Responses to the comments of Referee #1:

Comment: This paper attempts to address the uncertainty around sources of aerosol WSON in the marine atmosphere of the subtropical North Pacific. They report data from an east to west cruise transect across the N. Pacific including WSON aerosol concentration and surface ocean chl, primary productivity and N2 fixation rates. Their main approach is to compare east to west trends in the measured parameters. They conclude that since N2 fixation and aerosol WSON are both higher in the eastern N. Pacific than the western N. Pacific, then N2 fixation must be the source of aerosol WSON. The mechanism the authors invoke is that N2 fixation increases ammonium and DON concentrations in the surface ocean which then flux to the atmosphere and lead to secondary WSON. This paper is an example of "correlation does not equal causation." It is undeniable that the east to west trends are similar in N2 fixation and WSON aerosol concentrations, as in, they are both higher in the east than the west. But that could be due to multiple factors, and in no way suggests that one is causing the other. The authors proposed mechanism is completely untestable as they do not present ammonium or surface ocean DON concentrations. They suggest the WSON must be secondary as it does not correlate with sodium, but it also does not correlate with MSA, a classic indicator of secondary processing. I have chosen not to present a detailed review of the manuscript as the general framework presented is not supported in the literature, nor do the authors present a mechanism that can be tested by the existing data. The conclusions drawn are therefore based on a single correlation and are not supported in any way by what is presented in the paper.

***Reply*: We appreciate the referee's valuable comments on our work. Based on the comments, we added substantive discussion to construct compelling arguments by showing additional data as below. Specific points of our major revisions are:**

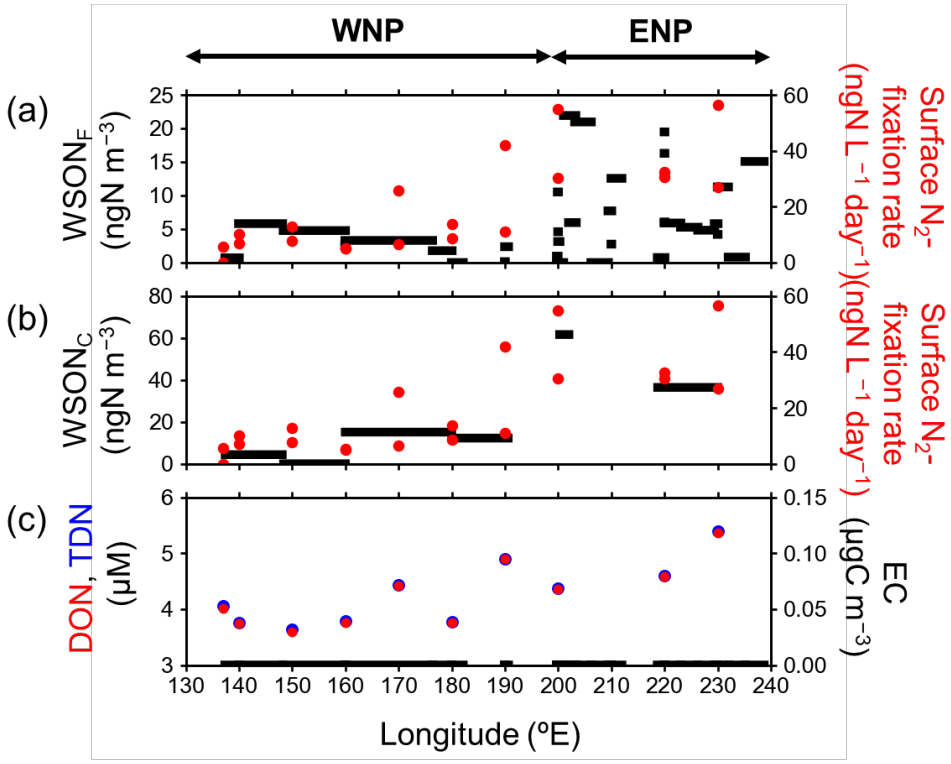
(1) To make a compelling argument on the causal relationship between N_2-fixation rate in surface seawater (SSW) and aerosol WSON concentrations, we have additionally shown and discussed the data of dissolved ON (DON) and total dissolved nitrogen (TDN) in SSW in the revised manuscript (Figure 5 has been revised as shown below). The figure clearly shows that DON concentrations in SSW of ENP (4.77±0.53 μM) were larger than those of WNP (4.03±0.47 μM). Furthermore, N_2-fixation rate and DON concentrations in SSW showed a positive correlation with R^2 of 0.63, where DON concentration accounted for >98% of TDN concentration, showing the dominance of DON in TDN during the cruise. The result suggests that DON were released associated with the recently fixed N_2 during the growth of N_2-fixing microorganisms. Based on these results, we have further discussed the linkage among N_2-fixation rate, DON, and aerosol WSON (P.6, L.22). This point is also discussed in terms of points below ((2) and (3)).

