Authors' response to the review comments #1

"Regional Contributions to Particulate Matter Concentration in the Seoul Metropolitan Area: Seasonal Variation and Sensitivity to Meteorology and Emissions Inventory" by Kim et al.

General response

The authors express their appreciation to the two reviewers and the editor. We believe that their comments are very productive and substantially contributed to improving the manuscript. We offer general responses and point-by-point responses to the issues and comments addressed by the reviewers. Reviewers' comments are shown in italics.

Anonymous Referee #1

1. The findings are framed as being robust and representing uncertainty. However, this limited number of cases does not constitute a comprehensive ensemble or represent the range of uncertainty that may exist in the emissions inventory. The modeling relies on a somewhat arbitrary set of 4 emissions inventories, which differ for domestic and foreign sources and which do not directly represent the year (2014) that is simulated. Thus, it is not justified to call the findings robust (p. 1, line 25 and p. 12, line 19) and more caution is needed in interpreting the findings.

Thanks for the comment. We understand the limitation of current study, and clarified the meaning of our findings in the manuscript. Our study intends to provide information of uncertainties of regional emission attribution depending on the selection of meteorology model and/or emissions inventories. We replaced the "robust" term with "mostly consistent".

"We also found that simulated surface PM concentration is sensitive to meteorology, but estimated contributions are mostly consistent." "While not a comprehensive ensemble, simulations using multiple combinations of emissions inventories all showed similar seasonal variation."

Selection of emissions inventory for regional air quality modeling is always an issue because we need to make a decision between "reliable but old emissions inventory" and "new but not fully tested emissions inventory". Current study is limited to the year 2014 due to the data availability. We are trying to extend this attribution estimation for longer period. Preliminary results during 2004-2015 show estimated contributions are mostly invariant year-by-year (Figure R*1*).

2. Source contributions are defined by doubling the impact of 50% emission reduction runs. However, if the reductions are applied only to anthropogenic emissions (this was unclear), then some of what is being termed "foreign" is actually resulting from biogenic emissions within Korea. Also, zero-out impacts are often larger than 2x the impact of 50% out cases, due to nonlinearities of the chemistry of pollutant formation. If that is the case here, it would systematically under-represent the domestic contribution, and hence over-represent the foreign share. One run should be conducted to test the linearity of response from 50% to 100% reduction, and language should be more cautious in defining source apportionment if it is based on 50% cases.

Sorry for the confusion. Emission reduction was applied to the total national emission (i.e. total biogenic and anthropogenic emission within South Korea). We have clarified it in the methodology section.

"Here, we used a 50 % reduction in the South Korean national emission (e.g. total biogenic and anthropogenic emissions within South Korea) as a test."

We also conducted an additional full-year simulation to test the linearity of the brute force method (BFM) of 50% domestic emission reduction. For the BFM method sensitivity test, we have compared two BFM runs using 50% reductions of domestic and foreign emissions. We think the concept of zero-out contribution could be ambiguous in the national emission level. Unlike local emission sources which can be practically zeroed-out, removing total national emissions is usually an unrealistic scenario. In an alternative approach, we tried to compare two 50%- emissions reduction methods for domestic emissions and foreign emissions. Seasonal variations of two BFM methods over the Seoul Metropolitan Area (SMA) and South Korea (SKR) are compared in Figure R2. As expected, responses to the reductions of domestic and foreign emission are not identical, showing non-linearity of responses. While estimated foreign emission contributions using different BFM methods show similar seasonal variations, high in winter and low in summer, month-to-month variations show a certain uncertainty range, ~10%, which is not much different from the uncertainties from meteorology model selection or emissions inventory selection.

3. The model substantially under-estimates observed PM. This raises serious doubt about the conclusions, since it could indicate error in either the domestic or foreign emissions inventory.

