Scattering and absorption by Aerosols during EUCAARI-LONGREX: can airborne measurements and models agree?

Author response to referee 2

We thank the referee for their helpful comments and recognise that they appreciate that the paper "tackles a highly relevant topic" and "makes a significant contribution to the topic of aerosol-radiation interactions and deserves publication in ACP". The majority of the revisions that they suggest address the concern that there is a mismatch between the broad title of the paper and the content which focusses on one platform from the EUCAARI-LONGREX campaign. The referee suggests that either the title of the paper be changed, or that the scope of the data used be widened. Considering this comment alongside those of referee 1, and the number of other papers already published or in preparation that detail the measurements from other instrument platforms, we take the view that it is more appropriate for this paper to consider the BAe146 measurements mainly, with more limited references to other platforms and therefore we have changed the title of the paper to:

"Aerosol scattering and absorption during the EUCAARI-LONGREX flights of the Facility for Airborne Measurement (FAAM) BAe146 : can measurements and models agree?"

The referee rightly makes the points that discussing closure with absorption measurements that have a 50% uncertainty is difficult, and that absorption measurements with better accuracy are available, although not from the BAe146 platform. We agree with both points, and have re-emphasised this throughout the results and conclusion sections. However, the referee recommends several papers that could be used as a benchmark for the results from FAAM measurements. We now provide a short summary of their relevance to the current work.

Fiebig et al (2002) does indeed measure absorption with greater accuracy than is current on the BAe146. However, it does not have black carbon (BC) mass measurements and uses absorption measurements to infer the soot volume. This is similar to the methodology that we have used previously in e.g. Osborne et al (2007) and Cook et al (2007), however, as we state clearly in the introduction of the paper – the approach we take is different in that we are independently measuring the BC mass and then assessing the degree of closure that can be achieved. We do not have the BC mass as a "free" parameter with which we can achieve closure. Thus, although Fiebig et al (2002) is a useful addition to our introduction, it is not suitable as a benchmark.

Petzold et al (2002) used filter samples and independent measurements of scattering and absorption but also varied the BC mass fraction to produce imaginary part of the refractive index which allowed a match with the measured absorption. The absorption and scattering from several types of measurements, and from using Mie calculations with the inferred refractive index resulted in an uncertainty of around 30% in aerosol optical properties. For scattering we obtain a similar level of uncertainty using our approach. However, our uncertainty in the absorption is around 40% due to measurement uncertainties and variability in SLRs. The uncertainty in absorption measurements is known to be a function of organic mass fraction (e.g. Lack et al, 2008) and we have very high OC mass fractions in our particular aerosol. Thus we believe that our estimates of uncertainty are valid. Additionally, as with the discussion above, our methodology for closure is different in that we use

the independent BC mass. We do not expect to get as good closure as those studies where the BC amount is varied to match the single scattering albedo.

Schmid et al (2006) find that remote sensing measurements of extinction tend to give higher values than in-situ ones, and that airborne nephelometer and PSAP measurements tend to be biased low compared to airborne sunphotometers. They conclude however that there is no definitive proof that anyone of the methods is fundamentally flawed. Systematic errors in extinction co-efficient are 15-20% at visible wavelengths, and random errors range from 26% to 98%. We believe our error estimates are not inconsistent with these values. We will add discussion of this paper to our uncertainty discussion.

Weinzierl et al (2009) considers a very different type of aerosol, Saharan dust and we do not feel that it is relevant to this paper.

We have added discussion of these important papers to the introduction and discussion sections.

Specific comments

Section 2.1 The overall meteorological situation is described extensively by Hamburger et al (2011). Please refer to this paper when discussing meteorological situations.

We do in fact already refer to this paper (albeit a previous version of the manuscript) when discussing the meteorology in section 2.1. We will correct the reference and expand the discussion of this paper in a revised version.

Section 2.2.1 Mass extinction is inferred from AMS data for the non-refractory components and from SP2 data for absorbing components. This approach misses any non-absorbing or weakly absorbing refractory particles like sea-salt or dust. Please discuss the expected uncertainties. Sea salt is of particular importance because some of the selected flights were conducted over sea.

Only 4 SLRs in the total data set used in this paper are at low level over ocean thus having the potential to be substantially affected by sea salt. Due to the meteorological situation (persistent high pressure), the wind speeds were in fact relatively low, suggesting less problem with sea salt than in other situations. However, there is some indication that the mass specific extinction for these low level “marine” runs is higher than for other runs, but in 2 cases the nitrate mass fraction is substantially increased, and in one the organic mass fraction is increased. We now discuss this in the paper.

