

General comment:

The manuscript presents how observed aerosol particle composition and concentrations are correlated with different meteorological and biological processes in the ocean upwind of the measurement location. The analysis is performed using meteorological data, comprehensive aerosol observations and the FLEXPART Lagrangian particle dispersion model. I find most of the results convincing and well justified by known physical/biological mechanisms. The paper is also generally well written and easy to follow. I suggest that the paper should be accepted for publication in ACP after minor revision considering all reviewers comments.

Specific comments:

Abstract: L39-41 “We hypothesized that the elevated total particle surface area associated with high PMA concentrations leads to enhanced rates of VOC condensation onto PMA” It is not the VOCs but their low- or semi-volatile oxidation products that condenses onto the PMA. Please, modify this sentence.

We have updated the sentence to state: “...enhanced rates of condensation of VOC oxidation products onto PMA.”

The observed negative correlation between aerosol submicron number and mass concentrations and wind speeds can as the authors suggest partly be a result of enhanced rate of condensation of condensable vapors onto wind generated PMA. However, other factors such as coagulation sink, and enhanced vertical mixing and dry deposition losses during high wind speed conditions may also contribute to the observed correlation.

We agree and have added the following sentence to section the section 3.2:

“Other factors associated with high wind speed conditions, such as coagulation, dry deposition, and enhanced vertical mixing likely also contribute to the observed negative correlation between wind speed and particle concentration.”

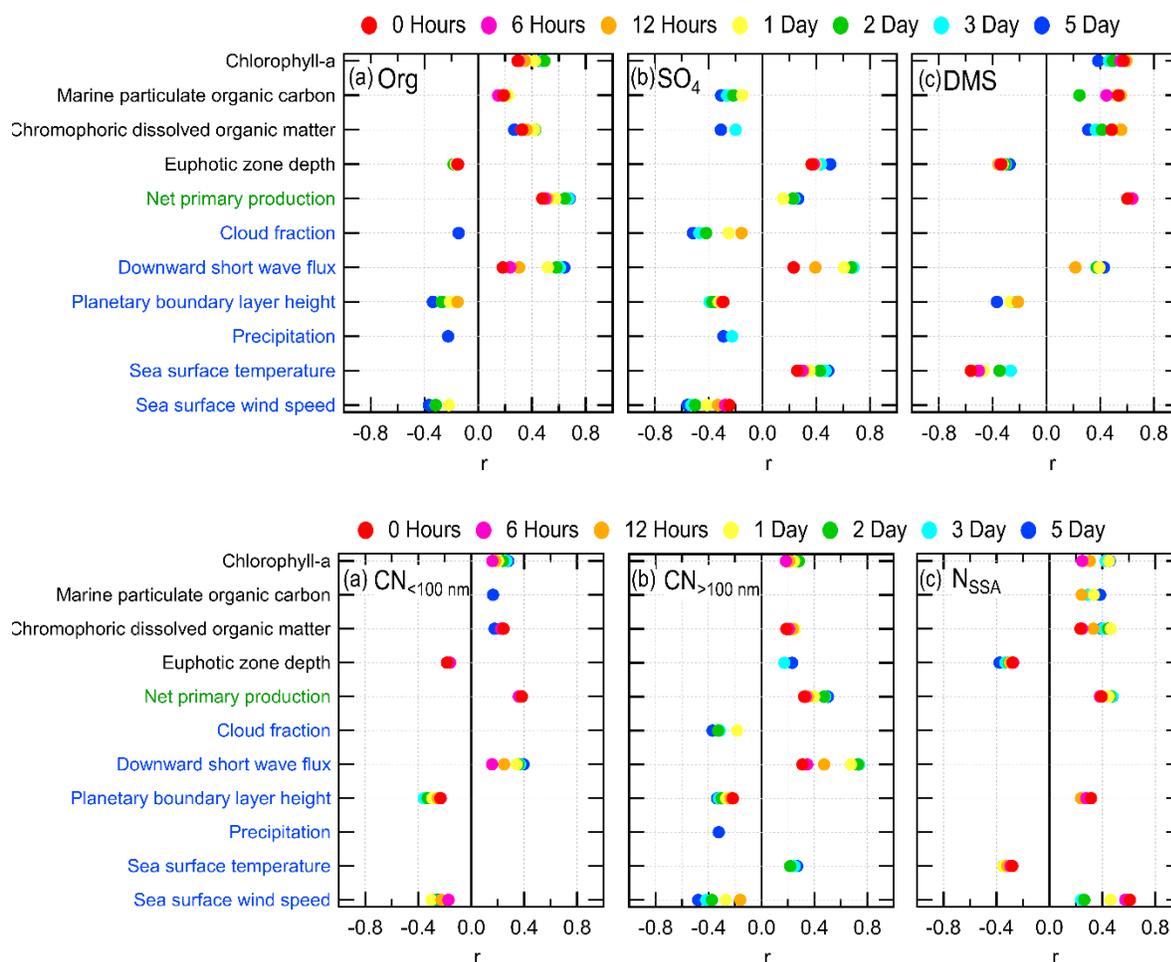
P8, L240-242 “For the remaining analysis in this paper, the vertical structure of the residence time is column integrated over only the vertical levels that are completely or partially within the MBL based on GDAS MBL heights. Remaining vertical levels were excluded from analysis.” Is it reasonable to exclude the air mass residence time above the MBL from the analysis? Does, this not also exclude the impact of free tropospheric air masses which may dilute the observed MBL aerosol concentrations. I would like to see result on how large fraction of the air mass residence time which is excluded because a fraction of the FLEXPART particles tracers is above the MBL. I would also like to see some analysis on if the fraction of air masses which is above the MBL correlates (anti-correlates) with the observed aerosol concentrations. I would expect that a large contribution from free troposphere air masses would result in lower PMA and aerosol particle mass in general, but possibly higher particle number concentrations.

