**We appreciate the referee's valuable comments on our work. Our responses to the specific comments and some changes made in the manuscript are given below.**

Responses to the comments of Referee #2:

*Comment 1*: Page 2, Line 22: Provide a reference for WSOC altering hygroscopicity.

***Reply 1*: Prather et al. (2013) has been added to the text as a reference.**

*Comment 2*: Page 4, Line 34; Page 8, Line 13: Report sample number when reporting correlations.

***Reply 2*: The number of samples has been provided in the revised manuscript.**

*Comment 3*: Figure 1 caption: what are the red traces in the right panel representative of? Clarify in caption, and thicken the red lines to allow for readers to see them more easily.

***Reply 3*: The red lines indicate the backward trajectory, which has now been mentioned in the caption of Figure 1 in the revised manuscript. The lines have been made bolder, as suggested.**

*Comment 4*: Page 5, Line 32: For the sake of comparison, it would be useful to provide info here about other archived datasets such as (for example) data collected in the Northeast Pacific Ocean showing both unimodal and bimodal characteristics for water soluble organic species: Maudlin, L. C., et al. (2015). Impact of wildfires on size resolved aerosol composition at a coastal California site, Atmos. Environ., 119, 59-68, doi:10.1016/j.atmosenv.2015.08.039. Also, some discussion about the sources of the two modes in this study would be helpful for readers.

***Reply 4*: A statement on the comparison with Maudlin et al. has been added to the text. In addition, we have added a discussion on the possible sources of the two modes in this study. (P.5, L.34–P.6, L.7) *"Bimodal size distributions of WSOC in marine aerosols were also observed in the western North Pacific (Miyazaki et al., 2010), whereas both unimodal and bimodal size distributions of water-soluble organic species were also reported in particles collected at a coastal site facing the Northeastern Pacific (Maudlin et al., 2015). The bimodal size observed in this study can be attributed to the difference in the formation processes of WSOC between the two size ranges. The two distinct size modes include (i) direct co-emissions associated with sea salt particles in both size ranges, (ii) aqueous-phase products in the submicron size range, and (iii) partitioning to the***

***surface of coarse particles (i.e., sea salt) and/or heterogeneous reactions in the supermicron size range (Mochida et al., 2002). Although it is difficult to provide a clear explanation by this data set alone, the observed WSOC size distributions might be explained by some combination of these possible origins and processes. Here we focus on the submicron size of WSOC relevant to its isotope ratios and several chemical tracers."***

*Comment 5*: Page 6, Line 11: Have other studies also shown a link between organics and high Chl A? if so, I suggest a discussion of those other studies too and what their chief results were to enrich the discussion in this section of the paper. Some suggested studies to look up are: Facchini, M.C., et al. (2008). Important source of marine secondary organic aerosol from biogenic amines, Environ. Sci. Technol., 42, 9116e9121. http://dx.doi.org/10.1021/es8018385. Sorooshian, A., et al. (2009). On the link between ocean biota emissions, aerosol, and maritime clouds: airborne, ground, and satellite measurements off the coast of California, Global Biogeochem. Cycles, 23, GB4007, doi:10.1029/2009GB003464.

***Reply 5*: The following discussion has been added to the text: (P.6, L.26–30) "*Previous studies have shown a linkage between organics and high Chl a concentrations on timescales of months (O'Dowd et al., 2004; Sciare et al., 2009). However, Quinn et al. (2014) found no well-defined relationship between instantaneous Chl a in seawater and organic-mass enrichment in sea spray, suggesting no significant variability in the OC content of freshly emitted sea spray aerosol, despite significant variability in seawater Chl a levels. This point will be discussed in section 3.3."***

*Comment 6*: Section 3.4: For the discussion about MSA and its lack of correlation with WSOC and Chl A, it may be useful here to bring up the point that other studies have observed that MSA may be enhanced in the presence of metals (mainly vanadium). Such catalytic effects that are speculated in past works may be at least one contributing factor as to why correlation is not observed between MSA and these other parameters:
Gaston, C. J., et al (2010), Real-time detection and mixing state of methanesulfonate in single particles at an inland urban location during a phytoplankton bloom, Environ. Sci. Technol., 44(5), 1566–1572.
Sorooshian, A., et al. (2015). Surface and airborne measurements of organosulfur and methanesulfonate over the western United States and coastal areas, J. Geophys. Res., 120, doi:10.1002/2015JD023822.

