably elevated layers; see Fig. 9 in original manuscript). The Ångström exponent of the extinction coefficient was used to extrapolate to the Sun photometer wavelengths. After a re-analysis of the data, we have reconsidered the way we present and discuss the findings and have removed Fig. 10 and added two extra panels to Fig. 9 showing now the temporal development of the relative differences between in-situ and Sun photometer AOD (see Figure 8 above). The reason is that the high values for R^2 were, as pointed out, largely determined by the elevated AOD values during the long-range transport event and thus it is questionable to compare all days together when determining R^2 or the linear regression coefficients. However, the overall finding that the in-situ AOD is highly underestimated remains. We agree that the extrapolation to 1600 nm is too uncertain, so we limit the discussion to wavelengths close to the nephelometer wavelengths (450 to 700 nm). But also here, it is clear that the agreement deteriorates at larger wavelengths. By now showing the temporal evolution of the relative differences, one can see that this is largely dominated by the period of potential elevated layers of mineral dust. We also tested the effect of using a minimum or maximum value for the Ångström exponents and the results are similar. The influence of the absorption Ångström exponent is a minor one due to the clear dominance of particle scattering. See also comments from reviewer #3. We removed the discussion related to Figure 10 and made the following changes:

We added to Sect. 5.4 (following Eq. 6):

"We have limited the extrapolation to SPM wavelengths that are close to the nephelometer wavelengths to reduce the involved uncertainties. The in-situ AOD's are therefore only calculated between 440 and 870 nm."

Discussion of Figure 8:

"The calculations were done for all SPM wavelengths between 440 and 870 nm which are close to the spectral region of the nephelometer. Figure 8d shows that the relative difference of the dry in-situ derived AOD to the SPM measured values increases for larger wavelengths. These differences are more pronounced for the period of potential long-range transported mineral dust."

We have changed the sentence in the abstract to:

"The in-situ derived aerosol optical depths (AOD) clearly correlate with directly measured values of the Sun photometer, but are substantially lower compared to the directly measured values (factor of ~2 lower in-situ values)."

And correspondingly in the conclusion:

"The in-situ derived AOD showed were correlated to the Sun photometer measurements, however, a clear underestimation of the AOD by at least a factor of 2 was found."

Discussion 6.1: without referring to other papers, it is not clear if the operational measurement (Neph cottage) are done at the same altitude than the container measurement or on the mast above the canopy. If the operational measurements are done above the canopy, it changes several points of your discussion.
The continuous in-situ measurements were performed below the canopy in the aerosol cottage within the forest, while the measurement containers for the campaign were located on a small parking lot next to the aerosol cottage. We have added a sentence in Sect. 2 to clarify this issue:

"The inlets of the measurement containers were ~ 4 m above ground, while the inlets of the aerosol cottage were ~ 5 m above ground and ~ 10-15 m below the top of the canopy."

Discussion 6.3: I would take the lidar data as measurement results influencing the analysis (see previous comments on the exclusion of these data for Fig. 10) and not really as a discussion point.
Unfortunately, the lidar measurements were not performed directly in Hyytiälä but in Kuopio, 200 km east-northeast of our site. In addition, only the 1064-nm-channel worked continuously without problems. Therefore we decided to keep the lidar measurements only as an add-on for the qualitative discussion on long-range transport of mineral dust. The percental difference of the AOD coming from above the maximum flight layer corresponds roughly to the relative difference found at Hyytiälä during the episode with long-range transported mineral dust. We have added a sentence that links these number to the new panels of Fig. 8 c and d:

“These percentages are in correspondence to the relative differences calculated for the in-situ derived AOD vs. the measured values by the SPM during the period of long-range transported mineral dust (see Fig. 8d). It should be mentioned that the comparison to the lidar profiles measured at Kuopio is only of qualitative nature to demonstrate the effect of lofted layers due to long-range transport.”

Is the size distribution at the maximum fly altitude similar to the one at the minimum altitude plotted on Fig. 12a ? If no, can the difference give you indication on fine/coarse particles losses as a function of altitude? If no and taking into account the CPC measurement, would it be possible/valuable to separate the scaling factor c into fine and coarse contributions?
The shape of the size distribution measured by the airborne SMPS does not significantly change with altitude, which can be seen in the Figure 9 below, where the median of the particle number size distribution shown (it is normalized to 1 since the concentration decreases with altitude). The SMPS is with its maximum diameter of 270 nm limited for our purposes and most of the scattering comes from particles above the maximum SMPS diameter bin (see Fig. 12b of the scattering size distribution in the original manuscript). Thus it is not possible in an easy and valuable way to constrain a scaling factor which differentiates between fine and coarse mode. It would cause additional uncertainty then already induced by the assumptions and instruments taken currently to calculate an in-situ AOD.

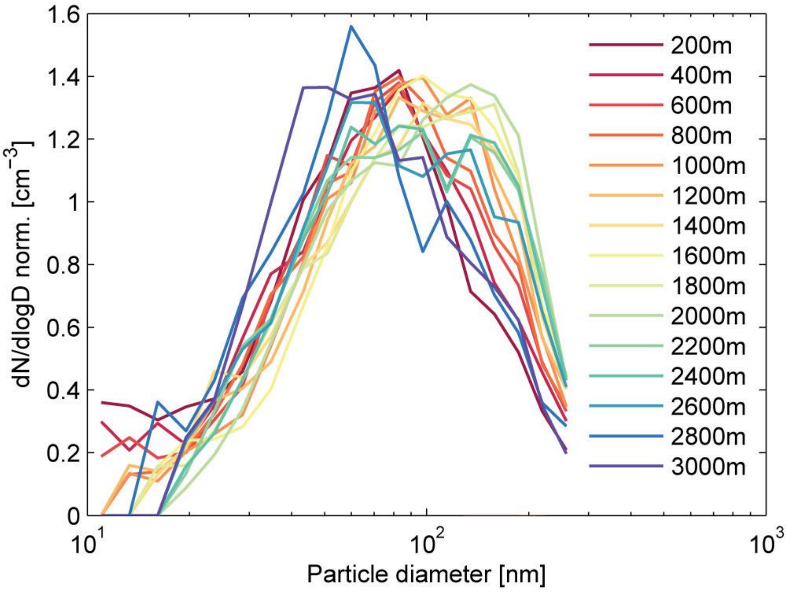

Figure 9: Normalized particle size distribution measured by the SMPS at different altitudes (median values for the profiling period).

Minor comments:
Abstract: p. 3329 line 27: The sentence is not clear, since it seems that the “direct measured values” are not the same than the “Sun photometer AOD”. When speaking of the Sun photometer AOD, I would emphasize the fact that it measures the whole aerosol column instead of using the adjective “direct”.
We have replaced the word “direct” by “columnar” and have changed the order of the second sentence:
“… and compared to columnar measurements of a co-located AERONET Sun photometer.”
and
“The in-situ derived aerosol optical depths (AOD) clearly correlate with directly measured values of the Sun photometer, but are substantially lower compared to the directly measured values (factor of ~ 2)”

Introduction p. 3332 lines 1-5: I find the questions not really pertinent. For the first question, the main point is more to compare the scattering enhancement at Hyytiälä with the ones measured at other sites than only to measure the value (what we know that you are able to measure regarding your previous publications). The second point is more about the quality and limits than about the feasibility of the columnar optical closure.

The columnar optical closure study can only be performed if the scattering enhancement is characterized for that site and aerosol type. Thus question 1 on the magnitude of *f*(RH) is a perquisite for answering question 2 on the columnar optical closure. We are aware that these two aspects cause the manuscript to be quite comprehensive. It could have been a possibility to split the work into two separate publication but we decided to keep the work together in a compact way.

Instrumental 3.7: p. 3339 line14: is the "level 2.0 data" self-explaining? A reference could perhaps be added to avoid a description.

We have added the following reference for the level 2.0 description to the manuscript:

Holben, B. N., Eck, T. F., Slutsker, I., Smirnov, A., Sinyuk, A., Schafer, J., Giles, D., and Dubovik, O.: Aeronet's Version 2.0 quality assurance criteria, doi:10.1117/12.706524, http://dx.doi.org/10.1117/12.706524, 2006.

Results: 5.1 p. 3342 first §: It seems curious that the maritime influence is not mentioned for Cabauw, but the answer is probably in your 2013 paper. However for Mace Head, it is not clear if the value f=2.08 is a mean for the station of if it corresponds only to time with anthropogenic emissions. Perhaps you could add f values for both maritime and anthropogenic influences. Finally it is written that Mace Head showed a "large variation in f(RH)". The variation (i.e. the f(RH) range with non zero PDF) is not larger than at Ny-Alsund or Cabauw, but is a convolution of three components.

