Responses to reviewer #2 comments

Anonymous Referee #2
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General Comments

The manuscript explores the chemical composition of water-soluble inorganic ion over a remote marine atmosphere in the western North Pacific. TSP aerosol samples are collected from Chichijima Island and analyzed for water-soluble ions such as Na+, NH4+, K+, Mg2+, Ca2+, Cl-, NO3-, SO42- and MSA-. The manuscript covers a 12-year observation and focuses on the impact of long range transport from East Asia on water-soluble ionic composition. Furthermore, the study identifies the decadal variability in nitrate, sulfate and nssK+ particularly considering anthropogenic emissions and biomass burning originating from East Asia. In this respect, it may be the interest of scientific community. Consequently I suggest acceptance of the manuscript. However, before that the manuscript should be revised.

Response: Thanks for the helpful comments on our manuscript, which improve the original scientific content.

Specific Comments

Title: I suggest modification of the title. Although the title implies that the water-soluble inorganic ions are of interest, the organic water-soluble methane sulfonic is also discussed in the text. For example, whole section (3.7) is dedicated to methane-sulfonic acid. It would be misleading to retain' inorganic' before ions. The discussion about the dust transport from the Asia is particularly limited (see comments below). Unlike the 'the title' (as it implies), the manuscript mainly focuses on the pollution transport from Asian continent. To illustrate, the impact of pollutants originating from Asia on the annual variability of chemical species is discussed in detail (in section 3.5). Thus, please modify the title according to the abovementioned statement.

Response: Based on the referee's suggestion, we modified the title by deleting the term of "inorganic". Thanks for the helpful suggestion.

Experimental

2.1. Sampling site and aerosol sampling: Please give more information about the sampling. Was the aerosol sampling carried out daily or weekly? What was the degree of blank contributions to water-soluble ions?

Response: Aerosol sampling was carried out on a weekly basis. Contributions from the field blanks varied between 0-8% and 0-2% of real samples for anions and cations, respectively, during the sampling period. These points have already been noted in the manuscript. Please see lines 122 and 147-149 in the revised manuscript.

2.2. Analysis of chemical species: In order to determine the concentrations of the water-soluble ions a punch of 2mm diameter from each filter was extracted. Was a punch of 2mm diameter representative for the collected aerosol sample when one considers the area of the filter? Please specify this issue.

Response: It seems that the reviewer might have confused with used filter area. We used a punch of 20 mm in diameter (not 2 mm) to determine the concentrations of the water-soluble ions. Please see line 133 in the revised manuscript.

Results and Discussion

3.1. Evaluation of Non Sea Salt Analysis: Please move this section to Experimental section, since this section simply defined the calculation of non-salt fractions.

Response: Following referee's suggestion, we moved this section to Experimental section. Please see section 2.5 in the revised manuscript.

3.3. Temporal Variation of Major Inorganic Species, MSA-/nssSO42- and S+/S-
3.4. Monthly Variations of Major Inorganic Species, MSA-/nssSO42-:

Please supply a table that shows relationship between the water-soluble species. Such an application would be useful to determine the (a) similar sources, (b) similar generation and/or removal mechanism and/or (c) similar transport patterns.

(Page 10, lines 286-289): It is claimed that drastic increase in the concentration of nssCa during spring was associated with dust transport originated from Asian continent. However, this claim should be supported by using dust episode. It would be useful to identify at least one dust event and discuss the episode in detail by applying ground measurements, back trajectories and satellite images.

Response: Regarding to the relation between water-soluble species, we added correlation coefficient matrix in the revised manuscript. Concerning the dust event, we discussed Ca episodes during the study period (in the year 2002) which can be related with variations in MSA. Please see lines 492-508 in the revised manuscript.

3.6. Percent Contribution of Major Ions to Total WSIM

The depletion of the Cl was ascribed to reaction between the alkaline sea-salt particles and the acidic species namely, sulfuric acid, nitric acid and oxalic acid (page 12, lines 349-367). Considering the written text, it is not clear which acid or acids responsible for the depletion of Cl. This claim should be clarified. For instance, the relationship between Cl depletion and acid species might be useful to address this issue.

Response: Following referee's suggestion, we have discussed the relations between chloride depletion and acidic species as given below.

