Dear reviewer 1:

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.

--

Regards,

Steve

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General comments: Lots of previous studies have indicated that The transboundary air pollution (TAP) constitutes one of the major contributor to the aerosol loading in Korea and Japan. However, it remains elusive to separate out the contribution from local emission and TAP. This study examined the spatiotemporal variations of TAP and sectoral contributions from China emissions and identified the contributions of TAP to acid depositions. The TAP's impact on acid deposition was found to be larger than TAP's impact on PM2.5 concentration. These findings have implications for the decision-making policy for emission control in the upwind regions. Overall, this manuscript is well written, and the analysis methods are scientifically sound. Apparently, this study could be a significant addition to the community of transboundary transport of air pollution, provided the following concerns have been fully considered. Therefore, I recommend its acceptance for publications in ACP pending minor revision.

Specific comments:

1. L44: Too many citation followed by "impacts on people's health, the environment, and economic costs: : :". I strongly suggested to cite these references separately. In addition, air pollution also exert influence on clouds and precipitation (Li et al., 2011, doi: 10.1038/ngeo1313; Koren et al., 2012, doi: 10.1038/ngeo1364; Guo et al., 2016, doi: 10.1002/2015JD023257)

Response: Thanks for your suggestion. We separate the citations to different aspects, including public health, environment, climate, and economic cost.

2. L60-61: Grammar error in "much of it transboundary in nature. "

Response: Thanks for your comments. We change this sentence to "The East Asian region has been suffering from air pollution for decades, especially transboundary air pollution.".

3. L110: Grammar error in "describe in the next section (2) details

Response: Thanks for your comments. We change this sentence to "The method details of the source apportionment analysis are provided in Section (2).".

4. In Fig.1, black cross representing the major cities is the same as the color of country boundary. This should be avoid.

Response: As suggested, we modified the color of major cities to be red, which is different with black country boundary.

5. L148: ", see Table 1"-> " (see Table 1)"

Response: Modified as suggested.

6. The titles of X-axis and Y-axis in in Fig.2 are suggested to indicate the PM2.5.

Response: We changed the titles of X- and Y- axis based on your suggestion.

7. L254: "accounted for in" -> "accounted for by"

Response: We changed the term from "accounted for in" to "contributed by".

8. L288: "Shown in Table 5"->"As shown in Table 5"

Response: We changed the term from "Shown in Table 5" to "As shown in Table 5".

9. Table 6 caption: "kg" is a typo? Is it supposed to be "tonne" or "Tg"?

Response: Kg is a typo error and thus changed to Tg. Thanks.

10. L387: grammar error in "..enhance increase soil N availability" .

Response: Thanks for your comments. We deleted "enhance" to avoid the duplication.

11. The fonts in Fig.4 are too small to be read easily.

Response: We increase the font size of Fig.4

12. Section 4: This study revealed a significant contribution (more than 50%) of TAP from Asia on surface PM2.5 in Japan and South Korea using one-year model simulation alone. Given that a large amount of previous observational studies have been involved in the TAP, especially trans-Pacific transport of aerosols, at the very

least, the authors are suggested to discuss more on the previous results from long-term observations, e.g., what is the difference of magnitude of the ratio of TAP to total pollution, what is the role that multi-scale circulation plays in the TAP, among others. As such, the readers can get a full picture on this topic.

Response: We cite an exhaustive list of research on TAP in this region throughout the manuscript, including those that have used back-trajectory analyses (Lee et al., 2013) (Lee et al., 2011), atmospheric models (Kim et al., 2017) (Koo et al., 2008), and/or measurements with positive matrix factorization and potential source contribution functions (Heo et al., 2009) in attempts to conduct source apportionment. These studies of both short-term episodic events and long-term average concentrations generally point to similar results, e.g. between 60% and 80% of local PM in South Korea is attributable to transboundary sources.

13. The journal name is missed in the reference of Gu et al., 2016b.

Response: We noticed that reference Gu et al, 2016b duplicated with Gu et al. 2016. Gu et al. 2016b is therefore removed.

References

Heo, J.-B., Hopke, P.K., Yi, S.-M., 2009. Source apportionment of PM2.5 in Seoul, Korea. Atmospheric Chemistry and Physics 9, 4957–4971.

Kim, H.C., Kim, E., Bae, C., Cho, J.H., Kim, B.-U., Kim, S., 2017. Regional contributions to particulate matter concentration in the Seoul metropolitan area, South Korea: seasonal variation and sensitivity to meterology and emissions inventory. Atmospheric Chemistry and Physics 17, 10315–10332.

Koo, Y.-S., Kim, S.-T., Yun, H.-Y., Han, J.-S., Lee, J.-Y., Kim, K.-H., Jeon, E.-C., 2008. The simulation of aerosol transport over East Asia region. Atmospheric Research 90, 264–271.

Lee, S., Ho, C.-H., Choi, Y.-S., 2011. High-PM10 concentration episodes in Seoul, Korea: Background sources and related meteorological conditions. Atmospheric Environment 45, 7240–7247. https://doi.org/10.1016/j.atmosenv.2011.08.071

Lee, S., Ho, C.-H., Lee, Y.G., Choi, H.-J., Song, C.-K., 2013. Influence of transboundary air pollutants from China on the high-PM10 episode in Seoul, Korea for the period October 16-20, 2008. Atmospheric Environment 77, 430–439.

Dear reviewer 2:

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.

--
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.

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

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

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

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

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 SO_4^{2-} and NO_3^- 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.

Dear scholar:

Thank you very much for your constructive comments. We have addressed them one by one below. Thank you again.

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Regards,

Steve

____

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Dear authors,
I have some short questions here:

1. **When you refer to the emission inventory, you cited Gu and Yim et al. (2016). There are two pieces of literature in the reference: Gu Y. and Yim, S. H. L.: The air quality and health impacts of domestic trans-boundary pollution in various regions of China, Environ. Int., 97, 117–124, doi:10.1016/j.envint.2016.08.004, 2016a. Gu, Y. and Yim, S. H. L.: The air quality and health impacts of domestic trans-boundary pollution in various regions of China, , 97, 117–124 doi:10.1016/j.envint.2016.08.004, 2016b. Actually, the inventory and its limitation were very briefly described in Gu and Yim et al. (2016), and the reviewer's comments were not shown. So would you please explain in detail the inventory and its limitation? I believe this is also very important and fundamental for the manuscript. And what about the natural dust part of the PM2.5 if is it not included in the inventory at all?**

   Respond: In Gu and Yim et al. (2016), the detailed emission inventory, including the source information, vertical and temporal allocation, and chemical speciation were discussed in the section 3 of supporting information (SI). Assumptions involved in the emission making were also described in the SI. For natural dust, we adapted a physical-based model to simulate mineral dust emissions, in consideration of land cover, wind speed, soil information, air density, etc.

2. **Can you get the components of the PM2.5? I think there are more elements other than SO4 2- /NO3- in the PM2.5 and the components of the PM2.5 may partly help to explain the seasonal variations.**

   Respond: Thanks for this question. We did consider other PM2.5 components when analyzing the seasonal variations such as ammonium, black carbon and organic carbon. Since we aim to assess acid deposition, SO_4^{2-} and NO_3^- are focused.

3. **Would you please also explain in greater detail the chemistry process regarding the PM2.5 and its wet deposition which was claimed to be the significant part of the deposition?**

   Respond: For the formation of $PM_{2.5}$, our air quality model involved a number of chemistry processes. In addition to primary species, $PM_{2.5}$ could be formed from inorganic precursors, e.g. SO_2, NO_x, NH_3, through gas-phase oxidation and aqueous-phase chemistry, and can be ultimately coagulated and deposited to secondary particulates. Also, some organic precursors, including VOCs and HCs, could be oxidized by O_3 and OH to form secondary organic aerosols, which can become a major component of $PM_{2.5}$. Refer to Binkowski, (1999) for details.

   Reference:
   Binkowski, F. S.: AEROSOLS IN MODELS-3 CMAQ, [online] Available from: https://www.cmascenter.org/cmaq/science_documentation/pdf/ch10.pdf, 1999.

