

## ***Interactive comment on “Sources of reactive nitrogen in marine aerosol over the Northwest Pacific Ocean in spring” by Li Luo et al.***

**Anonymous Referee #2**

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This manuscript describes ship-board measurements of marine aerosols collected during two cruises around the East China Sea and the northwestern Pacific Ocean in 2014 and 2015. In this manuscript, authors reported concentrations of water-soluble total nitrogen (WSTN), water-soluble organic nitrogen (WSON), nitrate ( $\text{NO}_3^-$ ) and ammonium ( $\text{NH}_4^+$ ), and values of  $\delta^{15}\text{N}$ -WSTN and  $\delta^{15}\text{N}$ - $\text{NO}_3^-$  in aerosols as well as dissolved organic nitrogen (DON) and  $\text{NO}_3^-$  concentrations and  $\delta^{15}\text{N}$ -DON and  $\delta^{15}\text{N}$ - $\text{NO}_3^-$  values in sea surface water, which provide good indications where future studies can understand possible sources of atmospheric WSON and air-sea exchange of N species. I believe that the contents, including data, of the manuscript should be eventually published because of scarcity of atmospheric WSON observation and its significance in biogeochemical N cycle. However, the manuscript is NOT publishable

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in its current format. I recommend publication of the manuscript after a major revision and the improvement of English. Overall, authors were successful in addressing their ideas in the Results and discussions section, though. A number of changes are recommended below.

Major points

1. The title of this manuscript is “Sources of reactive nitrogen in marine aerosol over the Northwest Pacific Ocean in spring”; however, the authors mainly described the spatial distributions and concentrations of atmospheric reactive N species and potential sources of WSON. I recommend the authors to describe in their manuscript the sources of atmospheric inorganic N species, although they are relatively well-known compared to that of WSON.

2. (Page 4, line 98–116) It is not clear that how many, what kind of aerosol samplers and filters were used during the cruises, and how avoid ed contamination from ship’s exhaust. Was a impactor used to separate  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$ ? More detail information on sampling method should be described in the manuscript, although the authors referred to Luo et al. (2016). In general, a pre-combusted glass fiber or a quartz filter is used for determination of WSON. If the authors used the same method for aerosol sampling described in Luo et al. (2016), the authors should explain about the treatment of aerosol filter samples and field blank concentrations and blank correction, because Luo et al. (20016) used a Whatman 41 cellulose filter. For determination of DON in sea surface water, the authors mentioned that a  $0.2 \mu\text{m}$  filter was used to remove particulate matters in sea surface water. Usually, a pre-combusted GF/F filter is used to remove particulate matter and minimize the influence of organic matter from the filters on DON concentration in seawater. Please update that what kind of filter was used for filtration of seawater sample. I am also wondering if any consensus reference material (CRM, e.g., deep Florida Strait water from Hansell lab, University of Miami) was used during DON measurement to check the accuracy of analysis.

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3. (Page 6, line 188–Page 7, line 191) Dry deposition velocity. It is unclear if marine aerosols are segregated into PM<sub>2.5</sub> and PM<sub>10</sub> during the aerosol sampling as mentioned in question 2. Although size distributions of atmospheric N species can vary on meteorological conditions, it is known that, in the marine atmosphere, both atmospheric NH<sub>4</sub><sup>+</sup> and WSON primarily exist on fine mode aerosols, whereas atmospheric NO<sub>3</sub><sup>-</sup> is predominantly associated with coarse mode aerosols (e.g., Nakamura et al., 2006). I recommend the authors to describe more detail that what dry deposition velocity was used for each N species.

4. (Page 8, line 228–232) The authors compared their NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> concentrations with those by Miyazaki et al. (2011) to explain why higher concentrations of inorganic N species were observed during the period of this study (spring). The authors mentioned that the study by Miyazaki et al. (2011) was carried out over the same regions. I doubt about it. The cruise by Miyazaki et al. (2011) was conducted from 44°N to 10°N along 155°E, which covers the subarctic to subtropical northwestern Pacific region. Although the study by Miyazaki et al. (2011) was carried out in summer, different sampling season is not the only reason why the authors observed high inorganic N species during their study period.

5. (Page 8, line 233–242) The authors described that “Likely the source of WSON in background aerosol did not share the same source with NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup>” (line 234–235), as if DON in sea surface water is the only source of atmospheric WSON in the open ocean. What is the grounds for this? Because high atmospheric WSON and inorganic N species concentrations were observed in the East China Sea and inorganic N was also detected in the open ocean, the long range transport of anthropogenic WSON to the open ocean should be considered.

6. (Page 8, line 252–Page 9, line 263) The results of characteristics of CHON molecular compounds shows that 13%, 3% and 19% of marine aerosols collected in the East China Sea, northwestern Pacific Ocean during dust period and northwestern Pacific Ocean during non dust period, were derived from biological sources, respectively.

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Does this mean that 87%, 97%, and 81% of marine aerosols collected in the same regions were affected by anthropogenic sources? It seems like the contribution of biogenic sources to atmospheric WSON is still low in the open ocean. What is the contribution of biologically-derived atmospheric WSON in the other oceanic regions?

7. (Page 11, line 331–Page 12, line 355) The authors described that atmospheric reactive N dry deposition flux can account for 14%–58% of the low  $\delta^{15}\text{N-NO}_3^-$  in the northwestern Pacific Ocean during the spring. It is surprising to me that atmospheric reactive N deposition has a significant influence on  $\delta^{15}\text{N-NO}_3^-$  values. My question is that dry deposition of atmospheric reactive N is strong enough to affect or change  $\delta^{15}\text{N-NO}_3^-$  values below the thermocline in the northwestern Pacific Ocean? What is the depth of thermocline in the northwestern Pacific Ocean in the spring season? I recommend the authors to estimate the contribution of atmospheric reactive N dry deposition to primary production in their study area. I think most primary production in the East China Sea and northwestern Pacific Ocean is controlled by nutrients in seawater, which implies that main factor for controlling  $\delta^{15}\text{N-NO}_3^-$  values in the ocean is marine N cycle.

#### Minor points

1. (Page 5, line 136–137) How did the authors obtain the recovery efficiency (i.e., 95–105% (n = 6)) of WSTN and TDN?

2. (Page 5, 155–156) The authors mentioned that the extraction efficiency on a carbon basis was on average  $46 \pm 24\%$  (n = 44). Does it mean that 64% of organic compounds in the extract was not identified?

3. (Page 6, line 159–163) The uncertainty of WSON estimated from propagating errors of WSTN, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> should be added.

4. (Page 6, line 175) The authors mentioned that [NH<sub>4</sub><sup>+</sup>] in sea surface water typically less than 0.05  $\mu\text{mol L}^{-1}$ . Is this a common condition in the East China Sea and the

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Northwestern Pacific Ocean during the sampling period (i.e., spring season)? Sea surface [NH<sub>4</sub><sup>+</sup>] can vary depending on sampling season and locations.

#### References

Miyazaki, Y., Kawamura, K., Jung, J., Furutani, H. and Uematsu, M.: Latitudinal distributions of organic nitrogen and organic carbon in marine aerosols over the western North Pacific, *Atmos. Chem. Phys.*, 11(7), 3037–3049, doi:10.5194/acp-11-3037-2011, 2011.

Nakamura, T., Ogawa, H., Maripi, D. K. and Uematsu, M.: Contribution of water soluble organic nitrogen to total nitrogen in marine aerosols over the East China Sea and western North Pacific, *Atmos. Environ.*, 40(37), 7259–7264, doi:10.1016/j.atmosenv.2006.06.026, 2006.

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