Interactive comment on "Significant wintertime $PM_{2.5}$ mitigation in the Yangtze River Delta, China from 2016 to 2019: observational constraints on anthropogenic emission controls" by Liqiang Wang et al.

Anonymous Referee #2

General comments:

This paper uses a data assimilation method to constrain the modelled $PM_{2.5}$ concentrations over the Yangtze River Delta (YRD) region and distinguish the impact on $PM_{2.5}$ from meteorology and emission variations. The results show that the emission reduction measures in G20 summit and long-term emission control strategies in YRD successfully curb the $PM_{2.5}$ levels both locally and regionally. This paper is good in general and within the scope of Atmospheric Chemistry and Physics. I recommend for publication once the specific comments expressed below are addressed.

Response: We thank the reviewer for the thoughtful comments on our paper and have addressed these specific comments as below.

Specific comments:

1. The author should provide more details regarding how to conduct data assimilation. First, the author needs to perform a sensitivity analysis in order to proof that choosing the fan-shaped quadrilateral (Figure 1a) minimizes the impact from outside on the YRD region. Second, how is the modelled $PM_{2.5}$ constrained spatiotemporally by observations, applying DA generated scaling factors to the whole fan-shaped quadrilateral region, the YRD region, city by city, or grid by grid, and hour by hour or day by day?

Response: Thanks. We have supplemented the additional discussions in Sect. 2.3 to explain why we choose the ground-level observations within the fan-shaped quadrilateral to constrain the model performance. As pointed by the reviewer, we aim to minimize the impacts outside the YRD region. Specifically, this was mainly due to the fact that this fan-shaped geographical scope covered almost all key regions that had potentially regional impacts on the YRD, involving the Beijing-Tianjin-Hebei region (BTH), the Pearl River Delta region, the Sichuan-Chongqing region, and the Shaanxi-Gansu region (Zhang et al., 2019). On the other hand, the ground monitoring sites within the fan-shaped quadrilateral were significantly denser than those outside, thus leading to much more effective DA in practice (Bocquet et al., 2015; Chai et al., 2017). Therefore, to assimilate the observations within the fan-shaped quadrilateral might be a sensible way to balance the DA effectiveness and computing efficiency. A resultant evidence lies in the model performance evaluation in Sect. 3.1, which would prove that this DA configuration can enable reliable $PM_{2.5}$ simulations. Collectively, we might eliminate the need of the associated sensitivity analysis.

In addition, we have supplemented the more discussions in Sect. 2.4 to further detail how to conduct observational constraints on the model simulations spatiotemporally. In short, we conducted hourly DA for grid cells. Note that the effective radius of each individual observation should be calculated in advance. When ground-level $PM_{2.5}$ measurements were assimilated, hourly observations were put into equation (1) to construct the new analysis fields. All-day state variables associated with aerosols in the model were adjusted from their background (simulated) to their analysis (constrained) states using the scaling factors ($\mathbf{X}^{\boldsymbol{a}}/\mathbf{X}^{\boldsymbol{b}}$). The adjusted model state variables were then used to initiate the model to predict the next background state ($\mathbf{X}^{b}$) in Equation (1). Therefore, the background state ($\mathbf{X}^{b}$) served as a prior model prediction before it was combined with the newly available observation ($\mathbf{Y}$) to generate a new analysis state ($\mathbf{X}^{a}$) using Equation (1).

