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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.

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Response to Referees' Comments

We would deeply appreciate the comments from the reviewer to improve the quality of our paper.

Anonymous Referee #1:

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 com-

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pute 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:

1. 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.

ANS) The primary objective of this study is to analyze the long-term trends in global SSA emissions and DMS fluxes during 2000-2015 (i.e., corresponding the recent global warming hiatus period). The secondary and third objectives are to evaluate the

long-term variations of separate aerosol loadings derived from primary (SSA) and secondary (DMS oxidation) marine aerosols and their contributions to satellite-observed AOD, respectively. Thus, we did not analyze causes of global warming hiatus because that is beyond the scope of this study. We changed the title of this paper, "Long-term trends of global marine primary and secondary aerosol production during 2000-2015." In addition, we removed two sentences in the “Introduction” to clarify our objectives. However, as the reviewer suggested, we further analyzed the correlation between aerosol properties (AOD) and physical conditions (SST and SSW) to find their connection during 2000-2015 (see Fig. 4 in the revised text and Fig. 1). For example, we analyzed the relation of SST with atmospheric aerosol loadings (MODIS AOD), as well as other parameters (e.g., SSW and chlorophyll). New discussion on this is imbedded in the revised manuscript.

Fig. 1. Distribution of correlation of (a) SST vs. U10, (b) SST vs. Chl-a, (c) SST vs. MODIS AOD, (d) U10 vs. MODIS AOD, (e) SST vs. AODSSA, (f) SST vs. AODDMS, (g) U10 vs. AODDMS, and h) U10 vs. Chl-a.

Note that the 1951-2012 period represents 61 years of climate data, while the 1998-2012 period represents only 15 years; a fifteen-year period is well known to be too short to reliably detect long-term climate trends in global mean surface air temperature. The slowdown in warming during this fifteen-year period, often termed a "hiatus" in global warming can be ascribed, either fully or in part, to natural variability in the climate system (Roberts et al., Nature, 2015; https://www.nature.com /articles/nclimate2531). Discussion on this is added to the middle part of page 9. In addition, we replaced U10 and SST data obtained from two satellites (QuikSCAT and ASCAT satellites) with ECMWF reanalysis data due to limitation of discontinuity of two satellite measurements for the trend analysis (see Supplementary Fig. 1). Relevant figures and tables were revised.

2. 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.

ANS) The sentences from lines 74 to 80 means that "the studies of the long-term variation in marine aerosol production, as well as its contributions to aerosol concentration over the ocean, are scarce", compared to the studies related to aerosol radiative forcing by changes in anthropogenic aerosol emissions. We revised the sentence to avoid confusion (in the upper part of page 4). Note that there is significant discrepancy in the magnitudes of SSA emission rates and DMS fluxes due to the difference in parameterization methods for the flux estimation. Our main objective was not to estimate these fluxes, but to identify the trend of temporal variation of SSA emission and DMS flux during 2000-2015 using the most widely used method, Gong (2003). Although there is large uncertainty in the magnitude of SSA emission rates, the estimation of long-term trend (e.g., upward, downward, on no change) is less uncertain because of the one parameterization method usage.

Specific comments:

3. 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)?

ANS) Additional references (Street, et at., 2009; Jaeglé, et al., 2011; de Leeuw, et al., 2011; Ovadnevaite, et al., 2014; Janssens-Maenhout et al., 2015) were added to the revised manuscript (in the lower part of page 3). In addition, some discussions on other sources of marine aerosol (e.g., glyoxal, isoprene) were newly added to the revised text (in the middle part of page 3). "In contrast to the importance of isoprene as a biogenic secondary organic aerosol (SOA) precursor in the continents, Arnold et al. (2009) suggested an insignificant role of isoprene in the remote marine SOA formation. SOA production from the oxidation of marine VOCs (dialkyl amine salts,

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marine hydrocarbon, glyoxal, etc.) was found to be insignificant, compared with natural SSA emissions (Fu et al., 2008; Myriokefalitakis et al., 2008; Myriokefalitakis, et al., 2010; Rinaldi et al., 2011; Mahajan et al., 2014)."

References: Ovadnevaite, J., Manders, A., de Leeuw, G., Ceburnis, D., Monahan, C., Partanen, A.-I., Korhonen, H., and O'Dowd, C.D.: A sea spray aerosol flux parameterization encapsulating wave state. Atmos. Chem. Phys., 14, 1837–1852, 2014. Jaeglé, L., Quinn, P.K., Bates, T.S., Alexander, B., and Lin, J.-T.: Global distribution of sea salt aerosols: new constraints from in situ and remote sensing observations. Atmos. Chem. Phys., 11, 3137–3157, 2011 de Leeuw, G., Andreas, E. L., Anguelova, M. D., Fairall, C. W., Lewis, E. R., O'Dowd, C., Schulz, M., and Schwartz, S. E.: Production flux of sea spray aerosol, Review, J. Geophys. Res., 49, RG2001, 432,doi:10.1029/2010RG000349, 2011. Street, D.G., Yan, F., Chin, M., Diehl, T., Mathwald, N., Schultz, M., Wild, M., Wu, Y., and Yu, C.: Anthropogenic and natural contributions to regional trends in aerosol optical depth, 1980-2006. J. Geophys. Res., 114, doi:10.1029/2008JD011624, 2009. Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G., Keating, T., Zhang, Q., Kurokawa, J., Wankmüller, R., Denier van der Gon, H., Kuenen, J. J. P., Klimont, Z., Frost, G., Darras, S., Koffi, B., and Li, M.: HTAP_v2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution, Atmos. Chem. Phys., 15, 11411–11432, 2015. Arnold, S.R., Spracklen, D.V., Williams, J., Yassaa, N., Sciare, J., Bonsang, B., Gros, V., Peeken, I., Lewis, A.C., Alvain, S., and Moulin C.: Evaluation of the global oceanic isoprene source and its impacts on marine organic carbon aerosol. Atmos. Chem. Phys., 9, 1253–1262, 2009. Rinaldi, M., Decesari, S., Carbone, C., Finessi, E., Fuzzi, S., Ceburnis, D., O'Dowd, C.D., Sciare, J., Burrows, J.P., Vrekoussis, M., Ervens, B., Tsigaridis, K., Facchini, M.C.: Evidence of a natural marine source of oxalic acid and a possible link to glyoxal, J. Geophys. Res., 116, D16204, doi:10.1029/2011JD015659, 2011. Fu, T.-M., Jacob, D.J., Wittrock, F., Burrows, J.P., Vrekoussis, M., Henze, D.K.: Global budgets of atmospheric glyoxal and methylglyoxal, and implications for formations of secondary organic aerosols, J. Geo-

