**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Enhancement of the aerosol direct radiative effect by semi-volatile aerosol components: airborne measurements in North-Western Europe" *by* W. T. Morgan et al.

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Firstly, we would like to convey our gratitude to the referee's for their helpful and encouraging comments on the manuscript. The following will detail our response to each referee's comments.

Anonymous Referee 1

The referee highlights areas where the quantitative detail of the manuscript could be improved. The following discussion will summarise the additional details that we feel address the referee's comments.

Inlet transmission: The manuscript states that the AMS inlet was not pressure controlled. Under these circumstances both the aerodynamic sizing and the inlet transmission will be a function of pressure. According to Appendix A, only the sizing was corrected. The statement that the mass measurement will not be affected "unless the lens performance is altered beyond the transmission envelope of the lens" is at best confusing. Since lens transmission is to first order controlled by the Stokes number and the Stokes number is pressure-dependent in the AMS inlet regime, the first-order response of the lens to pressure is that the entire transmission function will shift to smaller sizes at higher altitudes. The error in the mass measurement depends on how the size distribution overlaps the lens transmission.

We thank the reviewer for pointing out the confusing nature of the statement in Appendix A and we will clarify this in the revised version of the manuscript. We will discuss the relationship between the overlap of the size distribution and the lens transmission relating to the referee's final comment below.

Aerosol bounce: First, the collection efficiency factor used to reduce the AMS data must be stated. Different authors use varying factors. Second, collection of the moist, nitrate-rich particles at the top of the boundary layer is likely to be significantly higher than the drier particles with relatively more sulfate at the surface. Since the AMS collection efficiency for dry ammonium sulfate is 20 to 25% and is near 100% for moist ammonium nitrate, this could be significant. Organic coatings reduce but generally do not eliminate this spread. How was the collection efficiency assessed?

We stated in the manuscript on page 10660, line 15 that "Morgan et al. (2009b) included specific information regarding the quantification and analysis of the BAe-146 AMS data pertinent to this study". This included an in-depth discussion of the Collection Efficiency (CE) correction due to particle bounce that was applied to this dataset. We used the correction developed by Matthew et al. (2008) to estimate the CE of our AMS, which in our case is strongly determined by the ammonium nitrate content present in the aerosol. The AMS data was then compared with the PCASP estimated

volume and for the LONGREX flying period the agreement was to within 30%, which is tolerable given the uncertainties in estimating volume from the PCASP and uncertainties relating to the AMS mass measurement and subsequent conversion to volume. We also did not observe a dependence between the PCASP/AMS ratio as a function of the ammonium nitrate content of the aerosol sample.

Note that Morgan et al., 2009b has now been published in ACP as Morgan et al., 2010.

p. 10662 lines 5+: Did the size distributions show that the humidified particles were still small enough for the Anderson and Ogren nephelometer truncation correction to be valid? Also, the nephelometer truncation error is slightly sensitive to the assumed refractive index, and water uptake will lower the refractive index (Massoli et al., AST, 42, 1064, 2009).

The PCASP measurements indicated that the peak in the volume size distribution was located at approximately 300nm for B379 (B366 was not available), with the majority of the sub-micron volume located below 600nm. Potentially some of the larger particles at the high end of the distribution could grow above 1 micron at high relative humidity (>85%), which would impact the humidified nephelometer corrections. However, we also compared the ratio between the "dry" and "humidified" nephelometers without applying the truncation corrections and found negligible differences when comparing the f(RH) values at high relative humidity. Therefore we have not included a correction for this but we will highlight this uncertainty in the revised manuscript.

We thank the referee for pointing out the Massoli et al. (2009) reference and will include this in the revised manuscript. As noted in Massoli et al., water uptake will lower the refractive index of the aerosol sample and introduce a error in the f(RH) determination when using the nephelometer truncation corrections. However, this error is small (2%) and within the experimental error for the system. Furthermore, we would need to estimate the refractive index of the dried ambient aerosol which in itself is highly uncertain. Therefore we have not included a correction for the refractive index

change.