This paragraph is kept on purpose compact and we like to refer to our 2013 *f*(RH)-overview paper, where the other stations and aerosol types are discussed in much more detail. However, we agree that a few more clarifications are needed here. It is true that "large variation" is the wrong term here, since the distribution of *f*(RH) for Ny-Ålesund is much broader. Therefore we have replaced "large variation" by the more suitable term "distinct differences". Cabauw is one of the sites with a high variability of aerosol types. The clean marine aerosol (with distinct deliquescence) was only observed for a few cases when the air came directly from the Northern Sea. In the manuscript we only stated campaign average values for more details we refer once more to the Table 2 in the *f*(RH)-overview paper from 2013. We changed this sentence to:

"The f(RH)-values given above are campaign averages, however, each site had its characteristics for specific air mass types like marine aerosol, anthropogenic influenced one or desert dust. For example, Mace Head in Ireland showed distinct differences in f(RH) depending on the wind direction; if the air had a maritime origin generally higher values were observed (f(RH=85%,550nm)=2.28±0.19) in contrast to wind coming from the island or continent with influence of anthropogenic emissions (f(RH=85%,550nm)=1.80±0.26). A separation of different air mass types for the other sites are given in Tab. 2 in Zieger et al. (2013)."

P. 3343 last §: did you see any deliquescence with air from maritime origine ?

No distinct deliquescence was observed in contrast to other sites like Melpitz, Cabauw or Ny-Ålesund. This is already mentioned in Sect. 3.1, but we repeat it here once more because it is a relevant question the reader might ask at that point. The reason for the lack of deliquescence is probably the high contribution of organics (mass fractions above 0.5, see Fig. 6). We have added to this paragraph the sentence:

“However, no distinct deliquescence was observed in contrast to other sites like Melpitz (Germany), Cabauw (The Netherlands) or Ny-Ålesund (Spitzbergen), which can be explained by the high contribution of organic substances at Hyytiälä.”

P. 3344, line 9: the “slightly increase” is not obvious.
We agree and have changed this sentence to:

“The EBC values show no significant trend compared to f(RH) or the organic mass fraction.“

5.2: p. 3345 line 11: replace “α the Angtröm exponent” by “αsp the scattering Angström exponent”.
Done.

P. 3346 lines 4-8: I suppose that you assume that the absorption coefficient is not changing with RH also because it was already shown in previous publications that the humidity impact on the absorption coefficient is negligible or at least far lower than on the scattering coefficient.
Yes. Although the humidity effect on the absorption is not negligible, it has only a very small effect on the particle light extinction coefficient because on the one hand the RH effect on the particle light scattering coefficient exceeds the RH effect on the absorption by far and on the other hand, the scattering is by far the dominant contribution to the extinction at Hyytiälä (values for the single scattering albedo are on average at around 0.95, see new Tab. 2 in the revised manuscript). We have modified that sentence and added a citation (which explicitly deals with the absorption enhancement) to the revised manuscript:

“This is reasonable assumption at Hyytiälä due to the fact that the scattering enhancement exceeds the absorption enhancement (Nessler et al., 2005) and even more important, due to the dominance of the light scattering (i.e. campaign average for the single scattering albedo ω_0=0.94±0.03 at λ=525nm, see Tab. 2), which in total will only induce a small error.”

P. 3346 line 21: the value at the surface is also higher than on the 23 May and the ML is probably also higher. Or is the N concentration particularly high at 2700 m for that day ? It can be valuable to correlate this profil with Fig. 7a. It is however quite difficult to distinguish the profiles for that 2 days. Could you choose particular colors that evidence the 23 may and the 2 June profiles in Fig. 7a?
We have removed this figure for two main reasons: First of all, not much is actually gained from the profiles of total number concentration and two example profiles are then shown and discussed in the following figure. Secondly, the higher concentration measured by the aircraft in the lowest layer are later shown as average size particle size distributions (see Fig. 12 in the original manuscript) with the same message. In addition, it helps to reduce the number of figures in general, as reviewer #3 asked for.
For the reply, we have plotted in Figure 10 for the two example days the number concentration and integrated particle surface vs. altitude. I can be seen that for the first day (23rd of May, blue curve) the number concentration is almost double compared to the second example (2nd of June, green curve). The difference is not seen in the scattering properties (see example profiles in Fig. 8 of the original manuscript) because the particles are generally larger (increased scattering coefficient) on the second day with a decreased total number.

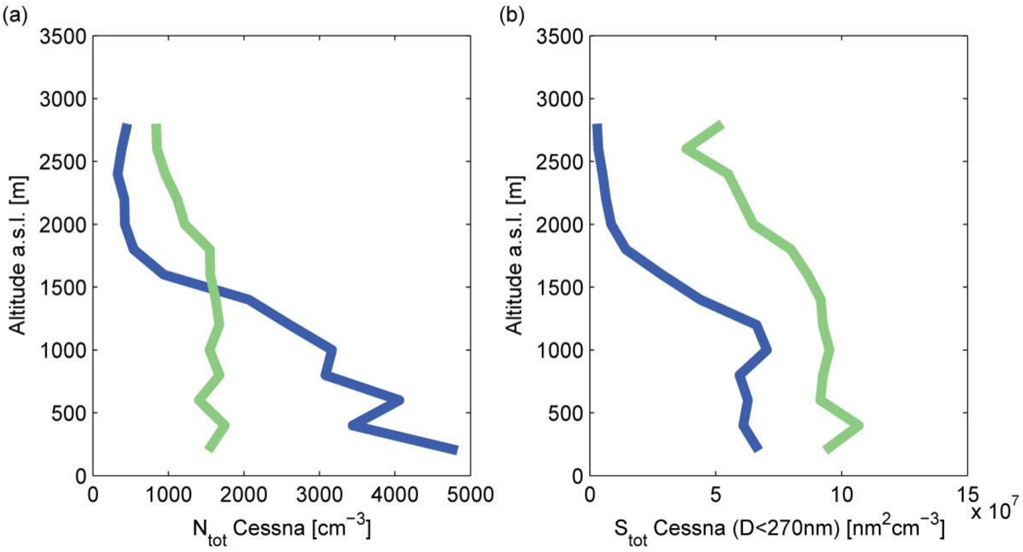

Figure 10: Median profiles of the number concentration and particle surface area measured by the SMPS (D<270nm) for the same days as discussed in Fig. 8 in the original manuscript (blue lines: 23 May; green lines: 2 June 2013).

P. 3347 line 15: really unexpected?

At the beginning we did not believe in this significant disagreement and thought for a long time to have some major bugs in the measurements or the analysis code. We agree that this is a judgmental statement and therefore we removed that word in the revised manuscript since it is not really needed here.

P. 3349 lines 7-9: Do you have an explanation for these larger variations of the WetNeph reference nephelometer due perhaps to the location on a parking place (traffic, wind, convection due to the ground material)?

The container site was on an open parking lot, next to the forest. However, there was no traffic during the campaign (only occasionally for instrument transport or maintenance), thus we believe that the influence of traffic is negligible. The container site could have been influenced by the loose gravel from the parking lot, but this should have been seen in both dry and wet nephelometers. The dry reference nephelometer showed a higher standard deviation (e.g. 3.3 Mm^{-1} at 535 nm at dry conditions) compared the nephelometer of the WetNeph system (e.g. 0.8 Mm^{-1} at 535 nm at dry conditions). The higher standard deviation is probably due to the fact that the reference nephelometer was a newer version of the nephelometer with a higher measuring rate than the nephelometer used for the WetNeph. The standard deviation of the data from the WetNeph is therefore biased low due to the repetition of the same values when polling the data from the nephelometer at a higher rate than its measurement rate.

P. 3351 line 6: does linear regressions have a spectral variability? What does mean the “optically decrease of a correlation coefficient”?

We originally meant that the coefficients of the linear regression for the different Sun photometer wavelengths changed (showing a degrading agreement with increasing wavelengths. We have replaced this sentence by

“The relative disagreement between in-situ derived and measured AOD values increased for larger wavelengths (see Fig. 8d) which points towards an influence of large particles which are not sufficiently sampled by the in-situ instruments.”

Table 1 and Figure 2 are redundant. I prefer the figure. Fig. 2c could have a smaller x scale

Although we agree that the both are slightly redundant, we would like to keep the Table 1 in the manuscript. The exact values help to directly compare the values and they are also needed to get an uncertainty of the gamma-fit.

Reply to reviewer #3

General comments:

This paper presents a detail study of aerosol hygroscopicity in a remote site using state of the art instrumentation. The authors combined many instruments which provide a valuable insight in the aerosol properties. The paper is of interest for the scientific community and it is clear and well written. The paper is suitable for publication in ACP after major revisions.