Measurements within the background-error correlation length scale were used to shape analysis states ($\mathbf{X}^{a}$). The background error covariance $\mathbf{COV_{ij}}$ between any two grid cells $\mathbf{i}$ and $\mathbf{j}$ was simulated as

$$\mathbf{COV_{ij}} = \boldsymbol{\varepsilon}_\mathbf{i}\boldsymbol{\varepsilon}_\mathbf{j}\mathbf{e}^{-\frac{\boldsymbol{\Delta}_\mathbf{ij}}{\mathbf{L}}} \quad (2)$$

where $\boldsymbol{\varepsilon}_\mathbf{i}$ and $\boldsymbol{\varepsilon}_\mathbf{j}$ referred to the standard deviations of the background errors in two grid cells and $\boldsymbol{\Delta}_\mathbf{ij}$ denoted the distance between the two grids. As a result, $\mathbf{L}$ was the background-error correlation length scale, which can be the Hollingsworth-Lönnberg method (Chai et al., 2017; Hollingsworth and Lönnberg, 1986; Kumar et al., 2012). Figure 2 shows the correlation coefficient, i.e., $\mathbf{COV_{ij}}/\boldsymbol{\varepsilon}_\mathbf{i}\boldsymbol{\varepsilon}_\mathbf{j}$, as a function of the separation distance between two grid cells, which was averaged over 10 km bins. The results indicate that a correlation length scale of ~ 180 km could be treated as the threshold by allowing the correlation coefficients to fall within the range of $\mathbf{e}^{-\mathbf{1}}$, defining the effective radius of each individual observation. Due to the intensive monitoring sites in our study domain, this threshold was applied uniformly for the YRD. In this study, observations beyond the background-error correlation length scale would have no effect on $\mathbf{X}^{\boldsymbol{a}}$.

Added/rewritten part in Sect. 2.3: As shown in Figure 1a, to consider regional impacts outside the YRD, the ground-level observations in the fan-shaped quadrilateral were used to constrain the model performance. This was mainly due to the fact that this fan-shaped geographical scope covered almost all key regions that had potentially regional impacts on the YRD, involving the Beijing-Tianjin-Hebei region (BTH), the Pearl River Delta region, the Sichuan-Chongqing region, and the Shaanxi-Gansu region (Zhang et al., 2019). On the other hand, the ground monitoring sites within the fan-shaped quadrilateral were significantly denser than those outside, thus leading to much more effective DA results in practice (Bocquet et al., 2015; Chai et al., 2017). Collectively, to assimilate the observations in the fan-shaped quadrilateral might be a sensible way to balance the DA effectiveness and the computing efficiency. A resultant evidence lies in the model performance evaluation in Sect. 3.1, which would prove that this DA configuration can enable reliable $PM_{2.5}$ simulations.

Added/rewritten part in Sect. 2.4: When ground-level $PM_{2.5}$ measurements were assimilated, hourly observations were put into equation (1) to construct the new analysis fields. All-day state variables associated with aerosols in the model were adjusted from their background (simulated) to their analysis (constrained) states using the scaling factors ($\mathbf{X}^{\boldsymbol{a}}/\mathbf{X}^{\boldsymbol{b}}$). The adjusted model state variables were then used to initiate the model to predict the next background state ($\mathbf{X}^b$) in equation (1). Therefore, the background state ($\mathbf{X}^b$) served as a prior model prediction before it was combined with the newly available observation ($\mathbf{Y}$) to generate a new analysis state ($\mathbf{X}^a$) using Equation (1).

Measurements within the background-error correlation length scale were used to shape analysis states ($\mathbf{X}^a$). The background error covariance $\mathbf{COV_{ij}}$ between any two grid cells $\mathbf{i}$ and $\mathbf{j}$ was simulated as

$$\mathbf{COV_{ij}} = \boldsymbol{\varepsilon}_\mathbf{i}\boldsymbol{\varepsilon}_\mathbf{j}\mathbf{e}^{-\frac{\boldsymbol{\Delta}_\mathbf{ij}}{\mathbf{L}}} \quad (2)$$

