Reply to Referee #1:

We deeply appreciate your helpful comments and suggestions, which enabled us to improve the quality of our present study. In our response, we use italicization in blue to indicate the reviewer's comments, and normal type in black for our response. Besides, we use boldface type to indicate changes in the manuscript.

General comments:

One additional analysis that I would like to see is a comparison among BECs per species. This would help show how each BEC constrains the spreads vertically and horizontally around the observation sites. Similar analyses are described in below:

Descombes, G., Aulign, T., Vandenberghe, F., Barker, D. M. and Barré, J.: Generalized background error covariance matrix model (GEN_BE v2.0), Geosci. Model Dev., 8(3), 669–696, doi:10.5194/gmd-8-669-2015, 2015.

Liu, Z., Liu, Q., Lin, H.-C., Schwartz, C. S., Lee, Y.-H. and Wang, T.: Three-dimensional variational assimilation of MODIS aerosol optical depth: Implementation and application to a dust storm over East Asia: AOD DATA ASSIMILATION, Journal of Geophysical Research: Atmospheres, 116(D23), doi:10.1029/2011JD016159, 2011.

Response:

Accepted. Following the references above, a comparison among the BECs per species has been added in the manuscript with a new figure as Figure 2, which shows the background error standard deviations. Also, analyses about the background error horizontal correlation length scales and vertical correlation have been mentioned in the manuscript with figures shown here as Figures R1 and R2 (not shown in the manuscript). The analyses are presented in section 2.3.3 in the manuscript as below.

"Following the analyses based on the GEN_BE v2.0 (Descombes et al. 2015), Figure 2 presents the background error standard deviations of each species at different vertical levels. For the aerosols in the first three size bins (Fig. 2a-2c), although the standard deviation errors vary across the species, the errors of NO_3^-, SO_4^{2-}, NH_4^+, OC, and OIN are generally larger than that of the others (BC, Cl and NA) in the three size bins. These results are consistent with the finding in Chen et al. (2019), which allows inorganic compounds (NO_3^-, SO_4^{2-}, NH_4^+), OC and OIN to be adjusted more in corresponding to their larger background errors. For the aerosols in the 4th size bin (Fig. 2d), the errors are unreasonably much smaller than that in the first three bins due to model deficiency. Under this circumstance, to get a reasonable bigger adjustment for the aerosols in the 4th size bin, it might need to enlarge their background errors in the DA procedure. As for the gaseous pollutants (Fig. 2e), CO has the biggest background errors in the middle and lower layers, followed by O3, SO2 and NO2.

For the background error horizontal correlation length scales, the results are similar as in Liu et al. (2011) (figure omitted). The length scales of aerosols are comparable in most of the species, which generally span from 1.5 to 2.5 times the grid spacing, while the aerosol species NA exhibits a smaller horizontal length scale than all the other species. For the

background error vertical correlations (figure omitted), the results are similar as in Descombes et al. (2015), in which the vertical correlations are bigger in the lower levels (where they are emitted) in most of the species. According to Descombes et al. (2015), the reactions with species emitted near the surface might create these strong correlations in the lower model levels."

