

Response to Anonymous Referee #2

Thank you very much for reading our manuscript, titled “A 15-year record (2001–2015) of the ratio of nitrate to non-seasalt sulfate in precipitation over East Asia,” (#acp-2017-848). We would like to thank for providing helpful and constructive comments. We have revised our manuscript according to the reviewer’s comments and suggestions. We believe that these revisions address all points raised by the reviewer. Our point-by-point responses are provided below, and revisions are indicated in blue in the revised manuscript.

Sincerely,
Syuichi Itahashi

The long-term trend of NO_3^- and SO_4^{2-} concentration in precipitation in China, Korea, and Japan was analyzed based on EANET monitoring data. The results showed certain correlation between China’s emission and the wet deposition in East Asia. The manuscript was quite well written. However, the method for trend analysis was not clear introduced. Since there are a lot of anthropogenic sources and other natural sources of Na^+ than sea-salt in China, the necessity of sea-salt correction for SO_4^{2-} concentration in precipitation need be reconsidered. Some detailed comments are:

In the revised manuscript, the methodology to calculate linear trend are explicitly introduced. Please refer the reply to the specific comment of 5). Regarding Na sources, we performed and checked the analysis of the correspondence between SO_4^{2-} and nss-SO_4^{2-} concentration in precipitation around Beijing in China. Please refer the reply to specific comments of 3) and 6).

1) Page 1, Line 15: The ratio is an index only for relative contribution of SO_2 and NO_x .

We have added a short note to clarify this point as follows on P1, L17:

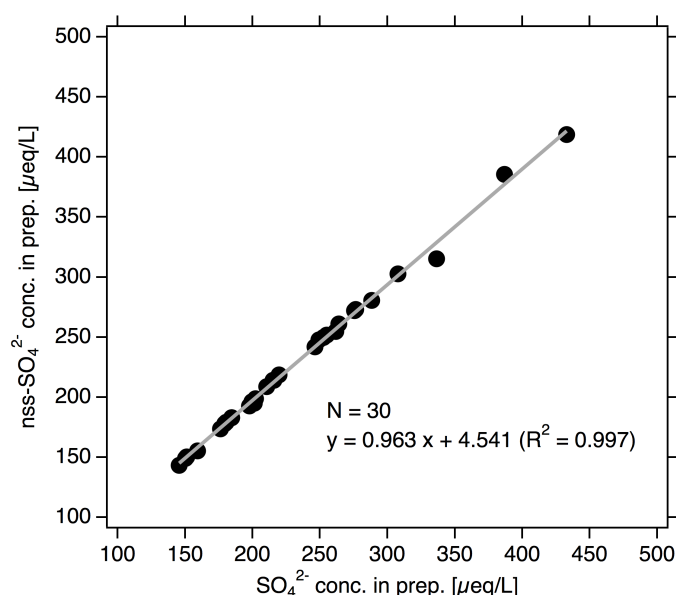
“...to investigate the relative contributions of these acidifying species.”

2) Page 3, Line 21: As I know, a long-term (2001-2013) monitoring of precipitation chemistry at one of the IMPACTS site (TSP) was recently reported by Yu et al. (ES&T, 2017).

Thank you for bringing this to our attention. We have added the suggested reference and a brief explanation of the study (e.g., P3, L22; P5, L28-29, P13, L21-22).

3) Page 4, Line 21: I suggest not to do sea-salt correction for SO₄²⁻, at least in China.

We agree that there are Na sources other than seasalt, particularly in China. Na might be affected by wind-blown dust in spring, biomass burning in autumn, and coal burning in winter. The impact of Na can be considered as minor on annual mean base because rainfall event mostly occurred during summer. The following figure shows the relation between annual mean (2008-2010) of SO₄²⁻ and nss-SO₄²⁻ concentration in precipitation analyzed on ten sites around Beijing in China. From this analysis, we can confirm that there are no significant differences between SO₄²⁻ and nss-SO₄²⁻ concentration in precipitation in China. On the other hand, in Korea and Japan, where surrounded with oceans, Na source from sea-salt would have impact. In this study, to ensure consistent data types for China, Korea, and Japan, we would like to show results for non-seasalt SO₄²⁻.



Supplemental figure: Scatter-plot of annual mean (from 2008 to 2010) SO₄²⁻ and nss-SO₄²⁻ concentration in precipitation at ten sites around Beijing in China.

4) Page 4, Line 24: Is the monthly mean concentration volume-weighted?

Yes, we have explicitly indicated that volume-weighted monthly means were used in the revised manuscript on P4, L29-30.

5) Page 5, Line 4: I can not see the introduction on the method for trend analysis.

Linear trends were determined based on a linear regression analysis. We have described these methods as follows on P8, L24-25:

“Linear trends during each of the three phases were analyzed using linear regression, and significance levels were calculated using the Student’s *t*-test.”

6) Page 5, Line 32: Na⁺ can not be used as sea-salt tracer, at least in China.

In this study, to facilitate comparisons among values for China, Korea, and Japan, we would like to use the same data types and thereby would like to present non-seasalt SO₄²⁻. Please also refer the reply 3).

7) Page 11, Line 7: Double 'Xiang Zhou'.

This was an error and should read "Zhuxiandong."

In the revised manuscript, the correct site classification has been included in Table 1.

8) Page 26, Line 4: Better use equivalence as the unit for the sum.

We have revised the calculation of the sum as suggested. Figure 7 and the relevant explanation in the main manuscript have been revised.

9) Page 28 and 29, Table 2 and 3: How to calculate the trend?

The trend was estimated using linear regression. We have clarified this point in the revised manuscript. Please refer the reply 5).