Authors' response to the review comments #2

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

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.

The article generally succeeds in what it attempts to achieve, limited mostly by the apparent absence of dust and wildfire emissions in all baseline and sensitivity simulations, the difference in vintage between emissions and simulation period, and the resultant low biases in simulated concentrations.

Thanks for the comment. Here are responses to three main concerns and minor comments from the reviewer.

(1) Fire and dust emissions

Episodic natural emissions from Asian dusts and wildfires have been seriously considered from the designing stage of the study. Technically, current modeling framework is capable of providing Asian dust and wildfire emissions, and we have studied several cases of fire and dust events (Bae et al. 2016). However, after initial assessment, we decided not to include modeling of Asian dust and wildfires in current regional emission attribution assessment. Estimated impact of dust and fire emissions in South Korea during 2014 was relatively small (<5%) and modeling of dust and fire emissions still have high uncertainties, especially in the threshold parameterization of friction velocity, for the dust emissions, and the magnitude and plume rise parametrizations, for the fire emissions.

Impacts of dust and fire emissions were investigated by filtering days with chances of dust or fire impact. For dust days, dust observations from the Korean Meteorological Administration (KMA) were used (http://www.kma.go.kr/weather/asiandust/observday.jsp). In 2014, 10 dust days were reported in the SMA. Discarding those days resulted in lowering the annual average of PM_{10} concentration over the SMA by 3.2% (51.6 $\mu g/m^3$ to 49.9 $\mu g/m^3$). For fire days, there is no official report of fire impact observations. We utilized chemical component measurements (e.g. PM, OC, EC, K and SO4) at the Bulkwang supersite (at Seoul, lon=126.9, lat=37.6), satellite wildfire emissions data (QFED, based on MODIS fire radiative power, https://gmao.gsfc.nasa.gov/research/science_snapshots/global_fire_emissions.php), and NOAA HYSPLIT backward trajectory simulations. Considering probable fire emission chemical signals (high OC, EC and K, OC/EC ratio, and low SO_4) and daily backward trajectory pathways together, we selected 8 days of fire impact, including July 27-28 episodes as reported in Jung et al. (2016). Impact of those days were 0.4% (51.6 $\mu g/m^3$ to 51.4 $\mu g/m^3$). Figure R1 summarizes time series of surface PM_{10} concentration over the SMA region in 2014. Days with (likely) dust and fire impacts are marked with "D" and "F". Examples of surface measurements and trajectory runs are shown in Figure R*2* and Figure R*3*.

Still being preliminary, these calculations suggest that the estimated dust and fire impact in South Korea during 2014 might be smaller than other uncertainties we have discussed in current study. We have clarified the use and (estimated) impact of dust and fire emissions in the manuscript. This analysis also will be prepared as a separate research article later.

Bae, C. H., H. C. Kim, B.-U. Kim, and S. Kim, 2016: Implementation of Near Real-Time Fire and Dust Emissions in Air Quality Forecast over Northeast Asia, *17th IUAPPA Conference*, Busan, South Korea

Jung et al., 2016: Impact of Siberian forest fires on the atmosphere over the Korean Peninsula during summer 2014, ACP, doi:10.5194/acp-16-6757-2016

(2) Date of emission inventory

Selection of emissions inventory brings another big dilemma because a decision should be made between "reliable but old emission inventory" and "new but not fully tested emissions inventory". Current study was conducted for the year of 2014 because the UM model simulations (provided by KMA) are only available for the period. We are trying to extend current regional emission attribution estimation for long-term period, and preliminary results suggest no significant inter-annual variations (Figure R*4*).

(3) Model bias

We agree that lack of Asian dust and wildfire emissions is one of the reasons for model low bias. However, we are hesitant to conclude that they can explain all the model biases. Simply, model biases happen regardless of dust or fire events, and current model bias is larger than our estimation of dust and fire impact. Currently, we have three hypothesis of model underestimation of PM concentration.
With 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 concentration. Based on discussion with meteorological modeler (S. Hong, personal communication), we may improve 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 develops.

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

A second set of simulations to assess the role of transported biogenic emissions would be valuable, but less critical.

Thanks for the suggestion. We will pursue this idea in our next study.