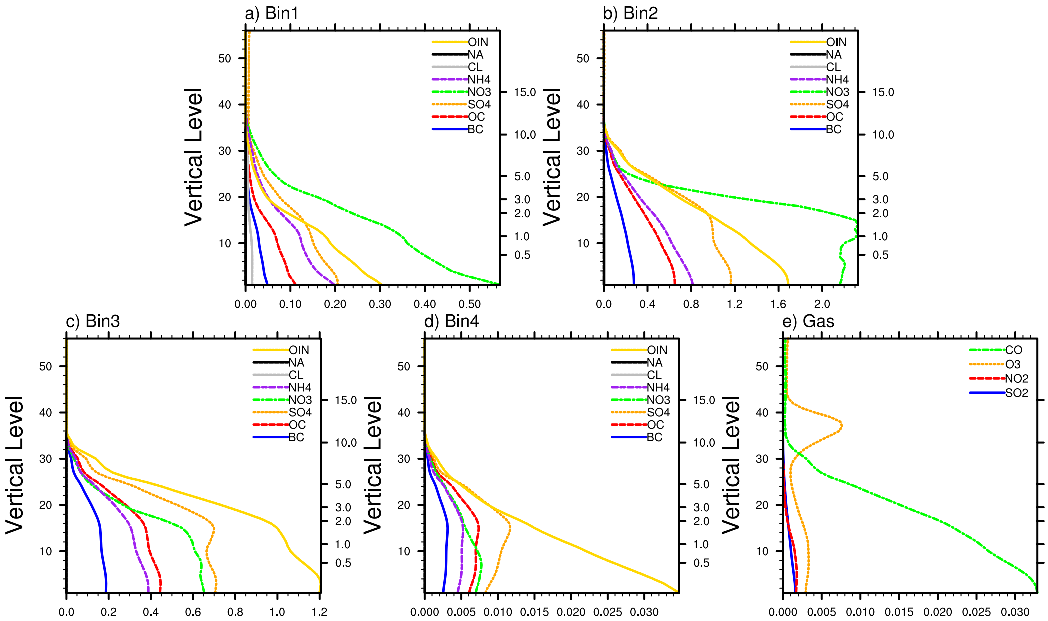

Figure 2. Background error standard deviations of aerosol species in the (a) 1st size bin, (b) 2nd size bin, (c) 3rd size bin, (d) 4th size bin, and of (e) gas pollutants. The units for the x-axis are μg m^{-3} for (a)-(d) and ppm for (e). The left y-axis denotes the model level, and the right y-axis denotes the vertical height (units: km).

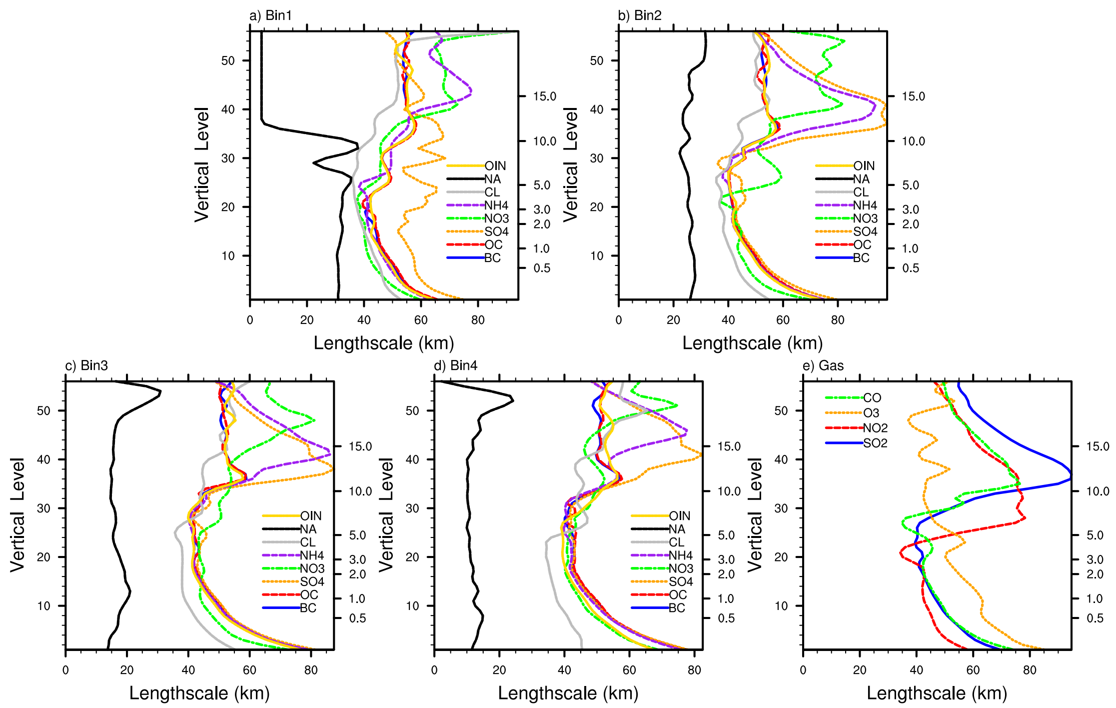

Figure R1. Background error horizontal correlation length scales of aerosol species in the (a) 1st size bin, (b) 2nd size bin, (c) 3rd size bin, (d) 4th size bin, and of (e) gas pollutants (units: km). The left y-axis denotes the model level, and the right y-axis denotes the vertical height (units: km).

