

## Scattering and Absorption by aerosols during EUCAARI-LONGREX: can airborne measurements and models agree?

### Response to Anonymous referee #1

We thank the reviewer for their detailed comments, and recognise that they expect the paper to be publishable following some revisions. Their comments are in blue below, our response is in black. Resulting changes that will be made for a revised version are in red.

The reviewer suggests that the paper **has value both in supporting other publications associated with the data set, and potentially as a recent overview of aerosol characteristics in the European lower troposphere**. To achieve the latter, the reviewer rightly contends that much more analysis and comparison with other data sets would be needed (a point also made by referee #2). However, much of this data has already been presented in studies by other studies already published (e.g. Hamburger et al, 2011, McMeeking et al, 2011, Morgan et al, 2010a and b), or known to be in preparation – we do not think it appropriate to include this material here. The scattering and absorption measurements from the FAAM BAE146 platform have not to date been published and the purpose of this paper was to address this gap. We suggest that a further motivation for the paper is to demonstrate the importance of accurate measurements of both black carbon mass and absorption by aerosols if airborne measurements are to be used to characterise the radiatively important properties of tropospheric aerosol. As such, we believe that the study has value without the suggested large scale additional analyses but we will address as many of the specific concerns raised here as possible

**The main conclusion of the paper – that correctly modelling absorption requires a narrow range of index of refraction for organics – is poorly motivated because of the dominating uncertainties in the BC and absorption measurements: in fact the uncertainty is large enough to render all associated discussion about the bias moot.**

The referee is correct to say that the uncertainty in absorption measurement is large, and restricts the conclusions that can be drawn about the refractive indices of OC and BC from the closure experiment. This is in fact reflected in both the last sentence of section 5 AND the ultimate sentence of the entire paper which urges the development of accurate and robust measurements of absorption. We disagree however with the discussion being moot. We believe that it is important to point out that in some air masses with relatively little black carbon but high organic carbon, the absorption is strongly affected by the refractive index that is assumed for the organic carbon – and this is not well known. In the course of responding to the reviewers comments we have changed figure 9 and added some additional analysis of potential absorption measurement problems. These changes demonstrate that there are multiple possible explanations for the model disagreements, and reinforces our view expressed in the original paper that it is imperative to consider reductions in the measurement uncertainty for aerosol absorption. **We have altered the first sentence of the 3<sup>rd</sup> paragraph in section 5 to read “Notwithstanding the large errors in the measured absorption, it is useful to consider reasons why the model could be over-estimating the absorption”, while the discussion of this issue in the final section of the paper has been rephrased to reflect the two possible ways of improving the degree of closure and to emphasise the need for more accurate measurements of absorption. In addition we have added similar text to the abstract.**

The underlying thrust of the paper is technical: to demonstrate the technical control over the airborne instrumentation necessary to achieve closure. Of itself, this is of value in supporting other publications associated with the data set, however the paper does not present the technical underpinnings of the comparison strongly enough to achieve this: better connection to the ground-stations is required for absolute validation.

We would like to emphasise that the closure that is the main focus of the paper is on the internal consistency between airborne measurements; we presented comparisons of optical depth with ground based sites merely for additional information. Apart from one occasion (Cabauw and flight B379) the matches of aircraft position and timing to ground based AERONET sites (of particular interest for aerosol radiative properties and impact) is poor due to combinations of meteorology and air traffic restrictions. We therefore agree with the reviewers later point that given the large variability in AOD spatially, and the difficulty in getting to very low altitude over the ground based stations, the comparison with most of the ground based values is of little value and **this has therefore been removed from a revised version of the paper, leaving only the AOD values from profiles and a discussion of their likely accuracy in section 4. The text there now reads as follows:** “

