Response to the Comments of Referees

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Title: Four-dimensional Variational Assimilation for SO_2 Emission and its Application around the COVID-19 lockdown in the spring 2020 over China

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We thank the reviewers and editors for providing insightful and constructive comments, which help improve the presentation of the manuscript. We have revised the manuscript according to the comments and suggestions of the referees.

The referee's comments are reproduced (black) along with our replies (blue). All the authors have read the revised manuscript and agreed with submission in its revised form.

< Anonymous Referee #2 >

Overall comments

This manuscript describes a four-dimensional variational (4DVAR) data assimilation (DA) system designed to update SO_2 emissions by assimilating surface SO_2 concentrations. The system was applied to study emissions reductions over China during the beginning of the COVID-19 pandemic. Additionally, the authors showed how obtaining SO_2 emissions from 4DVAR can potentially lead to better subsequent SO_2 concentration forecasts relative to using static SO_2 emissions.

Response： We thank the referee for the positive assessment of our manuscript. The manuscript has been carefully revised according to the referee's comments and suggestions.

I think the topics in this manuscript are interesting and important, and I have few concerns with the overall objectives of this work. However, I think there are shortcomings regarding descriptions of the DA system and the experiments, most of

which are related to lack of clarity. In addition, I don't think the OSSE part of the manuscript is necessary, and I instead suggest replacing this material with other metrics demonstrating that the 4DVAR DA system works as expected. Finally, the writing is mostly understandable, but there are many grammatical errors that should be fixed prior to publication in ACP.

Response: Thank you for your comments and suggestions. Following your advice, we have made systematic revisions on description of the DA algorithm, and the OSSE part has been removed. The 4DVAR system's efficacy due to the assimilation of real observations has been discussed to show the effectiveness of the DA system in emission optimization.

We have carefully corrected grammatical errors in the revised manuscript.

Below are our point-to-point response to your comments in detail.

Major comments

Commemt1: I think more descriptions about the mechanisms through which your 4DVAR DA system updates SO_2 emissions are needed. It isn't entirely obvious how this adjustment is achieved using 4DVAR. Did you somehow model cross-covariances between SO_2 concentrations and emissions? Or is the tangent-linear/adjoint model effectively altering the emissions? I was left unsure of exactly how you updated SO_2 emissions, and I think this key point of your method needs to be explicitly described. It might also be worth contrasting your method with how an EnKF works; in the EnKF it would be straightforward to update SO_2 emissions, as SO_2 emissions could simply be added to the state vector and be naturally updated by ensemble-based covariances.

Response 1: Thanks for your questions and suggestions. The cross-covariances is used for the EnKF method to update SO_2 emissions by assimilating SO_2 concentrations. But for the 4DVAR method, it is not necessary to use the cross-covariances between SO_2 concentrations and emissions. In the 4DVAR system, the SO_2 emission is the state vector and can be directly updated by assimilating SO_2 concentration

observations.

In our 4DVAR system, the cost function is as follows:

$$J = \frac{1}{2}\left(c_0 - c_0^b\right)^T \mathbf{B}_{\boldsymbol{c}}^{-1}\left(c_0 - c_0^b\right) + \frac{1}{2}\sum_{i=0}^{n-1}\left(e_i - e_i^b\right)^T \mathbf{B}_{\boldsymbol{e_i}}^{-1}\left(e_i - e_i^b\right) + \frac{1}{2}\sum_{i=0}^{n}\left(y_i^o - \boldsymbol{H_i}c_i\right)^T \mathbf{R}_{\boldsymbol{i}}^{-1}\left(y_i^o - \boldsymbol{H_i}c_i\right) \quad (1)$$

where c_0 is the state variable that denotes the initial concentration vector, and e_i is also the state variable that denotes the emission at each time step. The c_0 and the e_i are assumed independent. Subscripts of the variables represent the time step. The c_0 at the initial time and the e_i at the time steps from 0 to $\mathrm{n}-1$ are directly updated, when the optimal J is obtained.

The SO_2 concentrations and the SO_2 emissions are connected by the differential equation of Eq. (4) in manuscript. The c_i depend on the emission e_{i-1}. When we minimize the cost function of Eq. (1), the SO_2 emission is used in the calculation of the observation term of $\frac{1}{2}\sum_{i=0}^{n}(y_i^o - H_i c_i)^T R_i^{-1}(y_i^o - H_i c_i)$. Thus, the SO_2 emission can be updated by the observation of SO_2 concentration. Although both EnKF and 4DVAR methods are based on a model to show the relationship between concentration and emission, different ways are used in two methods. For the EnKF method, the cross-covariances between concentrations and emissions are generated by an ensemble of model outputs. But the 4DVAR method depends on the dynamical equations of model to establish the relationship between concentration and emission.

This statement has been added in the revised manuscript.

Commemt2a: I believe that when harvesting synthetic observations from "truth" in an OSSE it is customary to add noise drawn from an "observation error" distribution to the synthetic observations. However, your methods don't mention doing this, which makes me wonder if the OSSE was properly performed. By not adding noise to harvested observations, you will likely get analysis fits much closer to "truth" than if you add noise to the synthetic observations you eventually assimilate.