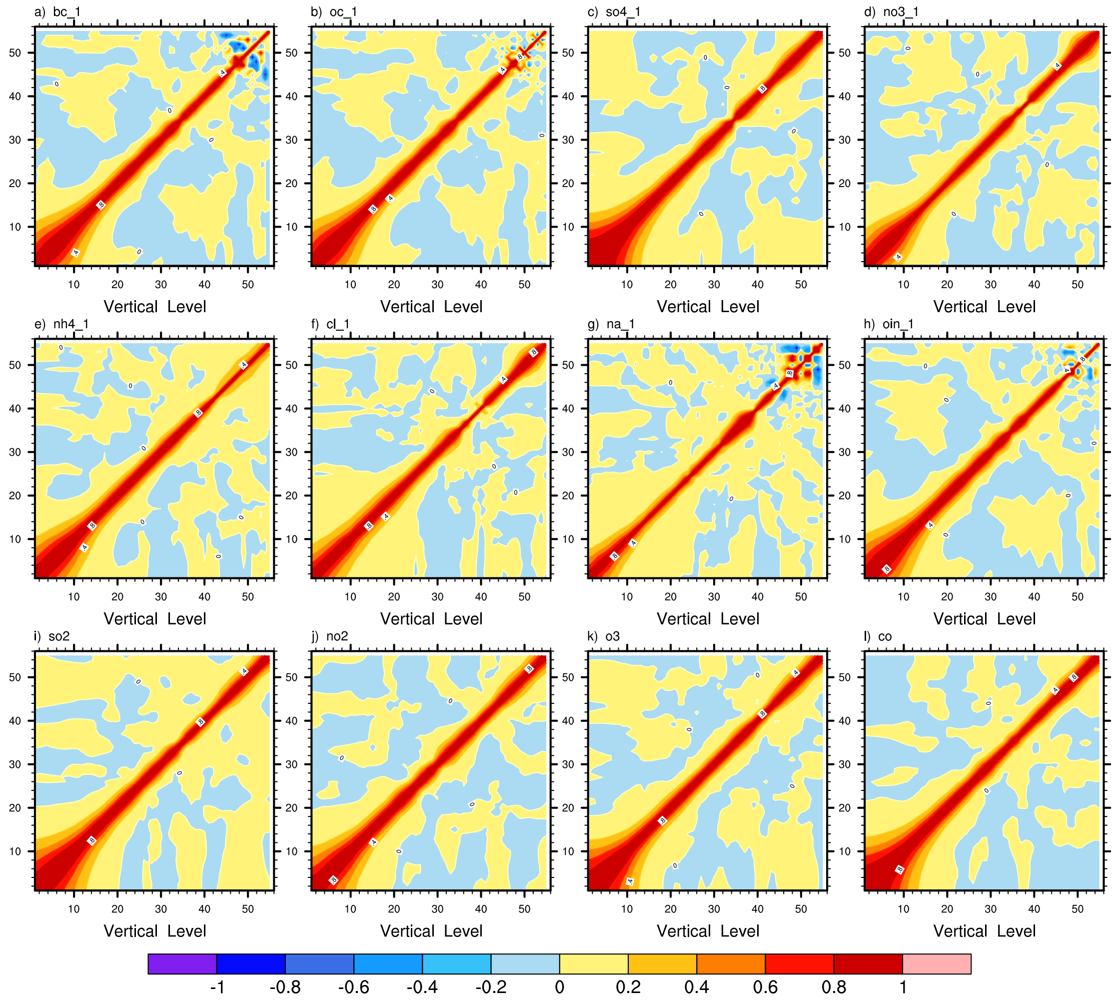

Figure R2. Background error vertical correlation of aerosol species in the (a-h) 1st size bin, and of (i-l) gas pollutants. The left x-axis and y-axis denote the model level.