phys. Res., 113, doi:10.1029/2007JD009505, 2008. Mahajan, A., Prados-Roman, C., Hay, T., Lampel, J., Pöhler, D., Großmann, K., Tschritter, J., Freiß, U. Platt, U., Johnston, P., Kreher, K., Wittrock, F., Burrows, J., Plane, J., and Saiz-Lipez, A.: Glyoxal observations in the global marine boundary layer, J. Geophys. Res., 119, 6160-6169 doi:10.1002/2013JD021388, 2014. Myriokefalitakis, S., Vrekoussis, M., Tsigaridis, K., Wittrock, F., Richter, A., Brühl, C., Volkamer, R., Burrows, J., and Kanakidou, M.; The inïňĆuence of natural and anthropogenic secondary sources on the glyoxal global distribution, Atmos. Chem. Phys., 8(16), 4965 –4981, 2008. Myriokefalitakis, S., Vignati, E., Tsigaridis, K., Papadimas, C., Sciare, J., Mihalopoulos, N., Facchini, M., Rinaldi, M., Dentener, F., Ceburnis, D., Hatzianastasiou, N., O'Dowd, C., van Weele, M. and Kanakidou, M.: Global modeling of the oceanic source of organic aerosols, Advances in Meteorology, 2010, doi:10.1155/2010/939171

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

ANS) This paragraph was removed.

5. 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.

ANS) Yet, direct long-term measurements of aerosol over the global oceans have not been. "A comparison of the large-scale spatial and temporal variability between satellite-based AOD and PM2.5 concentration using monthly mean measurements indicated that the AOD and PM2.5 agree well in terms of their interannual variability (Li et al., 2015)." SSA emission is also directly related to aerosol concentration, assuming the aerosol removal mechanism does not significantly change. Thereby, aerosol changes (trend) by direct emissions and secondary production during 2000-2015 were estimated in terms of emission and AOD changes (AODSSA and AODDMS) from sea spray aerosol and DMS flux. The temporal variation (trend) of MODIS aerosol optical depth (MODIS AOD) resulted from the atmospheric aerosol loading changes by

natural, and anthropogenic emission sources also indirectly reflected ambient aerosol changes during the study period. Discussion on this was added to the "Introduction."

Reference: Li, J., Carlson, B.E., Lacis, A.A.: How well do satellite AOD observations represent the spatial and temporal variability of PM2.5 concentration for the United States? Atmos. Environ. 102, 260-273.2015.

6. 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?

ANS) As the reviewer commented, DMS concentrations in seawater are not perfectly correlated with chlorophyll-a and the parametrization used in certain areas, sometime, may not be correct. The correlation between DMS and chlorophyll-a was re-examined using the updated monthly global DMS climatology dataset by Lana et al. (2012). Lana et al. (2012) showed the good correlation (r2 = 0.47) between DMS and chlorophyll-a concentrations and estimated the seawater DMS uncertainty of −25%/+15% (see Fig. 2). The current available data to estimate the global distribution of DMS concentration in seawater during a long-term period (16 years) are global chlorophyll-a data from the European Space Agency (ESA) GlobColor (www.globcolour.info/). We also performed the sensitivity test of chlorophyll-a to seawater DMS. 25% change in the chlorophyll-a concentration resulted in +1/-2% change in DMS. The related sentences were added to the upper parts of pages 6 and 13.

"A re-examination of the correlation between the DMS in seawater and chlorophyll-a using the updated monthly global DMS climatology data set showed a good correlation (R2 = 0.47) (Lana et al. 2012)." "In addition, a ïĆś25% change in Chl-a gave a +1/-2% change in DMS concentration in seawater."

Reference: Lana, A., Simo, R., Vallina, S., Dachs, J.: Re-examination of global emerging patterns of ocean DMS concentration. Biogeochemistry (2012) 110:173–182, DOI 10.1007/s10533-011-9677-9

Fig. 2. Linear regression of DMS concentration against Chl-a.

7. 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?

ANS) "Recently, Smith et al. (2018) directly measured DMS fluxes made using two independent methods: the eddy covariance (EC) technique and the gradient flux (GF) technique during Feb-Mar 2012 in the southwest Pacific Ocean. Their results showed good agreement between these two independent methods. We evaluated two parametrization methods (kw) of Liss and Merlivat (1986) and Wanninkhof (1992) using Smith et al. (2018) flux measurement data set. Our evaluation of the gas transfer velocity showed the lowest root mean square error (RMSE) between the calculated kw of Liss and Merlivat (1986) and the indirect measurements of kw by Smith et al. (2018) (Supplementary Fig. 2 and see Fig. 4). "According to Elliott (2009), there is also some indication that the lower end values of kw of Liss and Merlivat (1986) may ultimately be preferred." The above sentences were added to the middle-bottom parts of page 6. Thus, our DMS flux estimation is not likely to be significantly underestimated.