(2) Regarding (1), we have also added discussion on possible effects of anthropogenic sources on WSON by showing the data of elemental carbon (EC) concentration (Figure 5). The EC concentrations in all the samples were below the lower detection limit and did not show any statistically significant differences between WNP and ENP. This result suggests that possible effects of anthropogenic sources on the observed aerosols in ENP were small, which is consistent with the stable carbon isotope analysis in our study. We made additional statement on this point in the revised manuscript (P.7, L.6).

(3) In the revised manuscript, we also added literatures on dissolved nitrogen released by N_2-fixing microorganisms in the subtropical North Pacific, which support our discussion. Specifically, data obtained from Station ALOHA in the subtropical North Pacific has shown enrichment of N pools (i.e., NH_4^+, NO_3^-, DON) within *Trichodesmium* blooms (Karl et al., 1992; Letelier and Karl, 1994). It has been shown that majority of the recently fixed N_2 is released directly as DON during the growth of *Trichodesmium* (Capone et al, 1994; Glibert and Bronk, 1994). Indeed, our data showed that DON was a dominant component of TDN (>98%) in SSW, the longitudinal distribution of which was closely linked with that of N_2-fixation rate. In the revised manuscript, we made additional statement on these points (P.5, L.40).

(4) As we described in the original manuscript, the insignificant correlation between WSON and MSA suggests that the origin of observed WSON aerosols differed from that of DMS or that the formation pathways of WSON were different from the oxidation processes of DMS. We made additional discussion on this point with data of sulfate as below:

P.5, L.28:*...It is also possible that dependence of temperature and/or OH levels in the subtropics on the relative yields of MSA to sulfate in the oxidation of DMS (e.g., Mungall et al., 2018) might also partly affect the insignificant correlations between WSON and MSA. In fact, no relations were found between sulfate, known as an oxidation product of DMS, and WSON concentrations (R^2 < 0.02) in fine particles (data not shown). On the other hand, sulfate and WSON concentrations in coarse particles showed some positive relationships, although the number of data is limited. This may reflect overlapping processes of biogenic sulfate and WSON on a longer timescale (i.e., several days of sampling of coarse particles) even though the exact origins are different.*



Revised Figure 5: Longitudinal distributions of mass concentrations of (a) $WSON_F$ (black) and (b) $WSON_C$ (black), together with N_2-fixation rate in the SSW samples (red solid circle), (c) DON (red solid circle) and TDN (blue solid circle) concentrations in surface seawater and aerosol EC concentrations (black) during the entire cruise.

Another change:

▪Along with the addition of DON and TDN data in the revised manuscript, Fuminori Hashihama and Saori Yasui-Tamura, who measured those parameters in seawater, have been added as co-authors.

References:

Capone, D. G., Ferrier, M. D.,and Carpenter, E. J.: Cycling and release of glutamate and glutamine in colonies of the marine planktonic cyanobacterium, *Trichodesmium thiebautii*, Appl. Environ. Microbiol., 60, 3989– 3995, 1994.

Glibert, P. M. and Bronk, D. A.: Release of dissolved organicnitrogen by marine diazotrophic cyanobacteria, *Trichodesmium* spp. Appl. Environ. Microbiol., 60, 3996–4000, 1994.

Karl, D. M., Letelier, R., Hebel, D. V., Bird, D. F., and Winn, C. D.: *Trichodesmium* blooms and new nitrogen in the north Pacific gyre, p. 219-237, Marine pelagic cyanobacteria: *Trichodesmium* and other diazotrophs. Kluwer Academic Publishers, Dordrecht, The

Netherlands, 1992.

Letelier, R. M. and Karl, D. M: The role of *Trichodesmium* spp. In the productivity of the subtropical North Pacific Ocean, Mar. Ecol. Prog. Ser., 133, 263-273, 1994.

Mungall, E. L., Wong, J. P. S., Abbatt, P. D.: Heterogeneous oxidation of particulate methanesulfonic acid by the hydroxyl radical: kinetics and atmospheric implications, ACS Earth Space Chem. 2018, 2, 48–55, 2018.