Response 2a: In the OSSE, the "real" emissions (EM_real) included 273 (13 × 21 gridded) sources, and the value of emissions were random from 10 to 130 mol km^{-2} h^{-1}. The mean value of EM_real was 50 mol km^{-2} h^{-1}. The background emission (EM_back) also included 273 sources having the same spatial distribution as EM_real. The value of EM_back conformed to a Gaussian distribution with a mean value of 50 mol km^{-2} h^{-1} and a variance of 10 mol km^{-2} h^{-1}. That is to say, a random perturbation, which conformed to a Gaussian distribution, was added to a mean value of 50 mol km^{-2} h^{-1}. The correlation coefficient between EM_real and EM_back was 0.01, suggesting two emissions were unrelated.

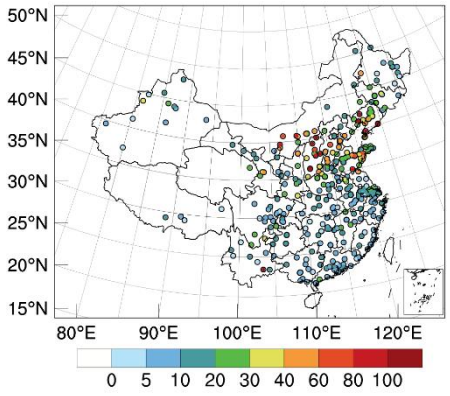
Commemt2b: Regardless, I don't feel the material about the OSSE adds much to the manuscript, and it is little more than a "sanity check" that I didn't find very convincing. I think a better demonstration of the 4DVAR system's efficacy would be to show plots of: 1) The cost function reduction from a 4DVAR analysis that assimilated real observations; 2) Analysis increments directly showing how SO_2 emissions were modified by assimilating SO_2 observations; and 3) "observation minus background" and "observation minus analysis" statistics. I think presenting these types of plots would increase confidence that your 4DVAR DA system works as expected and can provide stronger evidence of system robustness than the OSSE.

Response 2b: Thank you for your suggestion. The OSSE was deleted in the revised manuscript. As your advice, the performance of the 4DVAR system at 0000 UTC 17 January 2019 has been added in the revised manuscript to estimate the 4DVAR system's efficacy that assimilated real observations.

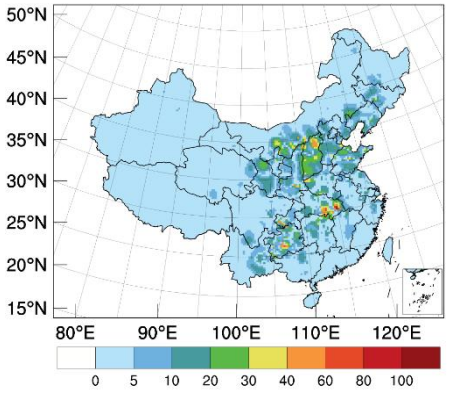
Figure R1 shows the result of Emi_2019 (Table 2 in manuscript) for the first day of 17 January 2019 to test the effect of the 4DVAR. The experiment employed MEIC_2016 as the background emission, and assimilated the hourly surface SO_2 observations during 0000–0600 UTC. In Fig. R1a, the observed heavy polluted areas with SO_2 concentrations exceeding 80 μg m^{-3} are most located in North China Plain and Northeast China, and the observed light polluted areas with SO_2 concentrations

below 40μg m^{-3} are most located in Southern China. Compared with the observed concentration, the background concentrations (Fig. R1b) underestimated in North China Plain, Northeast China, but overestimated in Central China and Sichuan Basin. Figure R1c shows the increment field of SO_2 concentrations that is the difference of analyzed field minus background field. There are positive values in most of Northern China and negative values in Central China and Sichuan Basin, suggesting that the optimized IC is more consistent with the observed SO_2 concentrations. The evaluation of the optimized IC and background concentrations are shown in Fig. R1d. Compared with the background field, the bias in analysis field improved from -2.8 to 1.8 μg m^{-3}, the RMSE decreased from 23.1 to 11.8 μg m^{-3} and the correlation coefficient (CORR) of analysis field increased from 0.2 to 0.8. The result indicates that there is an improvement in the accuracy of the SO_2 concentration of IC by using 4DVAR method. The forecast accuracy with optimized IC can be improved (Peng et al., 2017, 2018), but the most important influencing factor for forecast accuracy is the emissions. The emissions and concentration IC can be optimized simultaneously by EMI_2019 experiment using our 4DVAR system.

(a) Observations



(b) background concentrations



(c) the increment of SO_2 concentrations

(d) scatter plots

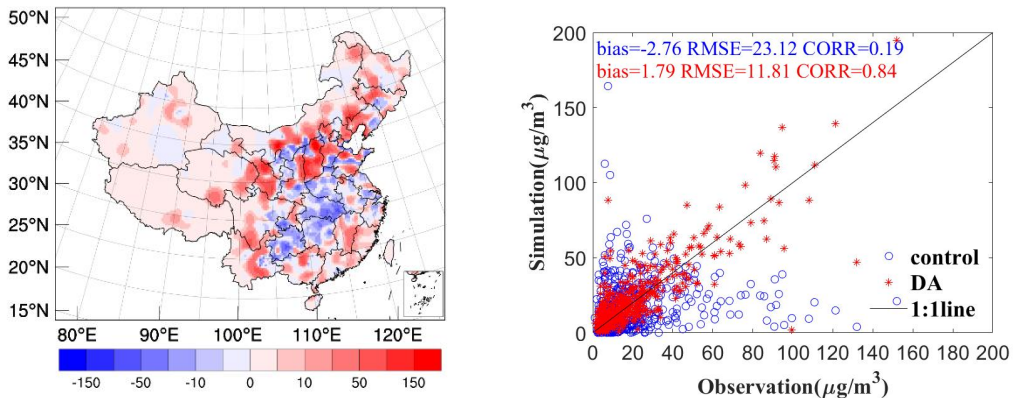


Figure R1: The simulated and observed SO_2 concentrations at 0000 UTC 17 January 2019. (a) Observations, (b) background concentrations, (c) the increment of SO_2 concentrations, and (d) scatter plots. Units: μg m^{-3}.