References: Smith, M.J., Walker, C.F., Bell, T.G., Harvey, M.J., Saltzman, E.S., and Law, C.S.: Gradient flux measurements of sea–air DMS transfer during the Surface Ocean Aerosol Production (SOAP) experiment, Atmos. Chem. Phys., 18, 5861–5877, 2018, https://doi.org/10.5194/acp-18-5861-2018. Elliott, S.: Dependence of DMS global sea-air flux distribution on transfer velocity and concentration field type, J. Geophys. Res., 114, G02001, doi:10.1029/2008JG000710, 2009. Nightingale, P. D., Malin, G., Law, C. S., Watson, A. J., Liss, P. S., Liddicoat, M. I., Boutin, J., and

Upstill-Goddard, R. C.: In situ evaluation of air-sea gas exchange parameterizations using novel conservative and volatile tracers, Global. Biogeochem. Cy., 14,373–387, 2000.

Fig. 3. Table for summary of environemntal conditions in air and water during gradient flux measurements. Table for Transfer Velocityies Applied in the Present work.

Fig. 4. Comparison of transfer velocity parameterization schemes with observations.

8. 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.

ANS) In this study, we analyzed SSA mass emission rates. However, Quinn et al. (2017) estimated the contribution of SSA to the number concentration of CCN (and total particle number concentrations) in the marine boundary layer using a lognormal-mode-fitting procedure (Aitken, accumulation, and SSA modes). Thus, direct comparison of our result with Quinn et al. (2017) is not reasonable because the number fraction of SSA depends on the Aiken mode, whereas the mass fraction of SSA on a coarse mode. We incorporated finding of Quinn et. al (2017) in the revised manuscript (in the upper part of page 10). "However, SSA emissions makes a contribution of less than 30% to CCN concentrations increasing with wind speed, on a global basis, with the exception of the high southern latitudes (Quinn et al., 2017)." As the reviewer commented, in the biologically productive areas, seawater DMS concentration can be more important than wind speed for sea-to–air DMS flux. However, since most regions of the world's oceans are oligotrophic, i.e., they contain very low levels of nutrients, DMS concentrations in seawater are low (see Fig. 5; Lana et al., 2011). DMS concentrations were typically in the range of 1–7 nM (Lana et al., 2011). According to Fig. 6 in Grythe et al. (2014), the histogram of wind speeds shows wide range with a long-tailed distribution based

on 25 yr of ECMWF data towards higher wind speeds (see Fig. 6 below). For example, if wind speed increases from 5 to 6 m/s, the transfer velocity of LM 86 increases by a factor 1.6. Thus, wind speed is more sensitive to the sea-to-air flux of DMS in global scale.

Fig. 5. Annual global mean climatology of DMS concentrations Histogram of global oceanic 3h, 10m wind speed in the 25yr of ECMWF data (gray bars)

References: A. Lana, T. G. Bell, R. Simó, S. M. Vallina, J. BallabreraâĂŘPoy, A. J. Kettle,J. Dachs, L. Bopp, E. S. Saltzman, J. Stefels, J. E. Johnson, and P. S. Liss. An updated climatology of surface dimethlysulfide concentrationsand emission fluxes in the global ocean. GLOBAL BIOGEOCHEMICAL CYCLES, VOL. 25, GB1004, doi:10.1029/2010GB003850, 2011. Grythe, H., Ström, J., Krejci, R., Quinn, P., and Stohl, A.: A review of sea-spray aerosol source functions using a large global set of sea salt aerosol concentration measurements, Atmos. Chem. Phys., 14, 1277. 2014.

9. 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?

ANS) We found that our DMS flux estimation and trends are not likely to be significantly underestimated (realistic) based on the recent flux measurements by Smith et al (2018). The relevant sentences are added to the upper part of page 6.

10. Lines 251-253: How was the comparison to previous work done? Was it precise by month/season and area?

ANS) In the previous papers (Kattle and Andreae, 2000; Lana et al., 2011), total global DMS fluxes were calculated using both surface seawater DMS measurement data, which are entirely from the Global Surface Seawater DMS Database

(http://saga.pmel.noaa.gov/dms/) plus 63 additional measurements in the South Pacific (Lee et al., 2010), and the gas transfer velocity derived from the Liss and Merlivat (1986) and Wanninkhof (1992) parameterizations for comparison. The annual DMS fluxes were also estimated using monthly data with a resolution of $1^\circ \times 1^\circ$ between -90° and 90° over the global ocean. Meanwhile, in our study, the global DMS fluxes were estimated using both surface seawater DMS concentrations estimated from a DMS empirical algorithm and the gas transfer velocity derived from the Liss and Merlivat (1986), including the monthly data with 25-km spatial resolution between -60° and 60°. Because there were somewhat differences (e.g., DMS measurement and covering area (km2)) between our and previous studies, we indirectly compared with the previous studies. Discussion on this was added to the lower part of page 11. "The flux estimates using the mean flux derived from the region of 60°N–60°S were similar to those derived from the climatological DMS database between 0° and 90° during 1972–1998 (15.0 Tg yr−1, Kettle and Andreae, 2000) and during 1972–2009 (17.6 Tg yr−1, Lana et al., 2011)."

11. 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?

ANS) As the reviewer suggested, we removed these sentences.

12. Line 290: What is the adjustment factor?

ANS) The adjustment factor (ïĄś) in Eq. (1) is an adjustable shape parameter that controls the submicron size distribution. The description of the adjustment factor is given in the section 2.1.

13. 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?

ANS) We added test results of the sensitivity of U10 to the DMS flux and AODDMS to

the middle part of page 13.

"The sensitivity study showed that a ïĆś25% change in U10 gave a +214/−37% change in annual SSA emission rates, +30/-23% change in AODSSA, and +51/-49% (+56/-44%) change in DMS flux for the method reported by Liss and Merlivat (1986) (and Wanninkhof, 1992) in AODDMS." "Finally, a 25% change in DMS flux resulted in a 15% change in AODDMS."

14. Lines 345-349: Did the authors look at forward trajectories for the anthropogenically influenced aerosols? How far can these aerosols go over the ocean?