"In order to investigate which acids are responsible for the depletion of chloride, we performed regression analysis between the Cl^-/Na^+ mass ratio and acidic species, nss-SO_4^{2-}, NO_3^-, MSA^- and oxalic acid for different seasons during 2001-2012 as shown in Figure 10. The regression analysis was verified by *t*-test. The results show that the differences between Cl^-/Na^+ mass ratio and acidic species are statistically significant with two tailed P value <

0.001 for each season during the study period. For all seasons, nss-SO_4^{2-} moderately correlated with Cl^-/Na^+ mass ratios with negative correlation coefficients (R^2) of 0.38, 0.29, 0.35 and 0.45 for winter, spring, summer, and autumn, respectively, whereas NO_3^- moderately correlated during winter (R^2= -0.30), weakly correlated in autumn (R^2= -0.22) and has no correlation in spring and summer. These results suggest that sulfate has more responsibility for the chloride depletion than nitrate. On the other hand, the biogenic tracer, MSA^-, moderately correlated during summer (R^2= -0.29) and has weak correlation in winter and spring. Freshly emitted MSA and H_2SO_4 (from oceanic biological productivity associated with the upwelling of nutrient rich water) are also little contribute to the chloride depletion by coating with sea salts, especially in summer. Interestingly, during spring, the Cl^-/Na^+ mass ratio did not correlate with NO_3^-, MSA^- but weakly correlated with nss-SO_4^{2-}. These results suggest that some other organic acids such as oxalic acid (because of its high abundance during spring) are responsible for the chloride depletion during spring. In fact, we found that oxalic acid significantly correlate with the chlorine loss in winter (R^2=-0.30), spring (-0.28) and autumn (-0.36) (see Figure 10d). These results confirm that oxalic acid plays an important role in a chlorine loss."

The above lines are briefly noted in the revised manuscript. Please see lines 419-439.

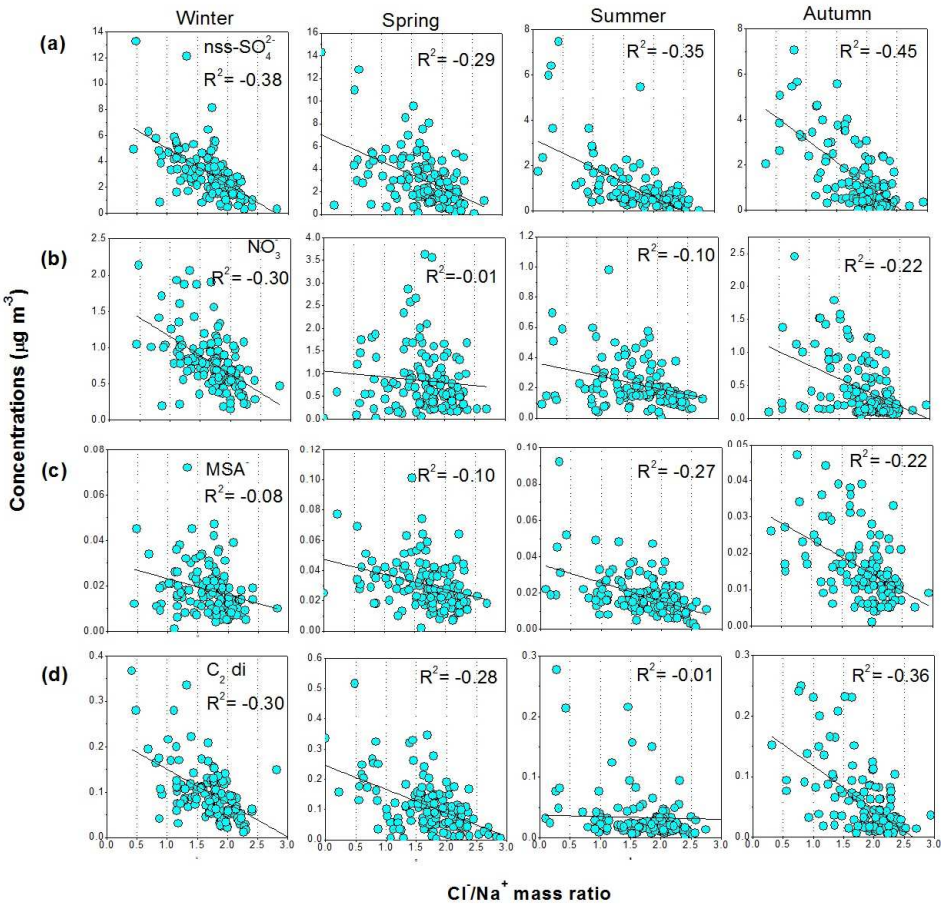

Figure 10. Relations between chloride depletion (Cl^-/Na^+ mass ratio) and acidic species (a) nss-SO_4^{2-}, (b) NO_3^-, (c) MSA^- and (d) oxalic acid (C_2di) for different seasons over the western North Pacific.