Figure R2a presents the background emission of MEIC 2016 at 0000 UTC. The heavy emission is found in North China Plain and Central China, Yangtze River Delta and Pearl River Delta. The largest emission values in these areas exceed 80 mol km^{-2} h^{-1}. But the emissions in Northeast China are relatively low and generally less than 40 mol km^{-2} h^{-1}. According to Fig. R1a and R1b, MEIC_2016 underestimated in most of Northern China and overestimated in Central China and Sichuan Basin. Fig. R2b shows the increment of SO_2 emissions at 00UTC 17 January 2019 by using the 4DVAR system. There are positive increment in North China Plain, Northeast China and negative increment in Central China and Sichuan Basin. Obviously, the distribution of the increment of SO_2 emissions is consistent with that of the increment of SO_2 concentration (Fig. R1c). There is a reasonable relationship between the two increments, since the underestimated/overestimated emission may result in underestimated/overestimated concentration for the simulation of SO_2.

(a) background emission (b) The increment of emission

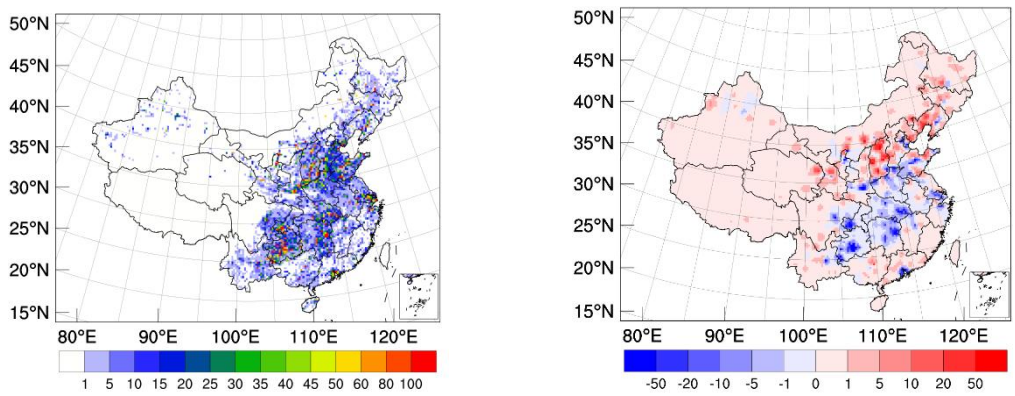


Figure R2: (a) The SO_2 background emissions and (b) SO_2 emissions increment at 0000 UTC 17 January 2019. Units: mol km^{-2} h^{-1}.

Figure R3 shows the size of the cost function for each inner iteration during the DA process of 0000–0600 UTC 17 January 2019. In this example, the maximum number of iterations was ten considering the balance between calculation time and result accuracy. It shows that the cost function quickly converges with an increase in the number of iterations. After eight iterations, the cost function was stable and close to minimum. The J at the end iteration was 12% at the first iteration.

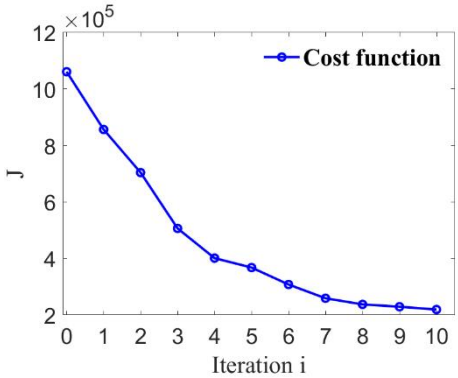


Figure R3: Cost function for each inner iteration during the DA process of 0000–0600 UTC 17 January 2019.

This statement has been added in the revised manuscript.

Commemt2c: Additionally, I'm somewhat concerned with Fig. 5b, which shows that the cost function increased between the 8th and 9th iterations. Variational cost

functions must monotonically decrease with each inner-loop iteration, so there is a chance that something went wrong. Please look into this or offer an explanation.

Response 2c: In our 4DVAR system, there are inner and outer loops. In an outer loop, there are ten (in our system) inner loops. During the run of each outer loop, ten cost functions are outputted to represent the minimum in a lower spatial resolution. For the different outer loop, the nonlinear trajectories and innovations are updated at high resolution, and the cost function are also updated. And the cost function after outer loop monotonically decrease. Note the outer loop is to provide some nonlinear information back into the minimization scheme, especially if the increment has move towards the limit of the viability of the tangent linear approximation about the current nonlinear trajectory.