Specific comments:

P3330-Lines 13-14: I wouldn't say that the aerosol hygroscopicity is "significantly lower" at the study site compared to other European sites. Of course, it depends on the sites that you are comparing with. For example, Carrico et al. (2000) reported a f(RH=82%) value of 1.46 for polluted air masses at Sagres (Portugal), Fierz-Schmidhauser et al. (2010) reported a value of 1.8 at Mace Head (Ireland) under polluted conditions and a value of f(RH=85%)=1.6 was reported by Titos et al. (2014) at Granada (Spain).

We agree that the wording is not optimal because lower values are also observed for example during dust episodes or highly polluted conditions at other sites. In fact, our lowest values also refer to mineral dust transported to the Jungfraujoch in the Swiss sites. However, if we look at the distribution of measured enhancement factors (Fig. 3) for entire measurement periods/campaigns, Hyytiälä strikes out a narrow distribution of low *f*(RH) values. We have replaced "*significantly*" by "*generally*" in the abstract.

P3330-Line 14: This is in fact a general comment. Why the authors use the 450 nm wavelength? I encourage the authors to focus on the 525 nm wavelength. The comparison with other sites will be more straightforward this way.

We agree and have replaced that statement with the values for the 525nm-wavelength. *"… (e.g. 1.63±0.22 at RH=85% and λ = 525nm) …"*. We have also replaced all figures and discussions from 450 nm to the 525 nm values of *f*(RH) or γ (see comments below).

P3333-Line14: State the duration of the humidification cycles (time scanning RH up and down).

We have added:

"One full humidograph cycle (hydration and dehydration) took three hours."

P3333-Line26 to 3334-Line10: It is not clear if this comparison is performed in this study or it was previously done by Fierz-Schmidhauser et al., (2010c). Concerning the comparison with the commercial humidified nephelometer by ECOTECH, it would be interesting to see how good the comparison was (slope and regression). This is a novel instrument and it will be great to see its performance compared with a well-tested humidifier (PSI humidifier).

Maybe this sentence was a bit misleading. We have actually performed those inorganic salt measurements directly in Hyytiälä and compared it to Mie calculations as described by Fierz-Schmidhauser et al., 2010c. Only the TSI nephelometer was replaced by an Ecotech one. For clarification we have modified the sentence to:

"In addition, measured humidograms of polydisperse ammonium sulphate particles measured at the site were compared to model predictions using the size distributions measured by a DMPS system (with a diameter range of 6 to 600nm, see below), theoretical growth factors of ammonium sulphate and Mie theory (Fierz-Schmidhauser et al., 2010c)."

Concerning the direct comparison of the WetNeph and the new commercially available ACS: Laborde et al. is currently preparing a technical paper on the instrument, which will show the direct comparison to the PSI WetNeph. We show the comparison already as a pre-look in this reply letter. In addition, we have added a sentence on the comparison in the revised manuscript (see Figure 2 and the comments for reviewer #1; page 3).

P3335-Line15: The correction factor for the aethalometer, was it determined for this campaign? This value might change depending on the predominant aerosol types…

The correction factor C mentioned is the multiple scattering artefact associated with filter-based absorption. The multiple scattering artefact was determined using a Multi Angle Absorption Photometer (MAAP) as a reference instrument. Although the MAAP is not an ideal reference instrument it is, however, well documented that the instrument performs better than the aethalometer in question and is more robust to multiple scattering than the Aethalometer. The referee is correct in pointing out that different aerosol types can influence the C value. The multiple scattering artefact is likely to depend on the size distribution of the aerosol and the backscatter fraction of the deposited aerosol and could therefore change with season which could be a study in itself. Since the main focus of the manuscript is not on this particular method, the site specific multiple scattering correction factor was used as such, in accordance with previous studies that have used the correction.

P3336-Line13: Can the heated inlet affect the measured size distributions?

The heated inlet and the temperature within the instruments can affect the aerosol measurements, which is a common issue for in-situ measurements. This is especially important for nitrate components which will be in the gas phase during daytime with warm temperatures. However, we believe that the partitioning of nitrate is a minor issue due to the low concentrations at Hyytiälä (see Tab. 2). Also semi-volatile organics could have evaporated (1-2% for each K of temperature difference according to Donahue et al., 2006). Although the temperature differences between inside the hut and outside (daytime) were not as pronounced as during winter months for example, we can't fully rule the effect of gas-phase partitioning out. However, this effect would have influenced all in-situ measurements in the same manner (note that the aerosol in-situ measurements themselves were shown to be consistent). Therefore, we added the following sentence in Sect 6.1 (Consistency of in-situ measurements: optical closure study):

"Partitioning effects of semi-volatile organics (Donahue et al., 2006) or nitrate components (due to the low concentration to a lesser extent, see Tab. 2) that could have caused a potential decrease in the overall particle properties cannot be ruled out completely. Although it is believed to have a minor effect during the summer months and daytime in-situ measurements at this site."

P3336-Line15: How are the DMPS and APS size distributions merged?

The size distribution were merged at 750 nm, which is the third last DMPS diameter bin. We have added this information to the revised manuscript:

"As input, the particle number size distribution measured by the DMPS and APS was used (the APS and DMPS size distributions were merged at the last DMPS size bin)."

Section 3.4: No specification about the inlet at which the ACSM is connected is given here. State if it is the same inlet than for other instruments, if it is PM10 or PM1, etc.
We have added a description of the ACSM inlet to the revised manuscript:

"The ACSM inlet line had a PM2.5 cyclone filter to stop dust and pollen contamination. The inlet line is dried using a nafion dryer, reducing sample RH below 30%. At the entry to the instrument itself the sample aerosol is concentrated into a beam by a standard AMS aerodynamic lens with a cut size of approximately 600 nm."

Section 3.5: Why the ecotech reference nephelometer is not used to retrieve the complex refractive index? Introducing the TSI neph in this study, which is measuring in a different cabin and with a different inlet system seems unnecessary from my point of view and it adds confusion to the manuscript. I suggest the author to focus on the neph tandem measurements.
The reason on why we used the TSI nephelometer is the following: The TSI nephelometer is at the same location as the size distribution and aethalometer measurements (aerosol cottage), which are needed to retrieve the complex refractive index. It is also an important discussion point since we find differences in the scattering coefficient measurements between the monitoring instrument (aerosol cottage) and the nephelometer located in the campaign containers which were probably due to losses in the canopy or in the common inlet of the aerosol cottage.
Secondly, the TSI nephelometer is the instrument which is used within the monitoring program and thus a long time series exists for further (long-term) studies and is needed to relate our findings to this instrument.

P3341-Line9: This statement is confusing, it is not clear if a humidogram cycle takes three hours or not. As mentioned before, state the duration of the RH cycles.
We agree and have added to the instrumental section:

"One full humidograph cycle (hydration and dehydration) took three hours."

P3341-Line14: To contextualize the aerosol properties at the measurement site, I recommend the authors to include a table with, at least, mean, std, min and max values of the dry scattering coefficient, absorption coefficient, single scattering albedo and scattering Ångström exponent.
We agree and have added a new table to the manuscript which state the mean, standard deviation and percentile values of all major optical properties and the chemical composition data (see Tab. 2 in the revised manuscript). All other optical properties were calculated to the nephelometer wavelengths as the main instrument used here. The values were calculated for the time period when the WetNeph was in operation (see Fig. 1). In addition we have added to Sect. 5.1 the following sentence:

"To bring our measurement results into a broader context, Tab. 2 shows the average values for the main aerosol optical parameters (all calculated to the nephelometer wavelengths) and the chemical composition measurements."

P3342-Line13: Why the f(RH) is given now at 450 nm? In the previous paragraph it is given for the 525 nm!!! Figure 1 also refers to the 525 nm wavelength. Are there any reasons why the authors use the 450 nm? This is very confusing and needs to be corrected. Additionally, using the 525 nm makes comparison with other studies more straightforward.
The reason why we originally showed the 450nm wavelength here is that this is the only common wavelength between the Ecotech nephelometer (used here) and TSI one (used in Melpitz) to which we compared our data. However, we agree that it is more consistent to stick to one wavelength

which is close to 550 nm. Therefore we have replaced all figures where the results for the 450 nm channel were shown by the results for the 525 nm channel.
In addition, we have improved the structure by summarizing the comparison to the chemical composition measurements in one subsection (Sect 5.2 *“Comparison to the chemical composition measurements*”) and the discussion on the parameterization into a second separate subsection (Sect. 5.3 *“A simplified parameterization of f(RH)*”). We have replaced Fig. 4 with Figure 11 (below) showing instead of *f*(RH) already the fit-parameter γ at 525 nm, which is the one related to the organic mass fraction in the next step. As reviewer #2 suggested, we now give the fit-coefficients for both data sets (Hyytiälä and Melpitz). We also replaced the (Matlab) robust fit with a weighted bivariate fit described by York et al., 2004, which allows to estimate an uncertainty for slope and intercept. See also next comment.

