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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Observationally-constrained carbonaceous aerosol source estimates for the Pearl River Delta area of China" *by* N. Li et al.

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We thank the two anonymous reviewers for their helpful comments and suggestions. We address below the major issues raised by the reviewers.

(1) One reviewer commented that the time of simulation should exactly match the dates of the measurement to avoid sampling biases.

We agree that, in general, the time of simulation should exactly match the dates of the measurements when doing model-observation comparisons to avoid sampling biases. However, in the present case our goal is to summarize the average seasonal and spa-

tial variability of the observed surface EC and OC concentrations and use that average seasonal/spatial variability signature to constrain PRD emissions. We do not attempt to capture the daily- to sub-seasonal-scale variability in either the observation or the simulation. Doing so would in fact be futile for the following reasons. Firstly, the measurements were conducted in different months and in different years and averaged here to represent seasonal mean conditions (Table 4 in the manuscript). Secondly, most measurements were conducted using filters accumulated for 24 hours and sampled every 1 to 6 days for short periods of time (Table 4 in the manuscript), such that sub-seasonal variability were poorly characterized by the measurements. Thirdly, at locations where the sub-seasonal variability were observed (characterized by, e.g., the standard deviation of the measured concentrations), the standard deviation is < $\pm$50% of the measured seasonal mean at most sites, indicating that the sub-seasonal variabilities are relatively small. Finally, daily-resolved emission inventories are generally not available (with the exception of satellite-based daily emission inventories for biomass burning, e.g., Wiedinmyer et al., 2011). Therefore, we think that comparing the observed and simulated results on a seasonal-average basis is reasonable, provided that both the observation and the simulation actually are representative of the seasonal-average conditions. We will add the above rationale to the revised manuscript.

(2) In relation to the (1) comment, both reviewers suggested that using four "7-day periods" to represent the four seasons may be unreasonable, and the spin-up time of one day may be too short.

We agree that simulating a "7-day period" may not be sufficient to represent the seasonal-average conditions. We will expand our simulations to at least one month per season, with at least 5-day spin-up time to better capture the seasonal-average conditions.

(3) One reviewer suggested that the two inner domains may be too small to account for lateral transport.

The boundary conditions from our two inner domains (D2 and D3) are provided by the outer domain (D1), which covers most of East Asia. The boundary condition for D1 is in turn provided by the monthly mean concentrations simulated by a global chemical transport model. Therefore the impacts of lateral transport are already accounted for. Nevertheless, we agree that later inner domains would help improve the simulation of short-term variability in aerosol concentration. We will enlarge the inner domain sizes in the revision. Modified domain settings are described in Figs. 1 and 2.

(4) One reviewer questioned whether Tap Mun should be considered an urban site and included in the calculation of the "Hong Kong" urban concentrations.

We appreciate the reviewer's concern. We will re-group the measurement locations in Hong Kong to represent an urban site and a rural site, the latter representing the average of "Tap Mun", "Hok Tsui", and "Tung Chung".

(5) One reviewer was concerned about the use of multiple regression on constraining EC and OC sources.

The use of multiple regression is appropriate for EC, since the surface concentrations are linearly related to the strength of emissions. As the reviewer pointed out, this method is not suitable for primary (POC), since it is not possible to unambiguously determine the POC concentrations in the measurements. This is why we use the "hybrid" method to constrain POC sources: we used the coefficients $\beta 1$, $\beta 2$, and $\beta 3$ from the multiple regression analysis for EC emission in Eq. (2) to scale the bottom-up OC emission estimates. This is equivalent to combining the POC/EC emission ratios from bottom-up inventories with the observed EC constraints. The uncertainties associated with this POC source estimate come from two traceable origins: (1) explicitly from the multiple regression analyses on EC observations, and (2) implicitly from the POC/EC emission ratios used in the bottom-up inventories.

(6) Both reviewers suggested that the uncertainties of the model and the top-down emission estimates be better quantified statistically.

We appreciate the reviewers' concerns and will add more detailed statistics to describe the model performance, the model/observation comparison, and the top-down emission constraints.

(7) One reviewer raised concerns about the characterization of the non-transportation source.

We agree with the reviewer that "non-transport" includes many sector, and that saying "non-transportation" sources are the main sources is somewhat misleading. We will include more detailed sectorial discussions in the revision.

