General Comments

This manuscript presents the linking of marine aerosols to oceanic biological and meteorological parameters that were estimated by residence-time-weighted air mass transport history. The paper appears to be original and to provide a valuable dataset obtained during four field campaigns. However, there are a few scientific issues to be addressed before the paper can be accepted for publication.

We sincerely appreciate the reviewer's feedback and believe their comments have led to an improved manuscript. We have responded to the reviewer's comments with the blue text below.

Specific Comments

As the authors highlighted, the biological activities of the surface ocean have an important influence on the physiochemical properties of marine aerosols. Further clear explanation of the biological characteristics of the study area would greatly benefit the paper. These biological characteristics may include the following: 'the main phytoplankton species, because the emission of biogenic VOCs is highly species-specific', 'differences in mean chlorophyll-a concentration and net primary production (NPP) for four field campaigns', and 'major biological pathway in oceanic VOCs production'.

We agree and have added the additional details that the reviewer is seeking to the Introduction, Methods, and Conclusions as follows:

In Introduction:

> "Analysis of phytoplankton taxonomy and its seasonal variability in the NAAMES region is presented by Bolanos et al. (2020). Bolanos et al. (2020) show cyanobacteria dominated subpolar waters during the winter and were a significant fraction in the subtropics, with taxa varying by latitude. In-addition, prasinophyta accounted for a significant contribution of subtropical species, with stramenopiles representing less than 30% of subtropical communities. Spring communities had significantly more diverse communities and significantly less cyanobacteria (<10%) relative to the winter, with the exception of one station. Prasinophyta dominated the spring phytoplankton composition, though taxonomic compositions differed from the winter period and between the subpolar and subtropical regions. Typically, diatoms are assumed to be the dominant phytoplankton species in blooms. However, diatoms only represent 10-40% of phytoplankton biomass in the spring bloom surveyed during NAAMEs. The phytoplankton functional groups present influence the overall isoprene production rate and, therefore, the marine atmospheric aerosol and VOC concentrations. Booge et al. (2016) compiled chlorophyll-normalized isoprene production rates from the literature to identify differences between phytoplankton species. The chlorophyll-normalized isoprene production rates varied from 4.56-27.6, 1.4-32.16, and 1.12-28.48 (µmol (g chlorophyll-a) $^{-1}$ day^{-1}) for cyanobacteria, prasinophyta and diatoms, respectively, indicating emissions for isoprene vary significantly with taxonomy. To further complicate the emission strength of VOCs, emissions can vary by production pathways, such as

photosynthetic byproducts, bacterial degradation of dissolved organic matter, and zooplankton grazing on marine phytoplankton (Gantt et al., 2009; Shaw et al., 2003; Sinha et al., 2006)."

In methods:

"For this study chlorophyll-normalized VOC production rates were not considered because of the large variability in observed values (Booge et al., 2016) and the overall unknown contributions from various VOC species to marine particle mass concentrations."

Updated text in conclusion:

"Future studies are needed to 1) understand how differences in subtropical and subarctic phytoplankton speciation may influence aerosol concentrations (Bolaños et al., 2020) and 2) quantify the contribution of transported aerosols to the marine CCN budget and how those may impact (or even dominate) the relationships we have identified in the remote North Atlantic."

Air mass transport history, combined with biological and meteorological parameters, was used to estimate environmental factors controlling marine aerosols and VOC in this study. Similar analyses have been conducted previously (Arnold et al., 2010; Park et al., 2018); hence, the authors should definitely explicitly explain what aspects of their work are novel and of significance.

