We thank the reviewer for their comments. Below we reproduce the comments in blue and provide our discussion in black.

Lines 62 – 63: This statement (". . .this view (the CLAW hypothesis) has been debated as primary sea salt aerosols. . ..have been hypothesized to be a more robust source of CCN than DMS-derived aerosols") is an inaccurate and oversimplified summary of Quinn and Bates (2011). There are at least three factors that prevent DMS-derived sulfate from being involved in a climate feedback mechanism – 1) there are additional sources of CCN in the MBL (e.g., sea spray), 2) nucleation of new particles from DMS sulfur occurs in the free troposphere prohibiting local feebacks, and 3) the connection between increased CCN and changes in aerosol-cloud interactions is more complicated than depicted in Charlson et al. (1987).

We agree with the reviewer that the original statement was oversimplified. We revised it to read, "More recently, it has been shown that there are factors, such as additional sea salt sources of CCN in the MBL or complex interactions between CCN and aerosol-cloud interactions, that prevent DMS-derived sulfate from being directly involved in a climate feedback mechanism (Quinn and Bates, 2011).

Lines 109 – 111: NAAMES took place in the western North Atlantic region – west of 30W.

This has been rewritten as "Other recent field measurements focusing on the aerosol chemistry of the North Atlatic region include the NASA North Atlantic Aerosols and Marine Ecosystems Study (NAAMES), which included both aircraft and shipborne observations focused on marine biological productivity of the Western North Atlantic (Behrenfeld et al., 2019)…"

Lines 123 – 124: Perhaps rephrase this statement as "A unique feature of the ACEENA aircraft deployments is the seasonally-resolved measurement IN THE EASTERN NORTH ATLANTIC. . ." as NAAMES also had seasonally resolved aircraft flights but in the western NA.

This has been revised according to reviewer's suggestion.

Section 3.1.1 and throughout: To avoid confusion, it should be pointed out that total particulate sulfate measured with the AMS is non-sea salt sulfate.

We have added the non-sea salt qualifier to sulfate.

Lines 190 – 209: It would be interesting to add a comparison with NAAMES seasonal sulfate values. See Saliba et al., JGR, 125, doi: 10.1029/2020JD033145 (2020) and Sanchez et al. Sci. Rep. 8, 3235, doi: 10.1038/s41598-018-21590-9 (2018).

We added the comparison, "The NAAMES cruises in the Western North Atlantic provide another point of comparison: in the winter, the loadings of organic, non-sea salt sulfate, ammonium and nitrate were 0.14/0.56, 0.15/0.48, <0.01/0.01 and 0.01/0.03, respectively, and in the late spring they were 0.61/1.62, 0.44/0.64, <0.01/0.13 and 0.02/0.14, respectively, for marine/continental airmass origin (Saliba et al., 2020)."

Lines 210 – 220: Seasonal concentrations of DMS and MSA from NAAMES could be compared to the values measured here. See Quinn et al., JGR, 124, 14240 – 14261, 2019. In addition, the MSA to non-sea salt sulfate ratio measured here during the summer (<10%) should be compared to previously reported ratios in remote marine regions during the spring/summer.

We now compare our reported MSA measurements to NAAMES measurements reported in Quinn, et al. (2019), "Similarly, during the NAAMES cruises, MSA concentrations measured with ion chromatography were reported as 0.07 μg m^{-3} in the late spring, and 0.01 μg m^{-3} in both March and September (Quinn et al., 2019; Saliba et al., 2020).".

We decided against comparing our DMS values to NAAMES, as our values potentially suffer from interferences in the summer, as explained in the paper, and we don't want to be misleading about the accuracy of our DMS measurements.

We also added the following discussion of MSA to non-sea sulfate ratio, "The MSA to non-sea salt sulfate ratio (MSA:SO_4) measured during ACE-ENA in the summer was 0.02 on average in the MBL (<1000 m), which is lower than historical estimates of the ratio. For example, Pszenny et al. (1990) reports the ratio as 0.05 in the North Atlantic in August-September, Berresheim et al. (1991) reports 0.033 in Western North Atlantic in September, Savoie et al. (2002) reports 0.06 in Bermuda in September and 0.05 in Mace Head in August. Huebert et al. (1996) found 0.07 in marine air masses and 0.02 in continental air masses in June in the Azores. The measurements reported in these earlier studies are based on analysis of filter samples, and they may not be directly comparable to AMS measurements reported here."