Revised and new Sect 5.2:

*“**5.2 Comparison to the chemical composition measurements***
The reason for the low f(RH) at Hyytiälä can be explained by the dominance of organic substances in the particle’s chemical composition, which leads to lower particle hygroscopicity. As an example, the fit-parameter γ (Eq. 2) is plotted in Fig. 4 as a function of the organic mass fraction F_{org} *for* $\lambda = 525$ *nm. The linear regression shows a clear anti-correlation (squared Pearson’s correlation coefficient:* $R^2 = 0.77$*) with a decrease in γ with increasing* F_{org} *(i.e.,* $\gamma(525nm) = (-0.71\pm0.15)\cdot F_{org} + (0.76\pm0.11)$ *retrieved from a weighted bivariate fit according to York et al., 2004, taking the SD of the average values as an input for the uncertainty calculation). The dominance of the organic mass fraction (mean ± SD: 0.7±0.11) clearly determines the low values of γ and thus the low f(RH) observed at Hyytiälä. For comparison, the values measured at Melpitz, Germany, are added to Fig. 4 (for more details see Zieger et al., 2014). The organic mass fraction at Melpitz of sub-micrometer particles was significantly lower than at Hyytiälä (mean ± SD: 0.23±0.10). Although the γ-values for Melpitz were measured at a different time of year (winter) and showed a higher variability (*$R^2 = 0.50$*), they almost line up linearly with the observations made at Hyytiälä. Due to measurement restrictions the total mass at Melpitz was only differentiated between black and organic carbon, while the total mass at Hyytiälä is determined from the elemental carbon of the EC/OC analysis (organic carbon is assumed to be included in the ACSM organic mass fraction). The ammonia mass fraction at Hyytiälä and Melpitz are also linearly correlated, while the sulphate mass fraction did not show a joint linear behaviour with the Melpitz data. The reason is that the aerosol found at Melpitz during the winter months also contained large amounts of nitrate which mainly formed ammonium nitrate (with a higher hygroscopicity than organic aerosol), while the nitrate contribution at Hyytiälä was very small and the sulphate mainly formed ammonium sulphate or ammonium bisulphate which lead together with the organic contribution to a generally lower hygroscopicity.”*

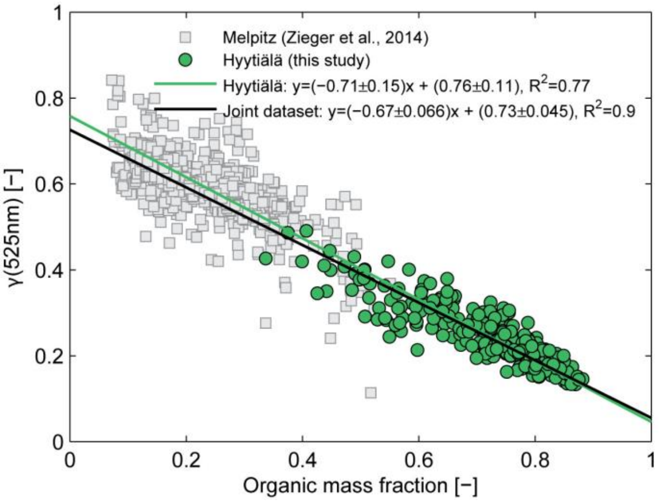

Figure 11: The fit parameter γ (for λ=525nm) vs. the organic mass fraction F_{org} measured at Hyytiälä (green bullets) and Melpitz, Germany (grey squares). The solid and dashed lines represent the corresponding bivariate weighted linear regressions. This is the revised plot for Fig. 4 in the manuscript.

P3342-line25: The authors should dig deeper into the literature regarding the relationship between organic fraction and f(RH). In particular, the paper by Quinn et al. (2005) should be mentioned here. Quinn, P. K., T. S. Bates, T. Baynard, A. D. Clarke, T. B. Onasch, W. Wang, M. J. Rood, E. Andrews, J. Allan, C. M. Carrico, D. Coffman, and D. Wornsnop. 2005. Impact of particulate organic matter on the relative humidity dependence of light scattering: A simplified parameterization. Geophys. Res. Lett., 32, L22809.

We agree. In the revised manuscript, we have compared our results to the parameterization by Quinn et al. (2005) and Zhang et al. (2015). In Quinn et al. (2005) the organic mass fraction is only calculated by using the organic and sulphate mass fractions. The gamma used in Quinn et al. (2005) is equivalent to the one used here, except that it is calculated using no pre-factor (a=1) and by considering the reference RH. However, their parameterization is based on PM_1 measurements and no specific wavelength dependency is given (the parameterization itself is given for 550 nm, P. Quinn, pers. comm., May 2015).

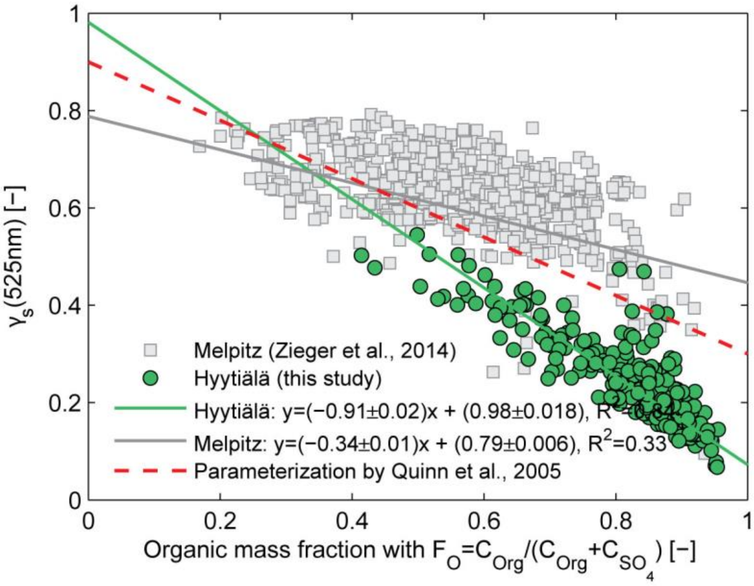

Figure 12: The fit parameter γ_s (calculated in the same manner as in Quinn et al, 2005) vs. the organic mass fraction only taking sulphate and organics into account. Both sites show a similar decreasing trend, however not the same linear behaviour as in Fig. 4.

Figure 12 shows the γ-parameter vs. the organic mass fraction calculated in the same manner as described by Quinn et al. (2005). Our results show a similar decreasing trend as the parameterization by Quinn et al. (2005) predicts (see red dashed line), however there are distinct differences. The Melpitz and Hyytiälä measurements do not show a common linear behaviour (in opposite to our analysis, see Fig. 4), which is due to the fact that the organic mass fraction is only calculated by using the organic and sulphate contribution. At Melpitz, however, nitrate and ammonia had a significant contribution to the total aerosol mass. Therefore, our parametrization using all the major chemical components is the favourable one. It should also be noted that the gamma calculated by Quinn et al. (2005) were determined by keeping a=1 and adding the dry reference RH into Eq. 2 ($\gamma_s=\ln(f(\mathrm{RH})) / \ln((100-\mathrm{RH}_{ref})/(100-\mathrm{RH}))$). However, the curvature is much better captured if this parameter is kept variable when fitting the measured humidograms, although a varied around 1 and showed no large variation (see Tab. 1). The intercept a can be used as an uncertainty estimate when calculating the f(RH) out of aerosol chemical composition measurements.

In a more recent study, Zhang et al. (2015) compared their gamma-values measured at Yangtze River Delta in China to AMS measurements. Similar to Quinn et al. (2005) they estimate the organic mass fraction by dividing the organic mass fraction by the sum of organic, sulphate and also nitrate. We have done the same and for the two data sets (see Figure 13). Adding the nitrate component to the organic mass fraction improves the comparison but there is still a clear deviation from linearity and our result. The reason is probably that the authors have not included ammonia and EC in their organic mass fraction as we have done.

