

Review of the manuscript “Linking marine phytoplankton emissions, meteorological processes and downwind particle properties with FLEXPART” by Kevin J. Sanchez et al.

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.

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.

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.

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 (E_t) set to zero or a value representing e.g. emissions over the continents?

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.

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.

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?

P13, L412-413 “Up to 25% of secondary sulfate formation has been shown to form from aqueous ozone oxidation of SO_2 to sulfate on PMA particles (Sievering et al., 1992b)”

Yes, this may be correct but generally the most important aqueous phase SO_2 oxidation mechanism leading to sulfate is the reaction between H_2O_2 and SO_2 .

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, O_3 and halogens. The fraction of DMS which is oxidized to SO_2 will delay the sulfate aerosol mass even further. In the gas-phase SO_2 has a relatively long lifetime (~ 1 week).