Response to Reviewer #2's comments:

We thank Referee # 2 for his thoughtful comments and suggestions that have helped to improve this manuscript. Our responses to comments (in bold style) and the corresponding changes to the manuscript are detailed below. In particular, we have added a simulation using the optimized emissions from 5 to 16 October 2014 according to his suggestions.

There is not much to criticize about the manuscripts as it relies on the assimilation methodology previously described by Peng et al. (2017). (1) Since the assimilation experiment was conducted over a ten-day period it is uncertain if the conclusions about different performance of forecasts for various species would hold in a general. The most interesting are results on emission factors. (2) Did you encounter negative lambdas and if so what did you do about them? (3) An ultimate test of the optimized emissions would compare a simulation using the optimized emissions with a control. (4) Would an ENFK run with concentrations as state vectors using optimized emissions be identical to the EnKF run with concentrations and emission factors as the state vectors? (5) Link http://113.108.142.147:20035/emcpublish (p. 3) would be a valuable data source on pollution over China for many users but the access requires installation of Microsoft Silverlight a software for watching videos. That seems odd and is not be allowed on government computers. Could that be ameliorated?

(1) Since the assimilation experiment was conducted over a ten-day period it is uncertain if the conclusions about different performance of forecasts for various species would hold in a general.

It is true that only a case was investigated in this work and it is uncertain if the conclusions about different performance of forecasts for various species would hold in a general. More case studies are needed to obtain general results in future works.

We have added the above paragraph in Lines 548-551, Page 19.

(2) Did you encounter negative lambdas and if so what did you do about them?

There are very few negative values for $(\kappa_{i,t})_{\text{inf}}$ after inflation (in Equation 3). A quality control procedure is performed for $(\kappa_{i,t})_{\text{inf}}$ before further appliance. All these negative data were set as 0 in this work. Then $(\kappa_{i,t})_{\text{inf}}$ were re-centered to ensure the

ensemble mean values of $(\kappa_{i,t})_{\text{inf}}$ were all 1. Besides, another quality control procedure is performed for $\lambda^{\text{a}}_{i,t}$ to keep them positive. Thus, all $\lambda^{\text{f}}_{i,t}$ and $\lambda^{\text{a}}_{i,t}$ could be positive.

We have added the above paragraph in Lines 158-163, Page 6.

(3) An ultimate test of the optimized emissions would compare a simulation using the optimized emissions with a control.

We have performed a simulation (fcEs) using the optimized emissions from 5 to 16 October 2014 to investigate the impact of optimized emissions on chemical simulations. Same as the control run, the ICs were the ensemble mean of the spin-up forecasts at 00:00 UTC on 5 October 2014. Thus the difference between the fcEs and the control run is the anthropogenic emissions. The results showed that the fcEs performed very similar to the control run in the whole in the BTH region (ReFig. 1). For $PM_{2.5}$, PM_{10} and CO, the values of the fcEs were a little smaller than those of the control run, which were consistent with the difference of the anthropogenic emissions. For SO_2 and NO_2, fcEs performed much better than the control run in most time though significant systematic overestimation still existed during the nighttime. For O_3, miner improvements were also gained due to the better simulation in fcEs for NO_2.