We thank the reviewer for pointing out this literature. The analysis performed by Arnold et al. (2010) and Park et al. (2018) are similar, however what really sets our manuscript apart is that we compared the influence of biomass on aerosol rather than VOCs (with the exception of DMS). We have added the references in the following relevant sentence in the introduction:

"Previous literature hints that phytoplankton activity is related to emissions of organic and sulfate aerosol mass precursors (Altieri et al., 2016; Arnold et al., 2010; Ayers et al., 1997; Bates et al., 1998; Brüggemann et al., 2018; Ceburnis et al., 2011; Facchini et al., 2008; Hallquist et al., 2009; Hu et al., 2013; Huang et al., 2018; Mansour et al., 2020; Ovadnevaite et al., 2014; Park et al., 2017; Quinn et al., 2019; Sanchez et al., 2018)."

We have also added the following statement in the results section:

"The DMS correlation with chlorophyll-a during the bloom period is consistent with results from similar analyses performed by Arnold et al. (2010) and Park et al. (Park et al., 2018), where DMS measurements were collected in the South Atlantic and Arctic, respectively."

The authors need to provide time series measurement results for key observation parameters (including atmospheric concentration of organics, sulfate and DMS).

The following figure has been added to the supplement and referenced in the main text:

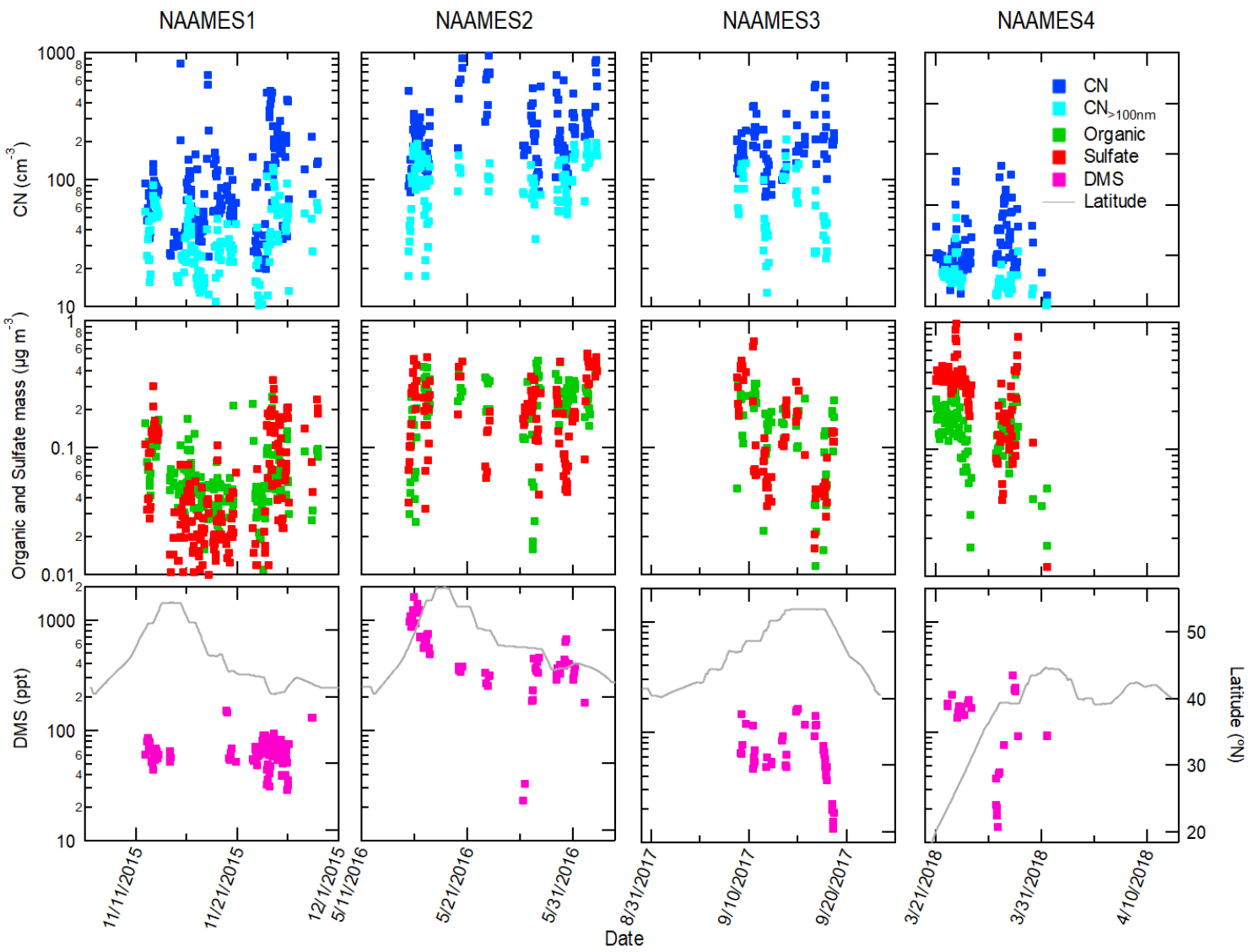

Figure S1. Time series of hourly CN and $CN_{>100nm}$, non-refractory organic and sulfate concentration, DMS concentration and *R*/V Atlantis latitude for each NAAMES campaign. Data has been filtered for clean marine conditions (see section 2.6).

Biogenic VOCs in the ocean can be produced via several pathways, including photosynthetic byproducts, bacterial degradation of dissolved organic matter, and zooplankton grazing on marine phytoplankton. The authors need to verify relevant explanations (e.g., lines 32-33).

We agree that we cannot link emissions to photosynthetic byproducts alone and have updated the text:

Updated text:

> "This result indicates non-refractory organic aerosol mass is influenced by biogenic volatile organic compound (VOC) emissions that are typically produced through bacterial degradation of dissolved organic matter, zooplankton grazing on marine phytoplankton, and as a byproduct of photosynthesis by phytoplankton stocks during advection into the region."

Provide relevant references for lines 46-48 and lines 54-55.

We have added relevant references for lines 46-48:

