

## Responses to reviewer #1 comments

**Interactive comment on “A 12 year observation of water-soluble inorganic ions in TSP aerosols collected at a remote marine location in the western North Pacific: an outflow region of Asian dust” by S. K. R. Boreddy and K. Kawamura**

### **Anonymous Referee #1**

Received and published: 13 April 2015

This is an interesting manuscript presenting 12 years of inorganics ions (TSP) measured at the remote location of Chichijima Island (western North Pacific) which I consider as suitable for publication in ACP after minor revisions.

**Response:** Thank you for the careful reading and valuable suggestions to improve the scientific content of our paper.

Main comments:

1) A comparison with similar measurements performed at other remote locations is missing. I suggest to add a short review of previous studies presenting inorganic ions aerosol measurements performed at other remote sites in order to compare them with the values reported in this manuscript. This kind of comparison could be interesting especially for the ratios (i.e.  $MSA/nss-SO_4^{2-}$ );

**Response:** Based on the reviewer’s comment, we added a short review on comparison of major inorganic ions over the North Pacific and results are summarized in Table 4. Thanks for the helpful suggestion.

**Table 4.** Mean concentrations of major ions at Chichijima Island from 2001-2012 and those at several other remote marine locations in the Pacific

Location (data set)	$NO_3^-$	$nss-SO_4^{2-}$	$MSA^-$	References
		<u>Present study</u>		
Chichijima (2001-2012)	0.58±0.07	2.12±0.42	0.02±0.00	
		<u>Other remote marine locations</u>		
Fanning Island (1981-86)	0.16±0.08	0.67±0.27	0.04±0.01	Savoie et al., (1989)
Nauru	0.16±0.09			Savoie et al., (1989)
Funafuti	0.10±0.07			Savoie et al., (1989)
American Samoa (1983-87)	0.11±0.05	0.34±0.14	0.02±0.01	Savoie et al., (1989)
Rarotonga	0.12±0.08			Savoie et al., (1989)
Midway (1981-2000)	0.29±0.16	0.56±0.45	0.02±0.01	Prospero and Savoie (2003)
N. Caledonia (1983-85)		0.42	0.02	Savoie and Prospero (1989)

We added the following sentences in the revised manuscript. Please see lines 517-546 and Table 4.

“The mean concentrations of  $\text{NO}_3^-$ ,  $\text{nss-SO}_4^{2-}$ , and  $\text{MSA}^-$  at Chichijima during the period 2001-2012 are compared with those from several other remote marine sites in the Pacific as summarized in Table 4. Results from the Chichijima data show that mean concentrations of  $\text{NO}_3^-$  and  $\text{nss-SO}_4^{2-}$  are higher than those from other remote marine locations. The mean concentration of nitrate ( $0.58 \mu\text{gm}^{-3}$ ) at Chichijima is more than 4 times higher than those from other remote marine sites (Fanning, Nauru, Funafuti, American Samoa, Rarotonga, and N. Caledonia) and more than twice higher than those from Midway. Whereas concentrations of  $\text{nss-sulfate}$  at Chichijima ( $2.12 \mu\text{gm}^{-3}$ ) is 4 times higher than at Fanning, Midway, and N. Caledonia and more than 7 times higher than those from American Samoa and Norfolk. The mean concentration of  $\text{MSA}^-$  ( $0.02 \mu\text{gm}^{-3}$ ) at Chichijima is comparable to those from other remote marine locations (see Table 4). These results suggest a similarity to that of the oceanic biological productivity in the North Pacific. In contrast, the mean  $\text{MSA}^-$  concentration at Fanning in the equatorial Pacific is about twice higher ( $0.044 \mu\text{gm}^{-3}$ ) than Chichijima. Savoie and Prospero (1989) have found high biological productivity associated with the upwelling of nutrient rich water near the equatorial divergence with mean DMS levels of  $3.8 \text{ nmol/l}$  in the surface ocean. They also documented that in the oligotrophic regions, the mean concentrations of  $\text{MSA}$  in the air and DMS in the seawater vary over the narrow range from  $0.02\text{-}0.03 \mu\text{gm}^{-3}$  and  $1.4\text{-}1.7 \text{ nmol/l}$ , respectively.