ANS) No we did not analyze forward trajectories in this study. We used global map of monthly AODs for Aqua MODIS satellite from NASA (https://neo.sci.gsfc.nasa.gov/view.php?datasetId=MYDAL2_M_AER_OD&year=2015, see Fig. 6) and anthropogenic emission strength of particulate matter (PM) from Klimont et al. (2017). We also added references describing the impact of continental outflow and trans-Pacific transport of Asian anthropogenic aerosols and their impact on surface air quality into North America. We added this information to the middle part of page 15.

“Several studies have documented the influence of Asian continental outflow on surface aerosols over the Indian Ocean and northwestern Pacific Ocean, even in North America (Heald et al., 2006; Hadley et al., 2007; Song et al., 2008; Kaneyasu et al., 2014; Srinjvas et al., 2014).”

Fig. 6. Monthly AODs for NASA Aqua Modis Satellite during 2015.

References: Kaneyasu, N., Yamamoto, S., Sato, K., Takami, A., Hayashi, M., Hara, K., Kawamoto, K., Okuda, T., and Hatakeyama, S.: Impact of long-range transport of aerosols on the PM2.5 composition at a major metropolitan area in the northern Kyushu area of Japan, Atmos. Environ. 97, 416-425, 2014. Srinivas, B., and Sarin, M.M.: PM2.5, EC and OC in atmospheric outflow from the Indo-Gangetic Plain: Tem-

poral variability and aerosol organic carbon-to-organic mass conversion factor, Sci. The Total Environ. 487, 196-205, 2014. Heald, C.L., Jacob, D.J., Park, R.J., Alexander, B., Fairlie, T.D., Yantosca, R. M., and Chu, D.A.: Transpacific transport of Asian anthropogenic aerosols and its impact on surface air quality in the United States, J. Geophys. Res., 111, doi:10.1029/2005JD006847, 2006. Hadley, O.L., Ramanathan, V., Carmichael, G.R., Tang, Y., Corrigan, C.E., Roberts, G.C., and Mauger, G.S.: TransâĂŘPacific transport of black carbon and fine aerosols (D < 2.5 μm) into North America, J. Geophys. Res., 111, doi:10.1029/2006JD007632, 2007. Song, S.-K., Kim, Y.-K., Shon, Z.-H., and Lee, H.W.: Influence of meteorological conditions on trans-Pacific transport of Asian dust during spring season, J. Aerosol Sci., 39, 1003-1017, 2008.

15. Results/discussion general: Again, the Quinn et al. (2017) paper was never cited. How do the general results compare with theirs?

ANS) See above reply. We added the findings of Quinn et al. (2017) to the upper part of page 10. "On the other hand, SSA emissions make a less than 30% contribution to the increasing CCN concentrations with wind speed on a global basis, with the exception of high southern latitudes (Korhonen et al., 2008; Quinn et al., 2017)."

16. Conclusion: This is basically just a summary and, again, provides no insight regarding the relevance of the hiatus.

ANS) Conclusion was revised as the reviewer suggested, including short discussion on the global warming hiatus.

17. Tables 1 and 2: Need better description of units

ANS) More description of units was added.

18. 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.

ANS) As the reviewer suggested, the reference (Quinn et al., 2017) was cited in the

revised text.

Reviewer 2

General comments:

After closer reading of this manuscript, I am unable to recommend this manuscript for publication in ACP. The subject of the manuscript is a calculation of inferred/estimated trends in DMS and sea spray aerosol emissions over the years 2000-2015, as well as trends in the AOD contributed by these aerosol sources, which may have occurred as a consequence of variability in wind speed and sea surface temperature during this time period. However, the methods used have such serious deficiencies as to make the results uninterpretable. I was surprised to find no discussion or evaluation of potential instrument biases, discrepancies, retrieval uncertainties, or long-term drifts in satellite measurements here. Without this information, it is impossible to determine whether any trends in the wind speed or SST time series used here are likely to be reliable reflections of real-world trends.

In particular, in the analysis performed here, for both wind speed and sea surface temperature, two different satellites were used with breaks in the long-term time series (U10 from QuikSCAT for 2000-2007, and from ASCAT for 2008-2015; SST from NOAA AVHRR for 2000-2002 and from Aqua MODIS for 2003-2015). Are measurements between these different instruments consistent? Is there a potential that a discontinuity or bias could be introduced in the time series by using different instruments during different periods? Since the analysis focusses on time series, this information is critical to understanding the causes of the inferred trends, and ruling out potential instrument bias.

ANS) Comparisons to independent mooring and shipboard observations by Bentamy et al. (2008), Verspeek et al. (2010), Guo et al. (2018) show that ASCAT wind speed has accuracies similar to QuikSCAT. Furthermore, Bentamy et al. (2012) examined a set of space-time collocated observations for QuikSCAT (QS) and ASCAT (AS) and triply

collocated satellite-satellite-buoy observations during their period of overlap (November 2008 through November 2009). An examination of the collocated winds shows a high degree of agreement in direction, but reveals systematic differences in wind speed that depend on rain rate, the strength of the wind, and SST. Time mean difference (ïĄĎW=WQS-WAS) between collocated QS and AS 10 m neutral wind speed estimates is generally less than 1 m/s (Bentamy et al. (2012), Figs. 3 and 4). After applying the correction function, 0.18 m/s positive bias in ïĄĎW was estimated (Bentamy et al. (2012), Figure 9 and Guo et al. (2018), Table 2). The biases of QS and AS between buoy-derived mean wind speeds and satellite-derived mean wind speeds at a height of 10 m above sea level are 0.23 and 0.09 m/s, respectively (Guo et al., 2018). The slopes between buoy-derived mean wind speeds vs QuikSCAT and that vs ASCAT are 1.03 and 1.01, respectively (Guo et al., 2018, Table 2). Thus, we compared European Centre for Medium-Range Weather Forecasts (ECMWF, www.ecmwf.int) reanalysis data with QukSCAT and ASCAT wind speeds (see Fig. 7). For accurate trend analysis with observation consistency, we reanalyzed entire trend analysis using monthly U10 and SST data at 25-km spatial resolution obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF, www.ecmwf.int) for 2000-2015.