The cost function in Fig. 5b (in the origin manuscript) was an attempt of the cost function in the inner loops, not the last cost function in this step (outer loop). The output of the real cost function was corrected in the 4DVAR system. Fig R4 shows the cost function of the OSSE. The cost functions decreased with each inner-loop iteration. After eight outer loop iterations, the cost function was stable and close to minimum.

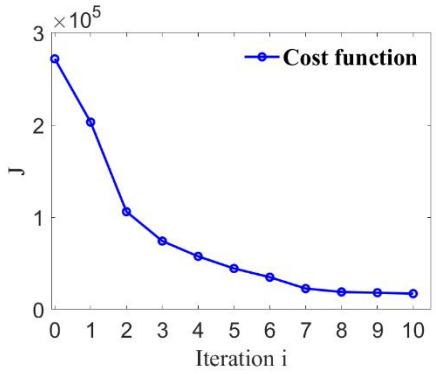


Figure R4: Cost function for each inner iteration in the OSSE.

Comment3a: I think section 2.4 needs to be more specific about what you actually did, rather than making fairly general statements. What specific observation error did you use? What were your values of ε_r and ε_o and how did you arrive at these values?

Response 3a: The observation errors include the measurement error and the representative error. The observation error of SO_2 concentration ε_{SO_2} is defined as below:

$$\varepsilon_{SO_2} = \sqrt{\varepsilon_r{}^2 + \varepsilon_o{}^2} \quad (2)$$

where ε_o is the measurement error, and ε_r is the representative error. The measurement error ε_o is the systematic error generated during monitoring by the instrument at each environmental monitoring station. The representative error ε_r represents the weight of observed data in the data assimilation system. Thus, the measurement error ε_o of SO_2 observation in this study is defined as $\varepsilon_o = 1.0$ μg m^{-3} following the result of Chen et al. (2019).

The representative error ε_r is caused by converting the model variable to the observation variable (Schwart er al., 2012) and can be expressed as:

$$\varepsilon_r = \gamma\varepsilon_o\sqrt{\frac{dx}{L}} \quad (3)$$

where γ is an adjustable parameter scaling ε_o. $\gamma = 0.5$ was used, which is same as Dai et al., (2021). dx is the grid spacing (27km in this study) and L is the radius of influence of an observation, which was taken to be 2km following Chen et al. (2019). Then, $\varepsilon_r = 1.8$ μg m^{-3} calculated from Eq. (3).

This statement had been added in the revised manuscript.

Comment3b: Similarly, in lines 201-204, please state how you produced these 48- and 24-h forecasts. What model configuration did you use? You should also cite the "NMC method" (Parrish and Derber 1992) for this approach. Moreover, in line 202, should "state variables" be "background errors"? How did you model the correlations in *C*, especially for the emissions? Overall, please be more specific about your background and observation error covariance construction.

Response 3b: Thank you for your suggestion. In Eq. (1), $\mathbf{B}_{e_i}$ is the background error covariance (BEC) of SO_2 emission that was estimated from the background emission. $\mathbf{B}_c$ is the background error covariance (BEC) of SO_2 concentration that was estimated

by national meteorological center (NMC) method. The details of estimate $\mathbf{B}_{e_t}$ and $\mathbf{B}_c$ had been added in the revised manuscript as followed.

The BEC is too large to be handled numerically, we thus followed the method used by Li et al. (2013) and Zang et al. (2016) to simplify $\mathbf{B}$

$$\mathbf{B} = \mathbf{DCD}^{\mathrm{T}} \tag{4}$$

where $\mathbf{D}$ is the RMSE matrix and $\mathbf{C}$ is the correlation matrix.

$\mathbf{C}$ can be simplified by the Cholesky factorization and Kronecker product method (Li et al., 2013) as:

$$\mathbf{C}^{\frac{1}{2}} = \mathbf{C}_x^{\frac{1}{2}} \otimes \mathbf{C}_y^{\frac{1}{2}} \otimes \mathbf{C}_z^{\frac{1}{2}} \tag{5}$$

The NMC method (Parrish and Derber, 1992) was used to estimate the BEC of SO_2 concentrations. The differences between 48 h and 24 h forecasts were generated from 17 January 2020 to 18 February 2020. The first initial chemical field at 0000 UTC on 17 January 2020 was obtained from a 10 d forecast to clear away the effect of spin-up. The subsequent initial chemical fields were derived from the former forecast one day prior. The horizontal length scale was used to determine the magnitude of SO_2 variance in the horizontal direction. This scale can be estimated by the curve of the horizontal correlation with distances, and the horizontal correlation is approximately expressed by a Gaussian function $\mathbf{e}^{\frac{(x1-x)^2}{2L_s^2}}$ ($\mathbf{e}$ is the base of natural logarithms equal to 0.272). Here, $x1$ and x are two points, and L_s is the horizontal length scale. According to Zang et al. (2016), when the intersection of the decline curve reaches $\mathbf{e}^{1/2}$, the distance can be approximately as the horizontal length scale in Fig R5(a). The horizontal length scale was 81 km, which is approximately three-times larger than the scale used in this study. The vertical variance of SO_2 concentrations was considered by vertical correlations in the BEC. A strong relationship was observed in the boundary layer (approximately below the 20th model layer) in the vertical direction (Fig. R5(b)). The standard deviation demonstrates the reliability of the forecasting model, and the standard deviation for the vertical distribution of SO_2 concentrations decreased with increasing height in the $\mathbf{B}_c$ (Fig. R5(c)).