Thanks for the comment. We agree that missing emission sources (e.g. dust and fire emissions and/or other unknown emission sources) might be associated with current model bias, but do not think that current model bias raises any critical issue in the quality of emission inventories. Currently, we suspect there are several issues that can cause model low biases. In current modeling system, modeling biases often occur by following reasons: (a) Uncertainty in emission inventory: Missing or old-dated emission emissions inventory. As the reviewer commented, lack of dust and fire emissions could explain a portion of model bias. (b) Missing chemical mechanisms: Unknown mechanisms of secondary organic aerosols formation or heterogeneous reactions (e.g. such as Sulfate formation on the surface of Asian Dust) (Baker et al., 2016; He et al., 2014; Xue et al., 2016). (c) Model wind overestimation. This issue is already mentioned in the current manuscript -- meteorological models sometimes fail to reproduce low wind speed (e.g. the stagnant condition) (Ngan et al., 2013), resulting in the underestimation of simulated particle concentration. Based on discussion with meteorological modeler (S. Hong, personal communication), we may improve the surface wind speed by adjusting background diffusivity, but he did not recommend it because it can ruin the predicted precipitation amount. We expect, wind bias issue will be improved as meteorological models further develop.
The bottom line is that current model is not perfect, and may be limited in generating absolute amount of particulate matter concentration, but performs pretty well to simulate spatial and temporal variations. In terms of relative attribution assessment, we do not see any serious limitation in the current modeling system we have employed.

Baker, K.R., Woody, M.C., Tonnesen, G.S., Hutzell, W., Pye, H.O.T., Beaver, M.R., Pouliot, G., Pierce, T., 2016. Contribution of regional-scale fire events to ozone and PM2.5 air quality estimated by photochemical modeling approaches. Atmospheric Environment 140, 539–554. doi:10.1016/j.atmosenv.2016.06.032

He, H., Wang, Y., Ma, Q., Ma, J., Chu, B., Ji, D., Tang, G., Liu, C., Zhang, H., Hao, J., 2014. Mineral dust and NOx promote the conversion of SO2 to sulfate in heavy pollution days. Scientific Reports 4. doi:10.1038/srep04172

Xue, J., Yuan, Z., Griffith, S.M., Yu, X., Lau, A.K.H., Yu, J. Z., 2016. Sulfate Formation Enhanced by a Cocktail of High NOx, SO2, Particulate Matter, and Droplet pH during Haze-Fog Events in Megacities in China: An Observation-Based Mode ling Investigation. Environ. Sci. Technol. 50, 7325–7334. doi:10.1021/acs.est.6b00768

Ngan, F., H. Kim, P. Lee, K. Al-Wali, B. Dornblaser, 2013, A study on nocturnal surface wind speed overprediction by the WRF-ARW model in Southeastern Texas, *J. of App. Meteo. and Clim.,* doi:10.1175/JAMC-D-13-060.1

Minor suggestions: p. 2, line 28: "region's"

Corrected

p. 2, line 33: excess precision in numbers

Corrected

p. 3, lines 3-6: Meteorological uncertainty has been studied elsewhere, with larger ensembles than considered here

Thanks for the comment. We know that meteorological ensembles have been studied actively, but those uncertainties in emission attribution in South Korea are not well addressed. We have clarified our point in the manuscript.

"While many studies have addressed sources of uncertainties in the estimation of contributions or source apportionment, few have tried to investigate, whether qualitatively or quantitatively, the uncertainties resulting from the meteorological model, especially in the estimation of source contributions in South Korea."

p. 6, line 26: "compromising" is the wrong word

We have modified the sentence.

"However, the BFM still provides efficient and practical way of analyzing source contributions."

p. 9, lines 1-7: These explanations are not convincing, and the discussion of specific days is not helpful

Thanks for the comment. As discussed in the model bias comment, we do not have clear evidence to specify the reason of model low bias from multiple possibilities. Manuscript was modified to mention this point.

"One typical problem in the chemical modeling of surface PM in the SMA is that simulated surface PM concentration constantly underestimates observed measurements. Low bias can happen with several reasons: missing or old-dated emission sources, lack of dust and fire emissions, unknown chemical mechanisms, and meteorological bias, such as the wind bias discussed in this study. At this point, we do not have clear evidence to specify the reason. In general, surface PM concentration simulated using UM-CMAQ generates higher PM concentrations compared to the WRF-CMAQ system, which we suspect results from UM-CMAQ's weaker wind field, which results in a more stagnant and shallower boundary layer."

p. 12, line 25: How can results be "considerable but not significant"?

We replaced the sentence.

"We found that differences in meteorological model can lead to discernible differences in the estimation of contributions from regional (e.g., domestic and international) emissions sources although they still have similar seasonal patterns."