Anonymous Referee 2

1. Gas-aerosol partitioning of nitrate and organics The vertical profiles of total nitrate (gaseous HNO3 + aerosol nitrate) and organics (volatile organic compounds (VOCs) + aerosol organics) are unknown with the lack of the data of HNO3 and VOCs. Therefore, there are no observational evidences of gas-aerosol partitioning of nitrate and organics (temperature and RH are indirect evidences). The discussion of the gas-aerosol partitioning should be toned down in the manuscript.

While we agree with the referee that we have no direct evidence for gas-particle partitioning of ammonium nitrate and organics from their gaseous precursors, we have argued in the manuscript that our observations are consistent with this phenomenon.

Firstly, we observe clear increased concentrations in sub-micron mass and volume as a function of altitude in the vicinity of Cabauw and such increases are well above instrument measurement uncertainties. Secondly, the major feature of the increased aerosol mass is a substantial increase in the ammonium nitrate concentration along with a more modest increase in the organic mass. Thirdly, we see clearly different aerosol compositions when comparing between the aircraft AMS and the ground-based AMS, with organic matter being the major component at the ground and ammonium nitrate being more dominant aloft. The chemical composition of the organic matter is also significantly different, with the semi-volatile component being enhanced aloft. Fourthly, the peak in concentrations at the top of the boundary layer is repeatedly observed across the region and not just in the vicinity of Cabauw. For example, in Figs. 5b and 5d, increased concentrations are observed during continuous profiles between 1000m and 2500m during the along wind transects across Northern Germany to the Cabauw site.

We have highlighted in the discussion (section 6.1) that there are likely inhomogeneities

in the aerosol field in the vicinity of Cabauw, as we observed a slight increase in sulphate when comparing the airborne data with the ground-based data. We used the sulphate profile to calculate a normalised nitrate and organic profile in order to investigate this and found that the nitrate increase was well in excess of this normalised estimate, although the organic matter profile was quite similar.

We also showed in figures 3b and 3c that the thermodynamic profile of the boundary layer during the measurement periods was well mixed and that we could expect mixing throughout the boundary layer. This coupled with our efforts to account for concentration gradients in the horizontal suggests that a major factor that would have driven the enhanced mass would be partitioning of gaseous precursors (specifically ammonia and nitric acid) to the particle phase at reduced temperatures and enhanced relative humidity typical of the top of the boundary layer.

We will modify the manuscript in order to satisfy the referee's concerns and limit the discussion of gas-particle partitioning to the discussion in section 6.1 (where our lines of evidence are set out) and then reiterate that our observations are consistent with and primarily attributed to partitioning of gas phase precursors to the particle phase in the abstract and conclusion.

2. Mie calculation Because the authors assume volume-mixing refractive indices, the absorption coefficient is probably overestimated, resulting in the underestimations of scattering coefficient and single scattering albedo in the Mie calculation. In addition, the treatment of same chemical composition and mixing states (well-mixed internally) for all particles is quantitatively problematic (especially for absorption coefficient and single scattering albedo) [e.g., Oshima et al., JGR, 2009; Zaveri et al., JGR, in press]. In section 5.3, the authors conduct the sensitivity calculation of water uptake by ammonium nitrate (NH_4NO_3) using the Mie calculations. However, the agreement between measurements and the Mie calculations is not so good (section 5.3). Since the two nephelometers were used (dry and ambient), the effect of water uptake by NH_4NO_3 (majority of total dry aerosol) can be derived solely from these measurements with-

out making sensitivity calculations. The importance of the Mie calculations should be explained.

We thank the referee for identifying that this section needs more explanation and we will do so in the revised manuscript. Also, the Oshima et al. (2009) reference is useful and we will add this to our discussion of the uncertainties inherent in our assumptions (we will also examine the Zaveri et al. paper once it has been published).

While we realise that the assumptions relating to the Mie calculations are idealised and that the closure for the ambient calculations is poor at high relative humidity, we have included them in order to explore the theoretical sensitivity of the increase in scattering associated with enhanced ammonium nitrate content and the scattering due to additional water uptake. The f(RH) calculated from the nephelometers does not provide us with this information as we cannot separate the contribution of the additional dry mass from the water uptake. This is possible using the Mie calculations. From these calculations we are able to show that changes in the mass fraction of ammonium nitrate can dramatically increase the water uptake by the aerosol, which underlines the importance of our observations.

