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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Marine boundary layer aerosol in Eastern North Atlantic: seasonal variations and key controlling processes" *by* G. Zheng et al.

Anonymous Referee #2

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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 of publication in ACP subject to my following 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.

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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 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 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 components.

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 SO2 concentrations measured at the ARM site? Have you any idea of what they might be? Are they too small to be routinely measured?

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

5) Abstract, line 28 – How is "the estimate based on major sources and sinks" made?

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

7) Page 2, lines 23-24 – "cloud coalescence scavenging" or "coalescence scavenging in cloud" or something with cloud factored in.

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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.

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).

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?

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

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

13) Page 6, line 2 - define “Pcb”

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

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.

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 SO2, it will be very difficult for the Ac particles to grow into the LA mode. While still a big ask, for relatively low SO2, 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?

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

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

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

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

21) Page 14, lines 25026 - 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.

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.

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

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?

25) Page 16, lines 14-15 – Can marine emissions be a factor here also, if they are lofted above the MBL somewhere and return to the MBL somewhere else?

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