“It would be desirable to compare AOD estimates with those measured from the ground by the AERONET network. Unfortunately in most cases this is not possible due to either a spatial or airmass difference. The most suitable match, B379 and the Cabauw station, showed that the AOD measured from AERONET was 0.26 whilst the combination of scattering profiles from the aircraft between 500-8500m gave an AOD of 0.20. A slight underestimate might be expected because the method used here neglects the absorption optical depth. An alternative method is to use an average single scattering albedo to estimate the contribution to the AOD from absorption. Using the boundary layer SLR average single scattering albedoes we can estimate that the true AOD should be between 3 and 9% higher than that calculated from the scattering profile alone. For B368 which has a very low SSA, if this is realistic, the total AOD would be 40% higher than that from the scattering alone. This is in addition to the uncertainty in AOD of 30% due to the uncertainty in the scattering measurements and combining these uncertainties would bring the aircraft value into better agreement with the ground based measurements. There may be an additional underestimate due to not sampling the lowest 500m with the aircraft.”

Further, we feel that our response to the specific points of both reviewers will strengthen the technical underpinnings of the comparison sufficiently.

The relative impact of BC on the conclusions and measurements here is never clearly presented. There are several comments about “BCs negligible impact” and “BC is a small fraction of the total aerosol” that, when coupled with the comparison of the scale lead me to believe that the BC measurement doesn’t influence the model/measurement comparison at any level near the uncertainties in the scattering/extinction comparison. Thus I encourage the authors to more explicitly present the relative contribution of BC to the total aerosol mass loading and AOD.

Details of the BC component as measured from the BAE146 during EUCAARI are presented in detail in McMeeking et al (2010) and we do not wish to repeat their analysis here. However, we agree that some quantification is necessary in this paper. From that study, refractory BC, as measured by the SP2 represented between 0.5 and 3% of sub-micron aerosol mass. **We have altered the sentence on page 18, line 3-5 to read “However, in most cases considered here, the black carbon mass fraction is**

small compared to the other components (being between 0.5 and 3% of total submicron aerosol mass, McMeeking et al, 2010)". The absorption contribution to optical depth cannot be calculated directly from the aircraft measurements since, due to problems with the flow meter, the PSAP data from profiles is not available. Thus the AODs presented in the original paper were derived only from the scattering measurements. An alternative method is to use an average single scattering albedo to estimate the contribution to the AOD from absorption. Using the boundary layer SLR averages we can estimate that the true AOD should be between 3 and 9% higher than that calculated from the scattering profile alone. For B368 which has a very low SSA, if this is realistic, the total AOD would be 40% higher than that from the scattering alone. **We have added this discussion to section 4 of the paper (see above).** However, this is the contribution from total absorption by aerosol, and as we discuss in section 5, the contribution of BC to this depends on the assumptions we make regarding refractive indices of BC and OC. Figure 9 can be used to assess the relative role of BC and OC – on average the BC contributes about 25% of the total absorption. Thus, BC is responsible for perhaps 2.5% of the AOD. The referee is right therefore to suggest that the BC measurement influence on AOD is generally small. However, we would point out that it is not only the total AOD that is important for assessing the radiative and climate impact of aerosol; the single scattering albedo is also very important and BC is important here as it contributes around 25% of the total absorption. **We have added a statement regarding this to section 5 of the discussion which now reads "The BC effect is relatively weak here as the mass is relatively small, and contributes around 2.5% of the total AOD. In regions with high BC loadings, the uncertainty in its absorption is expected to play a far greater role. However, the extent to which we can definitively constrain the refractive indices is significantly limited by the considerable uncertainties in measured absorption. Additionally, BC plays a significant role in the determination of the single scattering albedo even here, contributing around 25% of the total absorption".** We thank the reviewer for suggesting the inclusion of this information in the study.

#### **Specific points made by the reviewer**

The title suggests a narrower treatment of data than is presented. Please consider modifying the title to better represent this work.

