

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

5 We thank the anonymous referee #2 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 #2:

10 **General Comments:**

This paper uses two years of observations from the ARM program and a process-based analysis to estimate the factors that are most significant in defining the marine aerosol size distribution at Graciosa Island in the Azores. I find the approach to be systematic and reasonable, the large data set to be valuable and the results to be useful. I have a few little issues with some of the interpretation, but I think this work is worthy 15 of publication in ACP subject to my following comments:

Detailed Comments:

1. *The paper reads better at the beginning. The grammar begins to suffer in various places later in the paper. The authors need to carefully read and correct the grammar where needed.*

20 **Responses:** We've read through the updated manuscript and corrected the grammar carefully.

2. *In the abstract and final paragraph (Page 16, lines 11-19), the authors say that the free troposphere (FT) is the major source of CCN to the marine boundary layer (MBL) via direct insertion and insertion of Aitken particles that grow via condensation in the MBL to CCN size; the latter being responsible for about 60% of 25 the CCN in summer. That is a nice result. The authors then say that DMS oxidation is responsible. We cannot deny the likelihood of a contribution from DMS, but we know from work over the past few years that organics may play a significant role in Aitken particle growth. For example, in the Arctic we find growth of Aitken particles in the summer is related to (non-DMS) marine-derived organics at least as much and perhaps more than marine derived sulphate (e.g. papers in the NETCARE special issue of ACP; work done 30 at the Zeppelin Observatory; Willis et al., GRL, 2016; Burkart et al., GRL, 2017; Mungell et al., PNAS, 2017). The Arctic MBL differs from the Pacific and Atlantic MBLs in some respects, but there are similarities and various groups (O'Dowd et al.; Prather et al., etc.) have pointed to (non-DMS related) organics in Aitken particles in the MBL. The authors need to present a slightly more objective summary on this issue. I'm not suggesting to rule out DMS, but don't rule out the possibility of other organic 35 components.*

Responses:

We thank the reviewer for raising this point. We agree that while the ENA MBL differs from the Arctic MBL in some respects, it is possible that organics play an important role in the growth of Aitken mode particles. We've added relevant discussions in the updated manuscript. For example, we've added a comment in section 4.2, the condensation part as (see Page 9, Line 32-33):

5 “Here we assume that H_2SO_4 is the dominant condensate. However, recent studies suggest that organics may play an important role in growth of particles inside MBL, and this is discussed later in section 6.2.”

And we've added the discussion into section 6.2 as (see Page 14, Line 14-21):

“Common continental biogenic volatile organic compounds (BVOCs) such as isoprene and monoterpenes typically have very low mixing ratio, and SOA formation from these BVOCs is generally minor in remote

10 marine environment (Kavouras and Stephanou, 2002; Arnold et al., 2009; Gantt et al., 2009; Myriokefalitakis et al., 2010). However, recent studies suggest photochemistry or heterogeneous oxidation at the sea surface microlayer may represent a substantial source of oxygenated gas-phase organic compounds (OVOCs), which potentially plays an important role in SOA formation and particle growth in the Arctic MBL (Burkart et al., 2017; Willis et al., 2017; Mungall et al., 2017). It is possible that the SOA formation from these OVOCs can contribute to the growth of Aitken mode particles in ENA as well. If so, the contribution to CCN by the growth of Aitken mode particles would be even higher than the estimate here, which is based on condensation of H_2SO_4 only.”

20 3. *There is no mention of emissions from ships? What reasons do you have for excluding the possibility of shipping emissions? Related to ships and to the aqueous-phase chemistry and NPF processes, are SO_2 concentrations measured at the ARM site? Have you any idea of what they might be? Are they too small to be routinely measured?*

Responses:

We thank the reviewer for this point. Data impacted by local ship emissions are screened out and not 25 included in the analyses (SI S1). Langley et al. (2010) show that ship particle emissions, when present, can contribute substantially to particles and CCN. The contribution of the ship particle emissions averaged over large spatial area in remote marine boundary layer remains unclear, and therefore it is not directly treated in this study. As a result, the particles emitted by ships are implicitly grouped into the category of “entrained from the FT”.

30 Several studies (Langley et al., 2010; Corbett and Fischbeck, 1997; Capaldo et al., 1999; Corbett et al., 2007; Wang et al., 2008; Johansson et al., 2017) have shown ship emissions represent a significant source of SO_2 in MBL. In this study, the concentrations of SO_2 and H_2SO_4 are estimated using DMS- SO_2 - H_2SO_4 yields based an observation-based parameterization (Russell et al., 1994; Pandis et al., 1994). Therefore, H_2SO_4 formed from ship emitted SO_2 , and its contribution to condensational particle growth is implicitly 35 included.