Technical comment
The authors do not define the simulation period until the results section. Specific start and end dates for the simulation and any initialization period should appear in the first paragraph of section 2.

Simulation period and spin-up time were included in section 2.

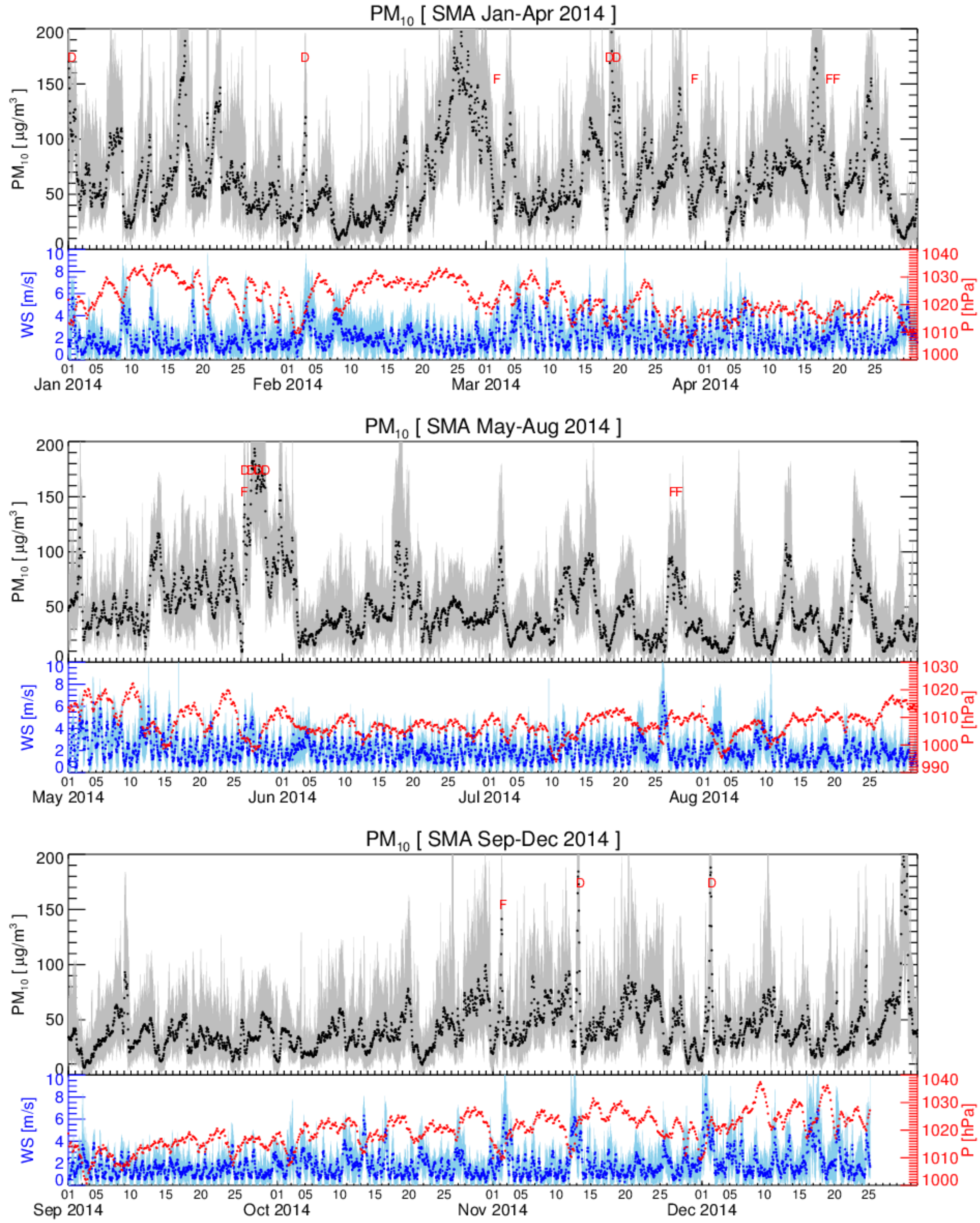

Figure R1 Time series of PM_{10} concentration from surface monitoring sites (112 sites) in the SMA, Korea. 10-m wind speed and surface pressure are also shown in blue and red lines. Dust and fire cases are marked with "D" and "F", respectively.