The title has been changed to reflect the contents of the paper although the comments of the two referees were somewhat contradictory in their recommendations. **Title changed to: "Aerosol scattering and absorption during the EUCAARI-LONGREX flights of the Facility for Airborne Measurement (FAAM) BAe146 : can measurements and models agree?"**

Please include uncertainties in the values presented in the abstract. **Done**

Line12-13 isn't accurately representing the findings of this paper. It is well known that the refractive index is a crucial parameter in modelling absorption. The point here appears to be that to achieve agreement between model and measurement, the imaginary part of the index for organic aerosol is fixed to a narrow range of values. This is a major conclusion at 550nm that must be defended. Please reword to more clearly present the findings of this manuscript to the reader.

We did not mean to suggest that this study was the first to point out the link to refractive indices, rather that in this study the assumed refractive index for OC was more influential than that for BC. **This part of the abstract has now been reworded in the light of this comment and the additional**

work regarding absorption measurement uncertainties, as follows: “Agreement to within 30% can be achieved for both scattering and absorption, but the latter is shown to be sensitive to measurement uncertainties for absorption and the refractive indices chosen for organic aerosols, and to a lesser extent black carbon. Agreement between modelled and measured absorption can be achieved if either the organic carbon is assumed to be weakly absorbing, or it is assumed to be completely non-absorbing and the absorption measurements are assumed to be in error due to the high organic aerosol loading in line with previous studies. Thus improvements in absorption measurement accuracy are required before any refractive indices for organic carbon could be inferred conclusively.

Line 16 “ratio of scattering” – altered “change” to “ratio”

The last sentence seems to be directed at the editor rather than the reader – sentence removed

Line 2 of 18491: The SP2 measures all aerosol that scatters sufficient 1064nm light, and additionally quantifies BC mass in all individual particles. I suggest the authors state something in this line “... as the AMS data does not detect mineral dust or sea salt aerosol and the SP2 was used only to detect BC, the analysis provided here is not likely to represent regions with high loadings of these undetected materials.”

The referees suggested wording differs only slightly from our original paragraph so we have made a small modification. The paragraph now reads: “As the AMS data does not detect mineral dust or sea salt aerosol, and the SP2 was used only to detect BC, the analysis provided here is not likely to represent regions with high loadings of these undetected materials”.

Instrumentation: It is clear that the AMS, SP2 etc have been documented at length in other papers, however, I suggest that the authors still include a brief description of the instrument parameters of interest here, especially those that bear on the discussion of the quality of the model/measurement closure

We agree that some additional information regarding the instrumentation is needed in section 2.2, particularly focussing on the size range sampled by the instrument and the inlets. Section 2.2 has been expanded with the following text (this replaces Table 2 and puts the instrument information in the main body of the paper). The wet neph description was also moved to this section from section 4, and the issue of drying aerosol moved from section 2.2.2:

### Section 2.2 Instrumentation and derived aerosol properties

The FAAM BAE-146 carries a considerable range of instrumentation. Only the instrumentation that is particularly relevant for optical property studies is described here. More comprehensive discussion of the full instrumentation can be found in Johnson et al, (2000) and Osborne et al (2007).

Aerosol scattering coefficients at 440, 550 and 700nm by (mostly) sub-micron aerosol are measured using a TSI3563 nephelometer. The resulting scattering is corrected for angular truncation, temperature and pressure as according to Anderson and Ogren (1998) and Turnbull (2010). It is assumed to be applicable to “dry” aerosol, although the sample was not actively dried during these flights. The sample is certainly at lower than ambient relative humidity due to the effect of heat from the nephelometer electronics and lamp, the dynamic heating through deceleration of the input flow

which reaches the instrument and the increased temperature of the sample lines compared to ambient air. However, the sample humidity depends on the ambient relative humidity and when this is high the sample may not be absolutely dry. During EUCAARI –LONGREX, a second TSI 3563 nephelometer, the “wet-neph” was operated in series with the standard nephelometer (e.g. Haywood et al, 2008). The sample flow through this nephelometer is humidified to a set value between 45 and 95%. During an SLR, the humidity is either cycled through a range between these values, or set at a fixed high level. The value  $f(\text{RH})$  is defined as the ratio of scattering co-efficient measured in the wet-neph to the scattering co-efficient measured by the original nephelometer. To minimise the uncertainty due to the unknown RH of the sample in the dry nephelometer, only sections of SLR where the relative humidity measured in the dry nephelometer is less than 30% are used to estimate the growth factors for scattering. For situations when the standard nephelometer recorded humidities higher than 30%, the growth curve for dry sections of the same flight have been used to adjust the standard scattering co-efficient to values at 20% and these are reported as “dry” scattering co-efficients.

