Author responses to comments of Anonymous Referee #3.

The authors would like to thank Anonymous Referee #3 for his/her detailed review of the manuscript. Many valid questions and comments were raised, and these will ultimately make the paper better. Here we will attempt to respond to all concerns and answer all questions adequately.

General Comments:

In-situ measurements are often conducted at low relative humidity. For comparison of data from in-situ measurements with data from remote sensing measurements have to be conducted at ambient or at controlled relative humidity. The reviewer thinks that it is worth to explicitly mention in the abstract and introduction that results for in-situ measurements were conducted at dry conditions and at controlled humidity.

This was pointed out by another reviewer also, and we intended to state this explicitly in the abstract and the Introduction of the paper. The measurements were made at the relative humidity inside our instruments (<40%) and most of the aerosol statistics presented in the paper are for dry conditions. This facilitates comparison with the surface measurements, which are also made under low-RH conditions. For the comparisons with satellite extinction measurements, our (scattering) extinction measurements were adjusted to ambient relative humidity conditions based on the measured aerosol hygroscopic growth.

Specific comments:

page 17189, lines 16 to 24: In-situ measurements are often conducted at low humidity. To avoid confusion the authors should state that the aerosol optical properties of interest are given for ambient condition.

This has been clarified in the revised manuscript.

page 17190 line 6: 'Covert et al. 1972' is a laboratory study investigating the hygroscopic growth of pure materials. The reviewer thinks that it is not a reference for atmospheric processing for different types of aerosols.

We have removed the Covert et al. (1972) reference. The statement this reference was attached to is a widely accepted fact, and in our opinion really needs no reference.

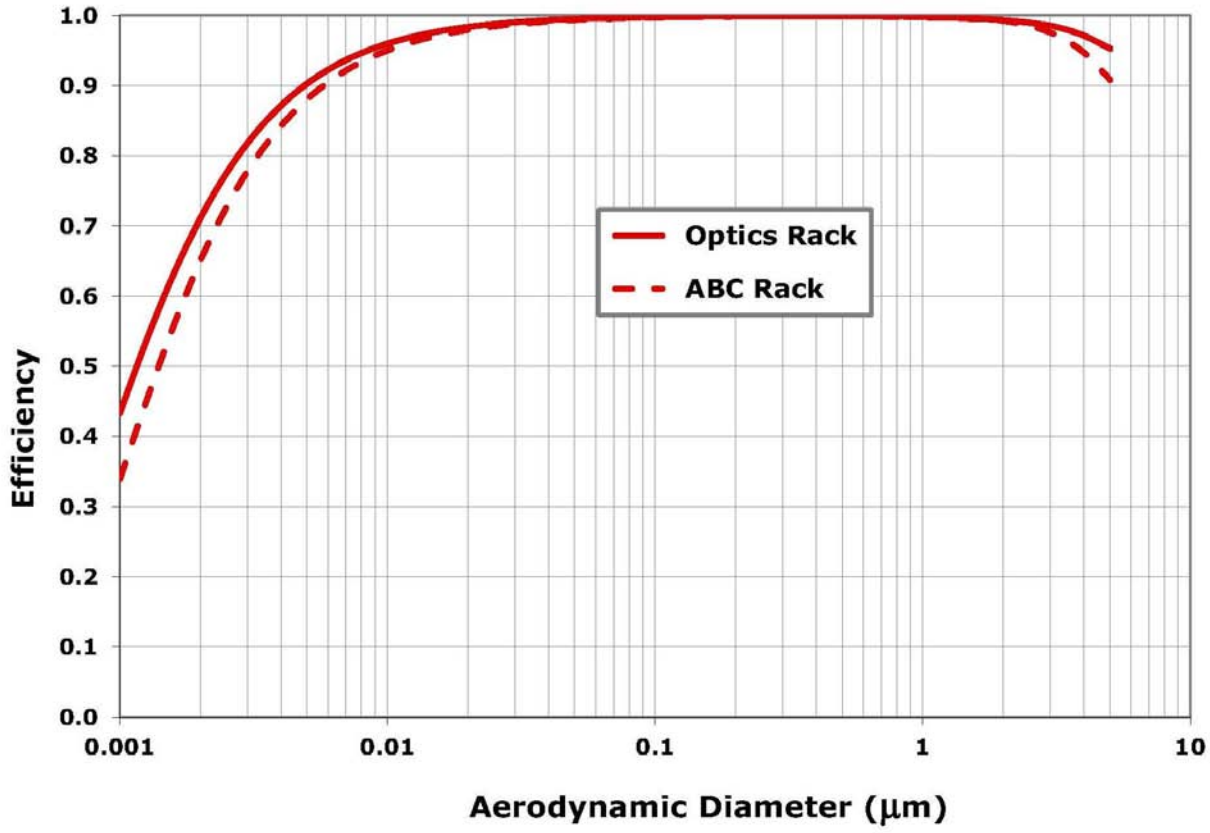
page 17195, line 18: The loss processes are not listed correctly. Gravitational settling, inertial and diffusion losses can be calculated for different flow regimes, laminar and turbulent flows, respectively.