For a potential different approach see the inversion scheme developed by Petzold et al (2009) for the combination of airborne PSAP and size distribution data using Mie theory.

We believe the reviewer wishes to point out that other, potentially more accurate approaches are possible with different combinations of instrumentation. Petzold et al (2009) refers to measurements of the optical properties of mineral dust and demonstrates nicely the use of a three wavelength absorption measurement and how this can be used to provide extra constraints on the refractive indices. However, that study takes a different approach to ours in that it does not use the chemical composition, for reasons explained in that paper. It is true that we could have used a similar approach using just one absorption measurement and that this might have accounted better

for the aerosol components that we cannot measure. However, it is our belief that for most of the cases considered here, the likely component of mineral dust or sea salt is small. We have added discussion of this to the paper in response to this comment, and a similar request from reviewer 1.

Further more a brief description of the inversion of particle size distribution data including resulting uncertainties is required.

THE PCASP is an optical particle counter. Each individual particle that enters the instrument is exposed to a laser beam and produces a scattered peak. The height and width of this peak is recorded, and the signal is binned based on the peak height. The inversion of scattering peak to size uses a calibration curve derived from Mie theory for calibrated spheres of known size and known refractive index. Very recent work by Rosenberg et al (2012) demonstrates that the bin edges can differ by an average of 13% and up to 30% from that provided by the manufacturer due to systematic changes, and the effect of differing refractive index of ambient aerosol compared to the calibration standards. The PCASP was calibrated in the laboratory on return from EUCAARI and bin widths used here are based on that calibration, however the refractive index used was of the calibration standard rather than that of the ambient aerosol. The refractive index correction becomes more important for aerosol with significant absorption. Based on the low mass of BC observed in this aerosol, we anticipate that our uncertainty in the size is less than 30%. Using a change in 13% of bin sizes on a limited number of runs, changes seen in modelled scattering were less than 5% and changes in absorption a maximum of 10%. These uncertainties are smaller than both the uncertainty in measured scattering and absorption due to variability within a SLR, and the estimated measurement uncertainty of both the PSAP and the nephelometer. We will add a brief statement about this in section 2 of the revised paper.

Please refer to Hamburger et al (2011) instead of McMeeking et al (2010) for the instrumentation of the DLR Falcon.

We intended to refer to the description of the intercomparison flight which is mentioned in both papers. In fact, in response to comments from referee 1, the relevant line has been removed and so this reference is no longer needed.

Section 2.2.3 and Section 3.2. I am surprised to see absorption contributions to extinction of 1% even over polluted regions. This is in contrast to other observations during EUCAARI. I suggest a discussion of the low level absorption in the presented data.

It is indeed interesting that the absorption contribution to optical depth (or extinction) is low. However, we have recalculated the contribution based on the single scattering albedos from the SLRs and find it to be closer to 3-9%. We apologise for the earlier error. We have checked the mass of BC used in our calculations against the original data reported in McMeeking et al (2010) and find that they are consistent. In addition, the BC mass fraction is similar to that found in the polluted environment of the Po Valley using the methodology similar to Fiebig et al (2002) and others (Cook et al, 2007). Interestingly, from a closer examination of Hamburger et al (2011), comparisons of BAE146 BC measurements with those measured at ground based stations suggest that the 146 measurements may be slightly low compared to the ground measurements. It is also true that the 146 on most flights did not sample the atmospheric regions forecast by the Flexpart model to contain the peak BC amounts, and therefore it is possible that there is less BC in these

measurements than in other measurements during EUCAARI. Section 2.3 and 5 in the revised version will have this calculation corrected and discussed further.

Furthermore, I suggest presenting observational data as median, 25 percentile and 75 percentile instead of average and standard deviation. This is particularly valid for the campaign average of the single-scattering albedo which is given as 0.93 +/- 0.37

This is an interesting idea, and it would be interesting to consider the in-run variability in this way. However, in considering this possibility it became apparent that the uncertainty quoted for the single-scattering albedo is calculated incorrectly. By considering the nature of the measurements and sources of uncertainty, and combining these properly, the campaign average is 0.93 +/- 0.02, which is much more in line with previous estimates of uncertainty and perhaps removes the necessity for altering the way in which we have quoted our results. Additionally, the uncertainties shown in the figures are a combination of measurement uncertainty and variability along a straight and level run and this is best achieved using standard deviation as a measure of SLR variability.

Section 5. Subsection 5.1 can be omitted because there is no subsection 5.2

We thank the reviewer for pointing this out.