We will expand on our reasoning for undertaking the Mie calculations in the revised manuscript in order to clarify their purpose.

3. Consistency between aerosol mass (section 4) and its optical property (section 5) In section 4, the authors discuss the increase in NH_4NO_3 with height within PBL using the vertical profile of B366 (Figure 6a). On the other hand, in section 5, the scattering coefficient does not so increase with height (Figure 9a). From the viewpoint of aerosol mass-optical closure, the consistency between Figures 6a and 9a should be explained in the manuscript. In addition, the authors mainly focus on B366 (the vertical profile of B379 is not shown) in section 4, while they use both B366 and B379 in section 5. Especially, B379 is quite important in terms of the increase in scattering coefficient with height within PBL. Therefore, to discuss the results of B379 in section 5, the vertical

profile of aerosol mass concentration of B379 should be also shown in section 4, even though the vertical coverage is not enough.

We thank the referee for highlighting that this area requires some clarification and we will do this in the revised manuscript. Firstly, the nature of the airborne operations and the differing sampling times for the nephelometer (1s) and the aircraft AMS (30s) are the major reason for the appearance of inconsistencies when comparing the optical and mass measurements in the vertical domain. The altitude track of the aircraft (see Figs. 5b and 5d) was made up of a series of Straight and Level Runs (SLRs) at constant altitude interspersed with short (less than 2 minute) vertical profiles to another constant altitude. This often meant that the AMS data from the profiles was unsuitable as the number of data-points was not sufficient to fully explore the vertical distribution (especially as the start and stop times of an individual measurement were not synchronised with the start and end of profile ascents or descents). Consequently, we chose to use the SLRs and plot these as a function of altitude, which is what is shown in Fig. 6a. The nephelometer on the other hand does not have such problems with vertical profiles due to its greater time resolution. It is more prudent to use the vertical profile data for the optical property comparison with the ground as this will minimise the impact of horizontal gradients in the aerosol field in the vicinity of Cabauw. Furthermore, plotting the nephelometer data for the SLRs, i.e. in the same way as how the AMS data is shown, displays a very similar trend to the AMS data due to their strong coupling.

We noted on page 10670, lines 10-17 that the enhancement for B366 was more modest between 1500-2000m and that this was due to some differences in the thermodynamic structure observed by the aircraft and the radiosonde at this height level. The scattering profile is enhanced relative to the ground and this can be clearly seen at the median percentile between 500-1500m. The enhancement is not as marked compared with B379 due to the reduced RH for B366, which was noted in the manuscript on page 10677, lines 1-9.

In order to demonstrate that the aerosol mass concentration and the aerosol scattering

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coefficient were strongly related, we included Fig. 8 as this includes the data from all of the flights discussed in the manuscript. This clearly shows that they are strongly coupled and that changes in total mass and scattering coefficient are typically a result of increased ammonium nitrate concentrations. It is this figure that links the profiles in Figs. 6a and 9a. We will highlight this in the revised manuscript.

The above points are the reason we did not show the vertical distribution of aerosol mass concentration from B379 as it had a low number of SLRs within the boundary layer close to Cabauw, compared with B366. The SLRs for B379 were at relatively lower altitude (approximately 500m and 1300m) compared with the top of the boundary layer (approximately 2000m) where the peak scattering was observed. The nitrate concentration was increased relative to the ground in this instance also, with concentrations at the ground from 1100-1200 UTC ranging from 2.8-3.4 μg/sm3. The aircraft AMS measured median nitrate concentrations of 6.9 μg/sm3 (500m) and 7.3 μg/sm3 (1300m). We will include these numbers in the revised manuscript but we will not include them in a figure due to the low vertical coverage.

4. Consistency between surface and aircraft measurements The consistency between surface and aircraft (lowest altitude) measurements should be discussed quantitatively in the manuscript. In Figures 6 and 9, the values of groundbased measurements and aircraft measurements around 500 − 1000m are very different. It is not clear whether this difference is fully from the increase in NH4NO3 with height or due to the uncertainties in the measurements (surface and aircraft measurements). This point influences the whole results presented in section 5.

