

Response to Reviewer #3

The authors of the present manuscript acknowledge the reviewers for carefully reading and providing constructive comments that have led to an improved paper. Responses are written in blue text.

1. Page 4, Line 23. What is the temporal resolution of N_a and aerosol optical properties used in this study?

Response: The temporal resolution is one minute. This information has been added to section 2.1.1: “The time resolution of the N_a , σ_s , and σ_a measurements is one minute.”

2. Page 6, Line 25. LTS is calculated at 1-min resolution?

Response: The temporal resolution is one hour. This information has been added to section 2.3: “The European Centre for Medium-Range Weather Forecasts model runs for ARM analysis with a one-hour resolution for a $0.56^\circ \times 0.56^\circ$ box centered on the site provided values of ω and potential temperature.”

3. Page 6, Line 29. Please specify how to collocate the datasets of different time resolutions (e.g. ACSM, AOS, LTS, large-scale vertical velocity and cloud properties) for the comparisons later on in the manuscript, particularly shown in Figure 10 & 11. And the final temporal resolution for collocated data.

Response: Most of the datasets, i.e., aerosol properties (scattering coefficients and number concentrations) from the AOS, cloud properties (COD, LWP, DER), and surface meteorology, have a 1-min temporal resolution. These data were first matched according to the observation time, and then matched with aerosol composition measurements and ECMWF simulations (LTS and large-scale vertical velocity) and integrated over 1-min time intervals. This means that the aerosol composition and LTS (vertical velocity) in the 1-min resolution datasets are assumed to remain constant within 30-min and 1-hour time periods, respectively.

We have added the sentence “To investigate the influence of aerosols on cloud properties, aerosol properties (N_a , σ_s , composition), cloud properties (COD, LWP, DER, boundary-layer height), surface meteorological parameters, and ECMWF simulations (LTS, large-scale vertical velocity) were matched according to observation time and averaged and interpolated over 1-min time intervals. “ to the revised manuscript.

4. Page 7, Line 23-25. For Spring season, AE values generally lower than other season, especially for April and May as shown in Figure 3 and Table 1. Also, σ_1/σ_{10} value is lowest in Spring. Which indicates aerosol plumes more enriched by larger particles, relatively. Please provide more evidences or paper citations to support the statement “due to the presence of a great number of smaller particles. . .”.

Response: We found that this statement is not fully supported by the current dataset and analysis. We have thus removed the sentence “The largest values of N_a corresponding to moderate values of σ_s are found in spring and are likely due to the presence of a greater number of smaller particles with less optical sensitivity.”

5. Page 8, Line 2-6. Please specify the exact months in this argument, and how to conclude that “This indicates that strong surface. . .”

Response: We have added the specific months to the sentence: “Months in summer and winter with the strongest mean surface wind speeds (e.g., June and January/February, respectively) ...”. The next sentence is not fully supported by the current dataset and analysis, so we have removed the “This indicates that strong surface ...” sentence.

6. Page 8, Line 25. Why only data of July and August 2012 are shown? How about particle size distribution in Spring and Autumn, since they are argued in section 3.1.1 as having discrepancies between Na and σ due to particle size distributions.

Response: Here, we intended to examine the relationship between particle size and particle chemical composition. However, data were not available in spring and autumn.

7. Page 9, Line 1. Please specify the bin sizes used for low and high AI condition. Is there any reason for the mismatched bins between those two conditions, as shown in Figure 7?

Response: We have added the sentence “The cloud properties were averaged over each 6-K LTS bin from 0 K to 30 K under low and high scattering AI conditions.”

The x-axis represents the mean values of LTS in each LTS bin. This is the reason for the mismatched bins between the two conditions.

8. Page 10, Line 8. “positive correlation each other” should be “positive correlation between each other”.

Response: The sentence has been changed to “The changes in DER with LTS possibly reflect the changes in LWP with LTS due to the high positive correlation between LWP and DER (Zhang et al., 2011; Sporre et al., 2014).”

9. Page 11, Line 30. “a narrower PDF a distinct peak” should be “a narrower PDF with distinct peak”.

Response: The sentence has been changed to “The high aerosol loading cases, conversely, have narrower PDFs with distinct peaks between 60 and 70 g m^{-2} .”

10. Page 12, Line 23-24. How about FIE under cluster III which has occurrence of 21.6%, and how to determine samples are not enough under cluster II.

Response: The air-mass clusters were determined for each day during the observation period. Although 15.9% and 21.6% of the daily trajectories belong to the clusters II and III, the number of combined cloud and aerosol samples passing the screening criteria (as described in section 2.2.2) are insufficient to do such an analysis. Furthermore, only cases with sample numbers greater than 50 and with calculated values of FIE that are statistically significant at the 95% confidence level ($P = 0.05$) are discussed in the study.

We have added the sentence “Only those cases with sample numbers greater than 50 per bin and where the calculated values of FIE are statistically significant at the 95% confidence level ($P = 0.05$) are analyzed here.” to the revised manuscript.

11. Page 13, Line 5-9. For ground-based assessments of FIE, Kim et al. (2008) and McComiskey et al. (2009) found decrease of FIE with LWP due to enhanced collision coalescence, please provide

the information of cloud droplet number concentration to support the statement “more droplets can get activated”.

Response: Conflicting findings regarding the dependence of the FIE on the LWP have been reported, i.e., a positive correlation in some studies (Pandithurai et al., 2009; Harikishan et al., 2016), a negative correlation in others (Kim et al. 2008; McComiskey et al., 2009; Liu et al., 2016), and an independence of the FIE on the LWP (Lihavainen et al., 2010; Zhao et al., 2012). Different mechanisms have been reported explaining the negative correlation (i.e., a decrease in cloud droplets due to enhanced collision-coalescence) and the positive correlation (i.e., an increase in cloud droplets due to enhanced aerosol activation). Thus, the dependence of the FIE on the LWP likely depends on which mechanism dominates during the study period in question.

Unfortunately, cloud droplet number concentrations were not available during the field campaign, but our results are consistent with some previous studies. The latter mechanism mentioned above possibly plays a dominant role.