> "Marine environments are sensitive to aerosol particle loading because particles can act as cloud condensation nuclei (CCN) on which cloud droplets form. The number concentration of cloud droplets can influence cloud optical properties and therefore affect the impact of clouds on climate (Leahy et al., 2012; Platnick and Twomey, 1994; Turner et al., 2007; Warren et al., 1988)."

Lines 54-55 are simply a statement. We have reworded the statement to prevent confusion:

> "Since ocean-emitted volatile compounds and particles can control the number, size and composition of marine aerosols (Brooks and Thornton, 2018), here we use satellite measurements of ocean biomass as a proxy for marine particle properties."

Line 56: dissolved organic matter also acts as an important contributor to marine aerosols.

We have updated the text to included DOM as a contributor to marine aerosols.

Line 66: In general, the abundance of marine phytoplankton reaches its maximum during the spring period, and the mixed layer depth is much shallower during summer than during spring.

We have updated the text below:

> "The bloom ends when phytoplankton division rates stop increasing (due to depletion of nutrients or annual maximum in mixed layer light intensity) and are matched by loss rates (Behrenfeld and Boss, 2018). When bloom termination is associated with nutrient exhaustion, mixed later depths may continue to shoal into summer (i.e., mixed layer light levels are still increasing), but phytoplankton biomass may decrease due to slowing division rates and excessive grazing (Behrenfeld and Boss, 2018)."

Line 93: Chlorophyll-a could be used as an indicator for the biomass of marine phytoplankton, but not for biogenic VOC emissions. The production of biogenic VOCs is highly species-specific and is controlled by a complex food-web mechanism.

We agree and have changed “biogenic VOC emissions” to “marine phytoplankton biomass”.

Lines 213-214: NPP is not the only process that is linked with biogenic VOC emissions; oceanic VOC production is also related to multiple biological processes. Please modify this sentence.
The text has been updated as below:

> “Net primary production is the formation of organic material through photosynthesis by phytoplankton. This process and correlated changes in other ecosystem rates lead to the emission of biogenic VOCs at the sea surface (Li et al., 2018). “

While NPP is not the only processes that produces biogenic VOC emissions, we are directly comparing NPP to aerosol and believe it is correct to state that any correlation is most likely related to VOCs that are produced as a byproduct of NPP and not other processes.
We have updated text elsewhere (as pointed out by the reviewer in other comments) to properly state the other sources of biogenic VOC emissions in more general statements.