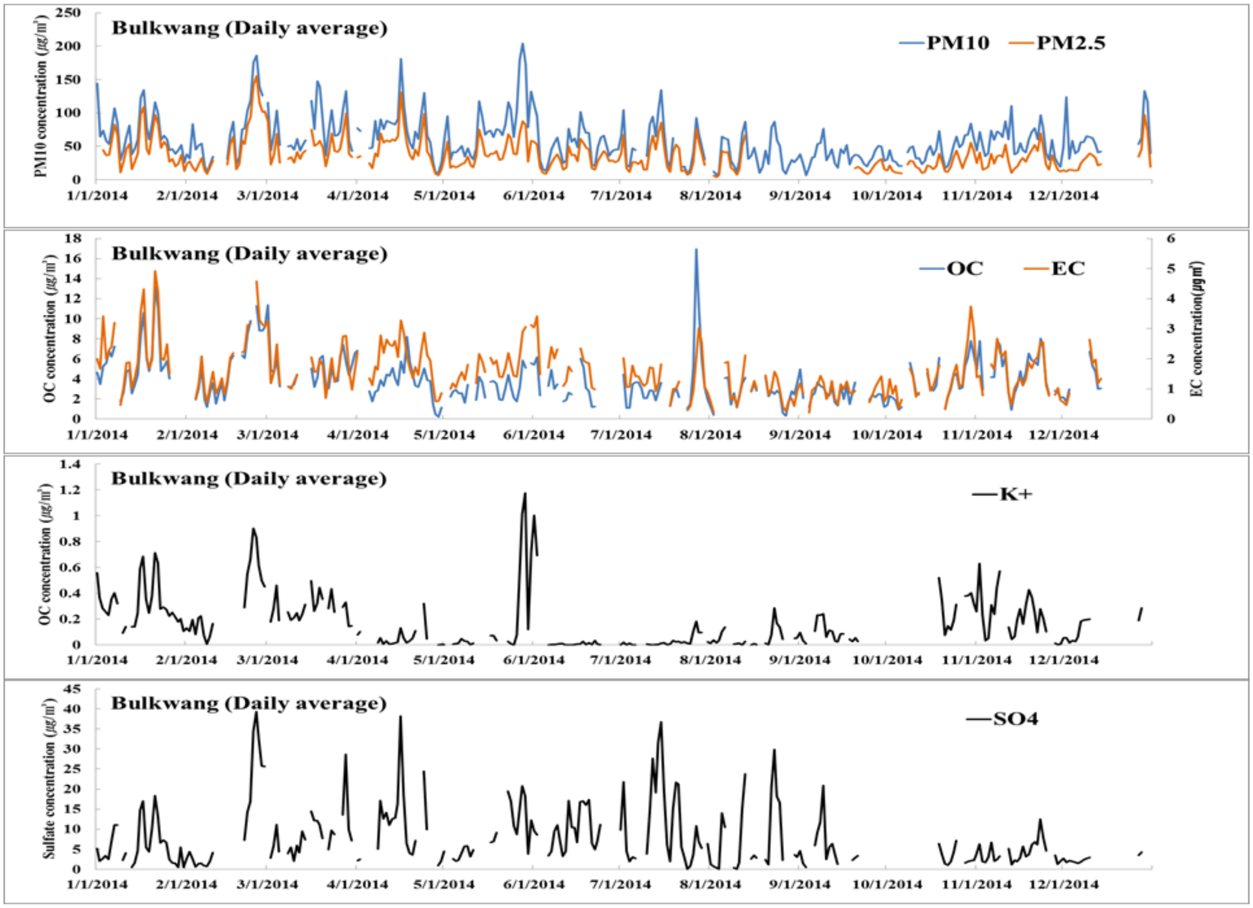

Figure R2 Time series of PM_{10}, $PM_{2.5}$, OC, EC, K and SO_4 measured at the Bulkwang super site in Seoul, Korea.