The Referee is correct in pointing this out. We did calculate the sum of gravitational, inertial, and diffusional losses for particles of different sizes. We performed the calculations for the proper flow regime, which was always turbulent in our system. We have clarified our description of the procedure,

corrected the wording in the revised manuscript, and modified Fig. 2 (shown below) to also show the transmission efficiency curve to the ABC rack system.

page 17195, line 21: The authors should quantify how much the inlet line losses for the racks A,B and C are higher compared to the inlet losses for the nephelometer rack. This is important for showing that the single scattering albedo is not affect by different transportation losses.

We have already done this calculation. A sentence in the text reads "A similar plot was obtained for aerosols passing to instruments in the ABC racks, with only slightly lower passing efficiencies at the low and high ends due to the longer run of tubing." We have modified Figure 2 to show the transmission curve for tubing to the ABC rack system along with the transmission curve for the tubing to the Optics rack (two curves now displayed instead of just one). This figure is shown below.



We chose tubing diameters and flow rates in the system to maximize particle transmission in the optically important size range of ~0.05 to 2.0 μm. This plot shows that gravitational settling is only important (perhaps a 5% effect) for the largest particles that pass the inlet diffuser. Differences in diffusional losses between the two branches are relatively small even below 0.01 μm. These data indicate that particle losses in the branch of the inlet system leading to the light absorption instrument are quite similar to those in the branch leading to the light scattering measurement, thereby lessening

concerns of a potential bias in the single-scattering albedo measurement. We have replaced Fig. 2 with the modified version in the revised manuscript.

page 17196, lines 9 to 14: The correction for the TSI nephelometer was mentioned few lines before. The reviewer thinks that this correction is for the so-called truncation error. The readers are left in the dark if the 1-wavelength nephelometers were corrected for truncation.

We thank this reviewer for alerting us to the fact that we did not explicitly mention the truncation corrections performed on the Radiance Research single-wavelength nephelometer scattering data. This was an inadvertent error on our part.

The single-wavelength nephelometer data were corrected for truncation using the correction factors presented in Table 4a of Anderson and Ogren (1998). These correction factors for submicrometer aerosols were considered appropriate for most atmospheric aerosol populations when the Ångström exponent was not available. Using the Ångström exponents derived from the TSI nephelometer would not be appropriate in this case because those scattering data were for the 'total' size cut (i.e., no 1-μm diameter upper size limit imposed by the sampling system).

The question of whether it is appropriate to use the Anderson and Ogren (1998) truncation corrections for the Radiance Research nephelometers has been answered in the recent study of Müller et al. (2009). This study shows that the measured illumination functions for the Radiance Research and TSI nephelometers are very similar, and these coupled with the internal geometry provide generic truncation correction factors for the Radiance Research nephelometer that agree quite closely (within a few percent up to 1-μm aerodynamic diameter) with the original Anderson and Ogren (1998) corrections we used in our study.

We have included this discussion in the revised manuscript, and have added the Müller et al. (2009) reference.

Müller, T., Nowak, A., Wiedensohler, A., Sheridan, P., Laborde, M., Covert, D. S., Marinoni, A., Imre,K., Henzing, B., Roger, J.-C., dos Santos, S. M., Wilhelm, R., Wang, Y.-Q.,and de Leeuw, G.: Angular illumination and truncation of three different integrating nephelometers: Implications for empirical, size-based corrections, Aerosol Sci. Technol., 43, #6,581-586, 2009.

page 17196, line 27: The influence of changing humidity on PSAP measurements is still under debate and not much is published concerning this issue. What is the relative humidity in the PSAP. Was the humidity controlled or measured?