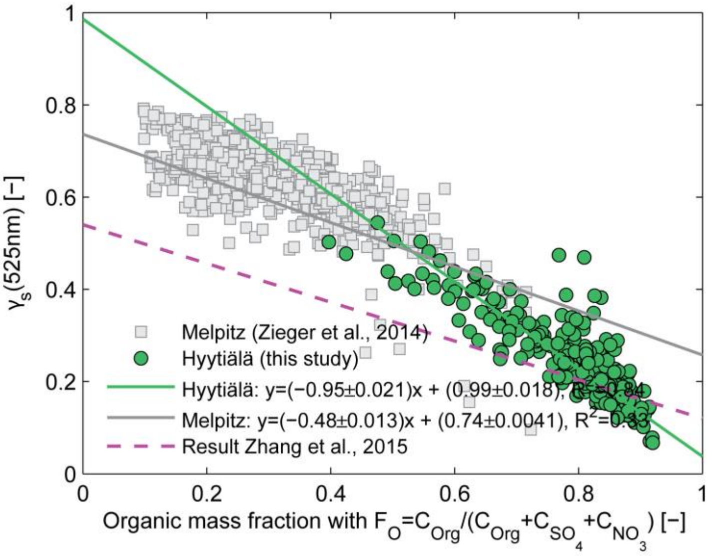

Figure 13: The fit parameter γ_s vs. the organic mass fraction only taking sulfate, nitrate and organics into account. The result by Zhang et al. (2015) is added to the graph (dashed magenta line). Both sites show a similar decreasing trend, however not the same linear behavior as in Fig. 4.

We have summarized the relation to the chemical composition data and the proposed parameterization (incl. the comparison to other studies) in a separate subsection.

“5.3 A simplified parametrization for f(RH)

A summary of the linear fit parameters of γ vs. the chemical mass fractions is shown in Table 3 for the components which showed a clear linear behaviour. The inorganic mass fractions, mainly sulphate and ammonia, are clearly positively correlated with γ and f(RH), in contrast to the organic mass fraction which is anti-correlated. This allows to use the continuous performed chemical composition measurements at Hyytiälä to predict f(RH) if no humidified nephelometer is operated. It can be done by taking the total organic or inorganic mass fraction as a proxy for f(RH) and using the linear regression parameters given in Table 3 to calculate γ for each wavelengths. f(RH) then follows by using Eq.

(2) assuming an intercept of a = 1. The variance of the intercept a can be used to estimate an uncertainty of the f(RH) prediction (see Table 1).

Parameterizations of f(RH) using the aerosol chemical composition are only sparsely published. Quinn et al. (2005) proposed a similar parametrization of γ using the mass fraction of organic matter and sulphate ($\gamma_s = -0.6 \cdot F_{org} + 0.9$ *with* $F_{org} = C_{org} / (C_{org} + C_{SO4})$ *and* $\gamma_s = \ln(f(RH))/\ln((1-RH_{ref})/(1-RH))$ *which is similar to Eq. 2 if a = 1;* RH_{ref} *denotes the dry reference H).This parametrization is limited to aerosol dominated by the accumulation mode and is only given for λ = 550 nm (P. Quinn, personal communication, May 2015). Our results (if calculated in the same manner as described in Quinn et al., 2005) show the same decreasing trend of γ s (for example for Hyytiälä:* $\gamma_s = -0.79 \cdot F_{org} + 0.96$ *and Melpitz:* $\gamma_s = -0.35 \cdot F_{org} + 0.81$ *at λ = 525 nm). However, both datasets do not show the same joint linear trend anymore because the organic mass fraction of the parametrization by Quinn et al. (2005) is calculated using the organic and sulphate concentrations only. The aerosol at Melpitz, however, had a significant contribution of nitrate, ammonia and black carbon which needs to be included in the parametrization to retrieve a reliable estimate on f(RH). In a more recent study, Zhang et al. (2015) parametrized their measurements of f(RH) from the Yangtze River Delta region in China in a similar way as Quinn et al. (2005) but adding also nitrate to the organic mass fraction. A linear relationship of* $\gamma_s = -0.42 \cdot F_{org} + 0.54$ *with* $F_{org} = C_{org} / (C_{org} + C_{SO4} + C_{NO3})$ *was found, which compares better to our results, however the ammonia and black carbon components are still missing in the linear relationship presented by Zhang et al. (2015).*

Table 3 also states the linear regression parameters for the joint Hyytiälä and Melpitz datasets. As mentioned above, the organic and the total inorganic mass fractions showed a common linear behaviour and thus a more general rule to predict f(RH) from aerosol chemical composition measurements can be derived. Individual inorganic components like sulphate or nitrate may show different functional dependencies individually for each site, however as the comparison to Quinn et al. (2005) and Zhang et al. (2015) showed, it is important to include all major chemical constituents when deriving a general parametrization of γ or f(RH) as has been done here. However, our parametrization for Hyytiälä is strictly spoken only valid for the summer months when the fine mode is clearly dominated by less hygroscopic organic substances. Verification during other seasons and adding other sites is needed to allow a generalization of these findings. The addition of the Melpitz findings from Zieger et al. (2014) should only be seen as a first step. Additionally, the parametrization may not be valid during periods with substantially different coarse mode contribution which can have a potentially large impact on the total f(RH) (Zieger et al., 2013).”

P3343-Line24: Do organics from marine origin affect f(RH) under maritime air masses?

This question is difficult to answer. The organic contribution within the particulate matter of marine aerosol will lower the overall *f*(RH) compared to pure inorganic sea spray for example. However, this effect is hard to differentiate with this dataset due to the general contribution of other biogenic and anthropogenic components like organic, sulphates or nitrate components at Hyytiälä. The organic contribution is less for marine air masses (see Fig. 6c) but still significant. Interestingly, if one calculates the *f*(RH) with no organic contribution, one would derive a values of approximately *f*(85%,525nm)=3, which is still higher than the one for pure inorganic sea spray as measured under controlled conditions in the laboratory (approx. 2 at 85%; manuscript in preparation). A dedicated field campaign with a sufficient long dataset of unperturbed marine aerosol using a WetNeph and an AMS could help to answer this question.

P3343-Line24-Figure5: This problem can be avoided if only concurrent measurements of chemical constituents and f(RH) are used. Doing so, it would be easier to see any relationship between these variables and their respective spatial patterns.

We agree and have re-done the analysis only for concurrent times and now for the 525 nm wavelengths (see comment and new figures below; page 28-29).

P3344-Line4: The manuscript has too many graphs (15!) and some of them are not necessary. This is the case of Figure 6, the information can be given in the text.
We have removed Fig. 7 and Fig. 10 (see comments above). However, our work presents a comprehensive study, which covers different aspects (incl. the scattering enhancement) needed to perform a columnar closure study. The large comprehension is due to the many different aerosol measurements and is also expressed in the large number of scientific contributors (# of co-authors). We have carefully re-evaluated each figure and believe that each remaining plot has its specific importance. Concerning Fig. 6: We do not agree. Figure 6 directly shows the influence of maritime vs. continental influence on the main parameters while the standard trajectory analysis which shows the spatial characteristics in a qualitative way (see Figure 5) is influenced by shadowing effects (i.e. if two trajectories from different origins with different aerosol parameters pass over the same grid box close to the receptor and are being averaged). In Figure 6, we have introduced a parameter which is calculated along each trajectory incorporating the residence time within the mixing layer over ocean or land to directly describe the maritime or continental influence.

P3344-Line25: How this comparison of the lowermost part of the profile with ground measurements is performed? The dots in figure 7b, are average values? How this comparison is done should be explained in more detail. Ntot Ground can be included in Figure 7a for better visualization of the agreement between the lowest part of the profile and the ground measurements.
We have removed this figure for two reasons: First of all, not much is actually gained from the profiles and two example profiles are shown and discussed in the following figure. Secondly, the higher concentration are then later shown as average size particle size distributions (see Fig. 12 in the original manuscript) with the same message. In addition it helps to reduce the number of figures in general, as the reviewer suggested. Concerning the previous figure in the original manuscript: As described in the figure caption, these values are median values of N_{tot} measured at the lowest layer by the aircraft and for the same time period at ground. The error bars denote the range to the 25th and 75th percentile values.

P3345: Many assumptions are done by the authors to calculate the AOD. I suggest the authors to soft this discussion and emphasize other parts of the manuscript since the conclusions driven from this AOD-comparison are subject to many errors and depend on the assumptions made.
We have soften this part by removing Fig. 10 and by now just focusing on the relative differences which we show as a timelines only for the wavelengths close to the TSI nephelometer ones (see Figure 8 above). In addition, we revised that section according to the specific comments given below and by reviewer # 1 & # 2.

- The use of the scaling factor c, has been previously used in the literature? The size of the particles (which is not included in the scaling factor) as well as the chemical composition of these particles will influence the magnitude of the AOD. This should be discussed in the manuscript as an additional source of error.