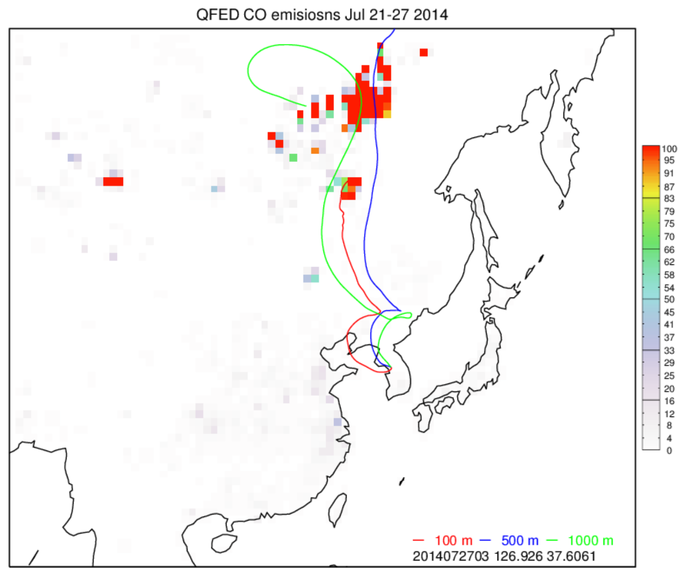

Figure R3 7-day HYSPLIT backward trajectory simulations overlaid with QFED fire emission (CO emission during July 21-27), arriving at the Bulkwang super site on July 27, 2014, 12:00PM local time.

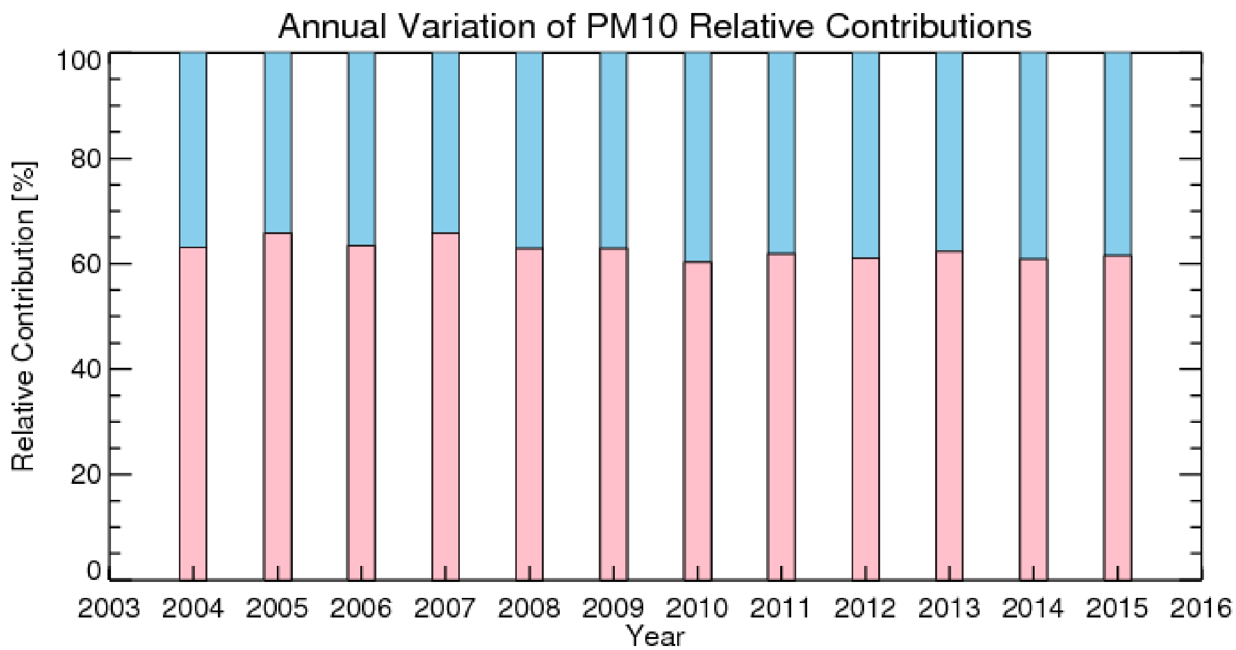

Figure R4 Inter-annual variation of estimated contributions from foreign emission sources (pink) and domestic source (blue). INTEX-B 2006 and CAPSS 2007 emissions inventories were used for international and South Korea, respectively.