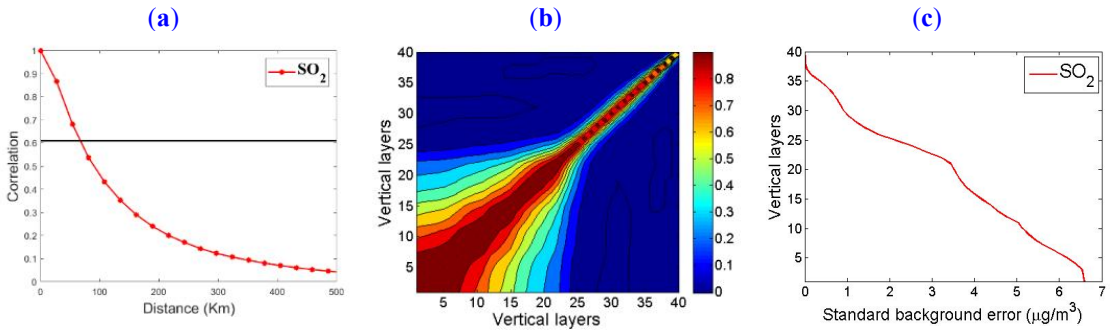


Figure R5. The background error covariation of SO_2 concentrations. (**a**) Vertical distribution of the horizontal correlation; the horizontal thin black line is the reference line ($\mathbf{e}^{1/2}$) for determining the horizontal correlation scales. (**b**) Vertical correlations. (**c**) Vertical distribution of the standard deviation.

For the $\mathbf{B}_{e_i}$, the standard deviation $\mathbf{D}_{e_i}$ is diagonal with a 200% error (Wang et al., 2012) and $\mathbf{C}_{e_i}$ is a block diagonal matrix, with the main diagonal blocks being the correlation matrices of SO_2 emission. The main diagonal blocks of $\mathbf{C}_{e_i}$ is 1.0 because the emission in each grid point is independent with other grids.

Comment4a: Several aspects of the experimental design were not initially clear to me and caused confusion. Although some elements became clearer with time, I think descriptions of the experiments should be clarified:

a. From Table 3, it appears that you performed DA experiments for ~3 weeks. What was the cycling period of your experiments (i.e., how often did you produce new 4DVAR analyses)? Was it 6 hours? Did you continuously cycle both the chemistry and meteorology, or did you periodically update meteorology from an external source, like the GFS model? What did you do for chemical boundary conditions (my apologies if I missed it)? Furthermore, Table 3 states 24-h forecasts were produced, but how often did you initialize these 24-h forecasts? Overall, the temporal aspect of the experiments should be clarified.

Response 4a: Thank you for your insightful and constructive question. Yes, the 4DVAR cycling period is 6 hours. We performed two sets of DA experiments to obtain optimized emissions of 2019 and 2020 (Table R1). For the set of experiments of Emi_2019, the first DA process started on 17 January 2019, and the observations during 0000–0600 UTC were assimilated by Eq. (1). The MEIC_2016 0000–0500 UTC emissions were the background emissions. The assimilated SO_2 concentration

initial field (0000 UTC) and the optimized SO_2 emission during 0000–0500 UTC were obtained.

Table R1 shows the details of DA emissions experiments. For the set of experiments of Emi_2019, the first DA process started on 17 January 2019, and the observations during 0000–0600 UTC of 17 January 2019 were assimilated by the 4DVAR system. Then, the optimized SO_2 concentration initial field (0000 UTC) and the optimized SO_2 emission during 0000–0500 UTC were obtained. Before conducting Emi_2019 experiment, 24 h forecasts were performed by WRF-Chem with MEIC_2016 emissions every 0000 UTC from 17 January to 7 February, 2019 to provide physical and chemical parameter. The chemical ICs of each day were obtained from the 24 h forecasting of the previous day. For the 24 h forecast, the meteorological initial and boundary conditions were provided by the 1° × 1° National Centers for Environmental Prediction (NCEP) Global Final Analysis data at a 6-hour frequency. The chemical boundary fields were not considered because the domain used in this study was wider than China. For the experiment of Emi_2019, the emission of 2019 were optimized by 4DVAR system every 6 hours with the background emissions of MEIC_2016. The physical and chemical parameter used in this DA process were obtained by the WRF-Chem forecast. For the experiment of Emi_2020, the DA process settings are similar with the Emi_2019 experiment. The optimized emissions of 2020 is obtained with the emission 2019 as background emission.

Table R1: Details of 4DVAR experiments to optimize the emission for 2019 and 2020.

Name	Background emissions	Optimized emissions	Study period
Emi_2019	MEIC_2016	2019 optimized emissions	Every 6 hours from17 January to 7 February, 2019
Emi_2020	2019 optimized emissions	2020 optimized emissions	Every 6 hours from 17 January to 7 February, 2020

To estimate the improvement of SO_2 forecasts using optimized emissions, three sets of forecast experiments were performed using the MEIC_2016 emissions and the optimized emissions for 2019 and 2020, respectively, and these were labeled

Ctr_2016, DA_2019, and DA_2020, respectively (see Table 3).. The three experiments were run daily with 24 h forecasts from 17 January to 7 February 2020. All experiments used the same WRF-Chem domain settings and physiochemical parameters. The SO_2 initial condition (IC) at 0000 UTC on January 17 was based on the spin-up forecasts initialized at 0000 UTC on January 7, 2020 for all three forecast experiments. The SO_2 ICs were later obtained from the 24h forecasting of the previous day for the three experiments, respectively. For example, the SO_2 IC of the experiment beginning at 0000 UTC on 18 January was from the 24h forecast result of the experiment beginning at 0000 UTC of 17 January, and so on. Meteorological initial and boundary conditions were provided by the 1° × 1° NCEP Global Final Analysis data at a 6-hour frequency. The chemical boundary fields were not considered.