where $\boldsymbol{\varepsilon}_\mathbf{i}$ and $\boldsymbol{\varepsilon}_\mathbf{j}$ referred to the standard deviations of the background errors in two grid cells and $\boldsymbol{\Delta}_\mathbf{ij}$ denoted the distance between the two grids. As a result, $\mathbf{L}$ was the background-error correlation length scale, which can be obtained by the Hollingsworth-Lönnberg method (Chai et al., 2017; Hollingsworth and Lönnberg, 1986; Kumar et al., 2012). Figure 2 shows the correlation coefficient, i.e., $\mathbf{COV_{ij}}/\boldsymbol{\varepsilon}_\mathbf{i}\boldsymbol{\varepsilon}_\mathbf{j}$, as a function of the separation distance between two grid cells, which was averaged over 10 km bins. The results indicated that a correlation length scale of ~ 180 km could be treated as the threshold allowing the correlation coefficients to fall within the range of $\mathbf{e^{-1}}$, defining the effective radius of each individual observation. Due to the intensive monitoring sites in our study domain, this threshold was applied uniformly for the YRD. In this study, observations beyond the background-error correlation length scale were assumed to have no effect on $\mathbf{X}^{\boldsymbol{a}}$.

2. The author used a statistical method to establish the correlation coefficients and chose separation distance of 180 km as a threshold. The author needs to give more explanations on the value of chosen. If the purpose is to find a correlation length scale to minimize the effect on $\mathbf{X}^{\boldsymbol{a}}$, based on Fig 2, it seems that separation distance of 600 km would be more appropriate.

Response: Thanks. The objective of identifying the background-error correlation length scale is to define the effective radius of each individual observation and thus to establish reliable analysis states ($\mathbf{X}^a$). Here the Hollingsworth-Lönnberg approach, wildly used for decades (Chai et al., 2017; Hollingsworth and Lönnberg, 1986; Kumar et al., 2012), is applied to calculate the background-error correlation length scale. Observations beyond the background-error correlation length scale were assumed to have no effect on $\mathbf{X}^{\boldsymbol{a}}$. Once observations far away are introduced, more background errors $\mathbf{COV_{ij}}$, larger than $\mathbf{e^{-1}}$, would be put into $\mathbf{X}^{\boldsymbol{a}}$ as calculated in Equation (2). Corresponding detailed information has been given in the response for the specific comment (2).

3. How did the author isolate the impact from emission reductions on $PM_{2.5}$ concentrations? Did the author use the constrained $PM_{2.5}$ subtract the impact on simulated $PM_{2.5}$ from meteorological variations? Even the modelled temperature, humidity, wind speed, and air pressure were also assimilated in this study, there are other parameters, for example, modelled PBL height, causing large uncertainties in the modelled meteorological field, and thus leading to bias and error in the calculated net impacts from emission variations. For example, figures c and f in Fig 5, show very small impact of anthropogenic emission control from 2016 to 2019 in most of Zhejiang province compared to the other provinces in the YRD region. Is it reasonable?

Response: Thanks. Yes, it is reasonable. We isolated anthropogenic impacts on $PM_{2.5}$ concentrations by subtracting the corresponding meteorological impacts from the constrained $PM_{2.5}$ fields. To further illustrate the process of meteorological assimilations, we have supplemented the additional discussions in Sect. 2.4. The ECMWF reanalysis datasets accounted for the hourly observational constraints on spatiotemporal meteorological evolutions. Therein almost all necessary meteorological factors (nine variables), involving temperature, U wind component, V wind component, pressure, relative humidity, precipitation, short-wave radiation, cloud cover, and boundary layer height, were assimilated (https://apps.ecmwf.int/datasets/data/interim-full-daily/levtype=sfc/, last access: 7 March 2020).

The model evaluation provides a more direct way to verify the corresponding model performance. As highlighted in Sect. 3.1, given the fact that the assimilated ERA reanalysis dataset has much wider spatial coverage than ground-based measurements, we also reproduced the spatiotemporal variations in the meteorological factors (e.g., temperature, relative humidity, wind speed, and air pressure) (Figures S5 ~ S8). Together with the comprehensive evaluation statistics as summarized in Tables S1 ~ S5, it has been demonstrated that the DA method can enable one to derive not only reliable $PM_{2.5}$ evolution but also accurate meteorological fields.

