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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemical Characteristics of Marine Fine Aerosols over Sea and at Offshore Islands during Three Cruise Sampling Campaigns in the Taiwan Strait– Sea Salts and Anthropogenic Particles" *by* Tsung-Chang Li et al.

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Review for "Chemical Characteristics of Marine Fine Aerosols over Sea and at Offshore Islands during Three Cruise Sampling Campaigns in the Taiwan Strait- Sea Salts and Anthropogenic Particles," by Li et al. submitted to ACPD This paper presents filter-based PM2.5 composition measurements area over Taiwan strait where is not well characterized and compares with the condition at an offshore island site in Penghu island. This is a high quality data set and an important contribution to the field. I do recognize that this is a data set over a large marine area with different cruises

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and as such it is difficult to analyze, but I do not think the analysis at present is as thorough and coherent as it could be. Overall comments: This paper presents almost all the compositions can be measured in PM2.5 in three courses over Taiwan Strait, including the major ions, heavy metals and OCEC. The data quality is good however the overall impression is the authors just showed so much data there and did not find any impressed or interesting findings. I suggest to submit to other popular journals such as Atmospheric Environment or Atmospheric Research, unless the author can provide any interesting findings after further analysis.

Detailed comments: A. The tables and figures A1: Units need to be added to the table 2 3 5.

» Thanks for the comments. We have added the units in Tables 3, 4, and 5 per request. (see P26-P28)

A2: Sample numbers should be added in Table 1. Actually I was confused about the sampling method. Totally how many valid filters were collected in different cruises? If the filters of PM2.5-10 were contaminated, it can be moved to the supplemented materials as there are no important results.

» Thanks for the comments. In this study, only one fine particle sample was collected during each sampling course. The number of PM2.5 samples over sea and at the offshore islands during three cruises sampling campaigns was 4, 5, and 4, respectively (see P4, L11-L12 and Table 3, L27).

A3: Tables can contain large information than the one can be illustrated in the manuscript. The authors do not need to mention every data in the tables, but need to add some comparisons with the data in other literatures to rich the contents. Some interesting findings can be discovered during this process.

» Thanks for the comments. We have added the comparisons of the mass concentration and chemical composition data with previous literatures around the Taiwan Strait

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and East China Sea per request. The sentences have been revised as "Table 6 compares the concentrations of TC, OC, and EC in PM2.5 with previous studies. The total carbon concentrations were close to those at the Penghu site located at an offshore island where clean marine air can dilute PM2.5 from long-range transport, resulting in local emission accumulation and lower OC and EC levels at the Penghu Islands. The OC/EC ratios ranged from 3.0-7.0 on the southeastern coastline of China and from 1.9 to 2.9 on the southwestern coastline of the Taiwan Strait, respectively. The OC/EC ratios obtained from this study ranged from 2.6 to 2.8 at the Taiwan Strait which were generally lower than those reported by Chou et al., 2010 (2.6 to 2.9) and Tasi et al., 2010 (1.9 to 2.9). The comparison of OC/EC ratios showed the variation of carbonaceous species analyzed with different analytical methods. The carbonaceous concentrations of particulate matter analyzed using thermal optical reflectance (TOR) and thermal optical transmittance (TOT) were generally higher than those using elemental analysis (EA)." (see P14, L12-L20).

B. Logic B1: The major discussion including seven sections, and less comparison with the data over other areas, which makes the reader feel that the author is just loading the data. It is better to find some internal connection between these data and name each section following the findings. Or the author could try 3.1.1 to including some sections into one section. e.g. 3.3, 3.4 and 3.5 are all about the sea salt particles, which can be in one section.

» Thanks for the comments. We have converged and further revised the original Sections 3.3, 3.4 and 3.5 as Section 3.3 "Chemical Characteristics of PM2.5 over Sea and at the Offshore Islands" per request. (see P9, L9-P14, L20)

B2:The outline really need to be reconstructed. I often have this problem with my paper that the closely related information is not discussed until much later in the paper. Please try to discuss, at least briefly, all the relevant information on a topic at one place. Otherwise, some issues sounds like mentioned several times. Language or content can be more condensed.

