## **Responses to the comments of R. Duce:**

## We appreciate the referee's valuable comments on our work. Our responses to the specific comments are given below.

*Comment 1*: Page 4, line 8 on - While the way the aerosol samples are collected should protect against local shipboard contamination in most cases, there are very few locations where contamination is greater than aboard ship. Were the authors able to use any specific organic tracers that would have indicated more clearly whether or not they were sampling any pollution from the ship? The results themselves do indicate that contamination was likely not a serious problem.

*Reply 1*: We did not analyze the samples for any specific organic tracers from local ship emissions. However, the aerosol data with a total air volume  $< 800 \text{ m}^3$  were omitted in the original manuscript, because these aerosol samples might have been affected by local contamination in stagnant air. Indeed, these data correspond well with time-series of substantially high aerosol number concentrations measured by a water-based condensation particle counter (CPC) (Mochida et al., 2011), presumably caused by the ship's exhaust. As a result of the data screening, we presented 32 samples out of the total 42 samples obtained in the present study. Based on the low concentration of EC and NO<sub>3</sub><sup>-</sup> in the aerosol samples, we believe that the aerosols presented in the manuscript were hardly affected by local ship emissions. Moreover, particles from local shipboard contamination generally reside in the nanometer size range. The mass concentrations of these particles appear to be relatively minor in our discussion about the bulk concentrations of aerosols.

*Comment 2*: Page 7, section 2.4 -I understand that the SeaWiFS data were used for chlorophyll a measurements, but were chlorophyll a measurements also made on board ship as the ship moved along its track? How similar were the SeaWiFS data compared with the measurements actually made on the cruise, if they were made? I note that from the mention of in situ measurements at the bottom of page 7 (which were apparently from a different cruise?) compared with the SeaWiFS data shown in Figures 2, the differences appear large.

*Reply 2*: In fact, the SeaWiFS data shown in Figure 2 represent the average concentrations of chlorophyll *a* exposure calculated along each trajectory point, as described in the text and figure caption. This means that the values are not necessarily consistent with those obtained at each sampling point, but they reflect the marine biological activity on the air masses transported over the ocean.

*Comment 3*: Page 8, paragraph beginning on line 5 - The explanation for the high nss-sulfate at low latitudes is given as volcanic activity, which may be correct. However, I note that the MSA concentrations are also higher near the equator, although not as much higher as the nss-sulfate. Could the higher MSA (and perhaps some of the nss-sulfate) be due to higher productivity in the equatorial regions?

Reply 3: We thank the Referee's suggestion for this important point. At low latitudes (10-20°N), the concentrations of cyanobacteria in surface seawater were larger than those at higher latitudes (20-45°N) from the same cruise observations (Suzuki et al., unpublished data). Moreover, Ooki et al. (2010) reported enhanced concentrations of methyl halide in surface seawater between 15°N and 20°N where SST values were high, which was also revealed from the same cruise. Although the chlorophyll a value was low at the subtropical regions, these results indicate that the increased MSA concentrations are likely due to marine biological productivity by bacteria rather than phytoplankton in the subtropical regions. We have added these statements in the revised manuscript, as follows: "We note that the measured concentrations of cyanobacteria in surface seawater at 10-20°N were larger than those at 20-45°N from the same cruise observations (Suzuki et al., unpublished data). Moreover, Ooki et al. (2010) reported enhanced concentrations of methyl halide in surface seawater between 15°N and 20°N where the sea surface temperature was high, which was also revealed from the same cruise. Although the satellite-derived chlorophyll a value was low at 10-20°N, these results indicate that the increased MSA concentrations appear to be due partly to marine biological activity by bacteria rather than phytoplankton in this subtropical region."

*Comment 4*: Page 9, lines 28 and 29 - This sentence is a bit misleading. If one looks at the plot of DEA vs latitude in Figure 4 it appears that if the 2 highest numbers for DEA at about 45N are removed, the concentrations are about the same at most latitudes. Note that 3 of the 5 highest values for DEA are at latitudes below 30N. Admittedly there are no really low values for DEA above about 35N, but their overall argument here is not particularly strong.