The AAO PSAP was modified to accommodate a small (~5W) heater on its internal inlet line just upstream of the optical block. The heating is gentle, typically only a few degrees, but the metal block stays warm relative to the sample so the RH stays low. Our own tests suggest that the heater kept the RH at the filter below 40% most of the time and an RH of 50% at the filter would have been exceeded only during sampling of very humid air, which was not common in this program. We did not actively control the heater to maintain a specific low RH, but unlike at high RH, RH variability at lower RH is not

thought to adversely influence the measurements. We inadvertently neglected to mention the small heater on the inlet of our PSAP in the manuscript. This has been corrected in the revised version.

page 17198, lines 11 to 14: The backscatter fraction was used to calculate the asymmetry parameter and the up scatter fraction using parameterizations given in Andrews et al 2006 and Grams 1976, respectively. Both parameterizations are limited in their use to special conditions and aerosol types. The authors should mention the limitations. Does this affect the calculated radiative forcing efficiency?

We mentioned in the paper that the hemispheric backscatter fraction (*b*) was related to the upscattered fraction (β) as described by Wiscombe and Grams (1976), who also related these parameters to the asymmetry parameter (*g*). We are not qualified to comment on how they derived the relationship, their assumptions and simplifications, or the limitations of their methods. We understand that the relationships assume a Henyey-Greenstein phase function, which they consider representative for the earth's atmosphere. We used a second-order curve fit to the points in their Fig. 3 to derive our equation for β in Table 2.

Haywood and Shine (1995) used β, along with the measured ω_0 and optical depth (δ), and made assumptions about cloud fraction, surface reflectance, and atmospheric transmission to calculate the global mean, annually-averaged, top of the atmosphere (TOA) aerosol radiative forcing (ΔF). We do essentially the same calculation to derive the aerosol radiative forcing efficiency (RFE, $\Delta F/\delta$). A major limitation in this method is that the calculation of RFE provides for no geographical variation in the assumed physical constants and no zenith angle dependence of β. Another limitation is that the dependence of ω_0 and β on RH has been ignored. This should not have a major effect on the reported RFE's in this study because they were calculated using our low-RH aerosol measurements.

The parameterization presented in Andrews et al. (2006) was used in this study to calculate *g* from our *b* measurements. This was also derived from a curve fit of data presented in Wiscombe and Grams (1976). Wiscombe and Grams (1976) suggest that the relationship between the upscattered fraction and asymmetry parameter is a function of the angle of the incident light, which we do not explicitly account for. In addition the refractive index of the aerosol also plays a role with more absorbing aerosol (higher imaginary part of the refractive index) complicating the relationship. The relatively high ω_0 of the BND/AAO aerosol is similar to that measured at the SGP site (~0.94) (Delene and Ogren, 2002) and because optical closure studies done on the SGP aerosol (Andrews et al., 2004) suggest that the imaginary part of the refractive index is relatively low (0.01-0.02), aerosol type/composition will not have a large effect on the parameterization.

Andrews et al (2006) show comparisons (Figs. 2b and 7d) of the calculated asymmetry parameter using 4 different, independent methods:

(i) the parameterization being questioned by the reviewer

(ii) calculated using Mie theory (assumption of homogenous spherical particles)

(iii) inversion of Aeronet spectral optical depth data

(iv) inversion of a suite of in-situ aerosol measurements (Fiebig and Ogren, 2005).

The figures suggest that all four methods agree quite well. This implies that, if the surface aerosol at SGP is similar to the BND/AAO aerosol (which we no reason not to believe), then the parameterizations of upscattered fraction and asymmetry parameter based on curve fits of the Wiscombe and Grams (1976) data are fairly representative of reality.

Given how little the RFE results contribute to the manuscript, this explanation is clearly too long and detailed to publish in the revised manuscript. We have added a few words of clarification on how the parameterizations were done and have referenced other papers for specifics. If readers are interested in the details and the limitations, we can direct them to look at the Author Comments in ACPD.

page 17198, lines 15ff: If scattering data of the 1-wavelength nephelometer have not been corrected for the truncation error, then the resulting growth factors f(RH) might be affected by instrumental artefacts since the truncation changes with particle size and thus with relative humidity. This could be important when comparing gamma values of different studies (page 17209 lines 27ff). Can the authors comment on that?