We thank the reviewer for highlighting this shortcoming in the discussion of the results and we will include a discussion of this in the revised manuscript. Unfortunately no direct side-by-side comparison of the instruments was possible during the campaign but we are confident that the observations are not purely a result of instrument uncertainties/errors.

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Regarding the AMS measurements, if we assume that the sulphate profile in Fig. 6a should be relatively constant when comparing between the ground and below 1000m then the concentration values are not markedly different. The sulphate concentration at the ground was 1.02 μg/sm3, while the median concentration at 700m was 1.33 μg/sm3. Such concentrations are within the uncertainty limits for AMS mass concentrations (approximately 25%, Canagaratna et al., 2007). Consequently, the observed increases in nitrate, ammonium and organics aloft are well above the AMS instrument uncertainties. Furthermore, we observe clear vertical trends in the airborne AMS data which we would expect to be robust i.e. comparing trends from a single instrument. The ground-based AMS data is useful as it provides an additional data-point at the surface where flight restrictions meant we could not fly.

Regarding the nephelometer measurements, it is important to remember that the profiles shown in Fig. 9 are for the ambient scattering coefficient, hence there is a change in relative humidity with height. For B366, the ambient RH at 1000m is approximately 40%, while for B379, the ambient RH at 1000m is approximately 60%. Thus there is a stronger divergence between the airborne measurements below 1000m and those at the ground. In the dry condition, the nephelometers have an estimated instrument uncertainty of approximately 10% (Anderson et al., 1996). The changes in dry scattering coefficient below 1000m are typically above this uncertainty (some points fall within this 10% band for B366 due to variability in the vertical domain) and are consistent with the observed increases in dry mass observed by the AMS.

To summarise, we are confident that the observed changes are well above instrument uncertainty when comparing between the ground and the aircraft and we will make this point clearer in the revised manuscript.

5. AMS instrument (P10660, Line 10) The information of measured size range should be given.

This will be given in the revised manuscript.

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6. Nephelometer measurement at Cabauw (P10662, Line 11) It should be clarified that the Nephelometer at Cabauw measured aerosol scattering coefficient on dry condition.

This will be clarified in the revised manuscript.

7. Vertical profile of SV-OOA (P10667, Line 21) If the gas-aerosol partitioning of SV-OOA is important, it should be directly shown in Figure 6a (not using LV-OOA mass fraction).

We will add the SV-OOA mass fraction profile to the figure in the revised manuscript.

8. Number of data points of individual altitudes in Figure 6a The number of the data points should be given for individual altitudes. Especially, it is important to show the number of the data points between 1700 – 2200 m (the layer of nitrate enhancement) to confirm statistical reliability.

We will include this information in the revised manuscript in Fig. 6.

9. Number concentrations at surface (L10669, Line 1) Is the aerosol number concentration derived from AMS measurement at ground-based site? The procedure to derive the number concentration should be mentioned.

We did not derive the aerosol number concentration from the AMS ground-based measurements. Point 2 of the AOD estimation procedure merely assumes that the number, mass and chemical composition measured at the ground is representative of the entire depth of the well-mixed boundary layer. We know from the airborne measurements that this is not the case as we have demonstrated that the mass and composition of the aerosol changes with altitude. We use the ground-based measurements to predict what the AOD would be if only water was added to the particulate burden i.e. there is no additional condensation of semi-volatile material at reduced temperature and enhanced relative humidity.

10. Caption of Figure 9 It should be mentioned in the caption of Figure 9 that the scale of x-axis (Figure 9b) is different between aircraft and lidar measurements.

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This will be included in the revised manuscript.

11. P10672, Line 3: "with agreement to within 30% compared with the measured values" It is not clear that which parameter is discussed in this sentence (extinction, scattering, absorption coefficients, or others).

The sentence refers to the dry scattering coefficient. This will be clarified in the revised manuscript.

12. Fig. 5b should be changed to Fig. 5c. (P10667, Line 2)

We thank the referee for noting this error and we will correct this in the revised manuscript.

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