

Anonymous Referee #3

We would like to thank Reviewer #3 for her/his time devoted and the constructive and helpful comments to which we will respond point by point.

The authors analyse a remarkable event of combined desert dust and biomass burning aerosol transport over Europe using observations and model results, and in the process evaluate some aspects of the CAMS air quality forecast system. Studying such events is important because of their quite drastic impact on the European air quality, potential implications on clouds and weather, and impairment of solar energy yield. Their distinct air quality signatures are good benchmarks for forecast systems. The subject is relevant and within the scope of ACP (there is some overlap with the GMD scope regarding the model evaluation aspect), the article is well organised and written so that I recommend publication after addressing the following comments: We thank the Reviewer for the general comment.

Page 1, line 11: Throughout the manuscript you use the correlation (coefficient) r , here you present the shared variance (and I assume you are referring to r^2). Because the term is less common or even used with different meaning, and to be consistent with the main text, I suggest to stick to r .

We agree with the comment and thus we have replaced “shared variance of 60%” with “correlation coefficient of 0.77” in the Revised Manuscript (RM) P1, L11.

Page 1, line 13: Please expand IFS here.

Done.

Section 2.1: Some more information about the aerosol representation in the model would be helpful: does the model assume the different aerosol types to be externally mixed, both regarding the aerosol optical properties and regarding any chemical or physical interactions between different components? This would be interesting to know especially since the event under consideration involves dust, biomass burning aerosol and sea salt.

We agree with the comment raised by the Reviewer. Indeed, the IFS aerosol types are treated as externally mixed (separate particles). The following has been included in the RM P4, L7: “The different IFS aerosol types are treated as externally mixed (Inness et al., 2019a).”

Page 4, line 8 and 9: The units should be micro metres not metres and it has to be mentioned that the numbers represent radii not diameters. What bins are used for the other aerosol types, e.g., what is BC1 vs. BC2?

We thank the Reviewer for the comment, to provide a more explanatory description. We have replaced “size bins” with “radius size bins” (m were already replaced with μm in the discussion version of the manuscript). The two bins used for organic matter and black carbon aerosols stand for hydrophobic and hydrophilic. We have modified the respective discussion in the RM P4, L15-18 as follows: “where ρ the air density, SS1 the sea salt radius size bin 1 (0.03-0.5 μm), SS2 the

sea salt radius size bin 2 (0.5-5 μm), DD1 the desert dust radius size bin 1 (0.03–0.55 μm), DD2 the desert dust radius size bin 2 (0.55–0.9 μm), DD3 the desert dust radius size bin 3 (0.9–20 μm), OM1 the hydrophobic organic matter, OM2 the hydrophilic organic matter, BC1 the hydrophobic black carbon, BC2 the hydrophilic black carbon and SU1 the aerosol sulfate (ECMWF, 2020).

Page 4, Eqs. (1) and (2): I understand these equations follow recommendations elsewhere, but could you indicate in the text where the factors come from? Also, SS3 is not considered for PM_{2.5} (and PM₁₀), because the radius is > 5 μm , but according to the intervals provided above, not considering DD3 for PM_{2.5} seems to ignore dust particles between 0.9 and 1.25 μm radius.

The factors applied in sea salt bins are used to transform sea salt from 80% relative humidity ambient conditions to dry, while the rest correspond to the fraction of each aerosol type included in PM₁₀ and PM_{2.5}. More information on the description and evaluation of the aerosol scheme used in IFS is provided by Remy et al. (2019) which we have included in the RM P4, L18-19 as follows: **“A detailed description and evaluation of the aerosol scheme used in IFS can be found in Remy et al. (2019).”** As for the disregarded DD3 radius size range 0.9-1.25 μm in PM_{2.5}, this is a limitation of the definition.

Page 5, line 10: Please expand LT once
Done. See P5, L16 of the RM.

Page 5, lines 28ff: Which selection criteria for the stations were used exactly?

