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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-term trends of global marine primary and secondary aerosol production during the recent global warming hiatus (2000–2015)" *by* S.-K. Song et al.

Anonymous Referee #1

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This manuscript aims to understand how marine derived aerosols changed during the warming hiatus from 2000-2015. The authors consider primary sea spray aerosol (SSA) and secondary aerosol formation from dimethylsulfide (DMS) fluxes. They compute the SSA fluxes using a parameterization based on sea surface temperature (SST) and wind speed (U10) from Gong (2003). DMS fluxes are computed using chlorophyll and mixed layer depth parameterizations for seawater concentrations from several publications and the Liss and Merlivat (1986) gas transfer parameterization. The aerosol optical depth (AOD) from both sources was computed using the model Optical Properties of Aerosols and Clouds (OPAC). In addition, the authors compare their computed marine derived AOD values with MODIS AOD values. They find that the annual global

trends in SSA and DMS fluxes were stable and decreasing, respectively, with opposite trends in the respective AOD values. The authors also found regional trends. When compared to MODIS values, the computed AOD values show that up to 62% of total AOD can be explained by SSA and up to 38% can be explained by DMS derived aerosols. While the topic is interesting and the presentation/methodology seem generally solid, I am not clear after reading the article how these findings tie in with the warming hiatus. I am not sure there is any new insight here and I would say this is my major concern. In addition, I have several specific points that need addressing before this paper can go further. I recommend major revisions before this manuscript can be considered for publishing in ACP.

Main comments:

Hiatus relevance - The authors state on lines 43 – 46 that the hiatus may be due to several factors including aerosols, but never really reach a conclusion about this. They refer to the ocean heat uptake and regional changes in wind later in the text to discuss changes in fluxes leading to changes in AOD, but never make a direct connection between the AOD and the hiatus. Did the aerosols contribute to the hiatus? This reads more like changes in physical conditions (e.g. U10) due to the hiatus caused changes in the precursor fluxes and the AOD.

From lines 74 to 80, the authors state that there are many unknowns related to aerosol production/loading and claim that, therefore, their goal is to study aerosol trends during the recent hiatus. However, I am not sure one has anything to do with the other. They could study aerosol changes over any period to investigate these uncertainties.

Specific comments: Lines 54-57: Are there any other studies the author can cite to corroborate the findings cited (i.e. Klimot et al., 2017)? Also, how important are other sources of marine aerosol not included here (e.g. glyoxal, isoprene)?

Lines 58-65: I am not sure of the purpose of this paragraph.

Lines 76-78: In order to understand if there is any change in aerosols during the hiatus, one need to understand the trends before. It does not appear the authors did this.

Lines 122-123: It is very well known that DMS concentrations in seawater are hard to predict and not really correlated with chlorophyll. The values depend highly on the presence of DMSP producing phytoplankton and DMSP cleaving phytoplankton and bacteria. The parameterizations used have some value in certain areas, sometime, but may not be correct all of the time. Did the authors do any sensitivity tests to compare different parameterization methods before settling on this set of equations?

Lines 134-135: It has been shown many times, with eddy covariance measurements, that unilinear gas transfer relationships with wind speed are more correct for DMS than the parameterizations discussed here. Liss and Merlivat (1986) is also extremely low compared to values found in the field. Why did the authors choose this?

Paragraph starting at 189: How do the findings presented in this paragraph compare with the findings in Quinn et al. (2017)? I realize the timing of the two studies may not be identical, but Quinn et al. take a global view and discuss the controls on SSA and DMS derived aerosol. Also, evidence from direct DMS flux studies suggests that seawater concentrations are more important that U10 for fluxes.

Lines 204-207: The authors present here a lot of interpretation based estimates of DMS fluxes that may not be very realistic. The statements can certainly be made about their own calculations, but I think it would be helpful to put these assumptions/speculations into the context of the real world. How likely is it that these trends in fluxes are applicable to the real world? Lines 251-253: How was the comparison to previous work done? Was it precise by month/season and area? Lines 283-284: I am not sure I follow the “thus” logic; does the comparison of the trends before this sentence make sense in another way? Line 290: What is the adjustment factor? Lines 293-295: It seems that more information is given here for SSA than DMS. How was the U10 changed for the DMS fluxes? Or were the SSA and DMS fluxes treated similarly?

Lines 345-349: Did the authors look at forward trajectories for the anthropogenically influenced aerosols? How far can these aerosols go over the ocean? Results/discussion general: Again, the Quinn et al. (2017) paper was never cited. How do the general results compare with theirs? Conclusion: This is basically just a summary and, again, provides no insight regarding the relevance of the hiatus. Tables 1 and 2: Need better description of units

References Quinn, P. K., Coffman, D. J., Johnson, J. E., Upchurch, L. M., Bates, T. S. (2017) Small fraction of marine cloud condensation nuclei made up of sea spray aerosol, Nature Geoscience (10), http://dx.doi.org/10.1038/ngeo3003.

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