Other minor issues:

1. Please present other emissions such as dust, biogenic and fire emission used in your study.

Response:

Accepted. The description of the corresponding emissions has been listed in section 2.1 in the manuscript as below.

“The dust emission is the GOCART dust emission and the biogenic emission is calculated online by the Gunther scheme within the WRF-Chem model. Given the time period of this study (January) is not the period with massive fires (crop/biomass burning), the fire emission is not used in this study.”

2. Please describe the reason why cross correlations were not applied.

Response:

The statement related to the cross-correlation issue, “Cross-correlations between different aerosol/chemical variables were not considered”, has been replaced in the manuscript as below.

“Since it is both technically and scientifically challenging to model the cross-correlations between different aerosol/chemical variables in a 3DVAR framework, they are not considered in this study. We plan to introduce the cross-variable correlations with the ensemble-variational approach in the future extension of the system.”

3. Could you describe the other trials for the background error covariance of the PM-coarse? Did the inflation factor of 90 was applied along with all vertical levels?

Response:

The inflation for the background error covariance is actually controlled by a new “var_scaling” factor similar as the original “var_scaling” for meteorology assimilation in WRFDA, and thus the inflation factor of 90 is currently applied to all the vertical levels similar as the inflation in meteorology assimilation.

Regarding the other trials for the background error covariance, the statistics of PM_{10} in the forecast are presented by Fig. R3 as below (not included in the manuscript). Since the trials are for the background error covariance of the PMcoarse, the statistics for PM2.5 and gaseous pollutants are highly similar among the trials and thus are not shown. The PM2_V1, PM2_V30, PM2_V60, and PM2_V90 are the experiments assimilating PMcoarse and $PM_{2.5}$ simultaneously but with PMcoarse inflation factor of 1, 30, 60, and 90 respectively through the setting of "var_scaling" (the PM2.5 inflation factor all kept 1); the PM1 experiment is the same one as in the manuscript that only assimilate the $PM_{2.5}$. Without the inflation, the PM2_V1 experiment are close to the PM1 experiment, which suggests that assimilating PMcoarse without inflation does not bring significant improvements to the forecast of PM_{10} as originally expected. Therefore, as stated in the manuscript, the inflation factor is used to address this issue. Viewing from Fig. R3, the forecast PM_{10} generally improves with the enlargement of the inflation factor, especially for the forecast range within 0-9 hr. This result is corresponded to the analysis of Fig. 4 in the manuscript, suggesting that it could be better to enlarge the background error covariance of the PMcoarse. Given the PM2_V90 experiment exhibits the best forecast performance, and PM2_V90 is relatively close to PM2_V60, the inflation factor of 90 is finally used in the manuscript without further enlargement.