Reference Wiedinmyer, C., Akagi, S.K., Yokelson, R.J., Emmons, L.K., Al-Saadi, J.A., Orlando, J.J., and Soja, A. J.: The Fire INventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning. Geosci. Model Dev., 4, 625-641,doi:10.5194/gmd-4-625-2011,2011.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 33583, 2015.

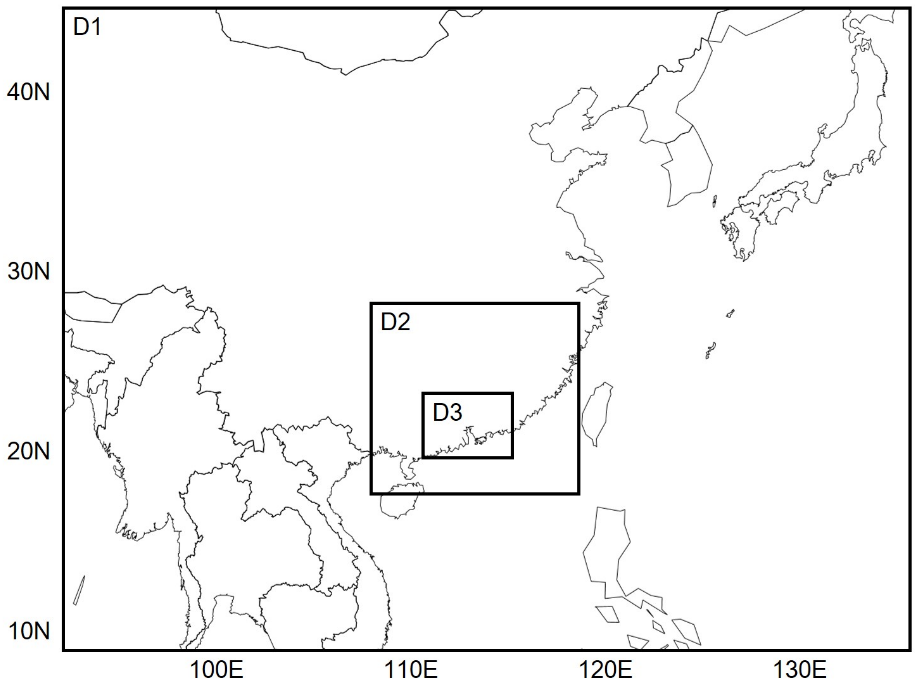

Fig. 1. The modified three nested domains in the CMAQ simulation

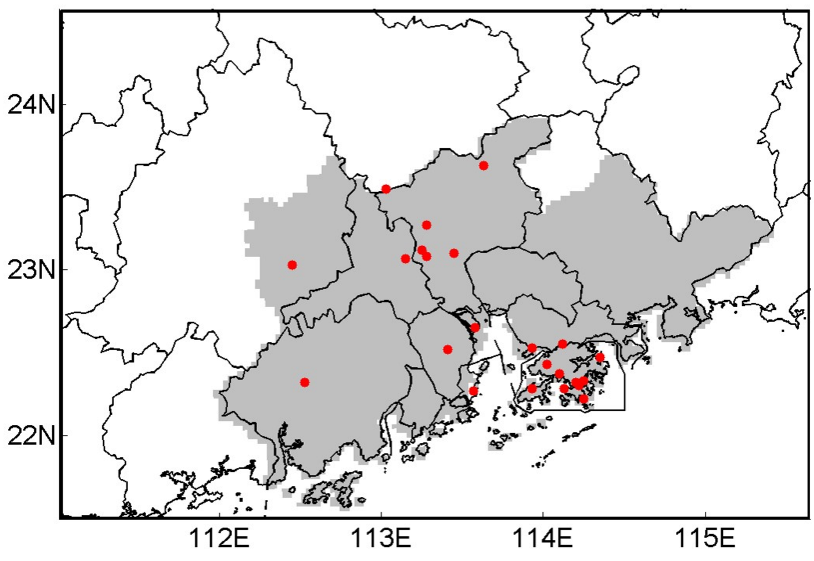

Fig. 2. The area of domain 3. Also shown are the PRD area (gray area) and observation sites (red points)