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Title: Marine boundary layer aerosol in Eastern North Atlantic: seasonal variations and key controlling processes

We thank the anonymous referee #1 for his/her valuable and constructive comments/suggestions on our manuscript. We have revised the manuscript accordingly and please find our point-to-point responses below.

Comments by Anonymous Referee #1:

General Comments:

The paper provides a thorough analysis of the processes controlling the number concentration of the Aitken, accumulation, and sea spray aerosol modes in the eastern North Atlantic based on a several year data record from a site in the Azores. The conclusions that the free troposphere is a significant source of the Aitken and accumulation modes in the MBL and that sea spray aerosol makes up a small fraction of the total particle number at this site are significant and consistent with recently published papers. One intriguing result, if I am interpreting the analysis correctly, is that a significant impact of biogenic sulfur on the CCN population requires the flux of continental Aitken mode particles from the FT to the MBL.

Detailed Comments:

1. Page 2, line 40: add the qualifier ": : :.long term observation IN THE ENA."

Responses: The expression has been corrected as suggested.

2. Page 4, line 3: change to ": : :the parameters of which ARE DERIVED from fitting"?

Responses: The expression has been corrected as suggested.

3. Page 4, line 35: The red trajectories in Fig.1 a, c, and d are all very similar, i.e., originating over the Arctic and passing over northern Canada. Why are they described as "air masses influenced by anthropogenic emissions from North America for fall and winter" and "contribution from Arctic" for Spring. Also – I don't see the "northern Europe air masses" in the trajectories for spring.

Responses:

The trajectories shown in Fig. 1 are average trajectories for each cluster. The individual trajectories are shown in Fig. R1. We've also added this figure as Fig. S3 in the updated SI, and referenced in the manuscript accordingly (see Page 5, Line 1-4).