The aerosol absorption co-efficient at 567nm is measured using a Radiance Research Particle Soot Absorption Photometer (PSAP). The air is sampled through the same inlet as is used by the nephelometer, and measurements are also therefore assumed to be representative of aerosol below 30% RH. The raw data is corrected for pressure and flow rate and for spot size effects as described by Bond et al, 1999, Ogren 2010 and Turnbull, 2010). The absorption at 550nm is found assuming an inverse dependence of absorption with angstrom exponent.

The aerosol number size distribution is measured using a Particle Measuring System Passive Cavity Aerosol Spectrometer Probe 100-X(PCASP) mounted in a wing-pod. This provides aerosol sized in 15 bins ranging from approximately 0.1 to 3 micron diameter. The bin widths had previously been calibrated in laboratory experiments. In this study we have not adjusted the bin widths for the refractive index as the difference from calibration standards was small.

The total non-refractory mass is measured by an Aerodyne time-of-flight mass spectrometer (ToF-AMS) and reported as the mass of groups of chemical composition (e.g. organics, nitrate, sulphate, ammonium and chloride). Only particles less than approximately 800nm aerodynamic diameter are sampled by the instrument. However, the sub-micron mass captured by the AMS agreed within 30% with the volume convolved number size distribution from the PCASP (Morgan et al, 2010) which suggests that the majority of the sub-micron mass is captured by the AMS.

The mass of sub-micron refractory black carbon is measured via incandescence of individual particles by the Single particle Soot Photometer (SP2). This instrument is described in detail in Schwarz 2008 and in the context of this measurement campaign and platform by McMeeking et al (2010). The SP2 captured between 80-100% of the sub-micron rBC mass based on log-normal fits to the measured size distribution (McMeeking et al, 2010). We did not apply a correction factor to account for the non-detected mass, as has been done in previous studies involving the SP2 (e.g. Schwarz et al, 2008) but rather report the mass observed within the instruments detection range.

Please include an estimate of the AMS lens efficiency with a comment about how well you expect the AMS to represent total aerosol mass for the size distributions observed in the dataset.

We recognise that the transmission window of the AMS lens (approx. 40-700nm vacuum aerodynamic diameter on the ground shifts to smaller sizes at reduced pressure. There is no specific experiment or correction made for this effect, but there has been no evidence that the AMS misses a large fraction of sub-micron mass. Page 18493, line 13-15 in the original paper discusses the extent to which AMS captures the sub-micron mass by comparing the mass from the size distributions derived from the PCASP (which are used in the closure study) and those from the AMS. These agreed to within 30% across the EUCAARI-LONGREX campaign. We have added the following text to the expanded section 2.2 (and removed something similar from section 2.2.1) **The sub-micron mass captured by the AMS agreed within 30% with the volume convolved number size distribution from the PCASP (Morgan et al, 2010) which suggests that the majority of the sub-micron mass is captured by the AMS.**

State how completely the SP2 captured total BC mass in the accumulation mode – what was the correction (if applied) for non-detected BC mass in this mode? This is significant for later in the paper when the bias in measured/modelled absorption is discussed.