Line 273: “because the phytoplankton cycle is fairly slow (1 year)”. I agree that the use of 8-day averaged values for sea surface chlorophyll-a and NPP is sufficient to evaluate the relation between aerosol parameters and oceanic biological activities. However, this explanation is inadequate because the life cycle of individual phytoplankton is not that slow (typically a few days). I believe that it might be better to demonstrate the variation in daily (or 8-day) chlorophyll-a concentration at a given domain for each cruise period to support the idea that the use of 8-day chlorophyll-a values is appropriate.
The reviewer is referring to the following text:

> “While not ideal, an 8-day average is still useful because the phytoplankton cycle is fairly slow (1 year) relative to the frequency of meteorological disturbances (days)”

We agree that demonstrating the lack of variation in consecutive 8-day average chlorophyll-a concentrations is an effective method to support the use of an 8-day average and have added the following text at the end of the sentence:

> “and consequently the low variation from one 8-day average in Chlorophyll-a values to the next indicates an 8-day average is appropriate (Figure S5).”

The new supplementary figure is shown below:

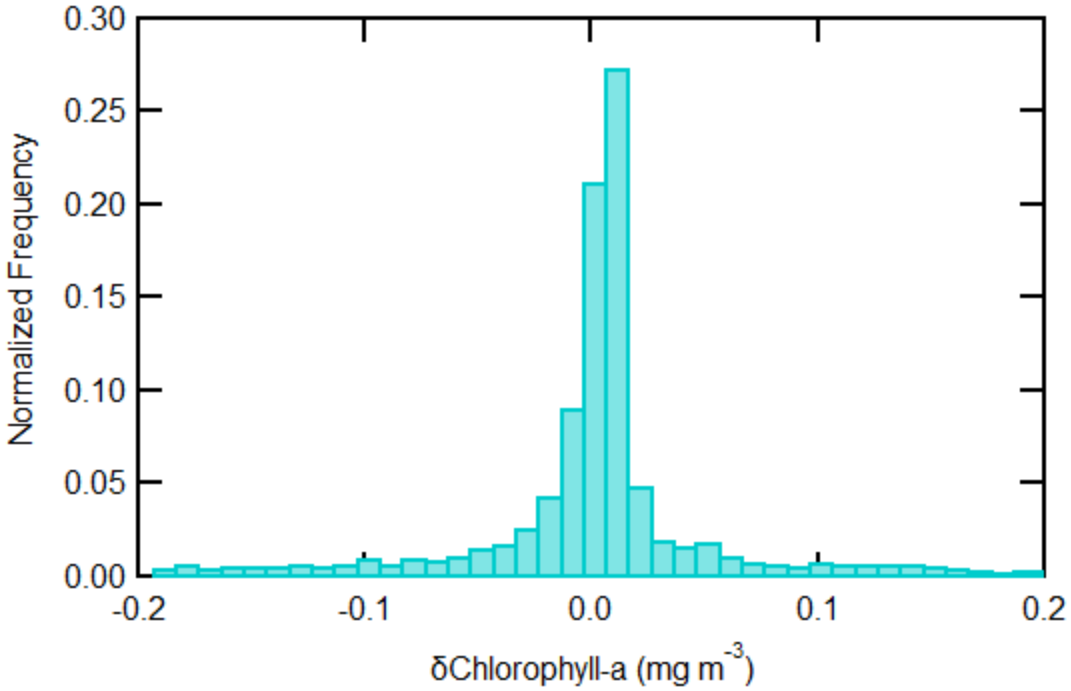

Figure S5. The normalized distribution of the difference in Chlorophyll-a between two consecutive satellite 8-day averages (24 May 2016 – 1 Jun 2016, shown in Figure 3b, and 1 Jun 2016 and 9 Jun 2016). The distribution includes the difference in chlorophyll-a from every 1° x 1° cell between 0° W and 90° W, and 10° N and 70° N, excluding cells on continents or with missing values.

