

Response to reviewer 1

We appreciate reviewer's thoughtful comments and suggestions, which are greatly helpful for us to improve our manuscript. The manuscript has been revised to accommodate the reviewer's comments and suggestions.

General comment This manuscript described a method using two different emission inventories to estimate background error covariance (BEC) for 3D-Var chemical data assimilation, and performed the corresponding sensitivity studies compared with the traditional NMC method generated BEC. One key issue is whether the better result achieved with the new method came from its better science or simply the larger BEC shown in Figure 5. If the difference between these two emission inventories was not so big, since they were just estimated emissions, could the new BEC still outperform the control run?

Response We agree with reviewer's comment that the differences between the two emission inventories determines the performances of data assimilation and short-term PM_{2.5} predictions. However, please also note that the differences in the two emissions are not artificially created, but they are derived based on two independent emission inventories established upon independent emission statistics and factors in East Asia. Certainly, the small differences from the independent emissions will lead to small uncertainties in emission inventory. Please, also note that the differences in these two emissions are not very big (relatively small), being less than ~10%, as shown in Table 1. Regarding this point, please refer to pp. 5:161–pp 6:167 in the revised manuscript.

Specified comments

Comment Line 63-75. You may need to re-write some contents. The NMC method used the later initialized field to reflect the more reliable prediction, or nearer the observation. Based on that difference, the model error was estimated. It is true that the NMC method may not be suitable for chemical DA, since the AQ model sometimes is more sensitive to emissions instead of the initial conditions, not because "emissions are not a state variable propagated in time" (line 67). In your later discussion (2.1 Model Configuration), there was nothing representing the observation for BEC estimation. How could you estimate the model error just based on difference between the two emission inventories?

Response We have re-written the sentences. Please, see pp. 3:71 – pp. 3:73. Main focus of this study was on how to improve our short-term PM_{2.5} prediction via new BEC using the emission perturbations. We think that the estimations of parameters in the BEC and model error against observations are a challenging topic and may be beyond the scope of this study.

Comment In the main description (line 105-130) about the new BEC construction, it stated where these two emission inventories came from, but did not mention the uncertainty in the emissions, which was more important. In fact, once the emission uncertainty is known, one may get the BEC from perturbing a single emission inventory instead of two.

Response Thank you for this opinion! Indeed, perturbations in single emission inventory can be used if we fully understand the sources of uncertainties in the emission inventory. It might be possible to estimate the uncertainty in emissions of selected chemical species having long lifetime or being measured intensively via inverse modeling techniques, but it is impossible to know the uncertainties of all chemical species in emission inventories. To address reviewer's comment, we have also added a recent study related to this issue. Please, see pp. 3:78 – pp. 3:81.

Comment Section 2.3. This section only mentioned how to filter out bad observation, and did not tell how to estimate observation error used in the DA. Figure 5, Besides the profiles of BEC standard deviation, it is better to have a regional map showing its horizontal distributions. Figures 6-9. These comparisons have issues. It should be avoided comparing DA results to the same observations used in DA. Otherwise, the higher BEC, the better results. All the comparisons should be made after certain hours of the forward simulation to make sure that the DA will not degrade the prediction or cause side effects, e.g. RMSE would not increase.

Response Please, see pp. 6:187 – pp. 6:189 describing the observations errors. We selected a method of binning as sampling all the horizontal grid points per each vertical level (i.e., an option of `bin_type = 5` in GEN-BE v2.0). Therefore, we have a single vertical distribution of the BEC parameters. We have added this information into pp. 7:219 – pp. 7:221. To answer the reviewer's comment, we carried out additional comparison of DA results with the 20% of independent observations which were taken out and were then used only for comparison

purpose. Regarding this point, please, refer to pp. 9:262–pp. 9:268 and pp. 11:333–pp. 11:340.

Comment The Figure 10 and all the statistics should follow the same comparison rule mentioned above.

Response Please, see the revised paragraphs mentioned above (pp. 9:262–pp. 9:268 and pp. 11:333–pp. 11:340).