The SP2 captured between 80-100% of the sub-micron rBC mass based on log-normal fits to the measured size distribution (McMeeking et al, 2010). We did not apply a correction factor to account for the non-detected mass, as has been done in previous studies involving the SP2 (e.g. Schwarz et al, 2008) but rather report the mass observed within the instruments detection range. **This text has been added to an expanded discussion of instruments and inlets in section 2.2 (see above)**

The McConnell paper (cited to support use of Rosemount inlets) appears to indicate very large (i.e. factor 3) discrepancies in extinction measured on the BAe146 and the DC8. This raises the fundamental question about sampling of urban aerosol, for comparison to ground sites, and also about sampling from different inlets for the internal closure experiments. Please describe inlet placement on the aircraft if the various instruments are sampled from different inlets.

Other than PCASP which is wing mounted, all the instruments used sample through Rosemount inlets towards the front of the aircraft. The nephelometers and PSAP sample through the same inlets, the AMS samples from an inlet close to this. The SP2 inlet is on the other side of the aircraft but in a similar position. Various studies have suggested that these inlets, and more specifically the combination of inlets and sampling piping do not allow coarse mode aerosols to be well sampled. This is a problem for studies involving mineral dust (e.g. that referred to McConnell et al, 2008) but less so for the generally smaller sized anthropogenic aerosol studied here. Previous studies (e.g. .... ) have used a correction factor of around 1.3 to allow for this. However, recently the inlets have been characterised more fully. The discrepancy between nephelometers on the B146 and the NASA DC-8 described in McConnell et al (2008) was indeed concerning, but the instruments have since been recalibrated and the consistency between column AOD measurements from AERONET and satellites in both this study, that of Morgan et al, (2008) and other projects since DODO suggest that whatever issue arose during the work described in McConnell et al (2008) has been resolved. In order to clarify the matters of interest for this work, the reference to McConnell et al (2008) has been removed from the text in section 2.2, and the inlets described in the following manner in section 2.2.

**All these instruments other than PCASP sample through Rosemount inlets. The nephelometers and PSAP sample through a common inlet. The AMS samples through an additional inlet close to this.**

The SP2 samples through a third Rosemount in a similar position but on the other side of the aircraft. Particle loss can occur both as a result of the inlet and the piping between the inlet and the instrument. Previous studies (albeit on a different aircraft) have suggested that the nephelometer and PSAP see particles only up to approximately 3 micron diameter (e.g. Haywood et al, 2003). This cut-off can prove problematic if there is considerable coarse mode aerosol, as is the case for mineral dust. It is less important for smaller anthropogenic aerosol. More recently the inlets have been characterised more comprehensively and this confirms that the particle transmission upper limit is in the region 3.25 to 3.75 micron (Trembath, personal communication, 2011). There is therefore some evidence that sizes sampled are not limited by the inlets themselves in the case of the AMS and SP2, and that the sizes for which scattering and absorption are measured are consistent with the sizes measured by the PCASP – this is important for the closure experiments discussed in section 5. There was found to be some enhancement of aerosol by the inlets. Below around 0.7 micron the enhancement is around 1.5 times and similar for both the AMS and nephelometer/PSAP inlets. At larger sizes the enhancement varied between inlets, but this is of less relevance to the aerosol measured in this study.

The Lack et al (2008) paper does not suggest correcting PSAP as a function of organic loadings, but merely points out the scale of possible artifacts when large loadings are present. To support conjecture about the possible artefact, please present the actual organic loadings in question.

We did not say or imply that the Lack et al (2008) paper suggested correcting PSAP as a function of organic loadings. We have considered the average organic loadings for each SLR and flight and compared these with the scales presented in the Lack et al (2008) paper. The ratio of Organic Carbon to Black Carbon ranges from 20 to 120. Using the relationships presented in Lack et al (2008) for “all data”, this would give a likely over-estimate of the absorption by PSAP of between a factor of 2.5 and 5.1 (though we accept that the OC/BC ratios we see here are beyond the scope of the data presented in that paper). Using the “high OOA” values gives a range of factors 2.2 to 4.4. It is interesting to note that in McMeeking et al (2010) and in this paper, the mass absorption efficiency of BC derived from the PSAP and SP2 instruments is around  $41 \text{ m}^2\text{g}^{-1}$ , a factor of 5.5 higher than the more usually quoted  $7.5 \text{ m}^2\text{g}^{-1}$  (Bond and Bergstrom, 2006). This enhancement is consistent with the implied enhancement of absorption measured by PSAP due to the presence of very high organics loadings. We have added a sentence to the paper to describe this in section 5.1 (replacing the conjecture in the original version of the paper) and added this uncertainty to figure 9 and its discussion.