4. **Have you ever traced the PM2.5 backward to see the trajectories and the origin from a Lagrangian perspective? If you did so, do the results agree with your current conclusions?**

   Respond: Thanks for your suggestion. It is true that the method of backward trajectories may to some extent explain the origin. However, the secondary particulates such as SO_4^{2-} and NO_3^- can form through chemical reactions that the backward trajectories can reflect these processes, especially heterogeneous reactions are critical for $PM_{2.5}$ formation and acid depositions in East Asia. We have cited several back trajectory studies of TAP in this region, and although those studies are typically limited to short-term episodic events, indeed our results align reasonably well (Lee et al., 2013) (Lee et al., 2011).

References

Lee, S., Ho, C.-H., Choi, Y.-S., 2011. High-PM10 concentration episodes in Seoul, Korea: Background sources and related meteorological conditions. Atmospheric Environment 45, 7240–7247. https://doi.org/10.1016/j.atmosenv.2011.08.071

Lee, S., Ho, C.-H., Lee, Y.G., Choi, H.-J., Song, C.-K., 2013. Influence of transboundary air pollutants from China on the high-PM10 episode in Seoul, Korea for the period October 16-20, 2008. Atmospheric Environment 77, 430–439.

Air quality and acid deposition impacts of local emissions and transboundary air pollution in Japan and South Korea

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Abstract

~~Recent~~ Numerous studies have reported that ambient air pollution, which has both local and long-range sources, causes adverse impacts on the environment and human health. Previous studies have ~~intensively~~ investigated the impacts of transboundary air pollution (TAP) ~~impact~~ in East Asia, albeit primarily through ~~in~~analyses of ~~episode~~ episodic events. In addition, ~~From the environmental perspectives, i~~it is ~~necessary~~ useful to better understand the spatiotemporal variations in TAP and the resultant impact on the environment and human health. This study is aimed at assessing and quantifying the air quality impacts in Japan and South Korea due to their local emissions and TAP from sources in East Asia – one of the most polluted regions in the world. We have applied state-of-the-science atmospheric models to simulate air quality in East Asia, and then ~~analyzing~~ analyzed the air quality and acid deposition impacts of both local emissions and TAP sources in Japan and South Korea. Our results show that ~30% of the annual average ambient $PM_{2.5}$ concentrations in Japan and South Korea in 2010 ~~was on average~~was contributed by local emissions ~~in Japan and South Korea~~within each country, while the remaining ~70% was contributed by TAP from other countries in the region. More detailed analyses also revealed that the local contribution was higher in the metropolises of Japan (~40-79%) and South Korea (~31-55%), and that minimal seasonal variations in surface $PM_{2.5}$ in Japan, whereas there was a relatively large variation in South Korea in the winter. Further, among all five studied anthropogenic emission sectors of China, the industrial sector represented the greatest contributor to annual surface $PM_{2.5}$ concentrations in Japan and South Korea, followed by the residential and power generation sectors. ~~In terms of acid deposition, our r~~Results also show that TAP's impact on acid deposition (SO_4^{2-} and NO_3^-) was larger than TAP's impact on $PM_{2.5}$ concentrations~~,~~ (accounting for over 80% of total deposition), and that seasonal variations in acid deposition were similar for both Japan and South Korea (i.e. ~~:~~higher in both the winter and summer). Finally, wet deposition had a greater impact on mixed forests in Japan and savannas in South Korea. Given these significant impacts of TAP in the region, it is paramount that cross-national efforts be taken to mitigate air pollution problems in across East Asia.

1. Introduction

Air pollution is one of the major environmental problems facing the modern world, ~~with~~ leading to adverse impacts on ~~people's~~ human health (Bishop et al., 2018; Brook et al., 2004; Brunekreef and Holgate, 2002; Cook et al., 2005; Dockery et al., 1993; Lelieveld et al., 2015; Nel, 2005; Pope III and Dockery, 2006; Samet et al., 2000), the environment (Gu et al., 2018; Lee et al., 2005; Rodhe et al., 2002), climate (Guo et al., 2016; Koren et al., 2012; Li et al., 2011; Liu et al., 2018) and economic costs (Lee et al., 2011b; Organisation for Economic Co-operation and Development, 2008; Pearce et al., 2006; Yin et al., 2017)~~(Bishop et al., 2018; Brook et al., 2004; Brunekreef and Holgate, 2002; Cook et al., 2005; Dockery et al., 1993; Gu et al., 2018; Lee et al., 2011b; Lelieveld et al., 2015; Nel, 2005; Organisation for Economic Co-operation and Development, 2008; Pearce et al., 2006; Pope III and Dockery, 2006; Rodhe et al., 2002; Samet et al., 2000; Yin et al., 2017)~~. This study focuses specifically on the phenomenon of transboundary air pollution (TAP), which creates problems of assigning attribution and thwarts the implementation of effective policies. There is a sense of urgency, though, given the significant implications of TAP on the environment and human health and the geographic breadth of the areas affected. Zhang

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et al. (2017) investigated the health impacts due to global transboundary air pollution and international trade, estimating that ~411 thousand deaths worldwide have resulted from TAP, while 762 thousand deaths have resulted from international trade-associated emissions. Lin et al. (2014) investigated the air pollution in the United States due to the emissions of its international trade in China, estimating air pollution of China contributed 3-10% and 0.5-1.5% to~~, respectively,~~ annual surface sulfate and ozone concentrations, respectively, in the western United States.

The East Asian region has been suffering from ~~the effects of~~ air pollution for decades, especially ~~much of it~~ transboundary ~~in nature~~air pollution. The extant literature reports significant impacts of TAP in Japan (Aikawa et al., 2010; Kaneyasu et al., 2014; Kashima et al., 2012; Murano et al., 2000), South Korea (Han et al., 2008; Heo et al., 2009; Kim et al., 2017a, 2017b, 2012, 2009; Koo et al., 2012; Lee et al., 2011a, 2013; Oh et al., 2015; Vellingiri et al., 2016), or East Asia in general and beyond (Gao et al., 2011; Gu and Yim, 2016; Hou et al., 2018; Koo et al., 2008; Lai et al., 2016; Lin et al., 2014a; Luo et al., 2018; Nawahda et al., 2012; Park et al., 2016; Wang et al., 2019; Zhang et al., 2017)~~(Gao et al., 2011; Gu and Yim, 2016a; Hou et al., 2018; Koo et al., 2008; Lai et al., 2016; Lin et al., 2014a; Luo et al., 2018; Nawahda et al., 2012; Park et al., 2016; Wang et al., 2019; Zhang et al., 2017)~~, emphasizing TAP's origins in China. For example, Aikawa et al. (2010) assessed transboundary sulfate (SO_4^{2-}) concentrations at various measurement sites across the East Asian Pacific Rim, reporting that China contributed 50%-70% of total annual SO_4^{2-} in Japan with a maximum in the winter of 65-80%. Murano et al. (2000) examined the transboundary air pollution over two Japanese islands, Oki Island and Okinawa Island, reporting that the high non-sea-salt sulfate concentrations observed in Oki in certain episodic events were associated with the air mass transported from China and Korea under favorable weather conditions. Focusing on an upwind area of Japan, Fukuoka, Kaneyasu et al. (2014) investigated the impact of transboundary particulate matter with an aerodynamic diameter < 2.5μm ($PM_{2.5}$), concluding that, in northern Kyushu, contributions were greater than those of local air pollution. In terms of China-borne TAP in Korea, Lee et al. (2013 & 2011) traced contributors to Seoul's episodic high PM_{10} and $PM_{2.5}$ ~~to Seoul's episodic~~ events, showing that a stagnant high-pressure system over the city led to the updraft, transport, and subsequent descent of PM_{10} and $PM_{2.5}$ from China to Seoul~~, resulting in high concentrations of both PM_{10} and $PM_{2.5}$~~. While TAP from China in Japan and South Korea was identified, the spatiotemporal variations of TAP and sectoral contributions from emission from China ~~emissions~~ have yet to be fully understood.