Lines 284-286: Comparing in-line chlorophyll-a with trajectory-weighted chlorophyll-a does not make sense. This is because the FLEXPART backward trajectories reflect the travel history of air parcels rather than ocean currents.

The reviewer is referring to the initial Figure S5 referenced in the following text:

> "When comparing measured quantities to 0-5 day FLEXPART-weighted-residence-time explanatory variables, the slope of the linear regression generally flattens (or decreases) with longer trajectories (Figure S5). This is because the trajectories cover more ocean surface area and thus they are more likely to be weighted by both high and low values (for example, chlorophyll-a in Figure 3b)."

Our intentions were to highlight the fact that there is less dependence on the local chl-a concentration when considering longer trajectory lengths. After consideration, we have decided to remove the figure to prevent confusion. The text has been updated as follows:

> "Over longer trajectories, the weighted parameter is less likely to be related to the local value because the trajectories cover more ocean surface area and thus they are more likely to be weighted by both high and low values (for example, chlorophyll-a in Figure 3b)."

As shown in Fig. 4, several key aerosol parameters are weakly and moderately correlated with FLEXPART-residence-time-weighted explanatory variables when all datasets obtained from the four separate field campaigns that were conducted in different seasons are gathered. However, to clearly support the author's explanation, a statistically valid relation between these parameters

should be observed for each cruise. This is because Environmental factors affecting the formation and growth of marine aerosols may vary from season to season.

We agree that, to some unknown degree, the statistics are likely influenced by difference in environmental factors from one season to the next. However, Figure 4 is already quite a lot of information to process and separating this analysis by season would generate 4 times the information. Furthermore, the dynamic range of observed marine biological parameters would be significantly lower for individual seasons, likely resulting in statistically insignificant relationships between biological processes and aerosol properties. Separating by season would also significantly reduce the sample size. Such an analysis may be appropriate by combining a number of campaigns that occurred during the same season.

It would be better to provide figures for 5-day FLEXPART residence-time-weighted values, since the authors insist that many of the correlation strength increased at longer trajectory lengths.

In Figure 5, 2-day FLEXPART residence-time-weighted values were used because the 2-day trajectory corresponded to the peak in the correlation between non-refractory organic aerosol mass and Chlorophyll-a. While Chlorophyll-a does not have the strongest correlation, its importance lies in the fact that it is commonly used as a proxy for marine biomass and marine biogenic particle production. Also the peak correlation between organic aerosol mass and NPP and DSWF (Figure 5b,c,e,f) is similar to the 2-day trajectory correlation value. In Figure 6, DMS was shown to correlate with shorter trajectory lengths when comparing to Chlorophyll-a and net primary production, so it made more sense to compare to a low trajectory length (0-hour).

Line 436: The use of 'satellite measured ocean surface biomass' is not correct. This is because colored detrital organic materials and euphotic zone depth do not reflect biomass at the sea surface.

We agree and the text has been updated to:

> "We studied the relationship between marine aerosols measured over the North Atlantic Ocean during NAAMES and back trajectories weighted by four metrics of satellite measured ocean biological and physical properties (chlorophyll-a, sea water particulate organic carbon, colored detrital organic materials, euphotic zone depth), modelled net primary production, and model reanalysis meteorological parameters."

Line 440: replace 'abundance' with 'biomass'

Fixed

Line 447: What does 'surface biomass' indicate? 'Net primary production'? NPP does not mean biomass.

We have changed 'surface biomass' to 'net primary production'.

Provide a clear explanation for 'refractory' and 'non-refractory' particles.