As discussed above, the scattering data for the 1-wavelength nephelometers have indeed been corrected for truncation effects. The correction used is admittedly a crude approximation, but it is much better than doing no truncation correction at all. Since the Anderson and Ogren (1998) and more recently the Müller et al. (2009) methods are commonly used nowadays to correct truncation errors in nephelometers, our data should be comparable to other studies, at least ones published after 1998.

page 17198, lines 16 to 25: Because of the hysteresis effect the scattering hygroscopic growth factor might depend on the direction in which the humidity changes, i.e. the aerosol is dried first and then humidified or vice versa (see Fierz-Schmidthauser et al. 2010). In the present study the aerosol is dried and afterwards humidified. Is this practice appropriate for investigating atmospheric processes?

We believe that it is. You have to go one way or the other. As the reviewer points out, we typically sampled a dry aerosol (<40% RH), performed a size cut at 1-μm aerodynamic diameter, and then fed it into humidified nephelometers (held constant at roughly 65% and 85% RH). The only times we have seen large differences in our scanning humidograph systems between the upscans and the downscans (i.e., hysteresis effects) is when we sample aerosols with a major inorganic salt component (e.g., sea salt near a coastal location, sulfate downwind of a major point source, etc.) or pure laboratory salt aerosols as discussed in the Fierz-Schmidhauser et al.(2010) reference. The aerosols over Bondville are fairly well-mixed and are comprised of many different aerosol components, some hygroscopic and some (e.g., soil dust, black carbon) not. This tends to damp out any deliquescent jumps at specific relative humidities from the pure components. In our limited operation of scanning humidograph systems at BND, we did not observe major deliquescence or efflorescence effects; therefore it should not make a major difference in which direction the humidification occurs. While the presence of some metastable particles in our system is likely and acknowledged, their effect on the growth functions is difficult to quantify.

page 17210, lines 11-13: A mean gamma value of 0.51 was chosen but Fig. 10 suggests that the gamma value depends on the altitude. How large is the effect in relation to the differences between AAO and CALIPSO shown in Figure 12.

The line in the text the Reviewer references states "A median γ value of 0.51 for the AAO aircraft measurements was derived by fitting curves as described in Table 2 to the humidified nephelometer measurements from the 89 valid AAO flight segments where all humidified nephelometers were working properly."

The Reviewer is absolutely correct that our data suggest that the gamma value depends on altitude. The sentence above was meant to show how our overall data set compared with the other known *f(RH)* measurements at Bondville (Koloutsou-Vakakis et al., gamma = 0.33 and Quinn et al., gamma = 0.67).

Two errors that we are aware of inexplicably made it into the submitted and reviewed manuscript. The first is the number of valid AAO flight segments; we have this listed at 89. The true number was 80, which can be found from summing the valid flight segments at each altitude in Fig. 10. This was the number of flight segments for the 12 'good' profiles (120 maximum possible flight segments) that exceeded 3.0 Mm^{-1} dry scattering coefficient at 550 nm. Many of the higher altitude segments did not exceed this threshold.

The second error was that, even though the manuscript is not clear on this point, we did in fact use an altitude-specific gamma to adjust our dry aerosol data to ambient conditions. We realized early on that this was necessary and performed the necessary calculations, but the old text was not modified in the paper. Thus, the comparisons shown in Figure 12 are valid… the aerosol extinction at each altitude was adjusted to ambient RH conditions using a gamma value appropriate for that altitude.

A median γ value of 0.51 for the AAO aircraft measurement program was derived by fitting curves as described in Table 2 to the humidified nephelometer measurements from the 80 valid AAO flight segments where all humidified nephelometers were working properly. This value of γ results in an estimated *f(RH)* (at 85% RH) of 1.87, intermediate to the values derived from the chemical composition and Koloutsou-Vakakis approaches (Esteve et al., 2012). For comparison, the median boundary layer (all flight segments conducted below ~1.8 km altitude) γ value was 0.53 and the median free troposphere (all flight segments above ~1.8 km altitude) γ value was 0.44.