Line 229 – 230: This result (MSA does not account for the majority of the particulate sulfate mass in the MBL) is not new and should be noted as such by providing appropriate references.

This sentence was misleading and was removed.

Line 236: Figure 9 is mentioned before Figures 5 to 8.

Discussion of Figure 9 was moved to Section 3.2

Lines 256 – 257: What was used to designate RF #9 and #19 as having lower and higher influence from long range transport?

The classification of those cases was done on the basis of vertical profiles, RF #19 has a clear layer of black carbon, sulfate and ammonium above the boundary layer (1000 – 3000 m), as seen in Figures 8 and 10. This is also correlated with increased methanol and acetone (Figure 11), all markers for continental transport. RF #9 looks very clean in comparison with very little black carbon, ammonium or trace gases other than DMS. We added this explanation, "To further test this, two representative research flights, RF #9 and #19 were selected to represent conditions with lower (RF #9) and higher (RF #19) influence from long-range transport on the basis of concentrations of black carbon, ammonium, methanol and acetone in the free troposphere, which are all markers for long-range transport."

Lines 275 – 276: Are measured levels of isoprene and monoterpene consistent with this statement, i.e., are they large enough to provide the third source of organic aerosol at ENA? Based on Figure 11, there is no significant surface (marine) source of isoprene even in summer.

It is correct that we did not measure significant concentrations of isoprene and monoterpenes. However, given that our detection limit for isoprene is approximately 100 ppt (quantified as 3σ of blank measurements) and that acidic aerosols significantly enhance the SOA production from isoprene and its oxidation products, we can't completely rule out this mechanism. We added the following explanation, "While the isoprene concentrations measured during ACE-ENA were low and close to the detection limit of 0.1 ppb (Table 2) SOA formation from acid-catalyzed IEPOX chemistry has been shown to be significantly more efficient than from non-IEPOX isoprene photochemical mechanisms (Surratt et al., 2010)."

Lines 278 – 286: The result that methanol concentrations are larger than DMS in the summertime should be provided with a caveat that reflects the results shown in Figure 11, i.e., methanol concentrations are higher aloft (~2000 m) while DMS concentrations are lower near the surface.

We added, "Methanol concentrations were higher above the boundary layer (>1000 m), but lower near the ocean surface, while DMS showed the opposite trend of high concentrations at the surface and low concentrations at high altitudes."

Lines 297 – 298: The winter surface source of sulfate appears to only be significant in RF 34. What would a winter source of sulfate be? DMS concentrations should be quite low. Figure 11b indicates that DMS surface concentrations were low during RF 34.

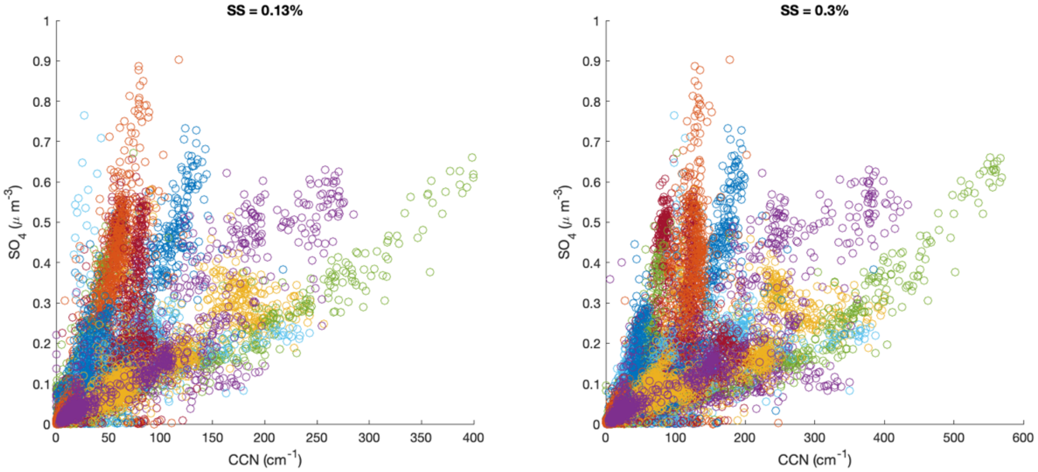
It is unclear what the source of sulfate is in RF #34, in the absence of DMS. Possibilities include remote transport that has mixed downward or an isolated local source, perhaps from a particularly polluting ship transiting through the area. The reviewer is correct to point out that sulfate has a surface source in the summer, and a high altitude source in both seasons. We clarified as follows, "Sulfate aerosol has two distinct sources, a marine source seen below 1000 m in the summer, and a high altitude source above 1000 m present in both seasons."