Fig.7. Comparison of QuikSCAT and ASCAT SSW (U10) with ECMWF reanalysis data.

1. L. 122-123: The first reviewer has already pointed out some of the (well-known) challenges in estimating oceanic DMS concentrations. Some comparison with observed ocean DMS concentrations and fluxes would be useful in evaluating how well this approach produces results that resemble the real world. The calculation of AOD associated with marine aerosol sources, as described in the manuscript, is also overly simplistic and will not produce interpretable results: - L.

ANS) As the reviewer commented, DMS concentrations in seawater are not perfectly correlated with chlorophyll-a and the parametrization used in certain areas, sometime, may not be correct. The correlation between DMS and chlorophyll-a was re-examined using the updated monthly global DMS climatology dataset by Lana et al. (2012). Lana

et al. (2012) showed the good correlation (r2 = 0.47) between DMS and chlorophyll-a concentrations and estimated the seawater DMS uncertainty of −25%/+15% (see Fig. 2). The current available data to estimate the global distribution of DMS concentration in seawater during a long-term period (16 years) are global chlorophyll-a data from the European Space Agency (ESA) GlobColor (www.globcolour.info/). We also performed the sensitivity test of chlorophyll-a to seawater DMS. 25% change in the chlorophyll-a concentration resulted in +1/-2% change in DMS. The related sentences were added to the upper parts of pages 6 and 13.

"A re-examination of the correlation between the DMS in seawater and chlorophyll-a using the updated monthly global DMS climatology data set showed a good correlation (R2 = 0.47) (Lana et al. 2012)." “In addition, a ïĆś25% change in Chl-a gave a +1/-2% change in DMS concentration in seawater.”

Fig. 2. Linear regression of DMS concentration against Chl-a.

"Recently, Smith et al. (2018) measured directly the DMS fluxes made using two independent methods: the eddy covariance (EC) technique and the gradient flux (GF) technique during Feb-Mar 2012 in the southwest Pacific Ocean. Their results showed good agreement between these two independent methods. . The two parametrization methods (kw) of Liss and Merlivat (1986) and Wanninkhof (1992) were evaluated using flux measurement data set reported by Smith et al. (2018). The evaluation of the gas transfer velocity showed the lowest root mean square error (RMSE) between the kw calculated from Liss and Merlivat (1986) and indirect measurements of kw by Smith et al. (2018) using the mean wind speeds, DMS concentrations, and flux measurements (Supplementary Fig. 2). According to Elliott (2009), there is also some indication that the lower end values of kw of Liss and Merlivat (1986) may ultimately be preferred. Therefore, in this study, the Liss and Merlivat (1986) approach was used to calculate the DMS fluxes during the study period." These sentences were added to the parts of pages 6 and 7. Thus, our DMS flux estimation is not likely to be significantly underestimated (see Fig. 3 and 4).

Fig. 3. Table for summary of environemntal conditions in air and water during gradient flux measurements. Table for Transfer Velocityies Applied in the Present work.

Fig. 4. Comparison of transfer velocity parameterization schemes with observations.

The AOD calculation procedure is not simplistic, which use the aerosol size distribution with lognormal-mode-fitting procedure, microphysical and optical properties of aerosols, etc. Description on this is given in the upper part of page 8 and detailed information the OPAC model is given Hess et al. (1998).

References: Lana, A., Simo, R., Vallina, S., Dachs, J.: Re-examination of global emerging patterns of ocean DMS concentration. Biogeochemistry (2012) 110:173–182, DOI 10.1007/s10533-011-9677-9 Smith, M.J., Walker, C.F., Bell, T.G., Harvey, M.J., Saltzman, E.S., and Law, C.S.: Gradient flux measurements of sea–air DMS transfer during the Surface Ocean Aerosol Production (SOAP) experiment, Atmos. Chem. Phys., 18, 5861–5877, 2018, https://doi.org/10.5194/acp-18-5861-2018. Elliott, S.: Dependence of DMS global sea-air flux distribution on transfer velocity and concentration field type, J. Geophys. Res., 114, G02001, doi:10.1029/2008JG000710, 2009. Nightingale, P. D., Malin, G., Law, C. S., Watson, A. J., Liss, P. S., Liddicoat, M. I., Boutin, J., and Upstill-Goddard, R. C.: In situ evaluation of air-sea gas exchange parameterizations using novel conservative and volatile tracers, Global. Biogeochem. Cy., 14,373–387, 2000.

2. L. 144-158: It's not possible to realistically calculate the AOD from emitted aerosol without treatment of its transport, microphysical evolution, and removal in the atmosphere. The distribution of SSA concentrations in the atmosphere is controlled by removal processes (e.g. the distribution of precipitation) at least as much as by emission processes.

ANS) The OPAC model used in this study needs aerosol number concentration to simulate the AOD. The aerosol number concentration is affected by emission, transport, heterogeneous reactions, microphysical evolution, removal in the air, etc. In this study,

we cannot use 3-D chemical transport model to simulate aerosol number concentration for AOD calculation over the global oceans during 2000-2015 due to computing time limitation. Thus, we used Lewis and Schwartz (2004) parameterization method for the aerosol number concentration. The OPAC model is the widely used method in AOD calculation (see Google search below).

Fig. 8. Literature searches for OPAC model application to the AOD calculation

3. L. 159-169: It's not possible to generate reasonable distributions of DMS derived sulfate aerosol in this way, with no consideration of atmospheric transport and chemistry (reaction with OH and subsequent reaction mechanisms), which occurs over time scales of several days to weeks. There is a large literature on the production of sulfate aerosol from DMS-derived SO2, which shows the critical role of transport and chemistry processes in determining the contribution of DMS-derived SO2 to global aerosol concentrations. As just one example of this, in a paper that is referenced by the authors, Korhonen et al. (2008), Figure 8 clearly shows that the geographic distributions DMS emissions, and of the increase in cloud condensation nuclei number associated with DMS emissions, are nearly uncorrelated. These are major flaws in the methodology that need to be addressed before any of the results will be interpretable.