*Reply 4*: We observed that all of the DEA concentrations at 40-44°N were greater than 0.25 ng m<sup>-3</sup>, whereas 64% of the data points at 10-44°N showed concentrations less than 0.25 ng m<sup>-3</sup>. Indeed, the median concentration of 0.52 ng m<sup>-3</sup> at 40-44°N was greater than that (0.20 ng m<sup>-3</sup>) at 10-44°N. Following the comment, the sentence has been changed, as follows: "*The DEA*<sup>+</sup> concentrations at 40–44°N (av. 0.52±0.19 ng m<sup>-3</sup>) were generally higher than those at 10–40°N (av. 0.23±0.11 ng m<sup>-3</sup>)."

*Comment 5*: Page 10, line 24 and following - As with DEA, the authors claim that the OC is higher at 40-44N compared with lower latitudes. Again, while this is clearly true for WSOC,

it is not at all as clear for OC. Two of the 3 highest values for OC are at 30N or below and 5 of the 9 highest values for OC are below 30N.

## *Reply 5*: We do not intend to say that the OC concentrations were higher at 40-44°N than those at 10-44°N. Indeed, OC showed a peak at 40-44°N, as well as at 10-20°N, and around 30°N. We have removed the statements about the "OC" latitudinal profile and have now focused on the WSOC profile.

*Comment 6*: Page 11, line 16 and following - The statement that WIOC on marine aerosols is produced from organic matter that accumulates in the microlayer of the ocean surface is only partly correct. That material also accumulates on the surface of the bubble as it rises through the water column, and that material "merges" with material already in the microlayer when the bubble breaks and ejects the particles. However, both soluble and insoluble surface-active material is concentrated in the bubble surface and the ocean surface microlayer. The higher WSOC at higher latitudes could partially at least be the result of more soluble OC in the bubble film/microlayer than at lower latitudes. What was the correlation between Na and WSOC? Also, the WSOC and the WIOC may be formed on different size sea salt particles due to different production mechanisms of droplet formation (i.e., film drops vs. jet drops), which in turn may have different amounts of WSIC and WSOC on them. There are many possibilities here.

*Reply 6*: In the first paragraph of Section 3.3, we discuss the possibility of primary emission of WSOC from sea surface. As stated in the text, WSOC showed no significant correlation with Na<sup>+</sup> ( $r^2 = 0.02$ ) or local wind speeds, supporting the idea that the majority of WSOC appears to be derived from secondary production. Additionally, for size-segregated aerosols (Miyazaki et al. 2010b), no significant correlation was found between WSOC and Na<sup>+</sup> ( $r^2 = 0.02$ ) in the submicrometer range of the particles, whereas WIOC and Na<sup>+</sup> showed a positive correlation ( $r^2 = 0.52$ ) in this size range. Furthermore, the peak diameter of WSOC (3.0–4.3 µm) was smaller than that of Na<sup>+</sup> (> 6.4 µm) (Miyazaki et al., 2010b), indicating that primary emission of WSOC with sea salts may be less significant in the present study. These points are now briefly mentioned in the revised manuscript.

*Comment* 7: Page 12, line 1 - Were the higher ON/OC ratios found on the larger or the smaller particles in the Miyazaki paper?

*Reply 7*: In Miyazaki et al. (2010a), we reported an average ON/OC ratio of 0.43±0.23 for the submicrometer particles and 0.53±0.31 for supermicrometer particles. Considering the large magnitude of the standard deviations, the difference between

these two ratios is not statistically significant. The sentence has been modified, as follows: "Miyazaki et al. (2010a) also reported higher ON/OC ratios (0.35–0.49) in both the submicrometer and supermicrometer size ranges of marine aerosols obtained farther north of the present study areas in the western Pacific."

*Comment* 8: Page 12, lines 4-7 - I am somewhat confused here. WSON/WSOC was 0.23 at a forest site, and WION/WIOC was 0.18 in this paper, but the authors indicate that there is more ON enrichment in marine aerosol compared with previous studies. I do not understand that conclusion.

