Response to Reviewer 2:

First of all, thank you for your constructive comments on this manuscript. We have tried to address all the comments you made in the manuscript and also in the replies. The line numbers mentioned in the following replies are based on the revised manuscript with track changes.

General Comments

This manuscript describes the initialization of ground-level PM2.5 for a chemical transport model using the ensemble Kalman Filter (EnKF) method. Authors implemented EnKF in the CMAQ model and claim this method improves PM2.5 predictability compared to the 3DVar and No DA. The PM2.5 predictability of South Korea is further improved if the EnKF is applied not only to the nest domain but also mother domain with ground PM2.5 observation. The manuscript looks like to have a reasonable structure for the paper. It will be available for publication if it is improved with a minor revision.

Specific Comments

- #1. In the manuscripts, ICs is an abbreviation for both initial conditions and initial concentrations. The same goes for BCs. Authors should only use the abbreviation "C" for either condition or concentration
- Reply: Thank you for this point. We first define the abbreviations of ICs and BCs (see lines 17, 19, and 38), and then use both abbreviations in the revised manuscript.
- #2. What are the background variables that are input to EnKF? I believe the EnKF uses background error covariance between PM2.5 and some meteorological variables. Is it correct that the experiments only update PM2.5, so other meteorological variables are the same as before DA? Adding a list of background and analysis variables is recommended.
- **Reply:** Yes, we updated only PM_{2.5} via DA. Therefore, there were no error correlations between meteorological variables and PM_{2.5}. We used the identical meteorological conditions for all the DA experiments. Because PM_{2.5} is a single control variable, we did not add a list of variables in the original manuscript. Regarding this point, we added same sentences into Section 2.1 (please, check out **lines 143–147**).
- #3. I am wondering about the observation operator for the PM2.5. Is PM2.5 one of the background variables? If not, the authors need to introduce the observation operator for it to calculate observation operator. Some descriptions for the observation operator would be better to be added in the manuscript.
- **Reply:** We agree with reviewer's comment that further descriptions of the observation operator are necessary. For a simple observation operator, we calculated PM_{2.5} before the DA, using aerosol-related species via a post-processing tool in the CMAQ software package. This could also be possible because we used PM_{2.5} as a single control variable. We have added same sentences about this process into the revised manuscript (please, refer to **lines 143–147**).
- #4. Line 174: What is the inflation parameter (alpha) for RTPS?
- **Reply:** Reply to this comment #4 is coming up with the following comment #5.

- #5. Since you have described the parameter for the localization, it would be better to also describe the parameter for RTPS.
- **Reply:** We will try to response to comments #4 and #5 together. We set α of the inflation parameter to be 1.0. We here assumed that the meteorological model was perfect. Therefore, no perturbations were made for the ensemble spread. Another reason was that perturbing meteorological variables could also break dynamical balances in the model simulations. Throughout the experiments, we used 1.0 for the value of α , and then inflated both the predicted ensemble (before DA) and assimilated ensemble (after DA) rather than using 1.2, the value used in Pagowski and Grell (2012) and Schwartz et al. (2014) (regarding this point, please refer to **lines 184–191**).
- #6. Line 175: Why do you apply the RTPS to ensemble before and after DA? The RTPS compares ensemble spread before and after DA because the amount of inflation in RTPS is proportional to the ensemble spread reduced by the DA. So theoretically, RTPS can be applied only once after DA.
- **Reply:** Comments #4, #5, and #6 are a series of questions about similar issue. More specific details have been added into the revised manuscript (please, refer to **lines 184–191**).
- #7. When you describe DA_icbc, can you show the pm2.5 field for domain1 which contains domain 2? For example, horizontal field of PM2.5 as in figure 4, but with domain 1. The distribution of PM2.5 over domain 1 can be clearer evidence showing the effect of boundary conditions.
- **Reply:** We agree with the reviewer's opinion. Instead of adding the PM_{2.5} field for domain 1 to Fig. 4, we added **Fig. S5** into the supplementary information to describe the impacts of the updated (or increased) transboundary PM_{2.5} by assimilating the concentrations from the ground stations in China. This additional figure could help readers to better understand the improved performances on 25 and 26 May, 2016 (high PM_{2.5} episode during the KORUS-AQ campaign) in Fig. 8 (also, refer to **lines 374–376**).

Technical Comments

#8. Line 245: sate --> stateReply: We have changed it (please, see line 266).