ANS) We evaluated the mass contribution of DMS to atmospheric aerosol loadings (column density). Thus, direct comparison of our result with Korhonen et al. (2008) is not reasonable because they examined the effect of DMS emission on CCN number concentration. Our results also showed low contribution (i.e., nearly uncorrelation) in the NH, where anthropogenic sulfur sources are overwhelming. A recent study by Quinn et al. (2017) found that Aitken-mode particles make up a large fraction of the CCN in the Southern Hemisphere at high supersaturations (>0.5%). Note that the timescale for oxidation of DMS is on the order of hours to one day. Longer temporal scale might not observe correlation of monthly data with CCN number concentration. Woodhouse et al. (2003) suggested that changes in the spatial distribution of DMS emissions (through changes in the phytoplankton population or wind speed patterns)

could exert a stronger control on climate (through CCN concentration change) than net increases in biological productivity. Seasonal covariance of aerosol and methane sulfonic acid (MSA), an aerosol phase oxidation product of dimethyl sulfide (DMS), was also used by Ayers and Gras (1991) and Ayers et al. (1991) as evidence supporting a major role of a marine biogenic reduced-sulfur source in driving the CCN seasonal concentration cycle. Thus, open questions remain on the source of MBL NSS-sulfate with possibilities including DMS oxidation in the MBL that leads to particle growth to the accumulation-mode size range through vapor condensation and accumulation of mass during cloud processing (Hegg et al., 1992; Hoppel et al., 1994). Note that our results are not number concentration, but AOD which indirectly semi-quantify the aerosol mass loading. As mentioned before, due to the limitation of computing time for 16 years simulation over the global oceans, we used our best approach to estimate the AODDMS based on DMS flux and sulfate aerosol number concentration. Our results showed similar result to Gondwe et al. (2003). For example, our mean contribution of DMS (∼50%) to AOD in the Southern Hemisphere were similar to mean annual contribution of DMS to the climate-relevant non-sea-salt sulfate column burden (43%) by Gondwe et al. (2003). Our mean contribution in the Northern Hemisphere were less (8% during MAM and JJA), where anthropogenic sulfur sources are overwhelming. Gondwe et al. (2003) reported 9% in the Northern Hemisphere. In a previous study, the contribution of biogenic sulfur (DMS) in fine particle mode over the Atlantic Ocean was less than 35% of the excess sulfur in the NH (0ïĆř–60ïĆřN), and approximately 60% in the SH (0ïĆř–35ïĆřS, Patris et al., 2000). Our mean DMS-derived sulfate contributions to atmospheric aerosol loading were similar to these values, 30% in the NH and 50% in the SH (51% for 0ïĆř–30ïĆřS). This suggested that out approach was not unrealistic.

References: Ayers, G. P. and Gras, J. L.: Seasonal relationship between cloud condensation nuclei and aerosol methanosulfonate on marine air, Nature, 353, 834–835, 1991. Ayers, G. P., Ivey, J. P., and Gillett, R. W.: Coherence between seasonal cycles of dimethyl sulfide, methanosulfonate and sulfate in marine air, Nature, 349, 404–406, doi:10.1038/349404a0, 1991. Gondwe, M., Krol, M., Gieskes, W.,

Klaassen, W., de Baar, H.: The contribution of ocean-leaving DMS to the global atmospheric burdens of DMS, MSA, SO2, and NSS SO42-. Global Biogeochem. Cy. 17, https://doi.org/10.1029/2002GB001937. 2002. Patris, N., Mihalopoulos, N., Baboukas, E.D., and Jouzel, J.: Isotopic composition of sulphur in size-resolved marine aerosols above the Atlantic Ocean, J. Geophys. Res., 105, 14449-14457. 2000. Quinn, P.K., Coffman, D.J., Johnson, J.E., Upchurch, L.M., and Bates, T.S.: Small fraction of marine cloud condensation nuclei made up of sea spray aerosol, Nat. Geo. 10, http://dx.doi.org/10.1038/ngeo3003.2017. Woodhouse, G.W. Mann, K.S. Carslaw, and Boucher O.: Sensitivity of cloud condensation nuclei to regional changes in dimethylsulphide emissions. Atmos. Chem. Phys., 13, 2723–2733, 2013. Ayers, G.P. and Gillett, R.W., DMS and its oxidation products in the remote marine atmosphere: implications for climate and atmospheric chemistry. Journal of Sea Research Volume 43, Issues 3–4, August 2000, Pages 275-286

In addition, I don't understand the underlying motivation for this study. The so-called "hiatus" period from 1998-2012 has been explained as most likely related to decadal scale internal modes of climate variability, in particular, the Pacific Decadal Oscillation. It's unclear to me why this period should be of particular interest for marine aerosol emissions. If the authors choose to revise this paper, they may also find the following specific comments useful:

4. L. 41-42: "However, the rate of warming (0.05 deg C per decade) over the last 15 years (1998–2012) was smaller than that (0.12 deg C per decade) calculated for 1951–2012 (IPCC, 2013)." This sentence needs to be contextualized to avoid potentially misleading readers. The 1951-2012 period represents 61 years of climate data, while the 1998-2012 period represents only 15 years; a fifteen-year period is well known to be too short to reliably detect long-term climate trends in global mean surface air temperature, and I think this should be explicitly noted here. For example, research at the UK Met Office on understanding the slowdown in warming during this fifteen-year period, often termed a "hiatus" in global warming, found that in model simulations of a

climate warming at a rate of 0.2 deg C per year, periods of cooling occur frequently. For example, a -0.10 deg/yr cooling period lasting fifteen years occurs more than 10% of the time (Roberts et al., Nature, 2015; https://www.nature.com/articles/nclimate2531). In light of such analyses, this “hiatus” period is more realistically viewed as a part of the expected natural variability in the climate system.