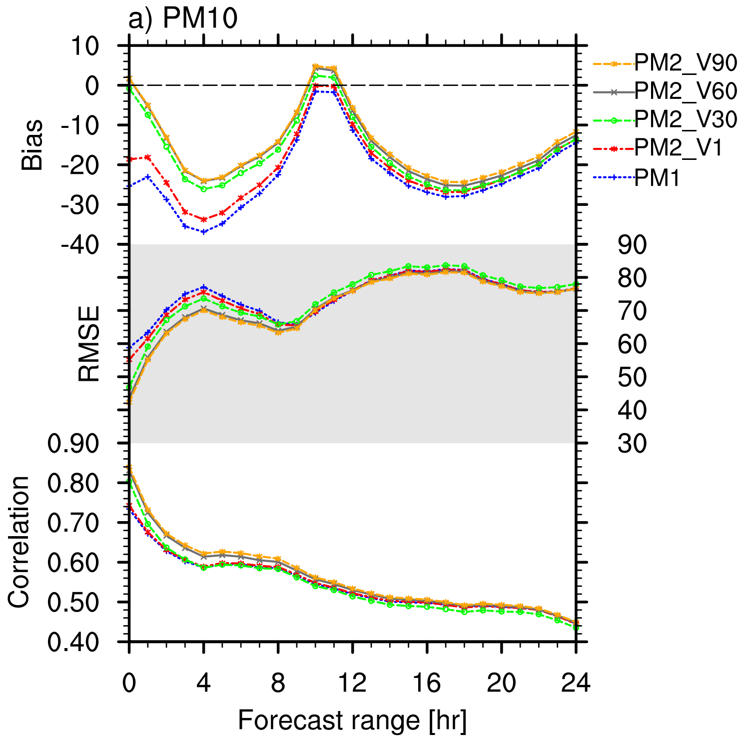

Figure R3. Averaged bias (units: μg/m^3), RMSE (units: μg/m^3), and correlation for PM_{10} in different experiments as a function of forecast range, verified against the surface observations of 531 stations in China. The blue, red, green, gray, and orange lines denote the results of experiment PM1, PM2_V1, PM2_V30, PM2_V60, and PM2_V90, respectively.

Specific minor issues:

Page 1. line 20: SO2 and CO -> SO2, and CO

Corrected.

Page 3. line 42: fastest growing -> fastest-growing

Corrected.

Page 3. line 43: extreme haze -> the extreme haze

Corrected.

Page 4. line 58: scientific community -> the scientific community

Corrected.

*Page 5. line 87: treatment -> treatment*s

Corrected.

Page 6. line 102: data assimilation -> DA

Corrected.

Page 6. line 108: recent -> recently

Corrected.

Page 6. line 113: extend -> extent or extends?

Accepted. The word “extend” has been revised to “extent” in the manuscript.

Page 6. line 116: observations and -> observations, and

Corrected.

Page 7. line 123: brief summary -> summary (tautology)

Accepted. The phrase “brief summary” has been revised to “summary” in the manuscript.

Page 8. line 151: capability -> the capability

Corrected.

Page 10. line 197: PM25 are -> PM25 is

Corrected.

Page 11. line 205: in first -> in the first

Corrected.

Page 12. line 233: each of the aerosol/chemical variable -> each of the *aerosol/chemical variables*

Corrected.

Page 13. line 253 – 256: needed to be tailored more clearly

The corresponding statement has been revised in the manuscript as below.

"In view of the cycling frequency is an important aspect in the DA strategy, especially for 3DVAR, two more experiments that assimilate all the six major pollutants with 3-h and 1-h cycling frequency are conducted respectively (experiment ALL_3h and ALL_1h)."

Page 15. Line 295: is slightly larger -> are slightly larger

Corrected.

Page 16. line 303: Due to lack -> Due to the lack

Corrected.

Page 16. line 305: As show -> As shown

Corrected.

Page 16. line 308: among -> to

Corrected.

Page 16. line 316: including bias, RMSE and correlation -> including bias, RMSE, and correlation

Corrected.

Page 16. line 318: model -> the model, poor -> poorly

Corrected.

Page 17. line 322: percentage -> percentages

Corrected.

*Page 17. line 328: lead -> lead*s

Corrected.

Page 17. line 329: individual -> the individual

Corrected.

Page 17. line 330: general -> generally

Corrected.

Page 18. line 346: applications, but also -> applications but also

Corrected.

Page 18. line 362, Page 19. Line 364, and Page 21. Line 425: heavy -> heavily

Corrected.

Page 19. line 379: ALL_3h and ALL_1h -> ALL_3h, and ALL_1h

Corrected.

*Page 20. line 397: becomes -> becom*e

Corrected.

Page 20. line 403: determine -> determines

Corrected.

Page 21. line 411: SO2 and CO -> SO2, and CO

Corrected.

Page 25. line 508: study -> a study

Corrected.

Page 25. line 509: contains -> contain

Corrected.