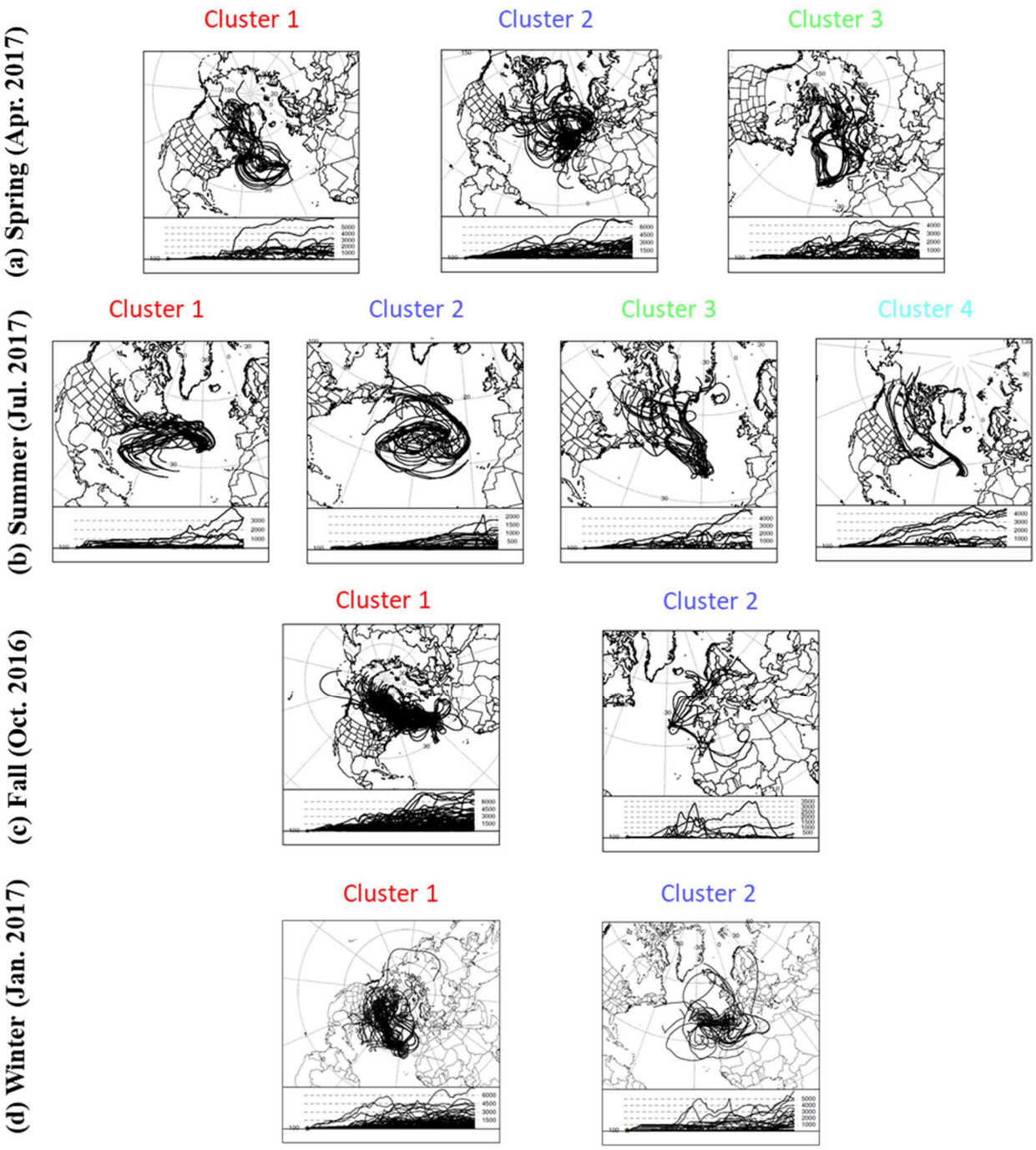

Fig. R1 *(added as Figure S3 in updated SI)* **Detailed trajectories for each cluster shown in Fig. 1.**

4. Figure 3.b2. and throughout: Figure 3.b2. clearly shows that what is termed here to be "Large Accumulation" mode is actually the sea spray aerosol coarse mode. To be in line with what it actually is and with published literature, it would be more appropriate to call it the SSA, PMA (primary marine aerosol), or primary aerosol mode.

Responses:

We agree, as shown in the Fig. 3b2, that the "Large Accumulation" mode is dominated by SSA and is essentially the sea spray aerosol coarse mode under vast majority of the cases. On the other hand, we don't want to simply call the mode "SSA" or "PMA" mode, without presenting any evidence to demonstrate the case (as shown in Fig. 6). We also note during some episodes (not shown in the manuscript), aged biomass burning aerosol and dust likely also contributed substantially to the large accumulation mode. We have added one sentence following the figure as (see Page 6, Line 26-28):

"Based on the average volume size distributions (Fig. 3b2), the "large accumulation mode" is essentially the sea spray aerosol coarse mode under vast majorities of the conditions."

5. Page 6, lines 23 – 24: The Ac mode Dp is 161 +/- 25 in summer and 155 +/- 31 in winter. Does the Ac mode really have a larger Dp in summer than winter given the fairly large standard deviations of the mean Dp?

Responses:

The standard deviation represents the variation of mode diameter during each season. We note that the mean, median, 25th and 75th percentiles of the Ac mode diameter all exhibit a higher value during the summer than that during winter (Fig. R2). The larger mode diameter is also evidenced by the seasonal average size distribution (Fig. 3a). We have modified the sentence to the following (see Page 6, Line 31-32):

"While there is substantial variation within each season, on average, the Ac mode exhibits higher number concentration, larger mode D_p, and higher occurrence in summer than in winter (Table 2)."

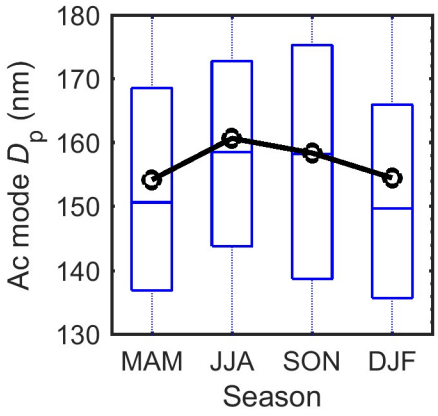

Fig. R2 Seasonal variation of Ac mode diameters.

6. Table 1: There is no instrument listed for MBL height or precipitation rate – unless they are included in the "Vertically pointing K-band: : :." list of instruments.

Responses: They are indeed included in the "Vertically …" list. We've added some solid lines to Table 1 to make it clearer.

7. Table 2: Why aren't modal volumes included in the table – especially since they are referred to in the text (e.g., page 6, line 25).

Responses: The modal volume information are added to Table 2 as suggested. In accordance with the data, we also modified the statement into (see Page 6, Line 34):

"In contrast, LA mode shows opposite seasonal trends, with the number and volume concentrations in winter exceeded 1.5 times those in summer (Table 2)."