We have clarified this in the revised manuscript. We've added the following in section S1 (Page 2, Line 11-15):

40 “With this filter, data impacted by local ship emissions are also screened out. Langley et al. (2010) shows that ship particle emissions, when present, can contribute substantially to particle and CCN concentration in

the MBL. That condition, if present in ENA, would also be screened out considering the high aerosol number concentration (1000 ~ 3500 cm⁻³). The contribution of the ship particle emissions averaged over large spatial area in remote marine boundary layer remains unclear, therefore it is not directly treated in this study.”

5

With regards to regional ship emissions, we've added relevant discussions in section 6.2 as (see Page 14, Line 22-26):

“Several studies (Langley et al., 2010; Corbett and Fischbeck, 1997; Capaldo et al., 1999; Corbett et al., 2007; Wang et al., 2008; Johansson et al., 2017) have shown that ship emissions represent a significant source of SO₂ in MBL. In this study, the concentrations of SO₂ and H₂SO₄ are estimated using DMS-SO₂-H₂SO₄ yields based an observation-based parameterization (Russell et al., 1994; Pandis et al., 1994). Therefore, H₂SO₄ formed from ship emitted SO₂, and its contribution to condensational particle growth is implicitly included.”

15 Unfortunately, SO₂ measurement is not available at the ENA site. Here we assume NPF is rare inside MBL, and estimated SO₂ concentration is used to qualitatively demonstrate that the aqueous-phase chemistry has negligible influence on the particle number concentration of each mode (see responses to comment #16).

4. *Abstract, lines 20-21 – Define the size ranges for At, Ac and LA modes here.*

20 **Responses:** The information has been added as (see Page 1, Line 23-24):

“Submicron aerosol size distribution typically consists of three modes: Aitken (At, diameter $D_p < \sim 100$ nm), Accumulation (Ac, D_p within ~ 100 to ~ 300 nm), and Larger Accumulation (LA, $D_p > \sim 300$ nm) modes,....”.

5. *Abstract, line 28 – How is “the estimate based on major sources and sinks” made?*

25 **Responses:** We've changed the expression into “generally agrees with the steady-state concentration estimated from major sources and sink.”

6. *Page 2, line 20 – For previous aerosol studies, consider Phinney et al., Deep Sea Research (2006) and Langley et al. ACP (2010).*

30 **Responses:** The references have been added as suggested.

7. *Page 2, lines 23-24 – “cloud coalescence scavenging” or “coalescence scavenging in cloud” or something with cloud factored in.*

35 **Responses:** The expression has been changed into “in-cloud coalescence scavenging” throughout as suggested.

8. Page 2, line 39 – Consider Leaitch et al., ACP (2010) that looks at two cases of aerosols and MBL clouds over the Western Atlantic. Also, they found the supersaturations to be between 0.1 and 0.2% depending on the aerosol concentrations.

Responses:

5 This paragraph focused on campaigns in Eastern North Atlantic only. We referred to the Leaitch et al., ACP (2010) paper in discussions of typical supersaturation levels for marine low clouds instead (Page 9, Line1-2):

“The maximum supersaturation near the cloud base where CCN activation occurs is typically 0.2 % for marine low clouds (Wood et al., 2012; Clarke and Kapustin, 2010; **Leaitch et al., 2010**).”

10

9. Page 3, lines 26-27 – Indicate how the inlet has been evaluated for transmission losses of the larger particles (i.e. 5-10 um diameter).

Responses:

15 The transmission losses for the 5-10 um size range are not corrected. However, as we only examine the relative trends of the coarse mode optical properties and their correlations with other aerosol properties, we do not expect this will affect the results or conclusions from this study.

We've clarified this by including following description in the revised manuscript (see Page 3, Line 35 to 37):

“Potential particle losses for large particles (i.e., in the diameter range of 5 ~10 μm) are not corrected.

20 However, we do not expect the losses affect the relative trends of PM_c B_{sca} presented here (section 5), or the correlation among PM_c B_{sca} and V_{LA} (Fig. 6c).”

25 10. Section 2.2.2 – Presumably, cloud base height and cloud thickness are used in the analysis, but at this point (Section 2.2.2) it is unclear why cloud properties are discussed. Maybe on page 3 (around lines 5-6) or at the beginning of 2.2.2?