Since here we examine the aerosol transport during Ophelia and the associated impacts on air quality in CAMS forecast systems, we only consider rural EMEP stations that fulfill the following criteria:

1. They are located over western Europe and away from the aerosol sources (i.e. Northern Africa and Iberian Peninsula).
2. They lie across the plumes of high AOD loadings during the examined event.
3. They exhibit remarkable increases in PM₁₀ and PM_{2.5} surface concentrations (visual inspection).

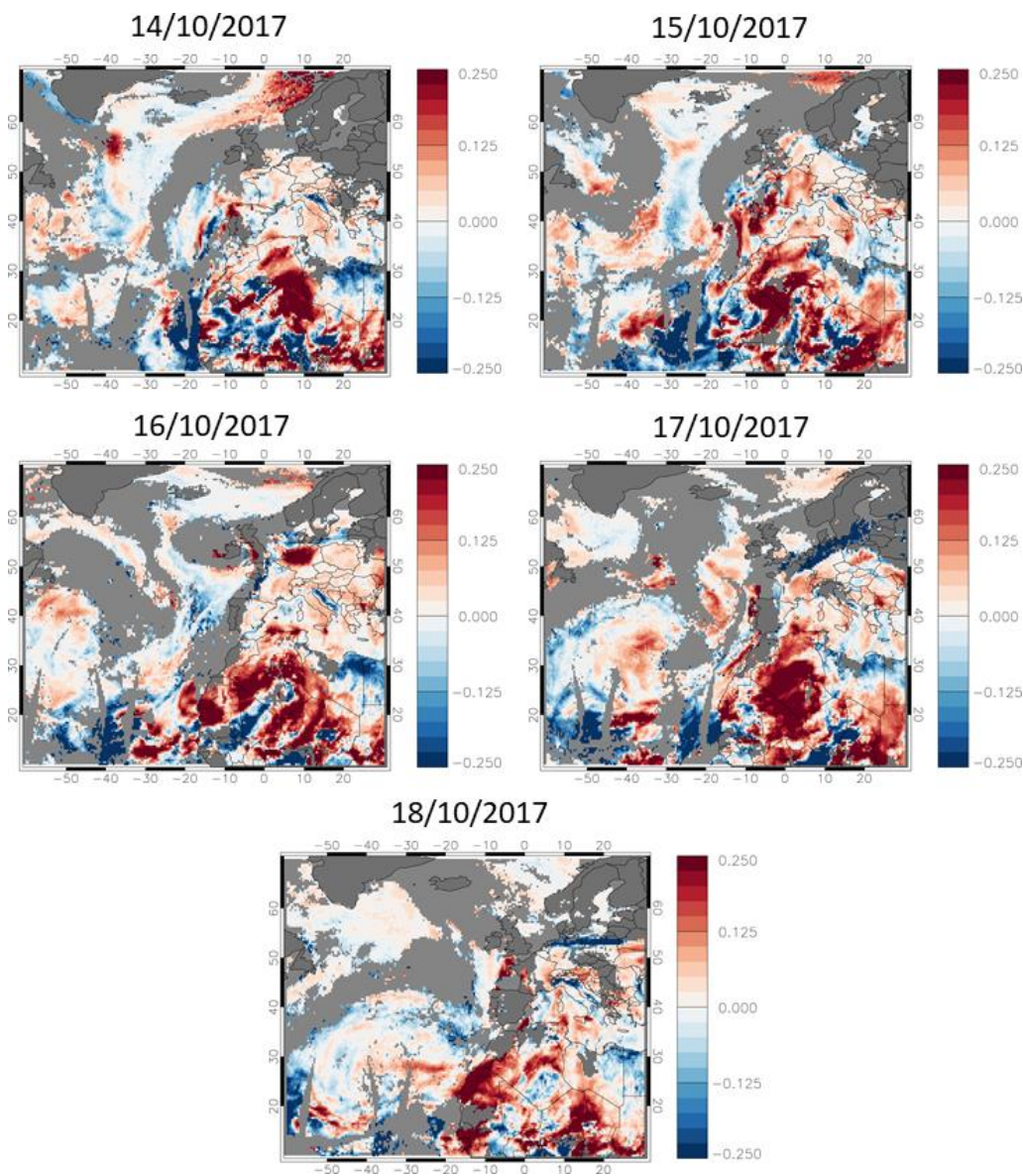
Accordingly, we have replaced the sentence **“The stations are located over the broader western European areas where dust and biomass burning transport occurred”** with the following in the RM P6, L2-4: **“The stations are located over western Europe and away from the dust and biomass burning sources, lie across the plumes of high AOD loadings, exhibiting significant increases in PM₁₀ and PM_{2.5} surface concentrations during the examined event.”**

Page 7, line 26: The FGE deserves to be introduced by an equation, moreover uppercase is more common.