8. Page 11, Line 9: should be Figure 6c.

Responses: Fig. 6c was discussed later (Page 11, Line 14). Here we are indeed discussing about Fig. 6b.

9. Page 11, Lines 10 – 12: Has a volume mode with a diameter of 0.6 to 0.8 um ever been observed in the remote marine boundary layer? It is not clear why it is discussed here as a possibility and why the "LA" mode is not simply called the "SSA" mode.

Responses:

Given the prevalence of marine low clouds, the brief discussion here is simply to eliminate the possibility that large accumulation mode observed is a result of in-cloud production of sulfate and/or organics (Pandis et al., 1990; Meng and Seinfeld, 1994; Seinfeld and Pandis, 2016). We agree the "LA" is essentially sea spray aerosol coarse mode under vast majorities of the conditions. Please also see the response to comment #4.

10. Page 11, Lines 15 – 16: There are many, many published papers that establish that MBL supermicron particles are dominated by SSA. Why is it being ebated/emphasized here?

Responses:

Here we're not emphasizing that supermicron particles are dominated by SSA. On the contrary, we are using that as a premise. What we stated is that since LA mode (D_p ~300 to 1000 nm) share the same source with supermicron aerosols (D_p 1~10 μm), and since supermicron aerosols are dominated by SSA in remote MBL, thus we speculate that LA mode should also be dominated by SSA. We've rephrased the description to avoid such confusion as (see Page 11, Line 14-19):

"This is also supported by the strong correlation between V_{LA} and PM_c B_{sca} (Fig. 6c). The PM_c B_{sca} is a surrogate for the supermicron mode (PM_c, D_p 1~10 μm) volume concentration (section 2.2), while supermicron particles are dominated by SSA in remote MBL (Campuzano-Jost et al., 2003). Therefore, the strong correlation suggests that LA particles are also dominated by SSA."

11. Page 12, Lines 7 – 8: There is no need to invoke a lack of correlation of Nat or Nac with wind speed to conclude that SSA is a minor contribution to those two modes. Figure 3 is evidence enough.

Responses:

Fig. 3 does show the fitted LA mode has a minor contribution to At mode or AC mode size range. However, LA mode likely presents the coarse mode of the SSA, and many studies have suggested that source function of SSA extends down to Aitken mode size range. We think lack of correlation provide additional evidence for the minor contribution.

12. Figure 8: Should make it clear in the caption that "(0.1%)" refers to supersaturation level.

Responses: The expression has been corrected as suggested into (see Caption of Fig. 8):

"Figure 8. Estimation of SSA contributions to *CN* and *CCN* (0.1%), namely CCN concentration at 0.1% supersaturation level."

13. Page 12, Line 33: should be ": : :fraction is consistent", not "in consistent".

Responses: The expression has been corrected as suggested.

14. Page 13, Line 2: "The SSA number concentration: : :" What number concentration is being referred to here? The present paper or Quinn et al., 2017?

Responses:

Here we mean the derived number concentration following the approach in Quinn et al. (2017). The sentence has been changed into (see Page 13, Line 2-3):

"In that study, the size distribution of SSA was derived by fitting aerosol size distribution. If we follow the same approach (Quinn et al., 2017), the estimated SSA number concentration is actually N_{LA} shown in this study, which represents 19 % of *CCN* (0.1 %)."

15. Page 13, Lines 3 – 7: Please clarify what the "above estimation" is. Numbering the equations and referring to them by number would help. Also, please define the f_ac,SSA and f_at,SSA terms. Are these the flux of SSA in the accumulation and Aitken modes, respectively?

Responses:

The sentence has been changed into (see Page 13, Line 5-9):

"Based on the estimated SSA contribution to *CN* and *CCN* (Eq. 8a and 8b), we can further estimate the SSA contribution to N_{Ac} and N_{At}, $f_{Ac,\,SSA}$ and $f_{At,\,SSA}$, as:

$$f_{Ac,SSA} = (CCN(0.1\%)_{SSA} - N_{LA}) / N_{Ac}$$
$$f_{At,SSA} = (CN_{SSA} - CCN(0.1\%)_{SSA}) / N_{At}$$

and the corresponding annual mean $f_{Ac,\,SSA}$ and $f_{At,\,SSA}$ are 21 % and 10 %, respectively (Table 4)."

16. Page 14, Lines 13 - 14: Please provide previously published fluxes of DMS in the ENA compared to the remote Southern Ocean. Also – this sentence is incomplete.