Responses:

We've added a paragraph at the beginning of section 2.2.2 as (see Page 4, Line 3-4):

30 “The cloud and MBL properties are needed to estimate some of the key controlling processes that drives aerosol properties (see section 4 for more details). Key parameter needed included the MBL height, cloud thickness and cloud fraction.”

11. Page 5, line 23 – due to changes in tropopause height.

Responses:

The expression has been modified into (see Page 5, Line 27-28):

35 “In contrast, CO and O₃ in ENA show a summer minimum and spring-winter maximum, **which is consistent with the FT entrainment as the dominant source and corresponding seasonal variations in**

tropopause height. This suggests minor contributions from local emissions and in-situ photochemistry (Parrish et al., 1998; Fischer et al., 2003; Mao and Talbot, 2004)."

12. Your BC is measured from estimated absorption based on a filter transmission technique. The standard
5 reference to this is Equivalent Back Carbon or EBC (Petzold et al., 2013) rather than BC. You should adopt
that nomenclature.

Responses:

The expression has been changed throughout as suggested, with a notion added in section 2 as (see Page 6, Lines 2-3):

10 "After these episodes are excluded, the **equivalent black carbon (EBC, following the naming convention suggested by Petzold et al. (2013))** mass concentrations were estimated from $PM_1 B_{abs}$ with an assumed mass absorbing cross section of $7.5 \text{ m}^2 \text{ g}^{-1}$ at 529 nm (Bond et al., 2013)."

13. Page 6, line 2 - define "Pcb"

15 **Responses:** The definition (precipitation rate at cloud base) has been added as suggested.

14. Page 6, line 35 - Particles "activate" and droplets "nucleate".

Responses: The word "activated" has been deleted.

20 15. Page 7, line 3 – Dry deposition may be slower on average, but you are discussing processes that happen over a week to 10 days. Please discuss further.

Responses:

In previous work comparing the contribution of dry and wet depositions (Henzing et al., 2006; Wood et al., 2012; Mohrmann et al., 2018), the time discontinuity in wet depositions are already considered, as also did 25 in our study by taking into account the cloud fraction. The results show that for submicron MBL aerosols, the contribution of dry deposition is very small, with a fractional contribution generally < 5% in total (dry and wet) removal processes (Henzing et al., 2006), and absolute loss rates "unlikely to exceed $2 \text{ cm}^{-3} \text{ d}^{-1}$ " (Wood et al., 2012) under most circumstances.

This point is further clarified in the manuscript as (see Page 7, Line 11-14):

30 "In addition, dry deposition is usually much slower compared to wet deposition for submicron particles, even after considering in the in-cloud possibility, f_{cloud} , which reflects the time and spatial discontinuity of the wet deposition processes (see the in-cloud scavenging part in section 4.2) (Lewis and Schwartz, 2004; Henzing et al., 2006; Wood et al., 2012; Mohrmann et al., 2018). Thus, it is neglected in further analysis."

35 16. Page 8, lines 32-34 - Using a limit of 100 nm diameter for the Ac particles, you eliminate the potential for some larger Aitken particles to grow into the Ac mode via S(IV) oxidation. Without relatively large

amounts of SO_2 , it will be very difficult for the Ac particles to grow into the LA mode. While still a big ask, for relatively low SO_2 , as seems more likely in the MBL you have constrained, the probability of growing 80 nm particles to 120 nm is more likely. What precursor concentrations do you use for your analysis?

Responses:

5 There may be some misunderstanding here. In this section we're discussing the aqueous-phase reactions inside cloud droplets. As has been clearly outlined in section 4 and depicted in Fig. 5, Aitken mode particles are not CCN and thus are not considered in aqueous-phase reactions. That is due to the much higher liquid water content and thus much higher aqueous-phase reaction rates in cloud droplets than in interstitial aerosols. In-cloud sulfate production can contribute to the growth of the Aitken mode aerosols, but that
10 only occurs after the Aitken mode particles have already grown to CCN (i.e. into Ac mode size ranges) through condensation. We've added a paragraph at the start of this section to further clarify this (Page 9, Line 9-14):

“The aqueous-phase reaction (i.e., in-cloud production of sulfate) rate is positively related the liquid water content (Seinfeld and Pandis, 2016; Meng and Seinfeld, 1994; Pandis et al., 1990; Cheng et al., 2016). As
15 the liquid water content of cloud droplets are orders-of-magnitude higher than that of interstitial aerosols, only aqueous-phase reactions inside the cloud droplets are considered here (Pandis et al., 1990). As a result, the aqueous-phase reactions only promote the growth of CCN (i.e., Ac and LA mode particles), the influence of aqueous phase reactions on the Aitken mode particles is neglected until they grow to CCN sizes through condensation (Hoppel et al., 1994; Pandis et al., 1990).”

