

Dear reviewer:

Thank you very much for your constructive comments. We have addressed them one by one below and incorporated your suggestions in our manuscript. Hope you find our revisions useful. Thank you again.

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Regards,

Steve

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This paper presents a study of local versus transported pollution in South Korea and Japan with emphasis on the impact of pollutant deposition to the ecosystem. This is an interesting perspective on a topic that has been studied extensively, but I feel there are significant changes required to this paper before it could be published.

This work uses the regional air quality model CMAQ to perform numerous model experiments, turning off emissions from various regions to quantify the impact of different source regions to aerosol distributions and deposition over Korea and Japan. The model configuration and design of the sensitivity experiments seems sound.

However, I feel far more model evaluation should be performed (and illustrated) before using the model to attribute source contributions. A more complete description of how the model bias statistics (e.g., Table 2) were determined is needed. For example, how was the ratio determined - is it the mean over the model divided by observation at each time of observations, or just the model mean divided by the observation mean? What is Index of Agreement - correlation coefficient? Also, it would be valuable to see time series of the model-observation comparisons: are there larger model differences in some seasons than others? As you show later, there is significant difference among seasons in the transport from China to Korea and Japan.

Respond: We noticed that the essential descriptions of those indicators are missing. The equations of the indicators are added to the manuscript (see L186-L199).

The indicators are calculated as follows.

$$r = \frac{\sum_{i=1}^n (M_i - \bar{M}) \times (O_i - \bar{O})}{[\sum_{i=1}^n (M_i - \bar{M})^2 \times (O_i - \bar{O})^2]^{\frac{1}{2}}},$$

$$\text{NMB} = \frac{\sum_{i=1}^n (M_i - O_i)}{\sum_{i=1}^n O_i} \times 100\%,$$

$$\text{RMSE} = \left[\frac{1}{n} \sum_{i=1}^n (M_i - O_i)^2 \right]^{\frac{1}{2}}, \text{ and}$$

$$\text{IoA} = 1 - \frac{\sum_{i=1}^n (M_i - O_i)^2}{\sum_{i=1}^n (|M_i - \bar{O}| + |O_i - \bar{O}|)^2},$$

where M is model predictions; \bar{M} is model output mean; O is observation measurements; and \bar{O} is observation mean.

Another issue with the model evaluation is that the satellite-derived PM_{2.5} is for 2014, while you model simulation is for 2010. Is the satellite product not available for 2010? If not, you need to explain how much error is introduced in not matching the years. At L.218 you discuss the discrepancy between model grid size and the observations, but I thought you were talking about comparison to the satellite product here, and you should be able to average the satellite grid to the model grid (or vice versa, if the model grid is smaller), so that you are comparing the same area. In Figure 2, what do each of the points represent (daily or hourly, each model grid)?

Respond: The satellite-derived PM_{2.5} was processed for 2010. We would like to clarify that it was just a typo error.

As presented in L177, each point represents annual averaged PM_{2.5} at each model grid.

It would be valuable to evaluate the model results to observed deposition rates. Aren't there some measurements available in Korea and Japan for this evaluation?

Respond: We extracted monthly wet deposition of SO₄²⁻ and NO₃⁻ across Japan and Korea from EANET datasets and compared with our model outputs. The evaluation results and corresponding discussion are added to as Table 4 and L238-L249.

It is not clear what is being shown in Figure 3 and discussed in Section 3.2 and onward. I guess this is only model results. Since there were significant biases in the comparison to observations, how well can we trust the source contributions based purely on model results that are presented.

Respond: Discussion about Figure 3 in Section 3.2 is based on the model outputs. We agree that dynamic modeling method may introduce biases when simulating air pollution concentrations, which is also the fact for other methods, such as satellite retrieving or statistical modeling. The model performance in this study is evaluated, and the results show that our model performance is comparable to that reported in other studies as we discussed in the Section 3.2. On the basis of currently available knowledge, we think our source contribution results are valid.

In section 3.4 (l.291), you write "implying that ... emissions ... remain relatively constant all year long." This conclusion is determined by the emissions inventory that you use to drive the model, but the way the sentence is written it suggests it is a finding from your analysis based on observations, but my impression is that you are just presenting model results here.

Respond: We agree for this sentence may leave an impression of findings from observations, rather than model outputs. The corresponding description was modified as follows:

“As well, there was little seasonal variance in terms of its contribution to Japan’s and South Korea’s PM_{2.5} concentration levels, which may be because industrial emissions from China remain relatively constant all year long.”

There are a number of typos or grammatical errors, for example:

l.25: perhaps you mean to say 'one of the most polluted regions of the world.'

Respond: Thanks for your suggestion. We changed the sentence accordingly.

l.110: 'with describe' needs to be rewritten.

Respond: We noticed that this is a grammar error, and thus modified the original sentence to “The method details of the source apportionment analysis are provided in Section (2).”.

l.149: 'Other two' should be 'Two other'.

Respond: Thanks for your suggestion. We changed the sentence accordingly.

l.367: use 'prevalent' instead of 'popular'.

Respond: Thanks for your suggestion. We changed the sentence accordingly.

l.393: 'enhance increase' (remove one word).

Respond: Thanks for your suggestion. We changed the sentence accordingly.