The cumulative dry aerosol σ_{sp} distributions shown in Fig. 7 were adjusted to ambient RH and were presented in Fig. 11. Individual flight segment-average σ_{sp} values at 550 nm were adjusted to ambient RH using the median altitude-specific γ value for all valid segments and the segment-average RH. The median γ value for each flight level was used because so few flight segments were conducted with valid hygroscopic growth measurements. It is implicit in this analysis that the median γ values at each flight altitude adequately represent the hygroscopic growth characteristics of the aerosols during flights when the humidified nephelometry system was not working properly. We observed no obvious long-term changes in aerosol characteristics over the duration of the program to lead us to believe that this

assumption was not valid. Additionally, the segment-average RH was typically low enough that small to moderate changes in the γ parameter would not result in major changes in the adjusted σ_{sp} values.

Even though the adjustment of scattering to ambient RH is nonlinear in RH, we felt using the segment average rather than the instantaneous RH was appropriate for two reasons. First, the RH for most of our flight levels was not very high, and RH variability at lower RH does not cause large changes in the optical properties. Figure 7(f) shows that median RHs for flight levels in the mixed layer were 50-60% and were lower at higher altitudes. Only a small fraction of all flight levels show segment-average RH values above 70%, where the growth function becomes steeper. Secondly, RH variability was consistently small over the 5 and 10 minute horizontal segments flown over the course of the program. Considering all AAO flight segments, the average standard deviation (SD) of RH measurements over a flight segment was 2% in RH (with a SD of the SD values of +/-2%. That is, on average, each AAO flight segment showed a relatively small variability (expressed as the SD) of 2% RH. While there were a small number of flight segments conducted at higher RH that also showed a larger RH variability, this was not a commonly observed feature of the data set. In general, the changes from the dry scenario for light scattering are relatively small, with median ambient scattering values in the PBL higher than the dry case (dry median values shown by red line) by only a few Mm^{-1}. This can be explained by the fact that flights typically occurred during sunny to partly cloudy conditions where ambient RH was relatively low (typically < 60%). Fig. 9(f) shows that even during the summer, median RH's in the PBL during AAO flights was ~57-65%. Relative humidities in this range are not high enough to increase the dry scattering values a large amount. No attempt was made to scale AAO light absorption data (or single-scattering albedo and extinction data) due to a lack of information on the hygroscopic growth function for light absorption.

The errors have been corrected and an expanded discussion of the procedure used for adjusting the dry measurements to ambient RH has been included in the revised manuscript.

page 17227, table 2: Equation for absorption Angström incomplete; "/log(lambda_1/lambda_2)" is missing

The reviewer is not correct here. We performed the scattering and absorption Ångström exponent calculations differently. For comparison with the historical literature and other current studies, we used the common two-wavelength method for calculation of the scattering Ångström exponent. For the absorption Ångström exponent, however, we used a power law fit of all three wavelengths. This provides more information than the two-wavelength fit, and there are not many historical studies to compare our data with anyway. We use the subscript *i* to denote all wavelengths. Here is the passage in the text that explains this…

"The $å_s$ data presented in Fig. 7 were calculated using the 550 and 700 nm wavelength scattering data from the TSI nephelometer, and the $å_a$ data were obtained using all three wavelengths from the PSAP."

page 17230, Fig2: The title "Cessna 206 Inlet penetration" is misleading since the efficiency of the 'main inlet' in not included in the figure.

We removed the title of the figure in the revised manuscript and clarified for the reader that these passing efficiency curves are for the inlet tubing downstream of the main inlet. The main inlet efficiency was characterized already in the DICE experiment, albeit under different sampling conditions. We did not perform a rigorous particle transmission efficiency study on this inlet.

pages 17233 and 17234: Figure captions: subscript "0" missing in "(d) ω for total. . ."

This has been corrected in the revised manuscript.

Page 17241: The authors should explain the x-axis label "f(RH) (85%/40%)"

As requested, we have explained the x-axis label in more detail in the figure caption in the revised manuscript.

References:

Fierz-Schmidhauser, R., P. Zieger, G. Wehrle, A. Jefferson, J. A. Ogren, U. Baltensperger and E. Weingartner "Measurement of relative humidity dependent light scattering of aerosols." Atmos. Meas. Tech. 3(1): 39-50.