The factor c is just a simple scaling factor and there is no need to search for literature equivalents. We have tested the difference between using the total number concentration (measured by the CPC, which could be biased too high due to freshly nucleated particles) and the particle surface area (which could be biased too low since the maximum diameter bin was 270 nm). As shown above (see Figure 5 and Figure 6 in this reply letter) the overall results due not change. The change in chemical composition with altitude had a smaller impact on the overall AOD compared to the particle concentration and size which is the dominant driver for the AOD, since the RH was moderate during the airborne profiling, thus effecting less the scattering enhancement.

- Why the SMPS size distributions onboard the aircraft are not used? Changes in the size distribution with height would affect the AOD estimation.

At the beginning, we have tested to use the integrated surface size distribution as a scaling factor, which decreases the influence of increased concentrations of nucleation mode particles (which are less important for scattering). However, the SMPS measured only up to 270 nm and thus misses particles important for scattering (see scattering size distribution in Fig. 12b in the original manuscript). We have tested both and also compared both scaling factors (number and surface based) and the overall results do not change significantly. At the end, we decided to use the total CPC, as the parameter which captures all particles including those above 270 nm. We added this information to the revised manuscript (see comments from reviewer # 1 and Figure 5 & 6 above).

- Assuming that the f(RH) is not dependent on the size distribution and chemical composition is in fact not consistent with the authors results and with the literature.

This is an unavoidable assumption which we had to make, since the *f*(RH) was only measured on ground and not on board the Cessna. Of course, *f*(RH) depends on the chemical composition and size distribution, which is sufficiently discussed within the manuscript. The magnitude of *f*(RH) is also generally lower compared to other sites. In addition, the dry summer months and thus low columnar RH values decreased the overall influence of *f*(RH) at Hyytiälä. Thus we believe that our assumption is justified for the here presented columnar optical closure study. We have added this point in the discussion (see comment below and new Subsection 6.1).

- A reference for the non-dependence of the aerosol absorption coefficient with RH is needed.

We have added the reference by Nessler et al. (2005):

"Note that the absorption coefficient is assumed not to change with RH. This is a reasonable assumption at Hyytiälä due to the fact that the scattering enhancement exceeds the absorption enhancement (Nessler et al., 2005), and, even more important, due to the dominance of the light scattering (i.e. campaign average for the single scattering albedo $\omega_0 = 0.94\pm0.03$ at $\lambda = 525$ nm, see Tab. 2), which in total will only induce a small error."

- Include in the Figures the AOD (AERONET and calculated) uncertainties.

We used as errorbars the difference between 25th and 75th percentiles to show the variability of the mean values, which were larger than the uncertainty coming from the AOD measurements of the Sun photometer. However, we added the reference in the instrumental part:

"The absolute uncertainty of the AOD for this instrument type was estimated by Eck et al. (1999) to be ~ 0.01 for the visible and near-infrared and ~ 0.02 for the ultra-violet region."

- The extrapolation to larger wavelengths is a source of uncertainty. The study should be limited to the 450-700 nm range, or at least a discussion about the errors of extrapolating from 700 to 1600 nm is needed.

We agree. We have changed the approach and now show only the temporal evolution of the relative difference between in-situ derived and measured AOD. In the revised manuscript we now focus on the wavelengths close to the nephelometer ones and have soften our discussion. In addition we have removed Fig. 10. See also comments of reviewer # 2 and the revised text above.

I’m very surprised that all the assumptions made for the AOD calculation are not considered as hypothesis of the disagreement. This should be included and carefully addressed.

We agree. We think that the influence of the assumptions being made are small compared to the four given hypotheses. The potential impact of the assumptions being made were in the original manuscript addressed in the section before (at the point where they were introduced), however, we have moved them to a separate section discussing the additional bullet point which we added to the hypotheses (“1. Assumption made to calculate $AOD_{in\text{-}situ}$”).

We have added:

*“**6.1 Influence of general assumptions being made***

The main assumptions that were made in Sect. 5.1 can all have a potential influence on the disagreement between in-situ derived and measured AOD values. The first main assumption is to use the total particle number concentration as scaling factor c(h) in Eq. 7 and Eq. 8. It should be noted here that the results are in a similar range if the particle surface is being used to calculate c(h), however this factor would miss optically active particles above the upper size limit of the airborne SMPS (see Fig. 8b below) and therefore we prefer to take the total concentration to determine c(h).

To calculate the ambient extinction, it was assumed in Eq. 8 that the particle light absorption enhancement is negligible. Again, this is justified for this site due to the low absorption enhancement effect compared to the scattering effect and the overall dominance of particle light scattering when determining the particle light extinction coefficient (Nessler et al., 2005).

For the ambient case, it was additionally assumed that the f(RH) is the same within the column as measured at ground and therefore only depends on the RH at different altitudes. This assumption implies that the chemical composition (hygroscopicity) and mean size is constant throughout the atmospheric column. This assumption is most likely fulfilled for a well-mixed boundary layer, however it will not be valid for lofted or separate layers during episodes with long-range transported air masses. During the summer months at Hyytiälä, however, the columnar RH was always moderate and low, in addition to the fact that particles are generally less hygroscopic at this boreal site and therefore, the overall effect of the constant f(RH)-assumption was probably small compared to the hypotheses discussed below.”

Section 6.1: This section is confusing. If you use the TSI nephelometer you are not looking for inconsistencies in your data since you used the ecotech neph as reference… The two nephs, wet and dry, were calibrated before the measurement campaign and they showed good agreement which each other (differences below 12% as stated by the authors). Therefore, this section is unnecessary. The direct comparison between both nephs is more reliable from my point of view than the comparison with the retrieved scattering coefficient using the size distributions measured in a different container (different location, inlet, and so on…).

Optical closure studies are useful and required tasks to identify measurement errors, especially if one keeps the large number of different instrumentation in mind. First of all, it gives confidence in the correct functioning of the humidified nephelometer, which fits to the predications of size and chemical composition measurements. Secondly, the optical closure study helped to identify that there is a real difference in the aerosol measurements between container and aerosol cottage (located within the forest). This could have not been identified by just comparing the dry scattering coefficient measurements, since the nephelometers from the containers (three Ecotech instruments) and the monitoring TSI nephelometer in the aerosol cottage were not compared directly during the campaign. Thus a difference could have also been attributed to calibration issues of the TSI nephelometer, which however agreed with the optical predications using the measured size distribution. The observed difference (which increase with increasing wavelength!) between the monitoring (forest) and the nephelometers in the containers (parking lot) are an additional hint that favourable large particles might have gotten lost by dry deposition within the canopy, which is also seen in the aircraft measurements and the wavelength-dependency of the AOD comparison. Therefore we argue that this is an important section which has to stay in the revised manuscript.

- Do you compare the reference neph (Ecotech) and TSI neph before or during the campaign but sampling from the same inlet?

The three Ecotech nephelometers (located in the campaign containers) have been compared among each other (and calibrated according standard procedures). Unfortunately, the dry Ecotech reference nephelometer has not been compared directly at the same inlet as the TSI nephelometer in the aerosol cottage. However, we are certain that all nephelometers have been measuring in a correct manner. This was also proven for the TSI nephelometer by the optical closure study (see Sect. 6.2).

Section 6.2: To avoid this problem the integration of the extinction coefficient to calculate the AOD can be done starting at 18 m. Thus, both AOD (AERONET and calculated) retrievals start at the same height.

The Sun photometer was located at 18 m above ground which corresponds to approx. 200 m above sea level, where the first level to calculate the in-situ AOD starts. It might have been confusing in the original manuscript (and it was also falsely plotted in Fig. 8, where the ground based measurements were shown at an altitude above ground, while the airborne measurements were plotted above sea level; we have corrected for the difference of ~180 m). Therefore we added “(a.s.l)” were needed and also added the altitudes in the first sentence of Sect. 6.2.:

“The Sun photometer was placed on a tower above the forest canopy (~198 m a.s.l.), while the in-situ measurements were performed on ground below the canopy (~180 m a.s.l.).”

P3350-Line27: According to the authors, particles below 100 nm in size are optically less important. However, these particles are included in the scaling factor c. It would be better to use the size distribution as scaling factor instead of the total number concentration.

See comments above (and also reply to reviewer # 1, page 6-7).

Section 6.3: How representative is the lidar data measured 200 km far from Hyytiala? In 200 km distance differences in the vertical distribution of aerosol particles is expected.