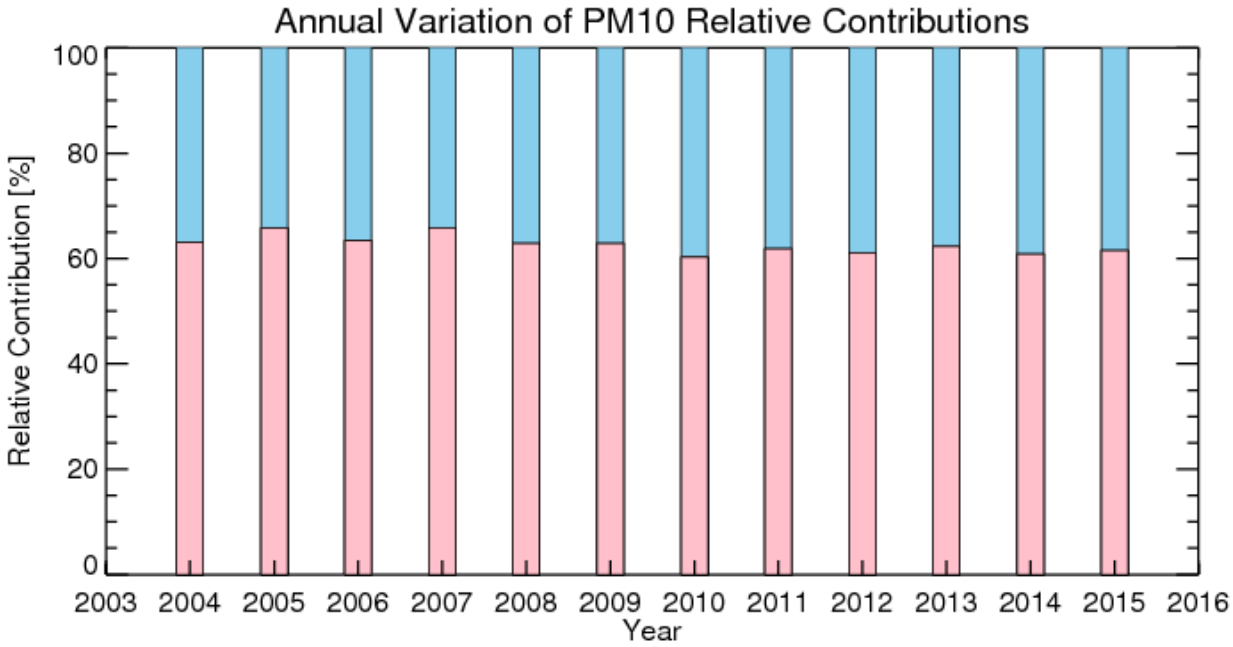

Figure R1 Inter-annual variation of estimated contributions from foreign emission sources (pink) and South Korean domestic source (blue). INTEX-B 2006 and CAPSS 2007 emissions inventories were used for international and South Korean emissions inventories, respectively.

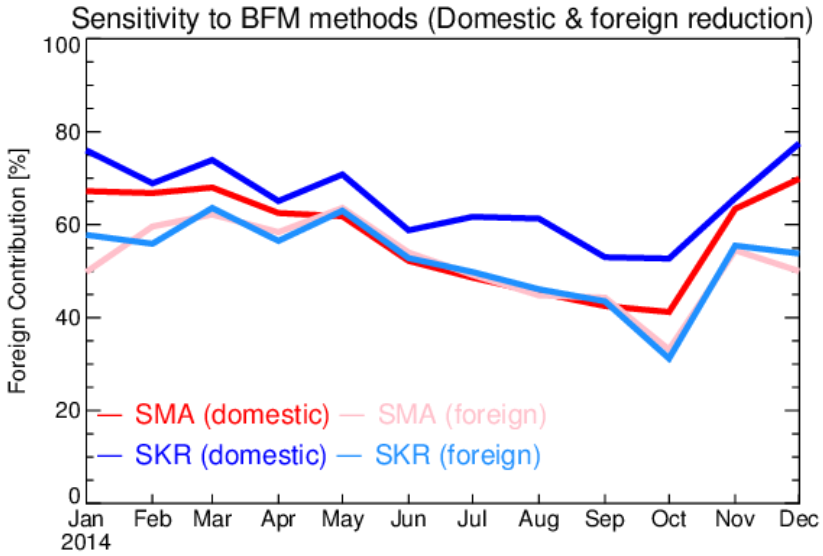

Figure R2 Estimated contributions from foreign emission sources in 2014 over the Seoul Metropolitan Area (SMA) and over South Korea (SKR). Two BFM methods, reducing 50% of domestic emissions or foreign emissions, were compared.