Table R2: Details of the forecast experiments with emissions of 2016, 2019 and 2020.

Name	Emission	Forecast duration	Study period
Ctrl_2016	MEIC_2016	24 h	Every day from 17 January to 7 February, 2020
DA_2019	The 2019 optimized emissions	24 h	Every day from 17 January to 7 February, 2020
DA_2020	The 2020 optimized emissions	24 h	Every day from 17 January to 7 February, 2020

b. Fig. 3 didn't seem clear to me. What specific field(s) are being updated? Just SO_2 emissions? Or both SO_2 emissions and concentrations? Additionally, this figure might be clearer if you annotated the mathematical symbols from Eq. (1) or (3) on it so readers can link this figure to the equations. It might also be nice if you added another panel to the figure showing the temporal progression of the DA system (per above comment). Finally, in the top-left box, there's a typo (it should be "field").

Response 4b: Thank you for your suggestion. Figure 3 in the original manuscript has been modified. In the 4DVAR system, both SO_2 concentration initial field (c_0) and SO_2 emissions (e_i) were updated as the state vector. The mathematical symbols from Eq. (1) had been added in Fig. R6.

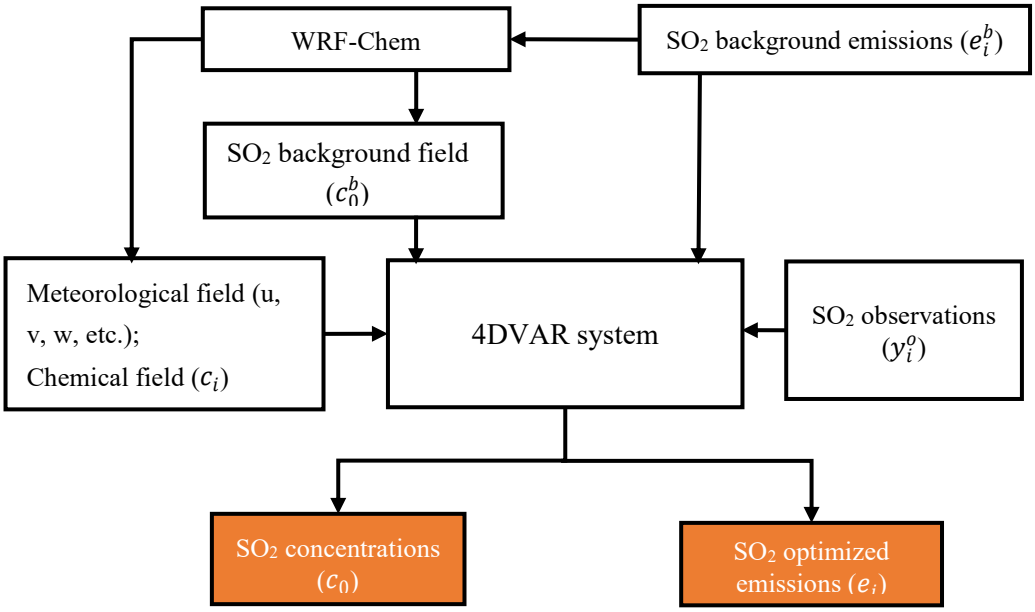


Figure R6: Flow chart of the SO_2 emissions optimization procedure in a single time step of i. The orange boxes represent the SO_2 optimized emissions and SO_2 concentrations of output. The c_0^b, c_0, e_i^b e_i *and* y_i^o are the mathematical symbols from Eq. (1).

c. It appears that you ran the 4DVAR DA system over two separate periods to estimate SO_2 emissions: 1) 17 Jan – 7 Feb 2019; and 2) 17 Jan – 7 Feb 2020. However, you never explicitly stated this! Thus, there are really two parts to this work. The first is estimating emissions in 2019 and 2020 from the 4DVAR DA system. The second is using different emissions estimates to drive various sets of forecasts over a common period in 2020. This distinction was not always clear, which caused me confusion.

Please explicitly state these various experiments and their purposes. It is important that you do so because of places like line 328, where you simply stated the *years* of the emissions, and not the names in Table 3 (which contain years); more distinction needs to be made for data *for a given year* versus various experiments *in 2020* that used emissions generated from various years.

Response 4c: Thank you for your suggestion. In this study, we performed two sets of DA experiments to obtain optimized emissions of 2019 and 2020 (Table R1) and three sets of forecasting experiments (Table R2) to estimate the improvement of SO_2 forecasts using optimized emissions. The statement of the experiments has been modified in the revised manuscript. Please see our **Response 4a** to previous comment

4a.

d. For the DA_2019 experiment, you effectively seemed to use a "pre-processing" step, where you ran the 4DVAR DA system over 2019 and then used those emissions when simulating a period in 2020. It might be worth noting that in contrast, the DA_2020 experiment did everything all at once without the need for a "pre-processing" step.