We have updated the text to clearly state the difference:

> "The AMS does not efficiently measure refractory particles (i.e. particles that do not efficiently vaporize at 600°C), such as sea salt particles."

Lines 31-35 (abstract), 445-453 (conclusion), and relevant explanation in the Results and Discussion section: The explanation in these parts is confusing and hard to follow. As the authors noted, the lifetime of DMS in the atmosphere (1-2 days) is longer than that of other trace gases such as isoprene and monoterpene (less than a few hours). Considering the typical growth rate of SOA particles in the marine atmosphere, the difference in the lifetime of these VOCs (DMS, isoprene, etc.) may not significantly affect their temporal contribution to the organic aerosol mass over the study period. Moreover, the North Atlantic Ocean is well-known for high sea water DMS concentrations (a few nM, and occasionally increasing up to hundreds of nM during the phytoplankton bloom period due to the high abundance of DMS-producing phytoplankton groups such as haptophytes). The seawater concentration of isoprene (a few pM) in the North Atlantic Ocean is much lower than that of DMS (e.g., Dani and Loreto, 2017).

We agree that the explanation provided requires greater detail, particularly the complexity of DMS and its link (or lack of) to particle concentrations. We understand that the levels of isoprene in seawater are relatively low when compared to DMS concentration. However, models have suggested there may be an undiscovered source of VOCs that leads to the formation of SOA. Being an unknown source, we can only put our results in context of what is known. We have included this caveat in the introduction:

> "While isoprene and monoterpenes are known precursors for secondary organic particle mass, models indicate previously observed particle yields and estimated air-sea fluxes of isoprene (2%, 13–38 μg $m^{-2}d^{-1}$) and monoterpenes (~32%, 0.27–0.78 μg $m^{-2}d^{-1}$) (Hu et al., 2013; Lee et al., 2006) are too low to account for the observed MBL organic mass, suggesting that there may be large undiscovered sources (Arnold et al., 2009; Myriokefalitakis et al., 2010)."

We have also added the relevant updated text below to the results section:

> "DMS also has a number of chemical pathways with various secondary aerosol yields, making a direct link to biological processes more challenging (Faloona et al., 2009) "

And

> "Also, SO_2, a DMS oxidation product, has a lifetime on the order of days to weeks."

We have also added updated the below text in the conclusion:

> "Furthermore, the longer lifetime of DMS and its oxidation products can delay the formation of sulfate aerosol mass, making sulfate precursors more likely to advect through long-range transport if vertically lofted into the free troposphere and re-entrained down into the MBL. MBL to free troposphere transport of DMS is not captured well by the FLEXPART model. In addition, there are numerous DMS chemical pathways with various secondary aerosol yields that can obscure any link between sulfate aerosol concentrations and biogenic processes (Faloona et al., 2009)."

Technical corrections

Line 68: replace SO4 with SO42-
Fixed

Line 168: provide full name for SEMS
We have reordered sentences so the SEMS was introduced before this sentence in line 168.

References

Arnold, S.R., Spracklen, D.V., Gebhardt, S., Custer, T., Williams, J., Peeken, I., Alvain, S., 2010. Relationships between atmospheric organic compounds and air-mass exposure to marine biology. Environ. Chem. 7 (3), 232–241. https://doi.org/10.1071/EN09144.

Park, K.-T., Lee, K., Kim, T.-W., Yoon, Y.J., Jang, E.-H., Jang, S., Lee, B.- Y., Hermansen, O., 2018. Atmospheric DMS in the Arctic Ocean and Its Relation to Phytoplankton Biomass. Global Biogeochem. Cy. 32 (3), 351–359. https://doi.org/10.1002/2017GB005805.

Dani, K. G. S., and Loreto, F. (2017). Trade-off between dimethyl sulfide and isoprene emissions from marine phytoplankton. Trends Plant Sci. 22, 361–372. doi:10.1016/j.tplants.2017.01.006