We have added a new subsection (2.4 Statistical metrics) in the RM, where we present basic information and formulae for the Pearson correlation coefficient and the fractional gross error. We have also replaced all instances of fge with FGE following the suggestion in both text and figures of the RM.

Page 7, lines 30 to 34: How can this conclusion be aligned with Fig. 4, where in many regions with low AOD the AOD is enhance by data assimilation?

What we mean is that generally over areas with low “observed” AOD the IFS forecast tends to overestimate and vice versa. This agrees with the comment raised by the Reviewer, as over several areas with low observed AOD the IFS (with DA) forecast mostly overestimates. We have replaced “**low AOD values**” with “**low observed AOD values**” in the RM (P8, L26) for clarity. In support of the above we present here the differences between IFS AOD₅₅₀ and MODIS/Terra and Aqua AOD₅₅₀, which along with the left column of Figure 4 (MODIS/Terra and Aqua AOD₅₅₀ fields) confirms our conclusion.



Figs. 9 and 10: I find the similarity of the PM10 and PM2.5 composition surprising, normally I would expect a higher dust and sea salt fraction in PM10 than in PM2.5. Does this indicate some

limitation of the model? After all, from Eqs. (1) and (2) it is clear that the contributions in Fig. 9 and 10 cannot be that different, and the largest particles (SS3 and DD3) are not relevant at all, but is that realistic? Do the stations provide the PM_{2.5} and/or PM₁₀ composition for comparison, or is there any other suitable data source to validate this aspect?

Indeed, as depicted from the PM₁₀ and PM_{2.5} equations the contributions are similar. This was also the case in some sensitivity calculations we performed using random values for each aerosol type and bin. The equations themselves may play a role in that. The applied formulae are mostly empirical and thus we agree that may insert uncertainties (e.g. large particles are underrepresented). However, it should be also considered that in enhanced dust and sea salt conditions the total PM₁₀ and PM_{2.5} concentrations are also higher likely resulting in similar relative contributions in PM₁₀ and PM_{2.5}. Unfortunately, from the examined stations only GB0048R and GB1055R EMEP stations provide fragments of PM₁₀ and PM_{2.5} composition data for the examined period. These data are not sufficient for a comprehensive estimation of dust and sea salt contribution to PM₁₀ and PM_{2.5} due to missing data and lack of all PM₁₀ and PM_{2.5} aerosol components (available with gaps: NH₄⁺, Ca⁺⁺, Cl⁻, Mg⁺⁺, NO₃⁻, K⁺, Na⁺ and SO₄⁻). From data for the GB1055R station during the 16-17 October PM peak, it seems that although the individual dust and sea salt component concentrations are higher in PM₁₀ compared to PM_{2.5}, it is the total PM₁₀ concentrations that are also higher compared to total PM_{2.5} that probably result in similar contributions.

Generally, I believe links in footnotes and within the text are supposed to be moved to the References section to comply with the journal standards.

Done. In the RM we have replaced all footnotes and links with References according to the Journal standards.

References

Inness, A., Ades, M., Agustí-Panareda, A., Barré, J., Benedictow, A., Blechschmidt, A.-M., Dominguez, J. J., Engelen, R., Eskes, H., Flemming, J., Huijnen, V., Jones, L., Kipling, Z., Massart, S., Parrington, M., Peuch, V.-H., Razinger, M., Remy, S., Schulz, M., and Suttie, M.: The CAMS reanalysis of atmospheric composition, *Atmospheric Chemistry and Physics*, 19, 3515–3556, <https://doi.org/10.5194/acp-19-3515-2019>, 2019

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