In terms of the issue associated with Zhejiang, we have supplemented the additional interpretations in Sect. 3.2. The impacts of anthropogenic drivers on $PM_{2.5}$ concentrations in the southern and eastern parts of Zhejiang were evidently weaker than those in other regions in the YRD. This divergence can mostly be explained by spatial distributions of anthropogenic emissions. Anthropogenic emissions in the southern and eastern parts of Zhejiang were also significantly less than those in other regions (Figure S9), thus leading to substantially low $PM_{2.5}$ concentrations (Figure 3). Besides, meteorological fields in the coastal regions, more conducive to $PM_{2.5}$ diffusions (Figure 5), might be another cause.

Added/rewritten part in Sect. 2.4: For all experiments, the prior anthropogenic emissions were kept consistent (i.e., MEIC), while the ECMWF reanalysis datasets accounted for the hourly observational constraints on spatiotemporal meteorological evolutions. The ECMWF reanalysis datasets accounted for the hourly observational constraints on spatiotemporal meteorological evolutions. Therein almost all necessary meteorological factors (nine variables), involving temperature, U wind component, V wind component, pressure, relative humidity, precipitation, short-wave radiation, cloud cover, and planetary boundary layer height (PBLH), were assimilated (https://apps.ecmwf.int/datasets/data/interim-full-daily/levtype=sfc/, last access: 7 March 2020).

Added/rewritten part in Sect. 3.1: In addition, given the fact that the assimilated ERA reanalysis dataset has much wider spatial coverage than ground-based measurements, we also reproduced the spatiotemporal variations in the meteorological factors (e.g., temperature, relative humidity, wind speed, and air pressure) (Figures S5 ~ S8). With the comprehensive evaluation statistics as summarized in Tables S1 ~ S5, it has been demonstrated that the DA method can enable one to derive not only reliable $PM_{2.5}$ evolutions but also accurate meteorological fields.

Added/rewritten part in Sect. 3.2: We recognized that the impacts of anthropogenic drivers on $PM_{2.5}$ concentrations in the southern and eastern parts of Zhejiang were evidently weaker than those in other regions in the YRD. This divergence can mostly be explained by spatial distributions of anthropogenic emissions. Anthropogenic emissions in the southern and eastern of Zhejiang were also significantly less than those in other regions (Figure S9), thus leading to substantially low $PM_{2.5}$ concentrations (Figure 3). Besides, meteorological fields in the coastal regions, more conducive to $PM_{2.5}$ diffusions (Figure 5), might be another cause.

4. How did the author consider the regional transport of $PM_{2.5}$ in this study? The regional emission control effect on $PM_{2.5}$ may have influence on calculated net impact of emission reduction in each city and the localized mitigation potential.

Response: Thanks. We agree with the reviewer that regional transport of $PM_{2.5}$ is central to our results and thus have considered it carefully. Using observational constraints on the state-of-the-art model, we have reproduced spatiotemporal variations in both $PM_{2.5}$ and meteorological factors, as illustrated in Sect. 3.1, and thus derived the reliable estimations of regional transport of $PM_{2.5}$. Hence, we have supplemented a sentence in Sect. 3.1 to highlight this point.

Considering the main objective of this work, we have not conducted source apportionments to predict the impacts of regional transport of $PM_{2.5}$. In theory, regional transport of $PM_{2.5}$ can be attributable to both anthropogenic and meteorological drivers. In turn, we provide paired experiment designs to isolate anthropogenic impacts by subtracting meteorological perturbations (i.e., the differences in simulated $PM_{2.5}$ concentrations between NO_2016 and NO_2019 and between DA_CON_G20 and NO_CON_G20) from the constrained $PM_{2.5}$ fields (i.e., DA_2016 and DA_2019 / DA_G20).

Added/rewritten part in Sect. 3.1: Regional transport of $PM_{2.5}$ can thus be captured reasonably in this way.

References:

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