***Reply 6*: The observed increases in the MSA concentrations were most evident in R2, in which the possible influence of anthropogenic sources appeared to be very low. Therefore, we believe that the effects of such a catalytic reaction on the increases in**

**the MSA concentrations in R2 were likely insignificant in our data. In response to this important comment, we have added the following sentences to the discussion: (P.9, L.37–40) *"Gaston et al. (2010) suggested a possible catalytic role of vanadium in MSA formation. The observed increases in the MSA concentrations were most evident in R2, in which the impacts of anthropogenic sources appeared to be very low. Therefore, the effects of such a catalytic reaction on the increases in the MSA concentrations in R2 are likely insignificant."***

*Comment 7*: Do the authors have any indication about how important cloud processing was during the research cruise in explaining any of the features in their WSOC data?

***Reply 7*: The back-trajectory calculation shows that relative humidity (RH) in the air mass along the back trajectories was typically within the range of 60–80%, with a maximum of 95%. This indicates that cloud processing or rainout of aerosols were likely insignificant over the period of transport in our study.**

*Comment 8*: Figure 2: Was precipitation important in explaining any of the reductions in concentrations observed during the time period shown after the local maxima values observed?

***Reply 8*: We observed almost no precipitation during the observational period, particularly after 10 February when the reductions in WSOC concentrations were observed. Therefore, it is unlikely that the observed decrease in the concentrations could be explained by precipitation.**

References

Gaston, C. J., Pratt, K. A., Qin, X. Y., and Prather, K. A.: Real-time detection and mixing state of methanesulfonate in single particles at an inland urban location during a phytoplankton bloom, Environ. Sci. Technol., 44(5), 1566–1572, 2010.

Maudlin, L. C., Wang, Z., Jonsson, H. H., Sorooshian, A.: Impact of wildfires on size-resolved aerosol composition at a coastal California site, Atmos. Environ., 119, 59-68, doi:10.1016/j.atmosenv.2015.08.039, 2015.

Miyazaki, Y., Kawamura, K., and Sawano, M.: Size distributions of organic nitrogen and carbon in remote marine aerosols: Evidence of marine biological origin based on their isotopic ratios, Geophys. Res. Lett., 37, L06803, doi:10.1029/2010GL042483, 2010.

Mochida, M., Kitamori, Y., Kawamura, K., Nojiri, Y., and Suzuki, K.: Fatty acids in the marine atmosphere: Factors governing their concentrations and evaluation of organic films on sea-salt particles, J. Geophys. Res., 107(D17), 4325, doi:10.1029/2001JD001278, 2002.

O’Dowd, C., Facchini, M. C., Cavalli, F., Ceburnis, D., Mircea, M., Decesari, S., Fuzzi, S., Yoon, Y. J., and Putaud, J.-P.: Biogenically driven organic contribution to marine aerosol, Nature, 431, 676– 680, 2004.

Prather, K. A., Bertram, T. H., Grassian, V. H., Deane, G. B., Stokes, M. D., DeMott, P. J., Aluwihare, L. I., Palenik, B. P., Azam, F., Seinfeld, J. H., Moffet, R. C., Molina, M. J., Cappa, C. D., Geiger, F. M., Roberts, G. C., Russell, L. M., Ault, A. P., Baltrusaitis, J., Collins, D. B., Corrigan, C. E., Cuadra-Rodriguez, L. A., Ebben, C. J., Forestieri, S. D., Guasco, T. L., Hersey, S. P., Kim, M. J., Lambert, W. F., Modini, R. L., Mui, W., Pedler, B. E., Ruppel, M. J., Ryder, O. S., Schoepp, N. G., Sullivan, R. C., and Zhao, D.: Bringing the ocean into the laboratory to probe the chemical complexity of sea spray aerosol, P. Natl. Acad. Sci. USA, 110, 7550–7555, doi:10.1073/pnas.1300262110, 2013.

Quinn, P. K., Bates, T. S., Schulz, K. S., Coffman, D. J., Frossard, A. A., Russell, L. M., Keene, W. C., and Kieber, D. J.: Contribution of sea surface carbon pool to organic matter enrichment in sea spray aerosol, Nat. Geosci., 7, 228–232, 2014.

Sciare, J., Favez, O., Sarda-Estève, R., Oikonomou, K., Cachier, H., and Kazan V.: Long-term observations of carbonaceous aerosols in the Austral Ocean atmosphere: Evidence of a biogenic marine organic source, J. Geophys. Res., 114, D15302, doi:10.1029/2009JD011998, 2009.