ANS) The main goal of this study is to examine whether there is a long-term trends of global marine primary and secondary aerosol production during 2000-2015 or not. We do not focus on the reasons of recent hiatus (1998-2012). As the reviewer suggested, we revised the text (adding a following paragraph). “The recent hiatus was attributed to some combination of external climatic forcing that are not represented adequately in the model simulation and the internal (natural) climate variation (IPCC 2013 and references therein). Roberts et al. (2015) estimated an approximate 10% probability of a 10-year warming hiatus due to internal variability given an expected contemporary warming rate of approximately 0.2ïĆřK per decade.”

5. L. 43-44: This sentence should be updated: “According to a recent study (England et al., 2014), the global mean SAT has remained flat since around 2001.“. It is now 2018; the global mean surface air temperature has clearly risen between 2001 and 2018.

ANS) The sentence was removed.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-322, 2018.

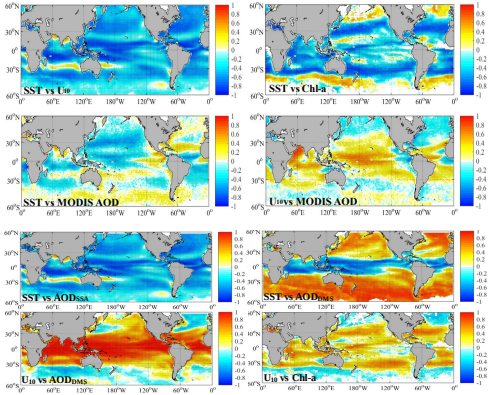

Fig.1 Distribution of correlation of a) SST vs. U_{10}, b) SST vs. Chl-a, c) SST vs. MODIS AOD, d) U_{10} vs MODIS AOD, e) SST vs. AODSSA, f) SST vs. AODDMS, g) U10 vs. AODDMS, and h) U10 vs Chl-a.

Fig. 1. Distribution of correlation of (a) SST vs. U10, (b) SST vs. Chl-a, (c) SST vs. MODIS AOD, (d) U10 vs. MODIS AOD, (e) SST vs. AODSSA, (f) SST vs. AODDMS, (g) U10 vs. AODDMS, and h) U10 vs. Chl-a.

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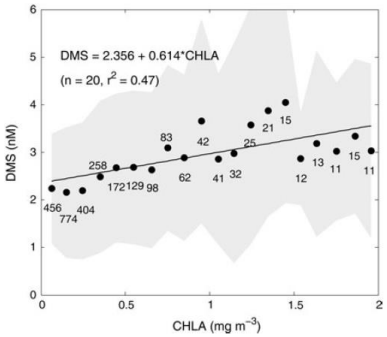

Fig. 6 Linear regression of DMS concentrations (L10 climatology) averaged by 10° latitude × 20° longitude boxes against Chla concentrations binned in 0.1 mg m^{-3} intervals

Fig. 2. Linear regression of DMS concentration against Chl-a.

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Table 1. Summary of environmental conditions and DMS concentrations in air (C_a) and water (C_w) during gradient flux measurements.

Deployment no.	DOY (UTC)	Time (NZST)	Mean wind speed (m s^{-1})	C_a (ppt)	Mean C_w (nM)	Mean flux (μmol m^{-2} d^{-1})	Wave Hgt (m)	Wave age	Distance ship–cat (km)
1	48.08–48.23	17 Feb 14:00–18:30	5.9	300–500	16.5	10	1.7	3.2	0.3–3
2	49.08–49.3	18 Feb 14:30–18:00	6.0	350–750	17.0	30 15–45	1.6	3.4	1–3
3	53.10–53.23	22 Feb 14:30–18:00	4.3	285–400	13.9	5	1.9	4.0	1–3
4	58.06–58.25	27 Feb 13:30–17:00	7.7		6.4	11	2.0	2.6	0.7–2
5	60.06–60.21	29 Feb 13:30–17:00	7.2	175–340	5.0	10	2.1	2.3	0.6–13
6	64.85–65.0	5 Mar 08:30–12:00	3.6	40–210		7.5	3.3	5.6	1–2.6

Table 1. Transfer Velocities Applied in the Present Work[a]

Scheme	LM86	W92	N00	B07
$U_{10} < 3.6$	$c_1U_{10}(Sc_t/Sc)^{2/3}$	$c_1U_{10}^2(Sc_t/Sc)^{1/2}$	$c_1U_{10}(Sc_t/Sc)^{1/2}$ $+ c_2U_{10}^3(Sc_t/Sc)^{1/2}$	W92
$3.6 \le U_{10} < 5.6$	$c_2(U_{10}-3.6)(Sc_t/Sc)^{1/2}$ $+ c_3(Sc_t/Sc)^{2/3}$	$c_1U_{10}^2(Sc_t/Sc)^{1/2}$	$c_1U_{10}(Sc_t/Sc)^{1/2}$ $+ c_2U_{10}^3(Sc_t/Sc)^{1/2}$	W92(5.6-U_{10})/2.0 + LM86(U_{10}-3.6)/2.0
$5.6 \le U_{10} < 13$	$c_2(U_{10}-3.6)(Sc_t/Sc)^{1/2}$ $+ c_3(Sc_t/Sc)^{2/3}$	$c_1U_{10}^2(Sc_t/Sc)^{1/2}$	$c_1U_{10}(Sc_t/Sc)^{1/2}$ $+ c_2U_{10}^3(Sc_t/Sc)^{1/2}$	LM86
$13 \le U_{10}$	$c_4(U_{10}-13)(Sc_t/Sc)^{1/2}$ $+ c_5(Sc_t/Sc)^{1/2}$ $+ c_6(Sc_t/Sc)^{2/3}$	$c_1U_{10}^2(Sc_t/Sc)^{1/2}$	$c_1U_{10}(Sc_t/Sc)^{1/2}$ $+ c_2U_{10}^3(Sc_t/Sc)^{1/2}$	LM86 format from regime $5.6 \le U_{10} < 13$ continued to >13
Constants	$c_1 = 0.17$ $c_2 = 2.85$ $c_3 = 0.612$ $c_4 = 5.90$ $c_5 = 26.79$ $c_6 = 0.612$	$c_1 = 0.31$	$c_1 = 0.333$ $c_2 = 0.222$	See LM86, W92
References	CO_2 in freshwater 20°C (Sc_t = 600)	CO_2 in seawater 20°C (Sc_t = 660)	See LM86	See LM86, W92
Data	Low U: Theory Mid U: Lake data High U: Tunnel experiments	Large-scale bomb and radiocarbon	He, SF_6 and an inert, involatile dilution tracer	Tropical eddy correlation