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» Thanks for the comments. We have rearranged the content of the Section “Results and Discussion” in the manuscript. In this Section, we discussed and interpreted the results obtained from this study in the following seven subsections. Subsection 3.1 presents the spatiotemporal variation of PM2.5 concentrations over sea and at the Offshore Islands. Subsection 3.2 introduces the transport routes during three cruise sampling campaign. Subsection 3.3 aggregates the chemical characteristic of marine fine particles during three cruise sampling campaign. Subsection 3.4 describes the distribution and source indicators of PM2.5 during three cruise sampling campaign over sea and at the offshore islands. Subsection 3.5 reconstructs the material balance equation for the gravimetric mass of PM2.5 during three cruise sampling campaign. Subsection 3.6 identifies whether the presence of certain metallic elements in PM2.5 were primarily due to natural or anthropogenic processes during the sampling cruise. Subsection 3.7 compares the chloride deficit of PM2.5 with previous studies, respectively. (see P7, L10-L18).

C. Detailed Comments C1: The weather condition should be also mentioned at first as the sampler number is limited and the reader need some general idea on the background air mass condition.

» Thanks for the comments. After checking the records of wind speeds and wind direction in the sampling boat, we found that the prevailing wind came from the northeastern direction. Additionally, the backward trajectories showed that air masses blown from the north generally had higher PM2.5 concentrations than those from the south during the cruise sampling campaign. (see P4, L6-L10).

C2: Page 6 line 30 section 3.2: It should be ammonium poor area that (NH4)2SO4 is not favored. NH4HSO4 is more likely.

» Thanks for the comments. We have recalculated the relationship between NO3-, SO42- and NH4+. NO3-, SO42- and NH4+ that were associated together in the same particulate system in the likely form of NH4NO3, and [NH4]2SO4 or NH4HSO4. Par-

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ticulate phase NH4+ concentrations can be calculated using the stoichiometric ratios of different compounds and compared with the measurements. Nitrate is in the form of NH4NO3, while sulfate is in the forms of either (NH4)2SO4 or NH4HSO4 which can be estimated by equations (1) and (2).

Previous study indicated that ammonia is known to neutralize sulfuric acid irreversibly, and then nitric acid. Additionally, hydrochloric acid may react with gaseous ammonia to form ammonium chloride aerosols. However, in thermodynamic equilibrium conditions ammonium chloride is reported to be 2-3 times more volatile than ammonium nitrate (Stelson and Seinfeld, 1982) and its formation occurs later. Thus, ammonia is believed to be neutralized firstly by sulfuric acid and forms ammonium sulfate and/or ammonium bisulfate (McMurry et al., 1983; Wang et al., 2005; Du et al., 2010). In this study, we assumed that both sulfate (SO42-) and bisulfate (HSO4-) could be neutralized by ammonia with various portions (see P.9, L12-L24). Results obtained from the calculation of nitrate showed that the predominant inorganic compounds of PM2.5 were ammonium nitrate (NH4NO3) and ammonium bisulfate (NH4HSO4) (see P9, L26-L27).

C3: Page 8 line 9. the data obtained by equation (1) is in the middle level of these three results, however it is not a reason that "the most accurate method to estimate the sea salt concentrations was equation (1)".

» Thanks for the comment. We have revised the sentence describing the equations of sea salt estimates (see equation (3), P10, L25-L27).

C4: Page 9 line 10-15 about the anthropogenic particle influence, it can be one important topic in this manuscript. Suggest the authors make two major concern: anthropogenic source and sea salts Cl deficit. Besides Section 3.5 title is missing in the manuscript.

» Thanks for the comment. We have rearranged the sections according to other reviewer's suggesting and merged the original Section 3.5 into Section 3.3 in the manuscript per request. Moreover, a new Section 3.6 describes the presence of spe-

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cific metallic elements in PM2.5 primarily emitted from natural or anthropogenic processes during the sampling cruises. Section 3.7 investigates and compares the chloride deficit of PM2.5 with previous studies. (see P16, L1-P18, L2)

C5: Page 12 line 30, an accurate (OC/EC)pri value used in this study should be mentioned in the manuscript. The discussion of OCEC is really poor.

» Thanks for the comment. We have removed the calculation of SOC and POC according to the suggestion from other reviewers. Moreover, we have also revised the title of the Section 3.3.4 as "Carbonaceous Contents of PM2.5 over Sea and at the Offshore Islands," per request. (see P13, L15-P14, L20)

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2016-384/acp-2016-384-AC2-supplement.pdf

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