Interactive comment on "One-year observations of carbonaceous and nitrogenous components and major ions in the aerosols from subtropical Okinawa Island, an outflow region of Asian dusts"

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Anonymous Referee #2

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This manuscript describes long-term measurements of OC, EC, WSOC and major ions in atmospheric aerosols from Cape Hedo, which is located in the northwestern edge of Okinawa Island (Japan) and in the outflow region of Asian dusts. Although the site was used before for air pollution studies, the manuscript describes the most extensive and comprehensive set of measurements about aerosol chemistry performed so far at Cape Hedo. Results are interesting and improve our knowledge about the composition of the atmosphere in southern Japan and about air pollution transport from China to the North Pacific. Interesting to note in this study is the time variation of aerosol composition which was strongly dependent on the seasonal variation of air trajectories and type and strength of emission sources. The overall quality of this study is good. The scientific approach is sound and the text is well written and is easy to follow. I recommend publication of the manuscript in ACP after the following comments are addressed.

Specific comments:

Line 219 - I have doubts whether these two sources can explain the spring maxima for OC and WSOC because photochemical activity and biogenic emissions from plants normally peak during summer. Emissions from fossil fuel and biomass burning in Chinacannot be neglected as significant sources of OC and WSOC.

Response: In growing season biogenic emissions are also significant. We agree with reviewer's opinion that emissions from fossil fuel and biomass burning in China are significant sources of OC and WSOC. Hence, we have added the following sentence in the revised manuscript. "...photochemical oxidation of various organic precursors emitted from fossil fuel combustion and biomass burning as well as biogenic volatile organic compounds (BVOCs) from terrestrial plants." Please see lines 224 to 226.

Line 228 - Solar radiation alone cannot explain the high SOC levels during summer. What are the sources of gaseous precursors? Back-trajectories suggest an ocean source of VOC's. Is there any evidence of significant marine VOC sources in the area?

The authors should explain it better.

Response: Thank you very much. We agree with the reviewer's opinion. There is no evidence found for the emission of VOCs in this region. However, emission of VOCs from marine source can be found in many literatures. So, we have added following sentence in the revised manuscript. "....as well as emission of marine volatile organic carbon (Shaw et al., 2010)." Please see lines 237 to 238.

Line 234 - Airborne pollen is not expected to be as important in summer as it is in spring. The increase in the contribution of WIOM to TSP seems to be the result of a lower abundance of inorganic species during summer.

Response: Thank you very much. Water insoluble organic matter is also higher in summer (Miyazaki et al., 2011). So, we have reworded the sentence in the revised manuscript. "....the contribution of WIOM (water insoluble organic matter) to TSP became highest in summer (6.9%), suggesting more emission of WIOM in summer probably from biological sources (Miyazaki et al., 2011)." Please see lines 257 to 260.

Line 271 - This statement seems to be unfinished. The authors should clarify the usefulness of WSOC/OC ratios.

Response: Thank you very much. Following the reviewer's comment, we have added following the following sentence. ".....to understand photochemical oxidation (Miyazaki et al., 2010b) and extent of gas to particle formation of secondary WSOC (Hagler et al., 2007)." Please see lines 299 to 301.

Line 291 - The argument that "the lower WSOC/OC ratio may be caused by the sea to-air emissions of water-insoluble organic compounds that are produced by marine phytoplankton" must be sustained by references to previous studies about these emissions.

Response: Thank you for your suggestion. Following the reviver's suggestion, we added the following sentence. "Miyazaki et al. (2010b) reported high abundance of WIOC in the western North Pacific during summer." Please see lines 325 to 326.

Line 296 - Section 3.2.3, describing water-soluble ionic components, should be placed after comparison of EC and OC with previous studies in East Asia (section 3.2.4). Response: Thank you for your constructive suggestion. Base on it, we have changed the sentence structures. Please see lines 330 to 338 and section 3.4.

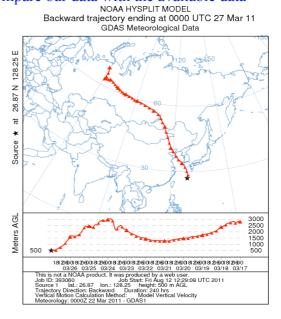
Section 3.2.3 - How do you explain the seasonal variation of MSA-? A comparison with other studies describing the seasonal variation of MSA- or DMS over or near the Pacific waters would be interesting.

Response: By comparing our data with literature, we found MSA⁻ concentration in literature during summer only. Thus, we compare our data with the available data

from the literature. Following the reviewer's comment, we have added following sentences in the revised manuscript.

"The summertime concentration of MSA⁻ in Cape Hedo is comparable with those in the western North Pacific during summer (Miyazaki et al., 2010a)." Please see lines 356 to 358.

Seasonal variation of MSA⁻ can be explained as follow. "During the longrange atmospheric transport, East Asian aerosols travelled over the marine regions (the East China Sea, Sea of Japan and Pacific Ocean) and resided over the marine atmosphere for 1 to 2 days. During the travelling, the aerosol from East Asia



is mixed with marine aerosols emitted from the ocean, which can be supported by individual backward trajectory." Thus, we added above sentences in the revised manuscript. Please see lines 352 to 356.

Lines 315 to 326 - Discussion about OC and EC concentrations should take into account that the size of sampled particles was different. TSP was sampled at Cape Hedo and PM2.5 or PM10 were sampled at the other sites. Therefore, a simple direct

comparison is not possible.

Response: Thank you for your suggestion. Following the reviewer's comment, we deleted the direct comparison. Please see lines 330 to 338.

Line 409 - How relevant are shipping emissions in the East China Sea? Are they significant contributors to $nssSO_4^{2-}$ concentrations at the Okinawa sampling site? Response: Only from the $nssSO_4^{2-}$ concentrations, it is not easy to estimate the shipping emissions from East China Sea. Because we cannot neglect the influence of shipping emission to Cape Hedo, we added the following sentence in the revised manuscript. "However, we also suspect a possible contribution of shipping emissions of $nss-SO_4^{2-}$ in the East China Sea to the Cape Hedo site during winter and spring." Please see lines 441 to 443.

Technical corrections Line 209 - The authors want to say "figure 4" instead of "figure 3". Response: Corrected.

Line 349 - The authors wanted to say "section 3.2.7" Response: Corrected.

Line 777 - What is the meaning of UF in figure 5? Response: UF means unanalyzed fraction. Please the caption of Figure 5.

References

- Hagler, G. S. W., Bergin, M. H., Smith, E. A., and Dibb, J. E.: A summer time series of particulate carbon in the air and snow at Summit, Greenland, J. Geophys. Res., 112, D21309, doi:10.1029/2007JD008993, 2007.
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- Miyazaki, Y., Kawamura, K., and Sawano, M.: Size distributions and chemical characterization of water-soluble organic aerosols over the western North Pacific in summer, J. Geophys. Res., 115, D23210, doi:10.1029/2010JD014439, 2010b.
- Shaw, S, L., Gantt, B., Meskhidze, N.: Production and emissions of marine isoprene and monoterpenes: A Review, Advances in Meteorology, Volume 2010, Article ID 408696, 24 pages, doi: 10.1155/2010/408696, 2010.