*Reply 8*: Both WSON/WSOC (0.23) and WION/WIOC (0.18) ratios were obtained in this study, which does not seem to provide useful information in this context. Thus, the sentence has been deleted in the revised manuscript. Moreover, because the comparison with the ON/OC ratio at a forest site is not useful, as pointed out by Referee #1, we also deleted this sentence. Overall, the sentences have been changed, as follows: "*The average ON/OC ratio was 0.19±0.11 during the study period, which is higher than typical ON/OC ratios (0.06–0.11) reported for oceanic dissolved materials (Hansell and Carlson, 2002). Miyazaki et al. (2010a) also reported higher ON/OC ratios for the oceanic materials in both the submicrometer (0.43\pm0.23) and supermicrometer (0.53\pm0.31) size ranges of marine aerosols obtained farther north of the present study areas in the western Pacific with high marine biological productivity. Based on the comparison with typical ON/OC ratios in oceanic dissolved materials, the present results indicate an enrichment of organic nitrogen in the marine aerosol collected in the area studied."* 

*Comment 9*: Page 12, lines 8-15 - The data on nitrogen isotopes does not appear to be very helpful here.

*Reply 9*: Although nitrogen isotopic analysis alone cannot provide conclusive evidence about the formation process of atmospheric nitrogen sources, there are limited data on nitrogen isotopes in aerosols in the literature. We believe that it still provides information on the sources of aerosol nitrogen. For example, the <sup>15</sup>N values in our study overlap with a range of marine phytoplankton, supporting our conclusion about the dominant source of aerosols. We've decided to keep the discussion on the nitrogen isotope data.

*Comment 10*: Page 12, lines 16 to 22 - In the Cachier et al. paper referenced those authors found that the small sub-micrometer particle had C isotope ratios indicating a continental sources whereas the larger particles has a carbon isotope signature of a seawater source, if I

remember that paper correctly. If correct, this is very important and should be mentioned here. Did the present authors make any C isotope measurements as a function of particle size? Where any samples collected at all as a function of particle size?

Reply 10: We have provided an additional description about Cachier et al. (1986), who reported stable carbon isotopic ratios in different size ranges of marine aerosols. Previously, we have reported the stable carbon isotopic ratios as a function of the size of aerosols collected in a similar region in a similar season (see Figure 3 in Miyazaki et al., 2010a). Accordingly, the average ratios of stable carbon isotope were similar in the submicrometer (-23.7±0.8‰) and supermicrometer (-23.4±0.7‰) size ranges, both of which are close to typical stable carbon isotope ratios of OC in seawater. In the revised manuscript, the following statements have been added to the text: "Based on the  $\delta^{13}$ C of particulate carbon over various remote oceanic areas, Cachier et al. (1986) found that most of the submicrometer particulate carbon was of continental origin, whereas coarse particles with diameter > 3  $\mu$ m were primarily of seawater origin. In contrast, Miyazaki et al. (2010a) found that the average  $\delta^{13}C$  values for the submicrometer  $(-23.7\pm0.8\%)$  and supermicrometer  $(-23.4\pm0.7\%)$  size ranges of particulate carbon were similar in a remote oceanic region with high biological productivity. Both of these values are close to the typical  $\delta^{13}C$  of OC in seawater. Likewise, the  $\delta^{13}C$  values of -22.1 to -20.8% obtained in the present study (Table 2) are similar to those of OC in seawater."

*Comment 11*: Page 12, lines 26-32 - I like the conclusions here. However, I note that in line 29 it says that Fig. 6 shows data where MSA is greater than 20 ng/m3, whereas on Figure 6 itself it says that MSA is greater than 30 ng/m3. Which is correct?

## *Reply 11*: "Twenty ng m<sup>-3</sup>" is correct. The number in Figure 6 has been corrected.

Comment 12: Page 13, line 7 - Couldn't one also add "and WSOC" after "(i.e., DMS)"?

Reply 12: We agree with the comment. The word "WSOC" has been added.