

## ***Interactive comment on “Fog composition at Baengnyeong Island in the Eastern Yellow Sea: detecting markers of aqueous atmospheric oxidations” by A. J. Boris et al.***

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*Dear Referee,*

*Thank you for your comments. Please see below for responses to your specific questions and suggestions (in italics). Several of your comments have been responded to by altering or adding figures; these relevant figures are attached at the end of this response.*

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### **General Comments from the Referee**

This manuscript reports measurements of chemical composition of fog water at Baengnyeong Island in the Yellow Sea. It provides a very rich set of measurements of fog water chemical composition in a region where fewer field measurements are available. The paper shows evidence that strongly suggest aqueous phase oxidation of organics upwind of the island, thereby, providing insight into the atmospheric chemical processing in this specific region. The chemical components of the fog water measured in this study indicate that the samples were influenced by emissions from anthropogenic activities, marine background and forest fires. The most dominant chemical species measured, ammonium, showed no correlation to wind direction at the measurement site, indicating long-range transport. The concentration of S(IV) was low relative to  $\text{SO}_4^{2-}$ , which suggests that oxidation may have occurred upwind of BYI. I think this article is written clearly with easy to interpret plots. I recommend this manuscript for publications, although I suggest a few minor revisions.

### **Specific Comments:**

1. Page 24881, Line 7: Could include more recent measurements of cloud water composition over the Eastern Pacific in the references here - Z. Wang, A. Sorooshian, G. Prabhakar, M. M. Coggon and H. H. Jonsson (2014): Impact of emissions from shipping, land, and the ocean on stratocumulus cloud water elemental composition during the 2011 E-PEACE Field Campaign, Atmospheric Environment, doi:10.1016/j.atmosenv.2014.01.020

*We have added the E-PEACE campaign results to our discussion of field campaigns during which aqueous atmospheric organic processing evidence has been detected. Unfortunately, the measurements did not include, to our knowledge, measurement of peroxides or S(IV). The following sentence has been added to the manuscript with regards to recent measurements of atmospheric constituents made during the E-PEACE campaign:*

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*“Successful approaches toward identifying the location and timing of AAOP reactions have included the use of coincident non-specific molecular tracers such as organic acids (Sorooshian and Varutbangkul, 2006; Sorooshian et al., 2013), the predominance of oxalic acid and SO<sub>2</sub> in a size mode generated from aqueous processes (the droplet size mode, Crahan et al., 2004), and high carbon oxidation states Chen et al. (2015).”*

*The following sentence has also been added to the manuscript with regards to shipping emissions impacts (line 414):*

*“International shipping lanes could also contribute to the measured SO<sub>2</sub> concentrations in BYI fog: some of the world’s largest shipping ports are located in the Yellow Sea (Streets et al., 2000). The contribution of fine particle ( $\leq 2.5 \mu\text{m}$  diameter) SO<sub>2</sub> has been estimated at  $\leq 15\%$  from ship oil combustion in this region (Lauer et al., 2007), and shipping routes in the Yellow Sea have been identified as major SO<sub>2</sub> source regions Kang et al. (2006). Shipping emissions have also been associated with elevated concentrations of other atmospheric constituents, including NO<sub>3</sub> Prabhakar et al. (2014).”*

**2.** Page 24884, Line 20: Are the r-squares reported here (and elsewhere in the paper) statistically significant?

*Statistical significance (p-values) has been added to the text. Specifically, the following section describing organic acids contributions has been changed (line 455):*

*“Concentrations of low molecular mass organic acids were strongly correlated with one another (air equivalent concentrations):  $r^2=0.83$  on average, with probability  $p \leq 0.01$  of random correlation, ranging from 0.47 to 0.99 ( $p \leq 0.001$  to 0.2) for C<sub>1</sub>-C<sub>6</sub> mono and di-acids. The predominance of succinate suggests a major anthropogenic source of organic acids at BYI (Kawamura and Usukura, 1993). While substantial contributions of oxalate to TOC*

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*suggest that AAOP reactions took place, they are not unequivocal evidence of it, since other atmospheric sources for oxalate have been documented (e.g., Kawamura and Kaplan, 1987; Yamasoe et al., 2000). The presence of MSA in the observed samples and its correlation with other measured organic acids ( $r^2 \leq 0.7$  and  $p \leq 0.02$  with all low molecular mass organic acids,  $r^2=0.88$  and  $p \leq 0.001$  with oxalate) additionally supports the occurrence of AAOP upwind of fog water collection.”*

**3.** References Missing: Page 24874, Line 24-25: Kim et al., 2011 Page 24875, Line 13: Yoo et al., 2010

*These references have been added to the manuscript reference section.*

**4.** Figure 2: Interpretation of the map would be easier if it included more labels.

*This figure has been re-made, and now includes regional labels, including many high-throughput shipping ports (Fig. 2).*

#### **Technical Corrections:**

Page 2, Line 10: There should be a comma before the word gradient for clarity. Page 24877, Line 24: Please explain the abbreviation CSU

*These corrections have been made.*

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## References

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Sorooshian, A., Wang, Z., Coggon, M. M., Jonsson, H. H., and Ervens, B. (2013). Observations of Sharp Oxalate Reductions in Stratocumulus Clouds at Variable Altitudes: Organic Acid and Metal Measurements During the 2011 E-PEACE Campaign. *Environmental Science Technology*, 47:7747–7756.

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## Relevant Figures

### Figure Captions:

**Figure 2: Map Back Trajectories.** Back trajectories of air masses intercepted during fog events (72 hours at one hour time resolution; HySPLIT). Locations labeled on plot include Mount Tai, where previous atmospheric water chemical measurements were made, and the highest throughput shipping ports in the region pictured. The BYI ARC is shown as a gold diamond. Sectors were defined to determine whether particular source regions existed for chemical constituents of the fog: northerly (30 June and 1 July); westerly (2, 14, and 15 July); southerly (18 and 20 July); and easterly (5 July). Each trajectory was initiated at the approximate beginning of a fog event. Imagery from NASA Blue Marble; plot generated using Python Matplotlib Toolkit BaseMap.

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, 15, 24871, 2015.

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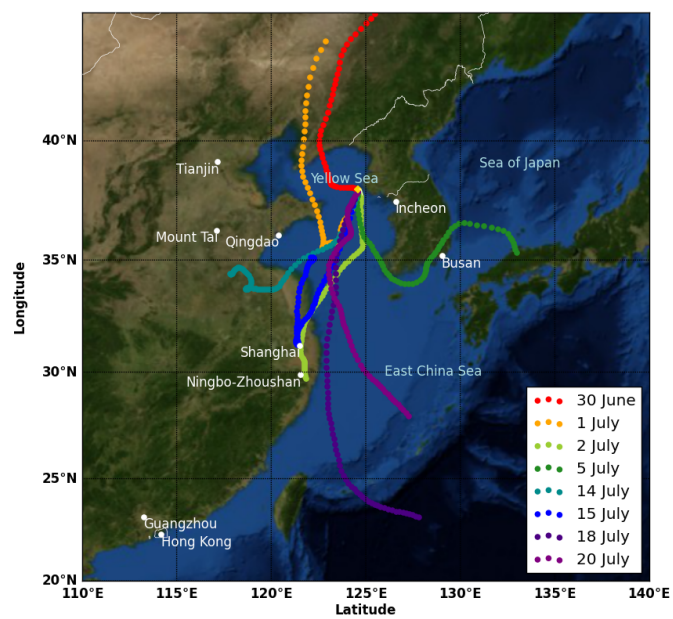


Fig. 1. Figure 2. Map Back Trajectories.

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