Wet acid deposition due to air pollution is also critically important given the risks to ecosystems. Adverse environmental impacts of wet deposition have been reported in Asia (Bhatti et al., 1992), and specific research ~~have~~ has investigated TAP's impact on wet deposition in East Asia (Arndt et al., 1998; Ichikawa et al., 1998; Ichikawa and Fujita, 1995; Lin et al., 2008). Within the East Asian region, Japan and South Korea are particularly vulnerable to acid rain (Bhatti et al., 1992; Oh et al., 2015)~~;~~. Arndt et al. (1998) reported that the contribution of China to sulfur deposition in Japan was 2.5 times higher in winter and spring than in summer and autumn, and that both China and South Korea have been primary contributors to the sulfur deposition in southern and western Japan. Ichikawa et al. (1998) found that TAP accounted for more than 50% of wet sulfur deposition in Japan. In

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their investigation of the contribution of energy consumption emissions to wet sulfur deposition in Northeast Asia, Streets et al. (1999) identified the impact of nitrogen oxides emissions on the region's acid deposition. Lin et al. (2008) reported that anthropogenic emissions of Japan and the Korean Peninsula had a larger contribution to wet nitrogen deposition than to wet sulfur deposition in Japan due to the substantial transportation sources of the two countries. This finding highlights the importance of assessing the contribution of various sectors to acid deposition due to their distinct emission profiles.

To mitigate air pollution in ~~the~~ East Asia~~region~~, it is critical to conduct a more comprehensive evaluation of the contributions of both local emissions ~~as well as~~and transboundary air pollution sources. Thus, this study assesses the spatiotemporal variations in the contributions of local emissions ~~—in Japan and South Korea, specifically—~~ and transboundary air pollution ~~—~~(from China) ~~—~~to air quality and thus wet deposition in Japan and South Korea. To identify which sectors are the largest ~~most significantly~~ contribut~~ing~~ors to ~~transboundary air pollution~~TAP and acid deposition in Japan and South Korea, we conduct a source apportionment analysis of China's sector-specific emissions. The method details of the source apportionment analysis are provided in ~~To these ends, with describe in the next s~~Section ~~(~~2~~)~~ ~~details regarding our method~~. Section 3 is divided into two parts: the first part presents model evaluation results and estimates of ambient $PM_{2.5}$ ~~level~~ concentrations and source ~~appointment of~~ ~~$PM_{2.5}$~~apportionment, while the second part discusses wet deposition results and its impact on various land covers in Japan and South Korea. A discussion in ~~section~~ Section ~~(~~4~~)~~ concludes this study.

2. Materials and Methods

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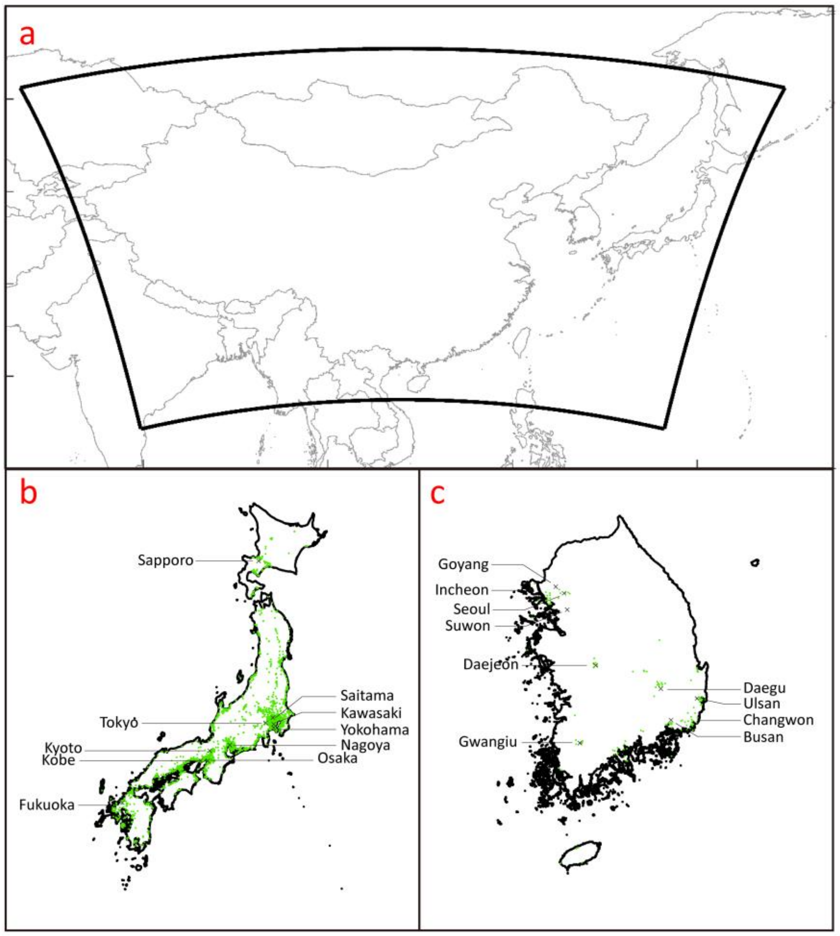
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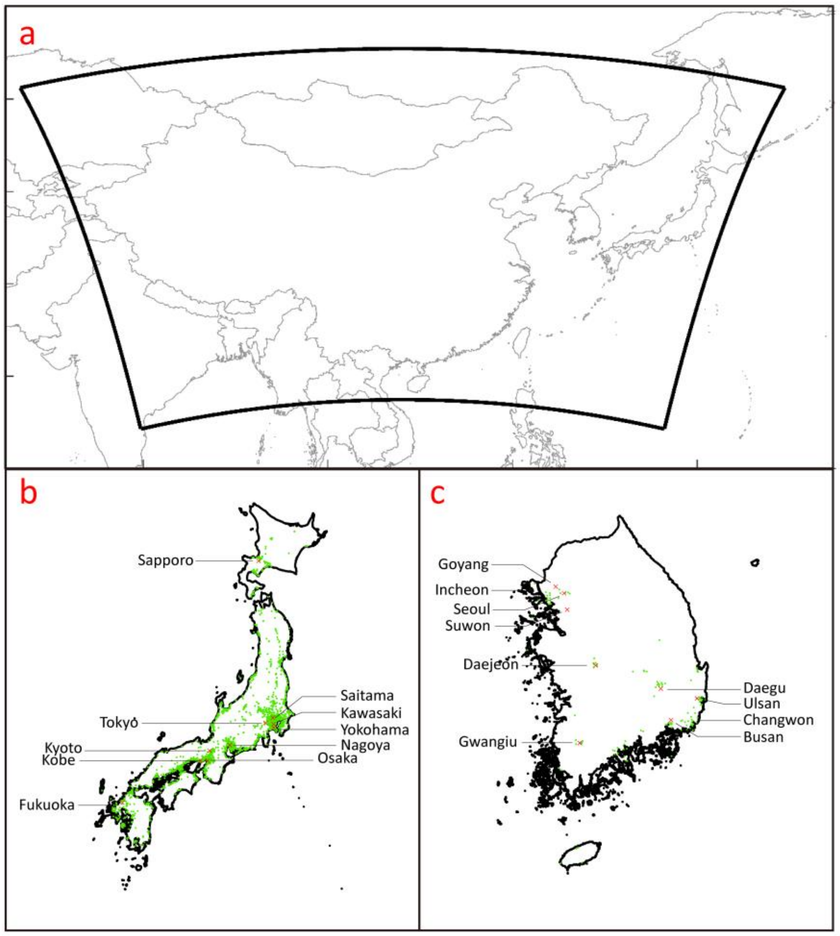

Figure 1. (a) Model simulation domain (solid black line). Monitoring stations (green dot) and major cities (black cross) with population ≥ 1 million in (b) Japan and (c) South Korea.