We have performed the analysis while including the residence time above the MBL and the results were not significantly different (See figure below after next comment). This shows that most air masses were largely confined in the boundary layer up to 5 days because of the stable nature of the MBL. Though mixing of FT air into the MBL can happen, the amount and occurrence is low enough that their influence cannot be seen in the statistics. Also, like all global

models, the GFS's coarse resolution prevents it from resolving some of the small scale mixing for FT and MBL air, but is well suited for resolving the large-scale advection. Furthermore, since we are examining how the ocean surface and boundary layer meteorology influence marine boundary layer particles, we do not believe it is reasonable to weight FT residence time by boundary layer quantities.

P9, L271 "Residence time over land is excluded from the integration of weighted trajectories"
Similar comment as above. Is it reasonable to exclude the residence time over continents. Should this residence time not be included in equation 1 but with the explanatory variable values (Et) set to zero or a value representing e.g. emissions over the continents?

We agree that including the continental residence time and setting the explanatory variable values to zero is an alternative approach. However, this approach is equivalent to stating that there is zero influence from continental processes and, for this reason, we believe our approach is better. Despite this choice, we have performed the suggested approach of including the continental residence time (and included FT residence time) and the results are not significantly different (see figure below). Furthermore, extensive filtering for continentally influenced air (and specifically high continental residence time) was performed to remove cases that were significantly influenced by continental processes.



Reviewer figure 1: Same as figure 4 of main text, but FLEXPART trajectories that extend into the FT and over land are included (explanatory marine biological variables = 0 over land).

P9, L277-279 “We define correlation strength by the calculated Pearson’s coefficient (r) following Devore and Berk (2012), where $|r| < 0.25$ indicates there is no correlation, $0.25 \leq |r| < 0.50$ is defined as a weak correlation, $0.50 \leq |r| < 0.80$ is defined as a moderate correlation, and $|r| \geq 0.80$ is defined as a strong correlation.”

What I miss in the main manuscript (at least I could not find it), but what is included in the supplementary tables, is a statement about if the correlation coefficient is significantly separated from zero ($r=0$). Please add a sentence stating e.g. that only statistically significant correlations on a 5 % significance level ($p < 0.05$) is presented.

We agree and have added the following sentence:

“All presented Pearson’s correlation coefficients are statistically significant ($p < 0.05$).”

P10, L296-299 “Comparisons of non-refractory organic aerosol mass with other net primary production models are shown in the supplemental Table S8. These results suggest a substantial

portion of non-refractory organic mass is from secondary biogenic VOC emissions, such as isoprene and monoterpenes and other unidentified biogenic VOCs (Altieri et al., 2016; Hallquist et al., 2009).”

What about MSA formed from DMS? In the AMS I expect that the MSA mass will be assigned both to the sulfate and organics non-refractory mass.