Response 4d: The "pre-processing" is to prepare background emission, chemical initial conditions, meteorological initial and boundary conditions. For the forecast experiments (Ctrl_2016, DA_2019 and DA_2020), all settings were the same in three forecast experiments, except for the emissions.

Comment5: Section 3.2: Please clarify that the data (i.e., emissions) discussed in this subsection were obtained from the 4DVAR analyses (I think), and not some other source. In general, please be more precise about from where the data on each figure come from.

Response 5: Thank you for your suggestion. The hourly SO_2 concentrations were taken from the China National Environmental Monitoring Center (http://www.cnemc.cn), and the 2016 SO_2 emission were obtained from the MEIC (http://www.meicmodel.org/). The 2019 and 2020 SO_2 emissions were obtained from the 4DVAR analyses.

The statement has been added in the Section 2.4 in the revised manuscript.

Comment6: The writing is understandable but there are many grammatical errors that I found distracting. Please carefully proofread the manuscript.

Response 6: The grammatical errors were corrected in the revised manuscript. Below are our point-to-point response in detail.

Minor comments

1. Line 48: "most explored algorithms" for what? Please be specific.

Corrected.

2. Lines 50-51: The second instance of "to estimate" should be removed, and perhaps "total regional and global emissions" should be moved to after the first instance of "to estimate".

   Corrected.

3. Lines 60-63: This statement is too broad. There are techniques to handle this problem, like inflation, that are well established at least for meteorological EnKF DA. Please refine this statement.

   Accepted. The statement had been deleted, and the previous EnKF SO_2 estimates had been added in the revised manuscript.

4. Line 89: Please add a reference for WRF-Chem.

   Corrected.

5. Line 105: Suggest "covering all of China" instead of "covering the entire country".

   Corrected.

6. Lines 105, 158, 159: "resolution" should be "grid spacing", as the two are not the same.

   Corrected.

7. Lines 111, Table 1: Please add a reference for the Grell-3D scheme.

   Corrected.

8. Fig. 1 caption: Please state that this figure also shows the WRF modeling domain!

   Corrected.

9. Lines 122-125: Somewhere in here, please specifically define *n*.

   Corrected.

10. Somewhere in section 2.2, please be more precise about which "control variables" are included in c_o. Is it just SO_2 concentration?

    Corrected. c_0 and e_i are the control variable that denotes the initial concentration vector and the modified emission.

11. Line 131: Here, is *H* nonlinear? (probably it is). Please state.

$\boldsymbol{H}$ is linearization, which operates on the model grid (SO_2 simulated concentrations) to generate a best estimate of the observed value (SO_2 concentrations). The statement of $\boldsymbol{H}$ has been revised.

12. Lines 131-132: Please state that *R* is the observation error covariance matrix.

Corrected.

13. Line 133: Please define *f*.

$f_{i,i-1}$ represents the model time integration for one time step from time $i-1$ to i. The statement had been added.

14. Eq. (3): Some more explanation is needed about how you go from Eq. (1) to Eq. (3) for the observation term. Specifically, please note the linearization about the background.

Thank you for your suggestion. Eq. (1) to Eq. (3) are only converted the equations from the solved of objective function to the incremental form. $\boldsymbol{H}$ plays a role of interpolation from the model grid to the observed value. Thus, $\boldsymbol{H}$ is linear. The tangent linear operators Γ, L (in Eq. (4-7) are derived from WRF-Chem are very complex and computational demanding, thus, we simplify the CTM to focus on SO_2.

Equation (8) in the manuscript is the governing equation for the concentrations:

$$\frac{\partial c}{\partial t} = -u\frac{\partial c}{\partial x} - v\frac{\partial c}{\partial y} - w\frac{\partial c}{\partial z} + \frac{\partial}{\partial x}\left(K_x\frac{\partial c}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_y\frac{\partial c}{\partial y}\right) + \frac{\partial}{\partial z}\left(K_z\frac{\partial c}{\partial z}\right) - \mathbf{e}^{-\Lambda}\frac{\partial c}{\partial t} - r\frac{\partial c}{\partial t} + V_m\frac{\rho_{air}}{\rho}\frac{\Delta S}{dz}e \quad (8)$$

In Eq. (8), the changes in concentrations are linear and only relate to the physicochemical parameters, such as $u, v, w, K_z, \Lambda,\ r, \rho$, and e.

We used the values ($u, v, w, K_z, \Lambda,\ r, \rho$, and e) within an integration step (10mins) to represent the mean of these variables in the 4DVAR system. This process would lead some errors due to the linear operators. But even we used a shorter integration step of 2mins, the result is close to that of the integration step of 10mins (Fig. R7). The average difference in concentrations between the two experiments was 0.3 in the grid of $i =$

$94, j = 152$ during 1 hour (Fig. R7a), which was also 1% of the total average concentrations. The mean difference in concentrations over China was 0.1 (Fig.R7b). Thus, it is concluded that the error from the linearization is very small and negligible.