This study applied the state-of-the-science atmospheric models [Weather Research and Forecasting Model (WRF)/The Community Multiscale Air Quality modeling System (CMAQ)] to simulate hourly air quality over Japan and South Korea in year 2010. The WRF model (Skamarock et al., 2008) was applied to simulate meteorology over the study area with one domain at a spatial resolution of 27 km and 26 vertical layers. Figure 1~~Figure 1~~a depicts the model domain. The six-hour and 1º × 1º Final Operational Global Analysis (FNL) data (National Centers for Environmental Prediction et al., 2000) was applied to drive the WRF model, and the land-use data was updated based on Data Center for Resources and Environmental Sciences, Chinese Academy of Sciences (RESDC) (Liu et al., 2014).

We applied CMAQv4.7.1 (Byun and Schere, 2006) to simulate ~~the~~ air quality over East Asia. The boundary conditions were provided by the global chemical transport model (GEOS-Chem) (Bey et al., 2001), while the updated Carbon Bound mechanism (CB05) was used for chemical speciation and reaction regulation. The hourly emissions were compiled based on multiple datasets: the HTAP-V2 dataset (Janssens-Maenhout et al., 2012) was applied for anthropogenic emissions; the FINN 1.5 dataset (Wiedinmyer et al., 2014) was utilized for fire emissions; and the MEGAN-MACC database (Sindelarova et al., 2014) was applied for biogenic emissions. The speciation scheme, temporal profiles, and vertical profiles adopted in our emission inventory were based on Gu and Yim (2016), while plume rise heights for large industry sectors and power plants were based on Briggs (1972). Details of the atmospheric models were further discussed in Gu and Yim (2016).

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Table 1. List of model simulations.

Simulation number	Scenario
1	Baseline
2	Baseline without Japan’s emissions
3	Baseline without South Korea’s emissions
4	Baseline without Japan’s and China’s emissions (to estimate the contribution of others in South Korea)
5	Baseline without South Korea’s and China’s emissions (to estimate the contribution of others in Japan)
6	Baseline without China’s agricultural emissions (AGR)
7	Baseline without China’s industrial emissions (IND)
8	Baseline without China’s power generation emissions (PG)
9	Baseline without China’s residential and commercial emissions (RAC)
10	Baseline without China’s ground transportation emissions (TRA)
11	Only include China’s, Japan’s and South Korea’s emissions (to compare with the baseline to assess the impact of emissions from other countries)

To investigate the contributions of local emissions and transboundary air pollution to air quality and acid deposition over Japan and South Korea, and in particular, ~~to~~ those originating from China sectoral emissions, a total of ten one-year simulations were conducted~~,~~ (see Table 1~~Table 1~~). The first simulation was a baseline case, in which all the emissions were included. Two ~~O~~other ~~two~~ simulations were performed in which emissions of Japan and South Korea were removed in-turn. Ano~~O~~ther five simulations were designed to apportion the contribution of various emission sectors of China. S~~ame as~~imilar to Gu et al. (2018), the sectors were defined as (AGR) agriculture, (IND) industry, (PG) power generation, (RAC) residential and commercial, and (TRA) ground transportation. Emissions of each China sector were removed in-turn. The difference of model results between the baseline scenario and another scenarios was used to attribute~~d~~ ~~to~~ the contribution of ~~the~~ emissions ~~of~~ from the respective country or Chinese sector. One

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additional simulation was performed in which only emissions of China, Japan, and South Korea were included. The differences between the baseline scenario and the last scenario was used to attribute~~d~~ ~~to~~ the contribution of emissions from all other ~~the~~ countries in the domain except China, Japan, and South Korea.

To examine the model capacity ~~in~~ for ~~performing~~ estimating ~~spatiotemporal~~ spatiotemporally-varied distribution of $PM_{2.5}$ in South Korea and Japan, we first employed ground-level respirable suspended particulates (PM_{10}) observation datasets in 2010 from Japan and South Korea to compare with respirable suspended particulates output gathered from our air quality model. Hourly measurements from 1678 valid observation stations in Japan were collected by the National Institute for Environmental Studies in Japan (http://www.nies.go.jp/igreen/); monthly measurements from 121 valid observation stations in South Korea were extracted from an annual report of air quality in Korea 2010 (National Institute of Environmental Research, 2011). The locations of monitoring are depicted by the green dots in Figure 1. Each measurement was compared with model outputs at the particular grid where the corresponding observation station are located. To further evaluate the CMAQ performance, we also compared our model results to satellite-retrieved ground-level $PM_{2.5}$ concentration data, which were fused from MODIS, MISR and SeaWiFS AOD observations in 2014 (van Donkelaar et al., 2016). We extracted concentration values of satellite-retrieved $PM_{2.5}$ at the center of each model grid within Japan and Korea, and then conducted grid-to-grid comparisons with annual-averaged model outputs. Model performance was specified by a series of widely used statistical indicators, including ratio (r), normalized mean bias (NMB), root mean square error (RMSE), and index of agreement (IoA). The indicators are calculated as follows.

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

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

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

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

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

To facilitate the discussion of ~~evaluation~~ model performance, evaluation results for different stations were gathered and averaged by the basic district division in different countries (i.e. prefectures in Japan, provinces in South Korea).

3. Results

3.1. Model evaluation

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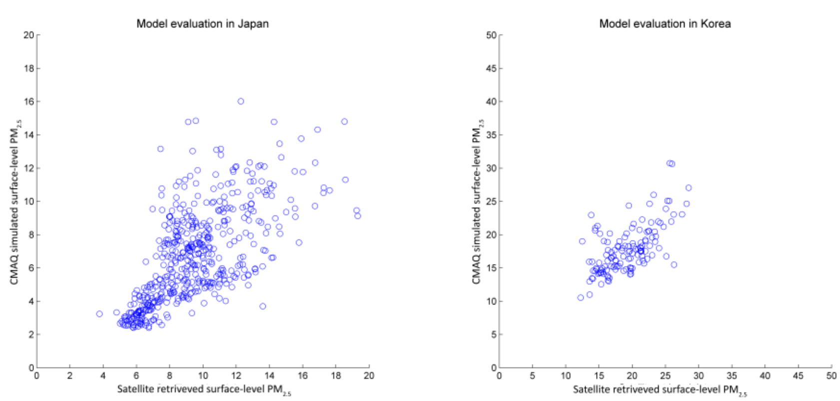
Table 2. Model evaluations of PM_{10} across Japanese prefectures and South Korean provinces where measurements are available. NMB refers to normalized mean bias; RMSE refers to root mean square error; and IoA refers to index of agreement. We note that the evaluation of Japan was based on hourly data, while that of South Korea was based on monthly data.

Prefectures (Japan)	***Ratio***	***NMB (%)***	***RMSE ($\mu g/m^3$)***	***IoA***
Aichi	1.71	0.69	19.88	0.59
Akita	1.09	-29.37	16.50	0.54
Aomori	1.13	-30.00	16.96	0.55
Chiba	1.43	-18.96	19.68	0.55
Ehime	1.33	-33.17	23.30	0.50
Fukui	1.35	-20.56	18.06	0.56
Fukuoka	1.26	-20.55	23.42	0.57
Fukushima	1.30	-21.19	16.12	0.59
Gifu	1.60	-7.64	16.54	0.60
Gunma	1.12	-33.45	19.39	0.56
Hiroshima	1.11	-21.81	20.59	0.59
Hokkaido	1.25	-23.94	14.63	0.54
Hyogo	1.37	-12.74	19.68	0.59
Ibaraki	1.16	-20.19	18.45	0.61
Ishikawa	1.20	-27.72	17.68	0.57
Iwate	1.04	-31.92	14.98	0.58
Kagawa	1.57	-18.78	22.88	0.55
Kagoshima	0.90	-42.71	22.05	0.52
Kanagawa	1.07	-20.32	19.08	0.55
Kochi	1.68	-15.86	17.54	0.52
Kumamoto	1.43	-27.99	21.08	0.55
Kyoto	1.50	-3.41	18.54	0.59
Mie	1.29	-15.02	17.82	0.59
Miyazaki	0.95	-41.11	24.90	0.46
Nagano	0.90	-41.86	15.24	0.58
Nagasaki	1.01	-31.19	23.23	0.54
Nara	1.56	-4.58	19.18	0.58
Niigata	1.07	-32.36	17.47	0.56
Oita	1.58	-16.94	19.68	0.54
Okayama	1.42	-7.04	22.06	0.58
Okinawa	1.10	-44.79	18.22	0.53
Osaka	1.28	-18.74	19.95	0.58
Saga	1.40	-8.41	18.63	0.61
Saitama	1.21	-27.16	19.72	0.57
Shiga	1.32	-5.93	18.26	0.60
Shimane	1.19	-18.81	23.32	0.53
Shizuoka	1.53	-20.73	17.43	0.55
Tochigi	0.97	-29.50	17.34	0.60
Tokushima	1.26	-21.04	17.31	0.57
Tokyo	1.18	-19.13	18.74	0.56
Tottori	1.52	-16.69	19.98	0.55
Toyama	1.20	-29.08	16.25	0.57
Wakayama	1.31	-24.63	18.02	0.56
Yamagata	0.94	-30.35	15.62	0.59
Yamaguchi	1.68	-3.96	20.39	0.58
Yamanashi	1.07	-41.42	17.05	0.52
Average	1.27	-22.44	18.98	0.56