We have added the above paragraph in Line 443-453, Page 15. For ReFig.1, the cyan line (refer to as "fcEs") was added in Figure 4 to save space.
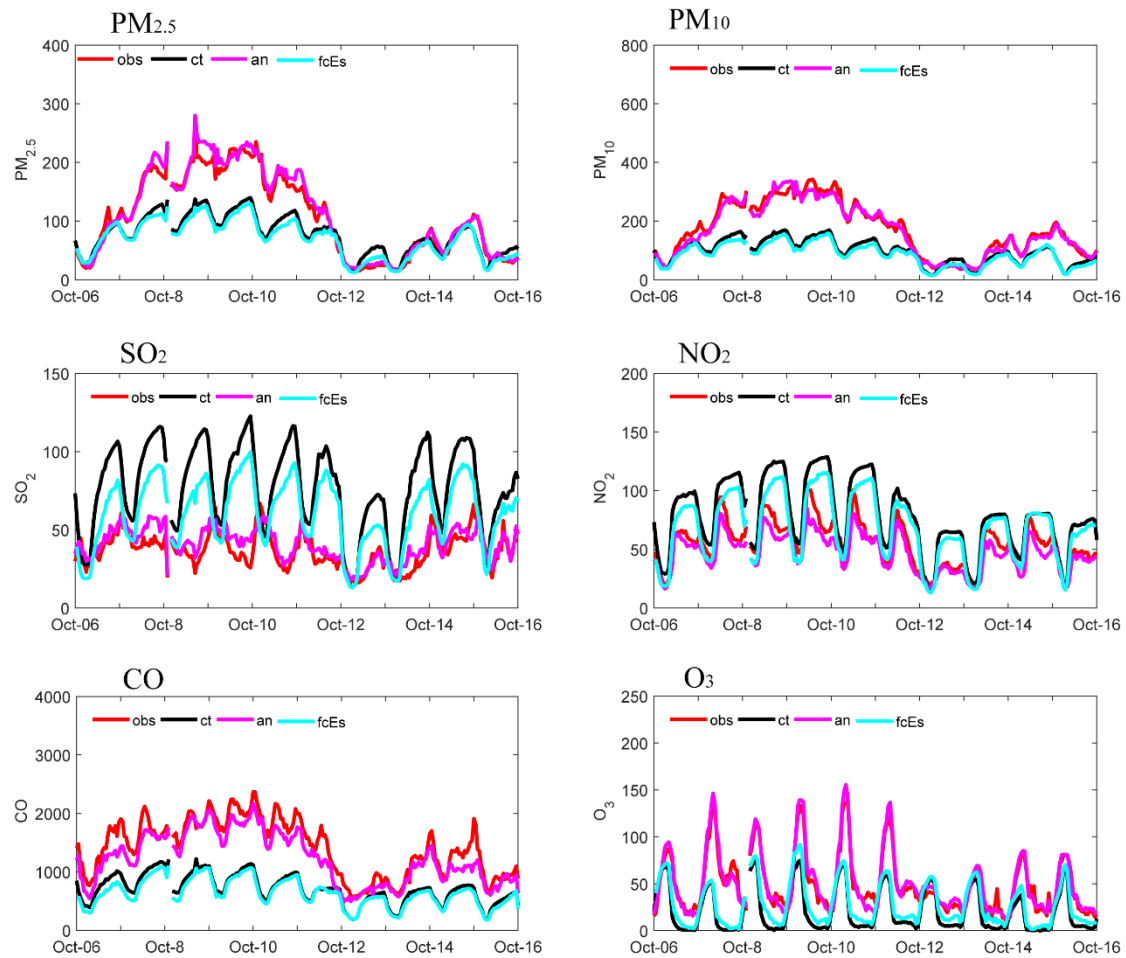
ReFig. 1. Time series of the hourly pollutant concentrations in the Beijing–Tianjin–Hebei (BTH) region obtained from observations (referred to as "obs", red line), the control run (referred to as "ct", black line), the analysis (referred to as "an", pink line), the simulation only using the optimized emissions (referred to as "fcEs", cyan line). The observations were obtained from the 47 independent sites in the BTH region. The modelled time series were interpolated to the 47 independent sites using the spatial bilinear interpolator method. Units: μg m^{-3}.

(4) Would an EnFK run with concentrations as state vectors using optimized emissions be identical to the EnKF run with concentrations and emission factors as the state vectors?

The optimized emissions are only the results of a mathematical optimum by utilizing observations. They are influenced greatly by model errors and observation errors. If the optimized emissions used in the EnFK experiment run with pure

concentrations as state vectors are identical to the emissions assimilated in the joint EnFK experiment run with concentrations and emission factors (representing emissions) as state vectors, identical results may be obtained.

We have added the above paragraph in Line 116-121, Page 4-5.

(5) Link http://113.108.142.147:20035/emcpublish (p. 3) would be a valuable data source on pollution over China for many users but the access requires installation of Microsoft Silverlight a software for watching videos. That seems odd and is not be allowed on government computers. Could that be ameliorated?

Yes, we agree with the reviewer that the requirement of installation of Microsoft Silverlight software to view the data is odd. There is another website for the data: http://www.resdc.cn/data.aspx?dataid=186. The data can be downloaded by request. If you are interested in the data, please contact the data manager of the website.