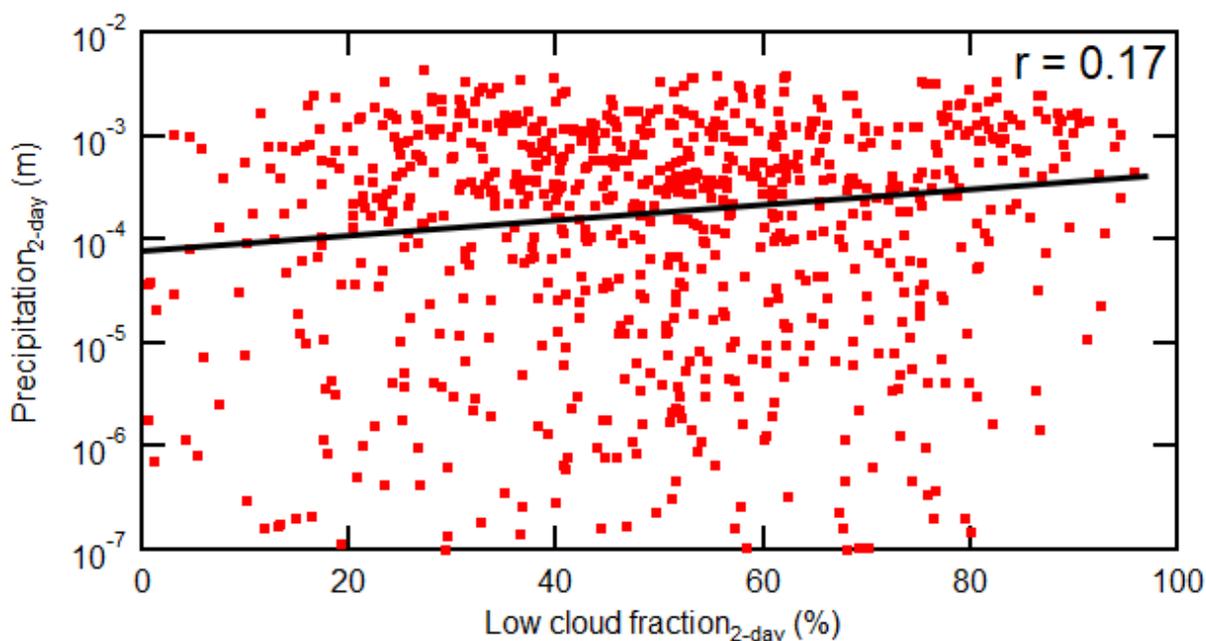
Yes, MSA will contribute to both organic and sulfate non-refractory mass for AMS unit mass resolution measurements. We have updated the sentence to also include MSA as a contributing VOC to the overall organic mass and added a relevant reference (Zorn et al., 2008).

P12, L373-374 “The negative correlation between low-level cloud cover and sulfate mass suggests the aqueous processing may be relatively less important than gas-phase photochemical mechanisms.”

I expect that low-level cloud cover also correlates with precipitation. Can this not also contribute to the negative correlation between the sulfate mass and low level clouds?

We have looked into the influence of precipitation on aerosol properties (see figure 4) and found no significant correlation with precipitation. However, as mentioned in the manuscript, the reanalysis precipitation has been shown to only moderately correlate with observations (Beck et al., 2019) and we believe the role of precipitation is possibly more important than what our analysis shows.

For reference, below is a figure showing the relation between the 2-Day trajectory averaged precipitation and low cloud fraction.



Reviewer figure 2: 2-Day trajectory weighted Precipitation and low cloud fraction from the GDAS reanalysis.

P13, L412-413 “Up to 25% of secondary sulfate formation has been shown to form from aqueous ozone oxidation of SO₂ to sulfate on PMA particles (Sievering et al., 1992b)”

Yes, this may be correct but generally the most important aqueous phase SO₂ oxidation mechanism leading to sulfate is the reaction between H₂O₂ and SO₂.

Yes under most conditions, H₂O₂ is the dominate oxidant for aqueous phase sulfate formation from SO₂. We have updated this statement to take into account this reaction:

“Secondary sulfate formation from SO₂ occurs rapidly on fresh PMA particles via uptake due to aqueous ozone reactions (Sievering et al., 1992). Subsequently, in-cloud sulfate formation from SO₂ continues by oxidation due to hydrogen peroxide (Jacob, 2000).”

P14, L450-453 “The longer lifetime of DMS 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.”

I agree. What complicates things with DMS is that the DMS oxidation is a multiphase process involving both gas- and aqueous phase and OH, O₃ and halogens. The fraction of DMS which is oxidized to SO₂ will delay the sulfate aerosol mass even further. In the gas-phase SO₂ has a relatively long lifetime (~1 week).

We agree and have added the following text 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, SO₂, a DMS oxidation product, has a lifetime on the order of days to weeks.”

We have also 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).”