AS discussed previously, DLR Falcon HSRL data for aerosol extinction should be used for comparison.
AS discussed above, we believe that the focus of this paper should be the measurements from the BAe146 platform and therefore large-scale comparison with the DLR Falcon HSRL data is beyond the scope of the paper.

The conclusions drawn from the highly uncertain absorption data should be discussed also in comparison to other observations because an uncertainty of 50% for absorption is not the ultimate limit achieveable with current instrumentation.

We are aware that uncertainty of 50% absorption is not the ultimate limit for current instrumentation. However, it is the current limit for this instrumentation and in this measurement campaign on this platform. Having changed the title of the paper, we hope this is clearer. The revised conclusion will include a revised statement making a clearer distinction between the uncertainty achieved with this combination of instrumentation and methodology, and other existing and potential future approaches. We can also report that changes will be made to the absorption measurement on the 146 in the near future, and that the accuracy of absorption measurements should be substantially improved.

Figure 9. Please add error bars to the data points.

Rather than add error bars (which can be seen in any case in Figure 8) we have included two flights in separate figures as well as some additional work on PSAP measurement uncertainties that have modified our conclusions.

Table 2, Please also reference the work by Virkkula et al (2005, 2010) on PSAP inversion.

Table 2 referenced only the techniques used in this study, which did not include that mentioned by the referee. In the revised version of the paper, this table has been removed as instrumentation and analysis details are in the main text.

References

Fiebig, M., A. Petzold, U. Wandinger, M. Wendisch, C. Kiemle, A. Stifter, M. Ebert, T. Rother, U. Leiterer (2002) Optical closure for an aerosol column: method, accuracy, and inferable properties applied to a biomass-burning aerosol and its radiative forcing. J. Geophys. Res., 107, doi:10.1029/2000JD000192

Hamburger, T., McMeeking, G., Miniken, A., Birmili, W., Dall'Osto, M., Flentje, H., Henzing, B., Junninen, H., Kristensson, A., de Leeuw, G., Stohl, A., Coe, H., Krejci, R., and Petzold, A.: Overview of the synoptic and pollution situation over Europe during the EUCAARI-LONGREX field campaign. Atmos.Chem.Phys.,11 1065-1082. Doi:10.5194/acp-11-1065-2011

McMeeking, G.R., T. Hamburger, d. Liu, M. Flynn, W.T. Morgan, M. Northway, E.J. Highwood, R. Krejci, J.D. Allan, A. Minikin and H. Coe (2010): Black carbon measurements in the boundary layer over western and northern Europe, Atmos. Chem. Phys, 10, 9393-9414

Petzold, A. m. Fiebig, H. Flentje, A. Keil, U. Leitner, F. Schroder, A. Stifter, M. Wendisch, P. Wendling (2002) Vertical variability of aerosol properties observed at a continental site during the Lindenberg Aerosol Characterisation Experiment (LACE 98) j. Geophys. Res., 107, 8128, doi:10.1029/2001JD001043

Petzold, A., K. Rasp, B. Weinzierl, M., Esselborn, T. Hamburger, A. Dornbrack, K. Kandler, L. Schutz, P. Knippertz, M. Fiebig and A. Virkkula (2009) Saharan dust absorption and refractive index from aircraft based observations during SAMUM 2006, Tellus, 61B, 118-130

Rosenberg, P.D., A.R. Dean, P.I. Williams, A. Minikin, M.A. Pickering and A. Petzold (2012) Particle sizing calibration with refractive index correction for light scattering optical particle counters and impacts upon PCASP and CDP data collected during the Fennec campaign, Atmos. Meas. Tech. Discuss, 5, 97-135

Schmid, B., R. Ferrare, C. Flynn, R. Elleman, D. Covert, A. Strawa, E. Welton, D. Turner, H. Jonsson, J. Redemann, J. Eilers, K. Ricci, A.G. Hallar, M. Clayton, J. Michalsky, A. Smirnov, B. Holben, J. Barnard (2006) How well do state-of-the-art techniques measureing the vertical profile of tropospheric aerosol extinction compare? J. Geophys. Res., 111, D05S07, doi:10.1029/2005JD005837

Weinzierl, B., A. Petzold, M. Essselborn, M. Wirth, K. Rasp, K. Kandler, L. Schutz, P. Koepke, M. Fiebig (2009) Airborne measurements of dust layer properties, particle size distribution and mixing state of Saharan dust during SAMUM 2006. Tellus, 61B, 96-117