Provincial divisions (South Korea)	*Ratio*	*NMB (%)*	*RMSE ($\mu g/m^3$)*	*IoA*
Bukjeju	0.48	-52.11	26.98	0.44
Busan	0.65	-36.60	22.05	0.45
Dae-gu	0.64	-37.28	22.84	0.52
Daejeon	0.72	-30.20	16.68	0.63
Geoje	0.65	-37.87	20.27	0.49
Gwangju	0.70	-32.57	18.44	0.63
Gyeongnam	0.63	-38.02	20.84	0.47
Incheon	0.74	-27.41	18.96	0.63
Jeju	0.49	-53.07	29.20	0.51
Jeonnam	0.84	-22.32	16.34	0.56
Kyungbuk	0.54	-46.42	28.78	0.00
Kyungbuk	0.77	-26.10	17.72	0.59
Seoul	0.86	-17.52	14.48	0.72
Taean	0.55	-45.07	26.84	0.48
Ulsan	0.63	-38.05	21.01	0.46
Average	0.66	-36.04	21.43	0.51

We conducted a model evaluation of PM_{10} to assess our model performance over the prefectures of Japan and over the provincial divisions of South Korea where measurements are available, see Table 1~~Table 1~~. On average, the annual mean ratio (normalized mean bias; root mean square error) for Japan and South Korea was 1.27 (-22.44%; 18.98 $\mu g/m^3$) and 0.66 (-36.04%; 21.43 $\mu g/m^3$), respectively. Their mean index of agreements was 0.51 and 0.56 for South Korea and Japan, respectively.

These results show that the model tends to underestimate PM, which is consistent with the results reported in other studies (Ikeda et al., 2014; Koo et al., 2012). For example, Koo et al. (2012) conducted an evaluation of CMAQ performance on PM_{10} over the Seoul and Incheon metropolises as well as the North and South Gyeonggi provinces, showing results similar to ours.



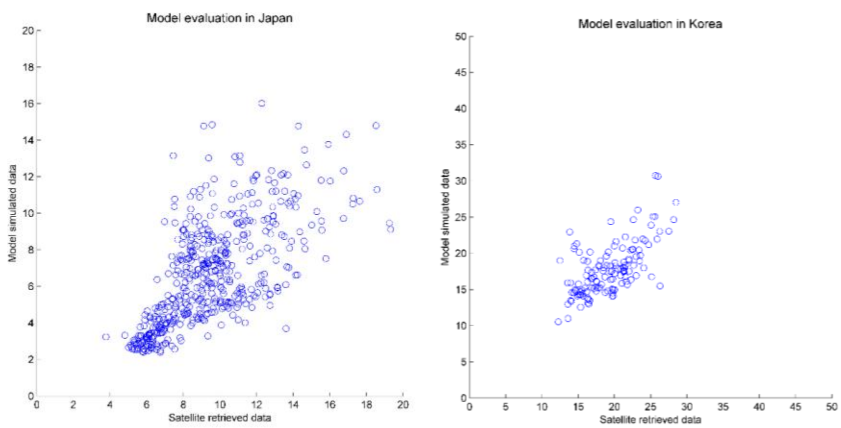


Figure 2. ~~The m~~Model evaluation using satellite-retrieval $PM_{2.5}$ over Japan and South Korea.

Table 3. ~~The s~~Statistical results of model evaluation using satellite-retrieval $PM_{2.5}$ over Japan and South Korea. NMB refers to normalized mean bias; RMSE refers to root mean square error; and IoA refers to index of agreement.

	Ratio	NMB (%)	RMSE ($\mu g/m^3$)	I
Japan	0.7	-29.3	3.6	0.7
South Korea	0.9	-7.3	3.4	0.8

Figure 2~~Figure 2~~ and Table 3~~Table 3~~ show the model evaluation using satellite-retrieval $PM_{2.5}$ over Japan and South Korea. The index of agreement is 0.7 and 0.8 for Japan and South Korea, respectively, while the normalized mean bias is ~-29% and ~-7%. Ikeda et al. (2014) reported that their CMAQ model tended to underestimate $PM_{2.5}$ over Japan with a monthly normalized mean bias of -24.1% to 66.7%. The underestimation may be because the model results were an average value over a model grid, while the measurements represented the local PM level at a specific location. Despite the underestimation, our index of agreement results indicate that the model can reasonably capture the PM variability over the two countries.

Table 4. ~~The m~~Model evaluation of acid deposition in Japan and South Korea. NMB refers to normalized mean bias; RMSE refers to root mean square error; and IoA refers to index of agreement.

Country	Stations	SO_4^{2-}				NO_3^-			
		Ratio	*NMB (%)*	*RMSE (mmol/m2)*	*IoA*	*Ratio*	*NMB (%)*	*RMSE (mmol/m2)*	*IoA*
Japan	Rishiri	1.30	20.35	1.09	0.75	3.42	181.22	3.54	0.17
	Ochiishi	0.68	-72.04	2.89	0.44	0.55	-62.51	0.71	0.58
	Tappi	0.66	-46.55	1.94	0.62	0.95	-16.59	1.24	0.72
	Sado-seki	0.81	-45.90	3.47	0.54	1.41	26.06	2.05	0.49
	Happo	1.49	30.20	1.04	0.61	1.37	21.49	0.67	0.91

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	Ijira	0.60	-44.84	2.01	0.65	0.54	-51.81	3.22	0.57
	Oki	0.54	-63.01	5.21	0.49	1.00	-8.91	1.41	0.85
	Banryu	1.35	-15.06	1.79	0.83	1.34	19.84	2.20	0.90
	Yusuhara	0.83	-31.54	0.99	0.72	1.08	2.52	0.39	0.96
	Hedo	0.29	-79.99	5.48	0.43	0.80	-15.26	0.58	0.91
	Ogasawara	0.13	-93.44	4.92	0.31	0.20	-75.13	0.43	0.56
	Tokyo	1.24	10.29	0.41	0.93	1.10	-28.88	0.94	0.79
	Average	**0.83**	**-35.96**	**2.60**	**0.61**	**1.15**	**-0.66**	**1.45**	**0.70**
South Korea	Rishiri	1.60	-29.51	3.18	0.42	1.85	-16.68	4.06	0.30
	Ochiishi	1.51	-11.54	1.66	0.39	2.31	7.75	2.33	0.38
	Tappi	0.77	-40.55	2.36	0.68	0.68	-51.10	3.43	0.49
	Average	**1.29**	**-27.20**	**2.40**	**0.50**	**1.61**	**-20.01**	**3.27**	**0.39**

SO_4^{2-} and NO_3^- deposition simulated by CMAQ has been compared with monthly ground-level measurements from the Acid Deposition Monitoring Network in East Asia (EANET) (https://monitoring.eanet.asia/document/public/). The evaluation results are shown in Table 4. SO_4^{2-} and NO_3^- tend to underestimate the in Japan and South Korea, which may be associated with simulation bias of $PM_{2.5}$ concentration. Normalized mean biases of SO_4^{2-} and NO_3^- ranged from -93.44% to 30.20% and -75.13% to 181.22% in Japan, respectively, while ranged from -40.55% to -11.54% and -51.10% to 7.75% in Korea. Averaged index of agreement and ratio of SO_4^{2-} and NO_3 indicates that our model could basically capture the fluctuation and magnitude of acid deposition in Japan and South Korea. Slightly better performance in Japan was observed.