We use the lidar measurements from Kuopio only as a qualitative indicator of elevated layers due to long-range transport. We expect that these long-range transport phenomena’s at an altitude of 5000 m are similar at Hyytiälä compared to Kuopio. This assumption is also back-upped by the concurrent Sun photometer measurements in Kuopio and Hyytiälä which showed a similar temporal behaviour (not shown). Of course, local characteristics or sources will cause differences in the lidar profiles (especially in the lower parts of the troposphere), but this is not relevant for our purposes. We have added to the manuscript (Sect. 6.3):

“It should be mentioned that the comparison to the lidar profiles measured at Kuopio is only of qualitative nature to demonstrate the effect of lofted layers due to long-range transport.”

Figure 1: Split into two graphs scaling the axis appropriately.

We have re-plotted Fig. 2 split in two graphs (see Figure 14). However, there is no gain of further insights or knowledge. To keep the amount of figures and space to a minimum (as the reviewer also demanded) and thus the paper as compact as possible, we would prefer keeping Figure 1 as it is.

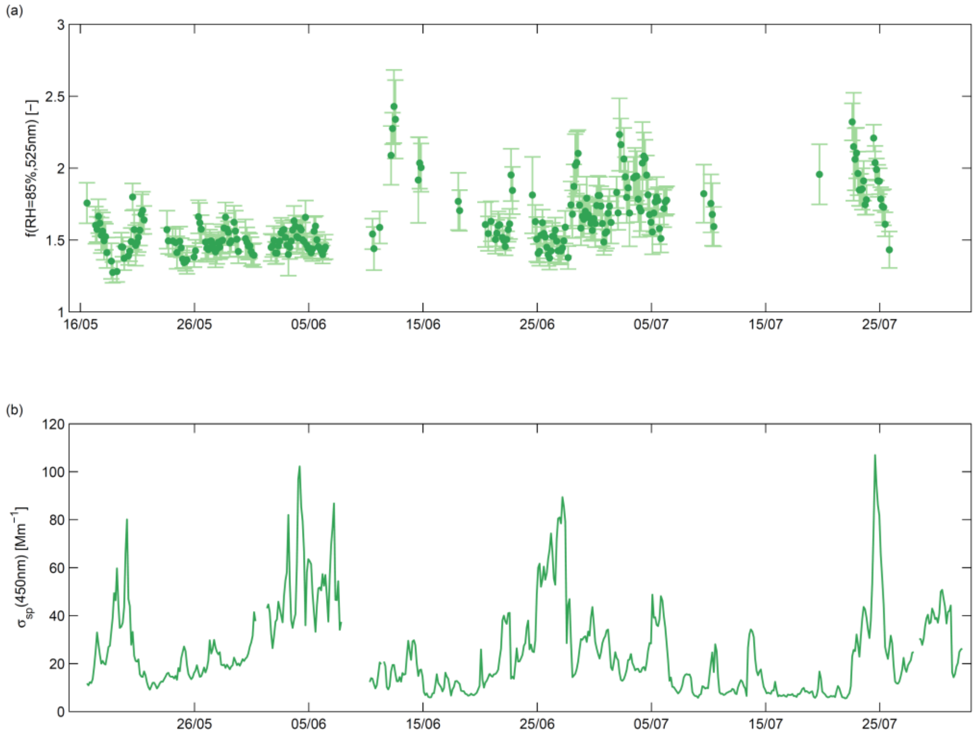

Figure 14: Figure 1 if plotted in separate panels.

Figure 2: At 450 nm the f(RH) values are lower than at 525 nm, which could be partially the reason why the authors observe low f(RH) values compared to other sites. On the other hand, the “a” parameter is closer to the ideal value of 1 at the 525 nm while it is slightly below 1 at 450 nm. This fact reinforce my opinion that the f(RH) values should focus on the 525 nm wavelength.

The difference between the different wavelengths is an effect of particle size and refractive index and was reproduced using Mie theory (a similar effect observed in Melpitz, see Zieger et al., 2014 and by other groups). It should also be noted that the effect of wavelength dependency of *f*(RH) is rather small (the red and blue *f*(RH) are less than 10% different from the green wavelength). Instead of only focusing on one specific wavelength we prefer to show the results for all three nephelometer wavelengths as we have done.

Figure 2c: reduce the x axis scale.

We have reduced the scales for panel c and b in the revised manuscript (see Figure 15).

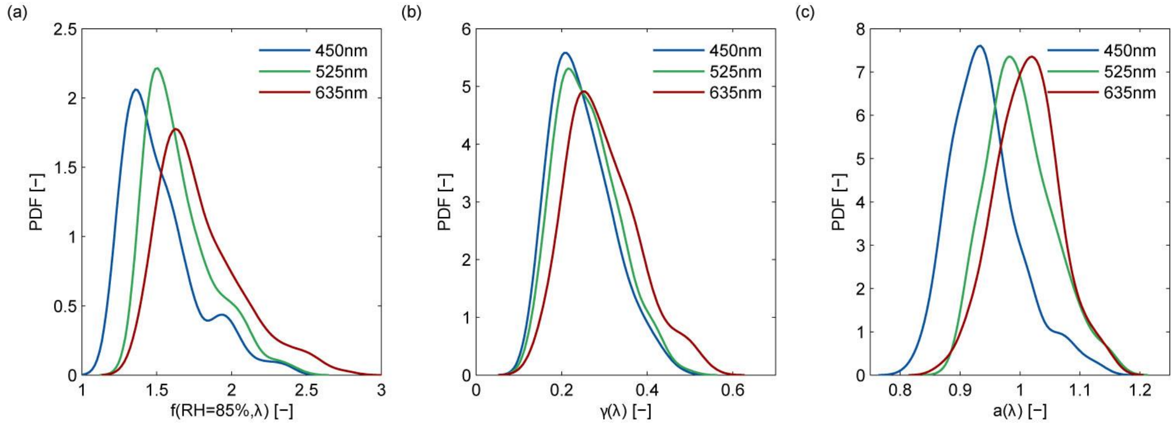

Figure 15: Revised version of Fig. 2 in the original manuscript (improved x-axis).

Figure 3: Figure 3b is very similar to that reported by Zieger et al. (2013) with the exception of Hyytiäla data. As I mentioned before, I think that there are many graphs. The authors should revise the manuscript and keep the more relevant ones.

We believe that this is a relevant graph and would like to keep in the manuscript. The figure relates our findings to other aerosol types measured throughout Europe using the same WetNeph system. Hyytiälä is in comparison a new exceptional site with its narrow distribution of small *f*(RH)-values compared e.g. to Arctic aerosol with large values and broad distribution. We have added in Sect. 5.1 the information that the same WetNeph was used at the other European sites:

"…in comparison to other European sites where the same instrument with a different nephelometer was used (Zieger et al., 2013)."

Figure 4: Change to f(525nm, 85%). The regression coefficients (slope, intercept and R2) for Melpitz differ from those presented by Zieger et al., (2014).

We agree. In the original manuscript we had decided to show only the 450-nm-channel because it is the common wavelength of both the TSI and the Ecotech nephelometer. In the revised manuscript we will show all three channels. We have linearly interpolated the Melpitz-data to the here used wavelength of 525 and 635 nm.

The regression coefficient differ from the Melpitz paper because they were given for 550 nm wavelength, while we show here the result for 450 nm as the common channel. In addition to the different channels, there are small differences because we used in the Melpitz paper an orthogonal regression technique described by York et al. (2004), while we now decided to use robust fit already implemented in Matlab technique (it is less influenced by outliers like an ordinary linear regression by using a iteratively reweighted least squares method). This fit-function is easier to use and easier to replicate for future users of the data. See revised figure and comments above (reply to reviewer #2).

Figure 5: I think that it would be more interesting to use only concurrent measurements. That way is easier to establish any relationship between the different spatial patterns.

We agree and have repeated the analysis and have re-plotted Fig. 5 only for times when all instruments (WetNeph, ACSM, aethalometer) were running at the same time. Now we also chose the 525 nm wavelength for *f*(RH) to be consistent (see Figure 16). We have also updated Fig. 6 using the 525 nm channel for concurrent times (see Figure 17). Since the first most maritime bin now only consists of two points, we add the number of points also to the figure. We also added to the manuscript the following sentence:

"Only concurrent times when all main in-situ instruments (WetNeph, ACSM, and aethalometer) were running in parallel were used."