20 The precursor concentrations are given in section 4.2, the condensation part. It was described as (see Page 9, Line 27-30):

(The annual mean H_2SO_4) “is assumed to be 1.0 ppt (Pandis et al., 1994), while being 1.4, 1.3, 1.1 and 0.2 ppt in spring, summer, fall and winter, respectively. This seasonal variation in v_i is based on the monthly dimethyl sulfide (DMS) fluxes (assumed to be 7.0, 5.4, 2.9 and 1.0 $\mu\text{mol m}^{-2} \text{ day}^{-1}$ in spring, summer, fall
25 and winter, respectively) given in previous studies in the North Atlantic Ocean (Tarrasón et al., 1995), and the proposed dependence of H_2SO_4 on DMS flux at the observed fluxes ranges (Pandis et al., 1994; Russell et al., 1994).”

17. Page 11, line 8-9 – Is the strong correlation between $N(LA)$ and WS a result of using equation 2?

30 **Responses:** Not likely. The N_{LA} is from fitting of the observation data, not calculated by Eq. 2.

18. Page 11, line 19 – “Given the large sizes of LA particles and that we have excluded dust, we do not: : :”

Responses: The expression has been corrected as suggested.

35

19. Page 11, line 21 – “: : : the concentration of LA particles from the FT is negligible: : :”

Responses: The expression has been corrected as suggested.

20. Page 12, line 7 – The decreasing At with increasing WS could indicate some wind associated dilution of oceanic sources of At particles.

Responses:

The MBL height likely play a more important role in the dilution of MBL aerosol than WS. A more plausible explanation may be that, the higher N_{LA} at higher WS will increase the coagulation sink of Aitken mode aerosols. As coagulation is the dominant sink for Aitken mode aerosols, this can better explain the negative correlation of N_{At} with WS.

We have added this potential explanation to the manuscript as (see Page 12, Line 7-9):

“Unlike N_{LA} , N_{Ac} is independent of the WS, and N_{At} decreases with increasing WS (Fig. 7), indicating relatively minor contributions from SSA to At and Ac modes. The negative correlation between N_{At} and WS may be due to the enhanced N_{LA} with increasing WS (Fig. 6), and thus enhanced coagulation loss for Aitken mode particles (see section 4.2 and section 6.3).”

21. Page 14, lines 25-26 - That does not mean there is no contribution from anthropogenic emissions. It could be a case of the contribution being high in spring and low in summer.

Responses:

We have further clarified this in the revised manuscript (see Page 14, Line 38 to Page 15, Line 2):

“ $\partial_t N_{At}|_{FT}$ 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 in 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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22. Page 15, line 19 – It is 70 nm in the figures, not 60nm. I assume 70 nm is correct due to the noise issues that appear to be common in the first one or two channels of the UHSAS.

Responses: The expression has been corrected to 70 nm throughout as suggested, and we added a notion below Table 1 as:

30 “^a In fact the lower size limit of UHSAS is 60 nm. Here we used only data larger than 70 nm to avoid noises sometimes observed in the first several channels of the UHSAS.”

23. Page 15, line 24 – Sources of LA particles are dominated by SSA.

Responses: The expression has been corrected as suggested.

35

24. Page 15, line 24 – “dilution by entrained FT air”. In order to dilute the LA particles with FT air, presumably some of the LA particles must enter the FT. Could that be an important FT source somewhere downwind?

Responses:

We don't think so. The entrainment process is not an exchange between FT and MBL. Instead, FT air is entrained into and stays inside the MBL (Stull, 2012). The entrainment counteracts the large-scale substance in ENA, and help sustain the MBL height (Stull, 2012; Mohrmann et al., 2018; Wood and Bretherton, 2004). While we agree that some LA particles may enter the FT through deeper convection, it is worth noting that these convections are often associated with precipitation that efficiently removes LA mode particles. Therefore the influence of such process on FT particle population is likely negligible.

25. *Page 16, lines 14-15 – Can marine emissions be a factor here also, if they are lofted above the MBL*

10 *somewhere and return to the MBL somewhere else?*

Responses: Please refer to our response to comment #24.

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