3.2. Annual and seasonal ambient $PM_{2.5}$ in Japan and South Korea

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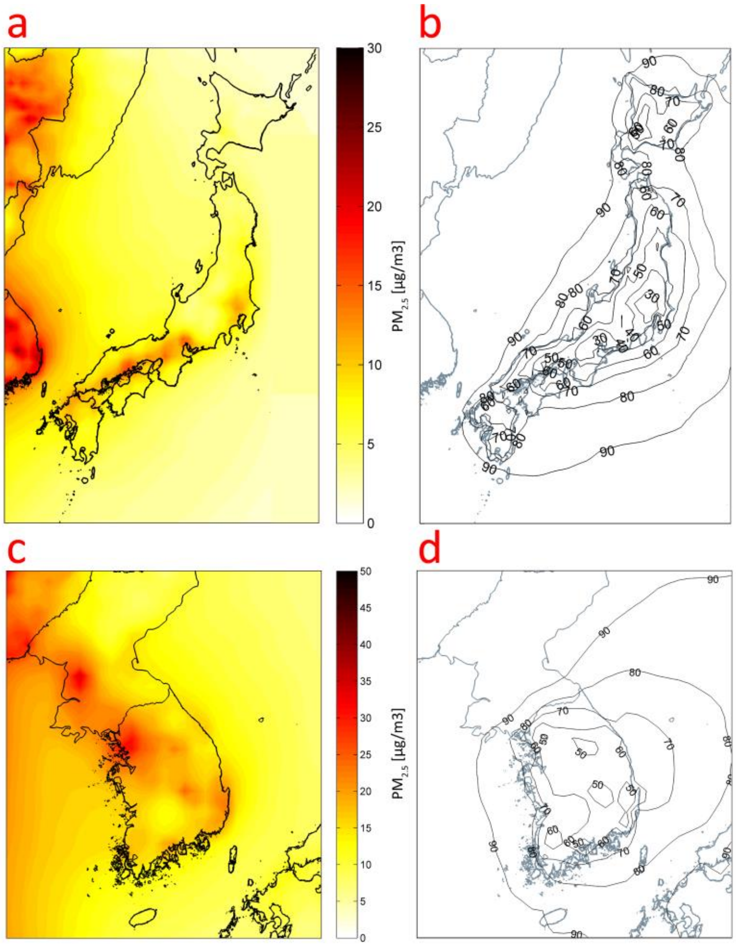


Figure 3. The modeled annual average surface $PM_{2.5}$ ($\mu g/m^3$) over (a) Japan and (c) South Korea in 2010, and the percentage (%) of total $PM_{2.5}$ due to transboundary air pollution over (b) Japan and (d) South Korea.

Figure 3~~Figure 3~~a and 3c show the annual average surface $PM_{2.5}$ over Japan and South Korea. The annual average surface $PM_{2.5}$ concentration over Japan was 5.91 $\mu g/m^3$, while that over South Korea was 16.90 $\mu g/m^3$. Higher $PM_{2.5}$ concentrations occurred in metropolises: in Japan, higher $PM_{2.5}$ levels occurred in Nagoya (13.48 $\mu g/m^3$), Osaka (12.07 $\mu g/m^3$), and Saitama (9.36 $\mu g/m^3$). ~~A h~~Higher $PM_{2.5}$ levels ~~was~~ were also observed at Okayama (14.78 $\mu g/m^3$), even though its population is not as large as the aforementioned metropolises, which may be due to its substantial industrial emissions in the region. In South Korea, higher $PM_{2.5}$ levels occurred in Incheon (23.90 $\mu g/m^3$), Goyang (27.05 $\mu g/m^3$), Seoul (30.64 $\mu g/m^3$) and Suwon (30.75 $\mu g/m^3$). Two additional high ~~concentrations~~

annual average levels of $PM_{2.5}$ can be identified in non-metropolis areas, which may also be due to those areas relatively high industrial emissions.

In Japan, seasonal variations in surface $PM_{2.5}$ did not vary significantly, ranging from 5.75 μg/m^3 to 6.09 μg/m^3. In South Korea, however, seasonal variations ~~was~~ were relatively larger. The winter surface $PM_{2.5}$ level was 18.53 μg/m^3, while the next highest levels occurred in spring (17.61 μg/m^3) and autumn (17.44 μg/m^3). The lowest level of $PM_{2.5}$ occurred in summer (14.02 μg/m^3) in South Korea.

3.3. Local and transboundary contributions

Table 4. Surface $PM_{2.5}$ concentration levels (μg/m^3) and source countries' contributions to $PM_{2.5}$ (%) in Japan and South Korea, annual and seasonal.

		Annual	Spring	Summer	Autumn	Winter
Japan	**surface $PM_{2.5}$ concentration level (μg/m^3)**	5.91	6.09	5.88	5.75	5.93
	local	29.3%	23.4%	29.0%	36.1%	32.2%
	transboundary air pollution (TAP)	70.7%	76.6%	71.0%	63.9%	67.8%
	TAP from South Korea	3.3%	3.7%	2.6%	4.1%	2.1%
	TAP from China	53.9%	61.4%	50.5%	44.0%	55.1%
	TAP from others	13.5%	11.5%	17.9%	15.7%	10.6%
		Annual	**Spring**	**Summer**	**Autumn**	**Winter**
South Korea	**surface $PM_{2.5}$ concentration level (μg/m^3)**	16.90	17.61	14.02	17.44	18.53
	local	29.4%	27.3%	33.8%	33.8%	24.0%
	transboundary air pollution (TAP)	70.6%	72.7%	66.2%	66.2%	76.0%
	TAP from Japan	0.4%	0.4%	1.9%	0.2%	-0.4%
	TAP from China	54.2%	55.5%	43.8%	51.7%	62.9%
	TAP from others	16.0%	16.8%	20.4%	14.3%	13.5%

Table 4~~Table 4~~ shows the contributions of emissions of different source countries to $PM_{2.5}$ in different receptor countries. On average, approximately 29% of annual ambient $PM_{2.5}$ in both Japan and South Korea were ~~accounted for in~~contributed by local emissions, while approximately 71% were identified as TAP. Of TAP's contribution, China was the key contributor, accounting for approximately 54% of annual surface $PM_{2.5}$ in both Japan and South Korea. The results of our analysis of the contributions of $PM_{2.5}$ between Japan and South Korea~~,~~ show that South Korea accounted for 3.3% of the annual surface $PM_{2.5}$ in Japan, whereas Japan's contribution to $PM_{2.5}$ in South Korea was marginal (0.4%). The contribution of other countries was non-negligible (i.e. 13.5% in Japan and 16.0% in South Korea).

Figure 3~~Figure 3~~b and 3d indicate that the local contribution was relatively higher in the metropolises of Japan (40.2 – 78.6%) and South Korea (31.4 – 55.2%), which is due to greater proportions of emissions being generated by local industry, transportation, and power generation. In Japan, the western areas showed a higher TAP contribution than the eastern areas, while, in South Korea, the western and northern areas showed a higher TAP contribution than other areas.