The wing-tip to wing-tip comparison only provides relative accuracy for 2 instruments. Please explicitly link to absolute uncertainty. The referee is absolutely correct that this does not describe the absolute uncertainty (although it was interesting to us at the time). There has been much discussion about the relative merits of PSAP instruments in measuring absorption in the literature. We have therefore estimated the measurement uncertainty at 30% which is at the top end of estimates by e.g. Bond et al (1999), Weingartner et al, 2003 and Virkkula et al, 2005) and consistent with some observations of over-estimate by 30-50% of absorption by PSAP instruments compared to other measurements when organics loading is high, as in this case e.g. Lack et al (2008). However, it is likely that this estimate is an underestimate, given the very high organics to BC ratios measured in this particular aerosol. We have inserted additional discussion of these issues to section 5 and the conclusion (see also response to point above)

Line 9, page 18494: please exclude biological aerosol too. Text changed as suggested

Section 3.1: please use the appropriate number of significant figures throughout the paper and include uncertainties in stated values

Although the referee does not say what they judge to be the appropriate number of significant figures, we assume that they object to overly precise values for the mass extinction co-efficient. In fact we have quoted our values to the same level of significant figures as other studies with similar values and uncertainties, so we believe that the original is appropriate here. We have added uncertainty values to section 3.2 and section 4 and section 6.

Section 3.2 Was the high SSA in the marine boundary layer runs also associated with higher mass extinction efficiency? Was BC/total mass different? Rather than merely narrating the figure, it appears that the authors wish to explain the observations. Hence they should present a wider range of possibilities considered.

We thank the reviewer for asking this question. Upon close examination, the low altitude runs over sea in B362, B365 and B374 correspond to the extreme outliers in figure 2 with a considerably elevated mass extinction efficiency (dry and ambient). In the case of B362 and B365, the nitrate mass/total mass increases dramatically. For B362 over the North sea the total mass also increases dramatically, whilst for B365 over the southern Baltic, the total mass increase is more modest. Since nitrate aerosol is both highly scattering and highly hygroscopic we might expect both an elevated mass extinction efficiency and an enhanced difference between dry and ambient aerosol optical properties, as is shown in our results. In the case of B374, the departure from the line of best fit is less extreme and although the mass increases somewhat compared to the other runs, it is the organic fraction that increases rather than the nitrate component. As organics are both less scattering and less hygroscopic than nitrates, this is perhaps why the departure is less extreme in this case. We have added some of this discussion to the appropriate section.

Line 11 page 18498 Why not include this broader result in the abstract for higher visibility?

We thank the reviewer for this suggestion. This result has been added to the abstract.

Section 4, line 4, page 18499 – I do not understand what is meant by a “hydrophobic effect on aerosol”

We thank the reviewer for pointing out our clumsy wording in this sentence. The sentence has now been altered to read “Black carbon, on the other hand, is hydrophobic in its pure form and is generally understood to reduce the hygroscopicity of aerosol mixtures in which it is a significant component. “

Line 5, 18500: I assume the nephs were operated in parallel. How was the aerosol humidified in the wet nephelometer? Are losses due to the humidification responsible for the low bias in figure 6. What was the timescale over which the humidity was varied?