Responses:

This sentence is based on Plate 2 in Kettle et al. (1999), so we can only give an estimated range. We've modified the expression into (Page 14, Line 9-11):

"Such difference is likely due to the much higher DMS sea surface concentration in ENA (~7.5 nM) than that in southern oceans (~2.5 nM) (Kettle et al., 1999), or due to the difference between observed and model-simulated aerosol size distributions, etc.."

17. Figure 9: What is meant by secondary processing rate? Isn't the SSA flux a primary process, i.e., direct mechanical production?

Responses: The word "secondary" is deleted throughout.

18. Figure 9b: This half of Figure 9 does not appear to be explained in the main text.

Responses: It was discussed in the second but last paragraph in section 6.2 (Page 14, Line 1-4), and section 6.3. We also added the citation in section 6.3 as (see Page 14, Line 20):

"$\partial_t N_{At}|_{FT}$ is higher in spring-summer while lower in fall-winter, and such seasonal variation is somewhat different from those of CO mixing ratio and EBC mass concentrations (Fig. 9b)."

19. Figure S5: Please provide r^2 values for these correlations to support the conclusion given on page 15, lines 20 – 21.

Responses: The values are added to Fig. S5 (see Fig. R3 below).

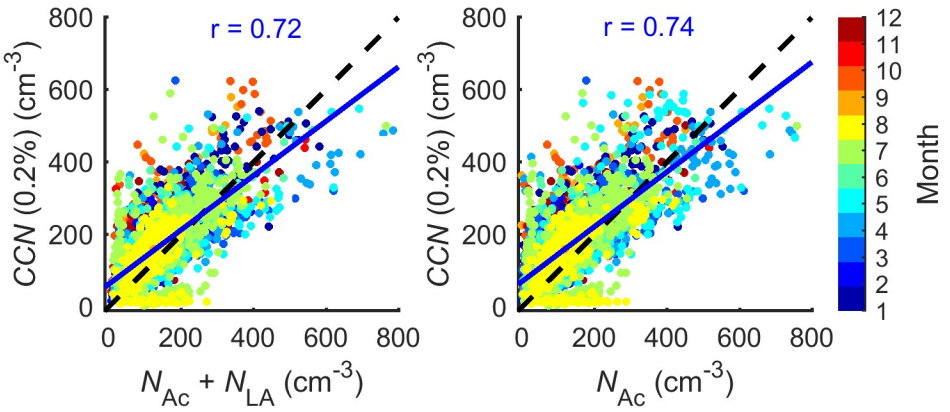

Figure R3. *(Fig. S6 in updated SI)* **Comparison of observed *CCN* concentrations with relevant modal number concentrations.** The black dash line is the 1:1 line shown for reference. The value of *r* given referred to the Pearson correlation coefficient, while the regression line based on York et al. (2004) is also shown for reference.

20. Page 16, Lines 11 – 19: Is the correct interpretation here that Aitken mode particles measured at ENA are continentally derived, while the growth of those particles to CCN size in the MBL is due to biogenic H2SO4? This implies that for ocean ecosystems (at least in the ENA) to have a substantial influence on the MBL CCN population, there must be Aitken mode continental aerosol for the required condensation and growth to occur.

Responses:

Both continental emission and NPF in the FT contribute to the Aitken mode population in FT. However, we cannot quantitively determine the contribution from each of the two sources. Nevertheless, Fig. 9b shows much higher entrained FT At mode particles in spring-summer than entrained CO and BC, suggesting an important contribution of NPF to FT Aitken mode at least during these two seasons. We've added this discussion (see Page 14, Line 37 to Page 15, Line 2):

"Contribution of SSA to the At mode is even smaller than it is to the Ac mode, and is estimated to be no larger than 10 % (Table 4). As a result, the entrainment of FT At mode particles represents the dominant source (Fig. 9a). $\partial_t N_{At|FT}$ is higher in spring-summer while lower in fall-winter, and such seasonal variation is somewhat different from those of CO mixing ratio and EBC mass concentrations (Fig. 9b). These differences may be partially due to stronger new particles formation from biogenic precursors in the FT during spring and summer seasons (Sanchez et al., 2018). The strength of new particle formation is not correlated with CO or EBC concentrations, which are tracers for anthropogenic emissions. The contribution of NPF versus anthropogenic emissions to FT Aitken mode particles cannot be quantitatively determined using data presented here alone, and will be a subject of future study."

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