Response to Reviewer #2

We thank reviewer for his thoughtful comments and suggestions that have helped to significantly improve this manuscript.

The reviewer's comments are shown in *blue italics* with the author responses in black.

Summary

This manuscript proposes an ensemble Kalman smoother to constrain the PM 2.5 emissions by incorporating the information of PM 2.5 observations. Results based on 5-year cycling assimilation provide quantitively estimates for annual and monthly variations of the PM 2.5 emission. By assimilating the observations with the ensemble Kalman smoother, the influences of COVID are clearly displayed. Moreover, diurnal variations of the PM 2.5 emission for each month are provided, which can be a valuable contribution to the PM 2.5 forecast. The manuscript proposed an advanced data assimilation method to update the PM 2.5 emissions by both present and future PM 2.5 observations. Overall it is well written and presented. It could be very beneficial to the community of chemistry data assimilation. I have several minor comments below.

1. An EnKS is proposed to update the emission along with the concentration. Are both the emission and the concentration updated by future observations?

Thank the reviewer for the valuable comments. The emissions are updated by current and future observations. But the concentrations are updated by current observations.

This discussion is added in the text in Lines 93-95, Page 4.

2. The lagged length for EnKS is an important factor because it determines how many future observations are applied to constrain the current state. The lagged length K is set to 6 in this study. How this parameter is determined?

Thank the reviewer for the valuable comments. The larger the K value, the more future observations are assimilated to constrain the current emission estimate. But the sample estimated temporal correlations could be contaminated by sampling errors and model errors, especially with increased lagged times. Thus, there is a tradeoff between the amount of future observations and accuracy of sample estimated temporal correlations. The choice of K (=6) is determined by sensitivity experiments. This discussion is added in the text in Lines 190-194, Pages 7-8

3. It is interesting to see the quick influences of COVID on PM2.5 (Figure 11). Can such a DA system be practical for real-time operations?

Thank the reviewer for the valuable comments. This DA system can be practical for real time operations.