The mean concentration ratio ( $\text{MSA}^-/\text{nss-SO}_4^{2-}$ ) at Chichijima is 0.02, which is lower than those of other remote marine locations by a factor of 5-7, indicating a substantial impact from continentally derived sulfate. At the tropical stations, American Samoa and Fanning Islands,  $\text{MSA}/\text{nss-SO}_4^{2-}$  ratios exhibit similar values with mean ratios of 0.07 and 0.06, respectively, indicating a cleanest locations regarding to the continental inputs (Arimoto et al., 1987). This result further supports our assumption that Asian dusts can act as an important source of nutrients that stimulate the DMS production in the ocean surface followed the emission to the marine atmosphere over the western North Pacific. However, it is rather less important that yield of  $\text{MSA}$  from DMS oxidation is enhanced as a function of temperature (Hyens et al., 1996).”

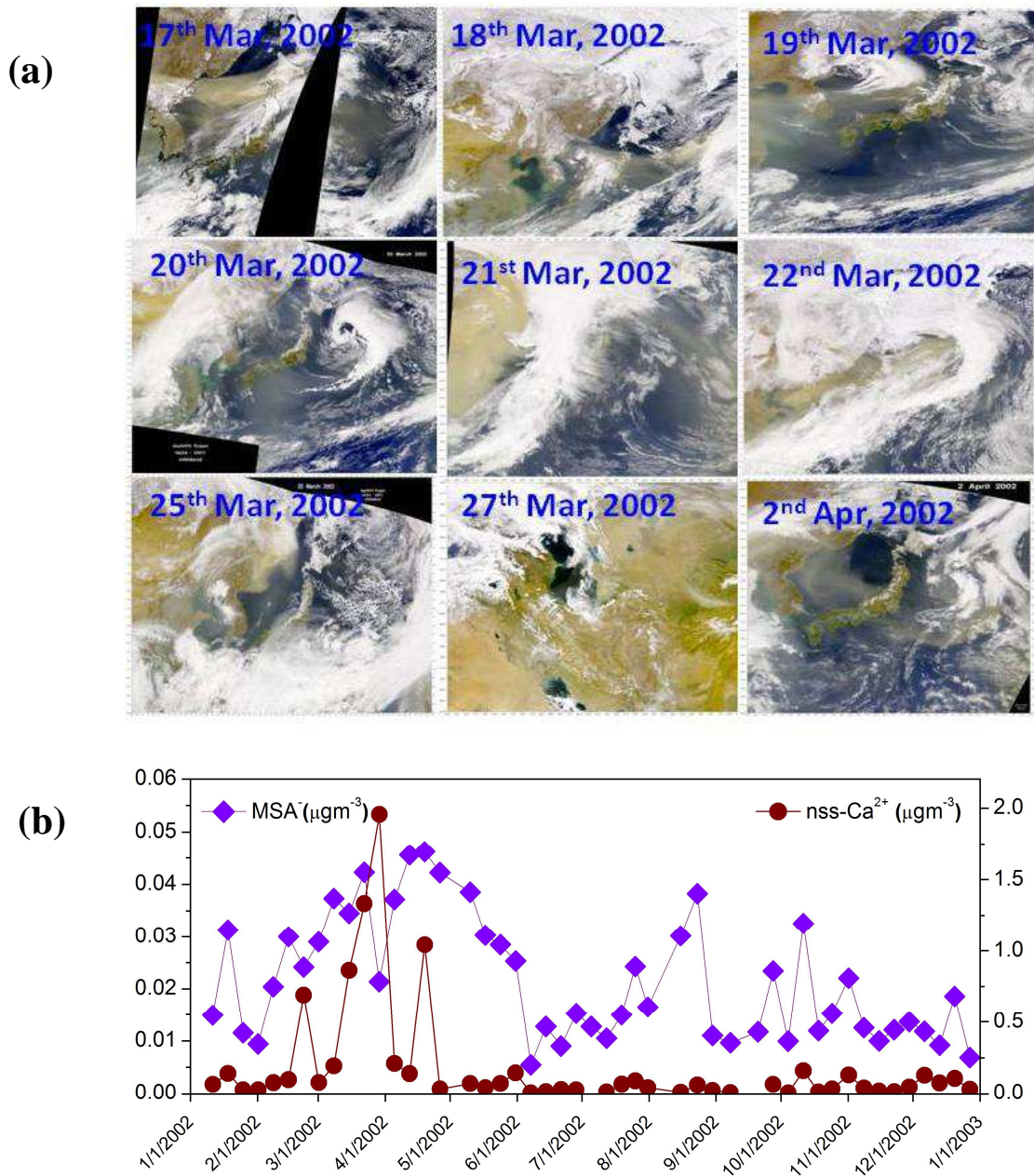
2) Some Figures need to be re-edited: y-axis missing in Fig. 2 and 3; Fig. 5:  $\text{S}^+/\text{S}^-$  is presented in the Figure 5, but  $\text{S}^-/\text{S}^+$  is described in the text; Figure 6h;

**Response:** We re-edited the two figures. Please see Figures 2 and 3 in the revised manuscript and also see lines 232, 234, and 235.