The TAP contribution varied with seasons. In Japan, the highest relative TAP contribution occurred in spring (76.6%) ~~in term of percentage~~, followed by summer (71.0%) and winter (67.8%). The lowest relative contribution ~~percentage~~ occurred in autumn (63.9%). In South Korea, the highest ~~percentage~~ relative contribution of~~due to~~ TAP occurred in winter (76.0%) and spring (72.7%), while the lowest ~~percentage~~ occurred in summer (66.2%) and autumn (66.2%). Seasonal variations in TAP were most likely due to varying emissions and prevailing wind directions across seasons.

3.4. Transboundary air pollution from China sectoral emissions

Table 5. Contribution of Chinese sectoral emissions to surface $PM_{2.5}$ ($\mu g/m^3$) in Japan and South Korea, annual and seasonal. Emission sectors include agriculture (AGR), power generation (PG), ground transportation (TRA), industrial (IND), and residential and commercial (RAC). Agriculture refers to agriculture and agricultural waste burning; power generation refers to electricity generation; ground transportation refers to road transportation, rail, pipelines, and inland waterways; industrial refers to energy production other than electricity generation, industrial processes, solvent production and application; and residential and commercial refers to heating, cooling, equipment, and waste disposal or incineration related to buildings.

(average)		**Annual (5.91)**	**Spring (6.09)**	**Summer (5.88)**	**Autumn (5.75)**	**Winter (5.93)**
Japan	**TRA**	4.0%	4.4%	2.3%	3.1%	6.3%
	AGR	4.2%	2.9%	1.1%	4.9%	8.4%
	PG	10.8%	11.7%	9.7%	9.5%	11.7%
	IND	20.4%	20.8%	21.0%	20.7%	18.9%
	RAC	14.5%	21.7%	16.3%	5.8%	9.7%
(average)		**Annual (16.90)**	**Spring (17.61)**	**Summer (14.02)**	**Autumn (17.44)**	**Winter (18.53)**
South Korea	**TRA**	5.4%	5.2%	2.3%	5.5%	7.9%
	AGR	7.0%	4.2%	1.8%	11.6%	9.5%
	PG	10.9%	10.6%	8.7%	10.1%	13.8%
	IND	20.2%	19.2%	19.5%	20.2%	21.9%
	RAC	10.7%	16.4%	11.6%	4.3%	9.8%

As ~~S~~shown in Table 5, among Chinese sectors, industrial emissions were a key contributor to annual surface $PM_{2.5}$ in both Japan and South Korea, accounting for approximately ~~one~~ one-fifth of annual average concentrations~~concentration levels~~. 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 ~~implying that~~ industrial emissions from China remain relatively constant all year long. For both Japan and South Korea, the second and third-most contributors to annual surface $PM_{2.5}$ were the residential/commercial (RAC) sector and the power generation (PG) sector, respectively. Unlike the industrial sector, seasonal variations in relative contributions for these two sectors were apparent. The southerly wind in Japan and Korea during spring and summer provided favorable conditions for pollutant transport of the Chinese RAC sector. We observed contributions of China's RAC sector to 12-22% of surface $PM_{2.5}$ in Japan and South Korea in spring and summer. In autumn, the relative contribution of the Chinese RAC sector was ~~minimum~~ minimal due to the northerly wind that was not favorable for ~~the~~ TAP from China. In spring and winter, the northwesterly wind was favorable for transporting pollutants from northern China, in which emissions from PG were substantial. The remaining Chinese contribution was from the ground transportation ~~sector~~ and ~~the~~ agriculture sectors. When combined, both sectors accounted for 8% and 12% of annual surface $PM_{2.5}$ in Japan and South Korea, respectively, with a maximum relative contribution in autumn and winter.

3.5. Effects of acid deposition

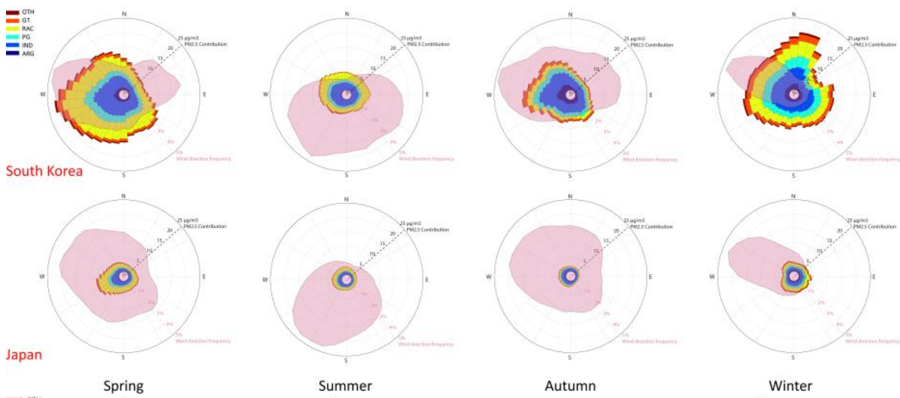
3.5.1 Annual and seasonal variations

Table 6. Acid deposition [sulfate (SO_4^{2-}) and nitrate (NO_3^-)] (T~~k~~g) in Japan and South Korea, annual and seasonal, including SO_4^{2-}/NO_3^- and local/TAP (transboundary air pollution) contribution ratios.

	Annual			Spring			Summer			Autumn			Winter		
	total (Tg)	SO_4^{2-}/NO_3^-	local/TAP	total (Tg)	SO_4^{2-}/NO_3^-	local/TAP	total (Tg)	SO_4^{2-}/NO_3^-	local/TAP	total (Tg)	SO_4^{2-}/NO_3^-	local/TAP	total (Tg)	SO_4^{2-}/NO_3^-	local/TAP
Japan	1.08	1.29	0.18	0.32	1.25	0.19	0.24	1.89	0.22	0.19	1.26	0.24	0.33	1.04	0.11
South Korea	0.37	1.33	0.17	0.09	1.18	0.21	0.13	1.88	0.15	0.06	1.25	0.22	0.09	0.96	0.14

Table 6 ~~Table 6~~ presents the annual and seasonal acid deposition in Japan and South Korea. We estimated that outdoor air pollution ~~respectively~~ resulted in 1.08 ~~tonnes~~ Tg and 0.37 ~~tonnes~~ Tg of acid deposition annually in~~, respectively,~~ Japan and South Korea, respectively. The local/TAP ratio was estimated to be 0.18 and 0.17 for Japan and South Korea, respectively, which ~~are~~ is lower than the respective ratios for $PM_{2.5}$ concentrations, highlighting TAP's larger impact on acid deposition. We note that $PM_{2.5}$ concentrations include both primary and secondary $PM_{2.5}$ species, while acid deposition focuses on SO_4^{2-} and NO_3^-, which are secondary species. As well, local sources may contribute disproportionately more primary $PM_{2.5}$ species, i.e. black carbon. Given that the annual SO_4^{2-}/NO_3^- ratio values were greater than 1 for both Japan and South Korea, sulfur emissions can be considered a key contributor to acid deposition.

The seasonal variation in acid deposition between Japan and South Korea was similar: higher in winter and summer and lower in autumn and spring. For Japan, the largest TAP occurred in winter and the smallest TAP occurred in autumn. For South Korea, the largest and smallest TAP occurred in winter and spring, respectively. Regarding the SO_4^{2-}/NO_3^- ratio, the seasonal variation in Japan and Korea suggests that SO_4^{2-} deposition was more important in summer and less important in the winter. For Japan, the value of these ratios ranged from 1.04 to 1.89; for South Korea, they ranged from 0.96 to 1.88. It should be noted that SO_4^{2-}/NO_3^- ratio is particularly lower in winter than in other seasons. Given minor local contributions, we conclude that TAP NO_x was significant in winter. Similar to the annual SO_4^{2-}/NO_3^- ratios, the seasonal ratios highlight the significant sulfate deposition in the two countries.