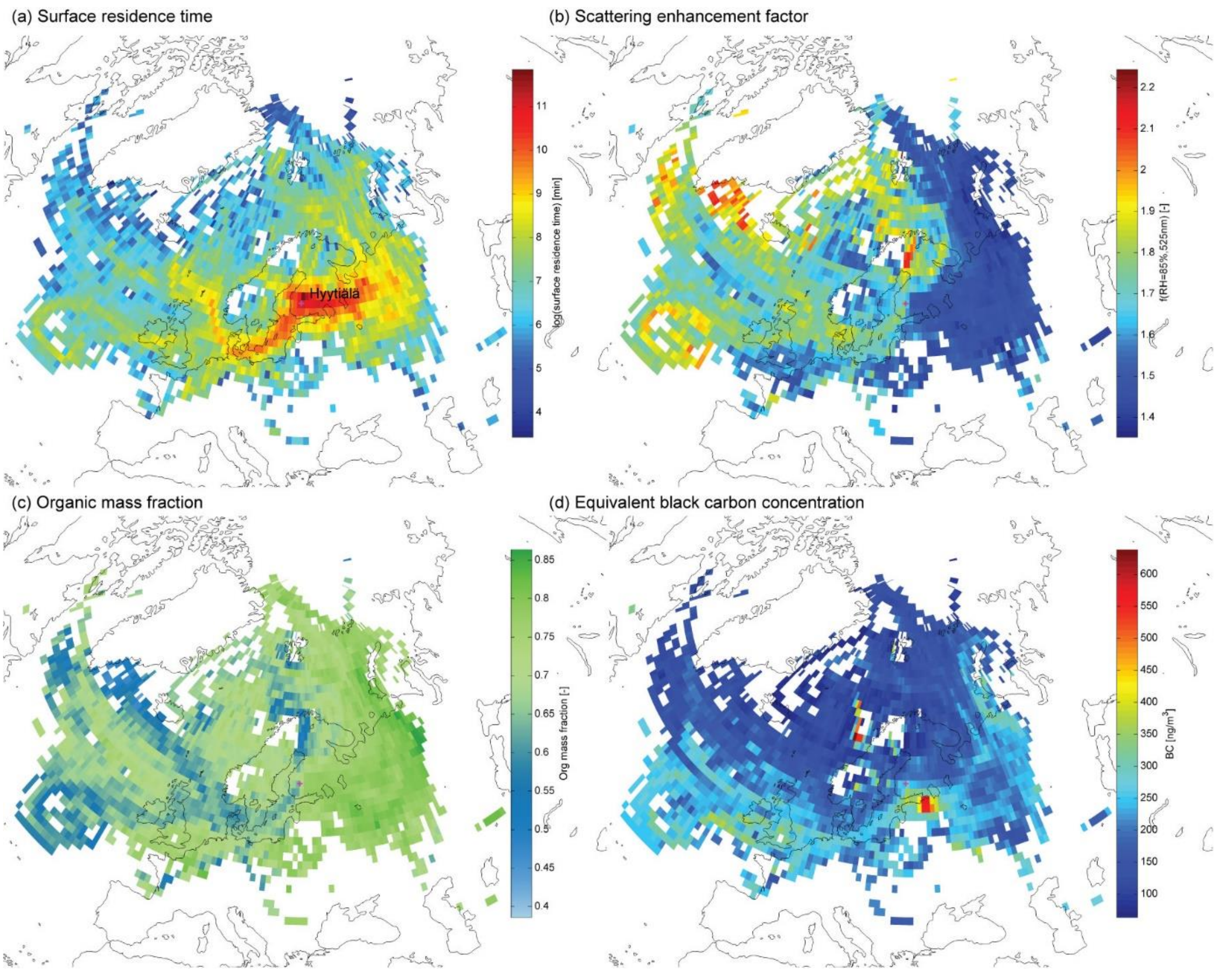

Figure 16: Updated Figure 5 using only periods when all three instruments were measuring at the same time. Now we also show the 525 nm wavelength for the scattering enhancement.

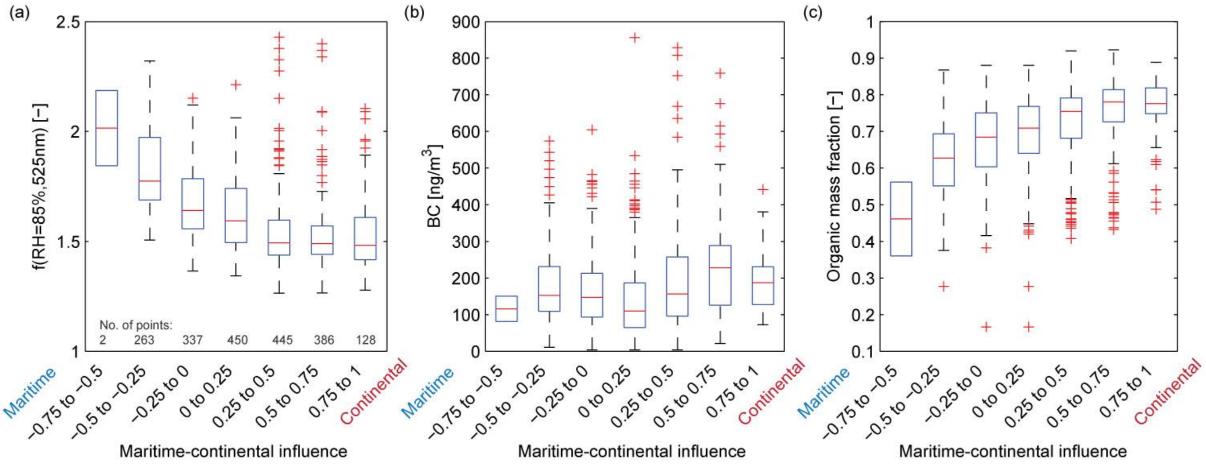

Figure 17: Updated Figure 6 using only concurrent times and the 525 nm wavelength for f(RH).

Figure 6 could be omitted.

Above we made the argument that the figure is important for the study and therefore kindly ask for the figure to be included in revised manuscript (see comment above, page 23).

Figure 7: include the ground measurement of Ntot in Figure7a.

We have removed this figure (see comments above, page 23).

Figure 10: This graph could be omitted too; the information can be given in the manuscript. In addition, consider limiting the AOD retrieval to 700 nm. The errors extrapolating to 1600 nm may justify partially the results.

We agree and have removed this graph. It is partially replaced by two additional panels for Fig. 9 showing the relative differences. See comments above and the ones from reviewer # 2 (page 6-7).

Further changes

We have added in the instrumental section (WetNeph) the following sentence: *"The relative measurement uncertainty of f(RH) as an upper and conservative estimate is 20% at RH=85% (Fierz-Schmidhauser et al., 2010c; Zieger et al., 2013)"*

Based on the reviewer's comments, we have separated the discussion on the parameterization of f(RH) using the organic or inorganic mass fraction in a separate subsection (see comments above). We have moved the 3rd and 4th paragraph of previous Sect. 5.1. to a new Sect. 5.1. ("*A simplified parameterization of f(RH)*").

Page 3333, Line 17: The RH given here (6.5±3.5%) is given for the dry nephelometer inside the aerosol cottages (TSI nephelometer), while the RH of the dry Ecotech nephelometer was 27.5±5.5% which we have corrected and have moved number for the dry RH of the TSI nephelometer to its correct position on page 3337, line 27 (referring to the ACPD manuscript page numbering).

In Sect. 3.4. (Particle chemical composition measurements): We introduced the variable F_i to define the chemical mass fraction (to be consistent with the comparison to Quinn et al. (2005) and Zhang et al. (2015). Revised sentences:

"The chemical mass fraction F_i was determined by dividing the concentrations of the individual components derived from the ACSM and EC/OC analysis by the sum of all (excluding the OC from the EC/OC analysis, which is covered by the ACSM measurement). The organic mass fraction F_{org} was determined by adding the EC part (which is known to have a low hygroscopicity) of the EC/OC analysis to the organic components of the ACSM. The mass fraction is representative for sub-micron particles only due to the experimental restrictions."

We have changed the first sentence of the 2nd paragraph in Sect. 5.1.:
"The differences in the scattering coefficients, as extensive properties, cancel out when the scattering enhancement as an intensive property is calculated."
to
"The differences in the scattering coefficients cancel out when the scattering enhancement property is calculated."
since the scattering coefficient is strictly spoken no extensive parameter.

Figure 8: We have re-done the analysis (based on the comments above) and have used now the 1-sec CPC data (before we used 2 min values), therefore the error bars are slightly different compared to the original version, but the overall results stay unaffected. In addition, while the profiles were shown at an altitude above sea level, we accidently plotted the ground based measurements at an altitude above ground. We corrected for the difference (180 m), which moved the green points slightly upwards.

We introduced SPM as an acronym for the Sun photometer and replaced AERONET when it was used to describe the Sun photometer by SPM (AERONET is strictly spoken a network and not an instrument).

We have added the reference Pappalardo et al. (2014) in the lidar section.

We corrected small typos and also updated the affiliations for some co-authors.

We updated the acknowledgements.

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