The nephelometers were operated in series, with the sample being humidified typically over a 10 minute period. For individual scans, the data shows some variability, therefore the recommendation is that 2s averages are produced as shown in figure 6, and in addition that several scans are

averaged together to give a more representative growth factor. Particle loss between the dry neph and the wet neph is potentially responsible for the low bias in figure 6. Tests on a number of flights during EUCAARI suggest that this is a maximum 10% loss in scattering. An alternative or additional possibility is that there is a problem with the zeroing of one or other nephelometer. A final consideration is that the original TSI sensors over-estimate RH by up to 15% RH at high RH (Fierz-Schmidhauser et al. 2010). In order to produce figure 7, the binned wet neph data for which dry neph RH < 30% was fit to the Model 2 formulation of Kotchenruther et al (1999). The degree to which this fit deviated from  $f(\text{RH})=1$  at 30% RH was assessed and used to scale the growth curves presented in figure 7. In all cases except for B374, the scaling needed was less than 10% which is consistent with independent estimates of scattering loss estimated from time periods when the two nephs were measuring the same RH. Therefore this approach is deemed valid. We now show the impact of this procedure in a new version of figure 6 which uses flights spanning the range of growth curves seen.

Figures:

Add the measurement wavelength to the captions for all relevant figures. **Done**

Figure 4 and discussion: it appears that only one ground station was below any vertical profiles. Given the relatively large variability in AOD spatially, what is the value of the comparisons to ground stations not co-located with the profile.

The referees assessment of the poor match with the ground stations is correct. **As discussed above we have removed this comparison and left only a brief discussion of the likely accuracy of AOD profiles estimated by this method – augmented by a discussion of the contribution of absorption and BC in particular as also suggested by the reviewer.**

Figure 5: Please expand these profiles figures to include more information. I suggest including total aerosol mass and BC mass profiles. If there is space, it would not be unreasonable to include a tracer such as CO to give more context to the reader. I understand that these are only 4 out of many profiles that contributed to the whole paper, but feel that a little context is better than none.

We agree that more context would be useful. We have included PCASP number concentration to show the relationship to scattering and humidity, and CO as a tracer. We have added appropriate discussion to the text to include these new variables.

Figure 6 and 7. Please comment on the discrepancy in  $f(\text{RH})$  measured dry (<30%) in figure 6:  $f(\text{RH})$  should be 1 at matching RH but is 1 at ~50% (note that the uncertainty in RH in each neph is only 5% for non-extreme values). Earlier in the text there was a comment about correcting wet neph data but it is not clear that it's appropriate here, as only dry (<30%) air was selected. Please clarify the source of the disagreement and state the range of error in RH measurement in the wet and dry portions of the neph measurement.

See discussion above for potential reasons why  $f(\text{RH})$  isn't 1 at low RH. In fact B374 is an outlier as the dry neph RH fell below 30% for only a few short segments of time and there is a concern that  $f(\text{RH})$  curves here are not valid. Therefore we have changed figure 6 to use two different flights that are more characteristic of the flights as a whole.

In Figure 7 it is clear that the curve for flight 374 does approach 1 at low RH, but this is non consistent with figure 6. The value at 90% also doesn't match.

This mismatch is due to the correction procedure for particle loss described in section 4 which has been performed on data in figure 7 but not on figure 6. B374 is an extreme example of correction needed as described above. The new figure 6 uses two different flights and the mismatch is much more modest. We have made it clearer in the text and the caption as to where raw data is used and where correction for hypothesised particle loss has been applied.

Figure 7 indicates intra-flight variability, but I suggest including a second flight in figure 6 to show this variability in the un-averaged data. Thus I suggest including flight B367 in figure 6. Done.

Figure 8: please enlarge the font used in the figure. Done

Additional changes made:

During investigation of some of the points concerning BC load which were raised by the reviewer, it became apparent that some of the data points in figures 2 and 8 were invalid due to an assumption about the BC mass in the absence of sufficient SP2 data to produce an average for some runs and flights. We apologise for this oversight. **In order to be scientifically rigorous, we have removed these points from revised versions of figures 2 and 8, and recalculated all quantities derived from these graphs. The number of data points is reduced, however, the conclusions are more defensible than before, and the agreement between measured and modelled absorption is actually somewhat better than when the data with an erroneous mass was used.**

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