[a]Definitions: Scheme abbreviations are as indicated in the text. LM86 [*Liss and Merlivat*, 1986], W92 [*Wanninkhof*, 1992], N00 [*Nightingale et al.*, 2000], and B07 (Blend, 2007). U_{10} is wind speed referenced to the 10 m level and expressed in m s^{-1}. Column summations give piston velocity in cm h^{-1}. The Schmidt number is dimensionless.

Fig. 3. Tables

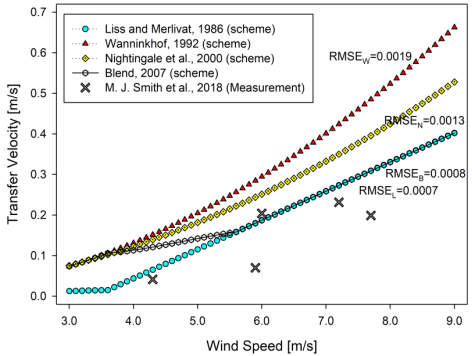

Fig. 4. Comparison of transfer velocity parameterization schemes with observations.

Fig. 4. Fig. 4. Comparison of transfer velocity parameterization schemes with observations.

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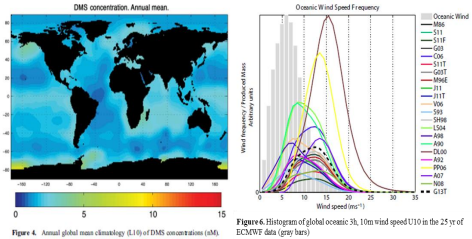

Figure 4. Annual global mean climatology (L10) of DMS concentrations (nM).

Figure 6. Histogram of global oceanic 3h, 10m wind speed U10 in the 25 yr of ECMWF data (gray bars)

Fig. 5. Annual global mean climatology of DMS concentrations and Histogram of global oceanic 3h, 10m wind speed in the 25yr of ECMWF data (gray bars)

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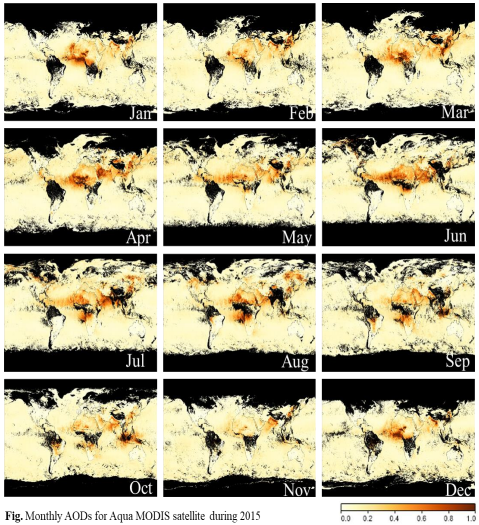

Fig. Monthly AODs for Aqua MODIS satellite during 2015

Fig. 6. Monthly AODs for NASA Aqua Modis Satellite during 2015

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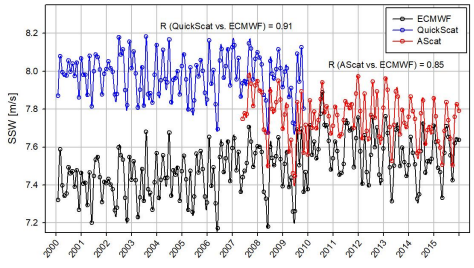

Fig.7 Comparison of QuikSCAT and ASCAT SSW (U10) with ECMWF reanalysis data.

Fig. 7. Comparison of QuikSCAT and ASCAT SSW (U10) with ECMWF reanalysis data.

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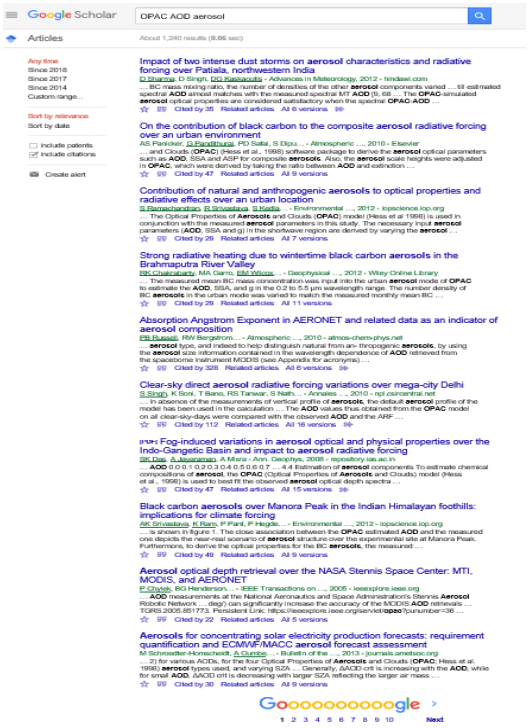

Fig. 8. Literature searches for OPAC model application to the AOD calculation

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