3) Paragraph 3.8. : Here the possible relationship between  $\text{MSA}^-$  and  $\text{nss-Ca}^{2+}$  is discussed comparing monthly averages. Are there any intense Ca episodes during the study period which can be related with variations in  $\text{MSA}$ ? For example Ca episodes of few days accompanied by statistically significant variations in  $\text{MSA}$ ?. This could help in identifying any  $\text{MSA-nssCa}^{2+}$  relationship.

**Response:** Following the referee’s suggestion, we revisited the data of Ca and  $\text{MSA}$  during the study period in which we found some relation in the variations of Ca and  $\text{MSA}$ . We added

few sentences in the revised MS. Please see Figure 12 and lines 498-515 in the revised manuscript. Thanks for the helpful suggestion.



**Figure 12.** (a) The Sea-viewing Wide Field-of-view Sensor (SeaWiFS) images that captured the large Asian dust storm visible over the Sea of Japan and North Pacific Ocean during March 17-April 2, 2002, (b) temporal variations of MSA<sup>-</sup> and nss-Ca<sup>2+</sup> concentrations during 2002 over the western North Pacific. The black regions in Figure 12a are the gaps between consecutive SeaWiFS' viewing swaths and represent areas where no data were collected.

“To further clarify the relations between MSA<sup>-</sup> and nss-Ca<sup>2+</sup>, we examined the intense nss-Ca<sup>2+</sup> episodes during the study period (March 2002), which can be related with variations

in MSA<sup>-</sup> as shown in Figure 12 as a typical example. Figure 12a shows the SeaWiFS (Sea-viewing Wide Field-of-view Sensor, flying aboard Orbview-2) images, which captured the large Asian dust storms over the North Pacific during March 17-April 2, 2002. Dust storms originate in the deserts of North China and Mongolia. The East Asian dust storm appears to have diminished somewhat on March 20, 2002, as compared to previous days. However, there seemed a new batch of dust rising toward the left side of this image. This scene spans from eastern Asia across Japan and over the western North Pacific, where the dust was partly entrained by a low-pressure system. On the other hand, the possible variations of MSA<sup>-</sup> concentrations related to the East Asian dusts are shown in Figure 12b. Interestingly, we found higher levels of MSA<sup>-</sup> after the Asian dust deposition over the ocean surface. This evidence strongly reveals that Ca episodes supply the nutrients to significantly stimulate plankton blooms accompanied by statistically significant variations in MSA concentrations in the atmosphere few days after the episodes. This result also demonstrates that Asian dusts can act as an important source of macro and micro nutrient including iron for phytoplankton and thus sea-to-air emission of DMS over the western North Pacific.”

4) Tables 1 and 2. Are the presented differences statistically significant between each pair of variables (seasons or year)? Please, clarify which differences are statistically significant.

**Response:** Considering the comment, we removed the result of t-test and applied new statistical approach for checking the annual and seasonal differences. The results are reported in Table 1 and Table 2. Please see Tables 1 and 2 in the revised manuscript.

5) The presence of any trend should be demonstrated by applying tests such as Mann-Kendall (for annual averages) or Theil-Sen (also for monthly averages).

**Response:** Based on the reviewer’s comment, we demonstrated annual and monthly trends using Mann-Kendall and Theil-Sen slope, respectively. The result of these tests are reported in Tables 1 and 2 in the revised manuscript and also given below for the reviewer reference.

“Monthly or seasonal mean concentrations of major inorganic ions (mean±SD) during 2001-2012 at Chichijima Island in the western North Pacific are reported in Table 1. The presence of monthly averaged trend is demonstrated by Theil-Sen Slope test (Sen, 1968; Theil, 1950). The results show that these differences are statistically significant with Theil-Sen slope values of less than 0.01.”

“Annual mean concentrations of major inorganic ions (mean±SD) for different seasons during 2001-2012 are reported in Table 2. The presence of annual averages trend is demonstrated by Mann-Kendall nonparametric test for each season during study period and the results are also reported in Table 2. The Mann-Kendall trend test (Mann, 1945 and Kendall, 1975) is one of the widely used non-parametric tests to detect significant trends in time series. In this test, the absolute value of Z is compared to the standard normal cumulative distribution to define if there is a trend or not at the selected level  $\alpha$  (=0.01) of significance. A positive (negative) value of Z indicates an upward (downward) trend.”

Please see lines 263-267 and 319-326 in the revised manuscript.