Line 327: 0.1 or 0.13%S?

It's 0.13%. This was fixed.

Figures S3 and S4: There appears to be two different populations showing up in Figure S3d and S4d. What is causing this split response between CCN and AMS SO4? It looks like it could be partially responsible for the low r^2 values.

The reviewer raises an interesting question. Unfortunately, it is difficult to know exactly why the sulfate/CCN plots appear to show different populations while the organic/CCN plots do not. Our hypothesis is that this is related to the aerosol size distribution and mixing state. As described in the manuscript, sulfate has both remote and local sources. New particle formation from DMS-derived sulfate would produce particles that are likely smaller and less CCN-active (due to their size) on average than particles from long-range transport. In contrast, the organic mass is likely to be largely from remote sources and therefore likely concentrated in larger sized particles. Unfortunately, we do not have size-dependent chemical measurements to test this hypothesis. The figure below, in which the colors indicate different flights, lends credibility to this hypothesis, as it appears that the SO_4 vs CCN correlations cluster by flight.



Lines 345 – 349 and Fig. S5: It is really difficult to see that winter concentrations of chlorophyll are higher than summer concentrations in this figure.

Added insets to the figure that show this better.

References

Berresheim, H., Andreae, M. O., Iverson, R. L. and Li, S. M.: Seasonal variations of dimethylsulfide emissions and atmospheric sulfur and nitrogen species over the western north Atlantic Ocean, Tellus B: Chemical and Physical Meteorology, 43(5), 353–372, https://doi.org/10.3402/tellusb.v43i5.15410, 1991.

Huebert, B. J., Zhuang, L., Howell, S., Noone, K. and Noone, B.: Sulfate, nitrate, methanesulfonate, chloride, ammonium, and sodium measurements from ship, island, and aircraft during the Atlantic Stratocumulus Transition Experiment/Marine Aerosol Gas Exchange, J. Geophys. Res., 101(D2), 4413–4423, https://doi.org/10.1029/95JD02044, 1996.

Pszenny, A. A. P., Harvey, G. R., Brown, C. J., Lang, R. F., Keene, W. C., Galloway, J. N. and Merrill, J. T.: Measurements of dimethyl sulfide oxidation products in the summertime North Atlantic marine boundary layer, Global Biogeochem. Cycles, 4(4), 367–379, https://doi.org/10.1029/GB004i004p00367, 1990.

Quinn, P. K. and Bates, T. S.: The case against climate regulation via oceanic phytoplankton sulphur emissions, Nature, 480(7375), 51–56, https://doi.org/10.1038/nature10580, 2011.

Quinn, P. K., Bates, T. S., Coffman, D. J., Upchurch, L., Johnson, J. E., Moore, R., Ziemba, L., Bell, T. G., Saltzman, E. S., Graff, J. and Behrenfeld, M. J.: Seasonal Variations in Western North Atlantic Remote Marine Aerosol Properties, J. Geophys. Res. Atmos., 124(24), 14240–14261, https://doi.org/10.1029/2019JD031740, 2019.

Saliba, G., Chen, C., Lewis, S., Russell, L. M., Quinn, P. K., Bates, T. S., Bell, T. G., Lawler, M. J., Saltzman, E. S., Sanchez, K. J., Moore, R., Shook, M., Rivellini, L., Lee, A., Baetge, N., Carlson, C. A. and Behrenfeld, M. J.: Seasonal Differences and Variability of Concentrations, Chemical Composition, and Cloud Condensation Nuclei of Marine Aerosol Over the North Atlantic, J. Geophys. Res. Atmos., 125(19), https://doi.org/10.1029/2020JD033145, 2020.

Savoie, D. L.: Marine biogenic and anthropogenic contributions to non-sea-salt sulfate in the marine boundary layer over the North Atlantic Ocean, J. Geophys. Res., 107(D18), 4356, https://doi.org/10.1029/2001JD000970, 2002.

Surratt, J. D., Chan, A. W. H., Eddingsaas, N. C., Chan, M., Loza, C. L., Kwan, A. J., Hersey, S. P., Flagan, R. C., Wennberg, P. O. and Seinfeld, J. H.: Reactive intermediates revealed in secondary organic aerosol formation from isoprene, Proceedings of the National Academy of Sciences, 107(15), 6640–6645, https://doi.org/10.1073/pnas.0911114107, 2010.