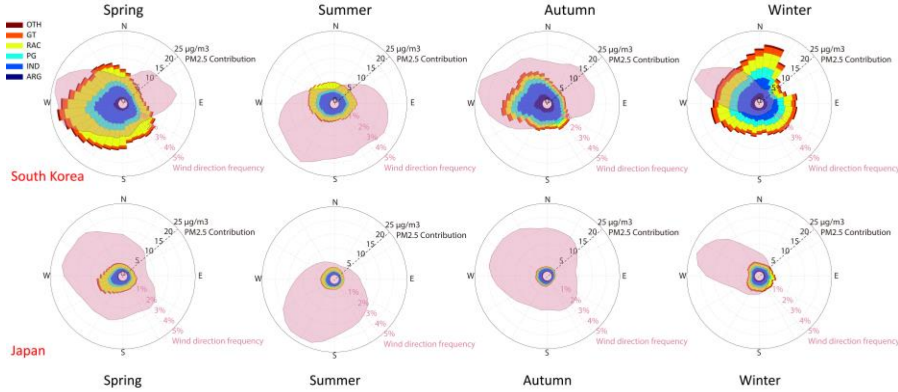


Figure 4. Seasonal wind roses for Japan and South Korea. Each direction bin presents the wind direction frequency.

3.5.2 Acid deposition over various land covers

Table 7. Percentage of land coverage (%) and air pollution-induced acid deposition (0.01Tg) across various land cover types in Japan and South Korea. 24 land cover types provided by the U.S. Geological Survey (USGS) were considered, including Urban and Built-up Land; Dryland Cropland and Pasture; Irrigated Cropland and Pasture; Mixed Dryland/Irrigated Cropland and Pasture; Cropland/Grassland Mosaic; Cropland/Woodland Mosaic; Grassland; Shrubland; Mixed Shrubland/Grassland; Savanna; Deciduous Broadleaf Forest; Deciduous Needleleaf Forest; Evergreen Broadleaf; Evergreen Needleleaf; Mixed Forest; Water Bodies; Herbaceous Wetland; Wooden Wetland; Barren or Sparsely Vegetated; Herbaceous Tundra; Wooded Tundra; Mixed Tundra; Bare Ground Tundra; Snow or Ice. The land covers with no acid deposition on them are not listed.

	Japan	
	% of grid represented by land cover type	**total acid deposition (0.01 Tg)**
Mixed Forest	55.28%	59.72
Water Bodies	11.88%	12.84
Savanna	8.15%	8.81
Irrigated Cropland and Pasture	5.53%	5.97
Cropland/Woodland Mosaic	5.04%	5.45
Shrubland	4.74%	5.12
Cropland/Grassland Mosaic	2.84%	3.07
Evergreen Needleleaf	2.15%	2.33
Dryland Cropland and Pasture	1.54%	1.66
Herbaceous Wetland	1.00%	1.08
Deciduous Broadleaf Forest	0.96%	1.04
Urban and Built-up Land	0.87%	0.94
	South Korea	
	% of grid represented by land cover type	**total acid deposition (0.01 Tg)**
Savanna	45.69%	17.1
Mixed Forest	20.86%	7.81
Irrigated Cropland and Pasture	11.02%	4.12
Water Bodies	9.06%	3.39
Cropland/Woodland Mosaic	6.36%	2.38
Dryland Cropland and Pasture	3.18%	1.19
Urban and Built-up Land	1.88%	0.7
Shrubland	1.04%	0.39
Deciduous Broadleaf Forest	0.92%	0.35

To assess acid deposition impact over various land cover types, Table 7~~Table 7~~ shows the percentage of each land cover type in Japan and South Korea along with its air pollution-induced acid deposition. We note that the land cover percentage refers to the percentage of the model grids that were dominated by each land cover type. For Japan, the land cover distribution shows that the most ~~popular~~ prevalent land covers (>5%) are mixed forest, water bodies, savanna, and irrigated cropland and pasture, and cropland/woodland mosaic. These land covers, when combined, account for ~87% of the land in Japan. Urban and built-up land occupies only ~1% of the land. In terms of the impact of acid deposition in the ecosystem in Japan, total deposition over mixed forest was 0.60 Tg, which may result in direct damage to trees and soil. In urban and built-up land, the acid deposition was estimated to be 0.01 Tg, representing ~1% of the total Japanese acid deposition.

For South Korea, the most ~~popular~~ prevalent land cover types are savanna, mixed forest, irrigated cropland and pasture, water bodies, and cropland/woodland mosaic. Together, they account for ~93% of the land, while urban and built-up land account for ~2% of the land. The acid deposition over savanna and mixed forest was estimated to be 0.17 Tg and 0.08 Tg, respectively. These two land covers share more than 66% of the total acid deposition in the country. Acid deposition on urban and built-up land was 0.01 Tg, which is comparable to that in Japan.

4. Discussion and Conclusion

This study estimated the ~~significant~~ contributions of both local sources and TAP from Asia on surface $PM_{2.5}$ in Japan and South Korea. ~~This~~ Our findings ~~was~~ were consistent with those reported by other studies (Aikawa et al., 2010; Koo et al., 2012). Among various emission sectors of China, our results show that, particularly with favorable prevailing wind, China's industrial emissions were the major contributor (~20%) to surface $PM_{2.5}$ as well as to acid deposition in Japan and South Korea. Our estimated wet deposition ratios of SO_4^{2-} and NO_3^- were still higher than 1.00, implying the need for further control of SO_2 emissions, particularly from China's industrial sector. Previous studies have reported a downward trend of SO_4^{2-} deposition in East Asia in recent years due to substantial SO_2 emissions reductions in China (Itahashi et al., 2018; Seto et al., 2004).

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In addition, wet deposition had significant impacts on mixed forests in Japan and the savanna in South Korea. It is noted that the dominant soils in Japan and South Korea have a low acid buffering capacity (Yagasaki et al., 2001). Acid deposition-attributable forest diebacks have been reported in Japan (Izuta, 1998; Nakahara et al., 2010) and South Korea (Lee et al., 2005). High acid deposition may cause soil acidification and eutrophication, which are particularly harmful in pH-sensitive areas such as forest and savanna. Despite the fact that N deposition may ~~enhance~~ increase soil N availability and hence photosynthetic capacity and plant growth in an environment with a low N availability (Bai et al., 2010; Fan et al., 2007; Xia et al., 2009), excessive N would suppress or damage plant growth (Fang et al., 2009; Guo et al., 2014; Lu et al., 2009; Mo et al., 2008; Xu et al., 2009; Yang et al., 2009), and also reduce biodiversity (Bai et al., 2010; Lu et al., 2010; Xu et al., 2006).

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In our analysis, we further revealed that higher TAP contributions from Asia occurred in spring in Japan and in winter in South Korea, due to the favorable weather conditions in the two seasons. While emissions of East Asia are projected to decline (Wang et al., 2014; Zhao et al., 2014), weather/climate may play a more important role under future climate change. Given the fact that summer and winter monsoons were weakening (Wang and He, 2012; Wang et al., 2015; Wang and Chen, 2016; Yang et al., 2018; Zhu et al., 2012), the frequency of favorable weather conditions for TAP from Asia is projected to decrease and TAP may be reduced subsequently.

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In conclusion, our findings highlight the ~~significant~~ significance of transboundary air pollution ~~depositing~~ affecting ~~in~~ Japan and South Korea as well as the impact of wet deposition on various land covers. In this way, this study provides a critical reference for atmospheric scientists to understand transboundary air pollution and for policy makers to formulate effective emission control policies, emphasizing the significance of cross-country emission control policies.

5. Competing interests

The authors declare that they have no conflict of interest.

6. Author contribution

S.H.L. Yim planned the research and sought funding to support this study. S.H.L. Yim conducted the analyses with technical supports from Y. Gu. S.H.L. Yim wrote the manuscript with discussions with all the co-authors.

7. Acknowledgment

This work was funded by The Vice-Chancellor's Discretionary Fund of The Chinese University of Hong Kong (grant no. 4930744). We would like to thank the Hong Kong Environmental Protection Department and the Hong Kong Observatory for providing air quality and meteorological data, respectively. We acknowledge the support of the CUHK Central High Performance Computing Cluster, on which computation in this work have been performed. The authors declare no competing financial interest.

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