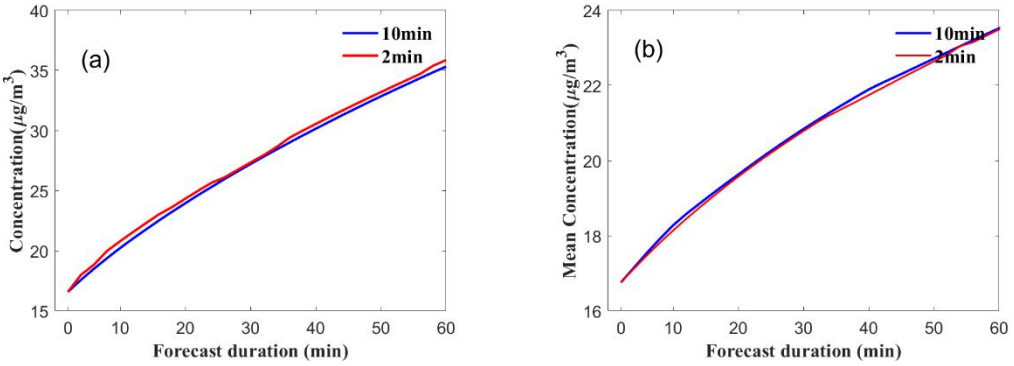


Figure R7: The forecast concentrations in the forward process by using different values in: (a) the grid of $i = 94, j = 152$ and (b) China.

15. Lines 150-152: How does this relate to the cycling period of the DA system? Does this mean you produced new analyses every 6?

    Yes, the 4DVAR system was performed every 6 hours to obtain the optimized emission. For example, the first DA process started at 17 January 2019, and the observations during 0000–0600 UTC were assimilated by Eq. (1). The MEIC_2016 0000–0500 UTC emissions were the background emissions. The assimilated SO_2 concentration initial field (0000 UTC) and the optimized SO_2 emission during 0000–0500 UTC were obtained.

16. Line 161: "large horizontal resolution study" is unclear. Are you referring to your specific study or something else? Please clarify.

    Sorry for the misleading. The horizontal resolution is 27km in this study. Thus, the $\frac{\partial}{\partial x}(K_x\frac{\partial c}{\partial x}) + \frac{\partial}{\partial y}\left(K_y\frac{\partial c}{\partial y}\right)$ can be neglect. The statement has been corrected.

17. Eq. (10): L_{turb} doesn't appear in the list of quantities in line 169, and L_{dry}, which does appear in line 169, doesn't appear in the list of equations.

Please clarify. Also please double check Eq. (14).

It should be L_{turb} in line 169. The statement had been deleted in the revised manuscript.

18. Lines 170-185: Should L_{trub} be L_{turb}?

    Corrected.

19. Line 180: Should it be Eqs. 9–13 instead of Eqs. 9–12?

    Corrected.

20. Lines 168-185: I'm not an expert about adjoint modeling, but I had the feeling that these lines aren't precise enough about the adjoint model formulation. Shouldn't there be more derivatives in there?

    In our 4DVAR system, the adjoint was applied to calculate Γ^T, L^T and $\boldsymbol{H}^T$ in Eq. (6) and (7), which are derived from the tangent linear model operator Γ, L, and observation operator $\boldsymbol{H}$. $\boldsymbol{H}$ plays a role of interpolation from the model grid to the observed value and is linear, thus $\boldsymbol{H}^T$ is easily derived using tangent linear coding techniques. The tangent linear operators Γ, L are simplified from the WRF-Chem model (Eq. (8)).

    Using tangent linear coding techniques, we derived the code for the discretized tangent linear operators Γ, L from the source code built in WRF-Chem. Once the source code is available for the tangent linear operators, we use the adjoint coding technique to derive the adjoint operator. The adjoint coding technique are detailed in Hoffman et al. (1992).

    This statement has been added in the revised manuscript in section 2.2.

21. Line 192: Please omit "the assimilation variable, which is the"…it's confusing, because that phrase is somewhat referring to a state/control variable, even though you're really talking about observation errors.

    Corrected

22. In lines 220-228, please be very precise about "emission" vs. "concentration" in your descriptions.

    Corrected.

23. Line 243: Please change February 6 to February 7 for consistency with Table

3.

Corrected.

24. Line 246: Typo: it should be “physiochemical”.

Corrected.

25. Line 249: Suggest “…based on the spin-up forecasts initialized at 0000 UTC…”.

Corrected.

26. Line 250: Please be more precise about “the previous day” (this comment relates to earlier comments about the cycling period).

Corrected.

27. Line 256: There seem to be more than 13 x 9 points in Fig. 4, so I was confused about this statement concerning “arrays and columns”. Please clarify.

Sorry for misleading, it is 13 x 21 points.

28. Fig. 5: The legend in the left panel is covering data and should be moved, and the y-axis in the right panel should probably be “J” not “J_b”.

Corrected.

29. Line 282: I’m not sure I agree with this statement, especially in (a) and (b); the 2019 SO_2 concentration decreases with time but the SO_2 emissions seem steady. Please revise.

Corrected.

30. Line 283: To my eyes, it looks like the lowest emissions were on 1 February, not 3 February (per Fig. 6b).

Corrected.

31. Lines 289-292: Please point to Figs. 6c,d here.

Corrected.

32. Fig. 6 caption: Please state the meanings of the vertical lines.

Corrected.

33. Line 311: I believes “rates” should be “ratios”.

Corrected.

34. Fig. 7: What are the insets in the lower right corner of each panel? Additionally, please be more precise about the subtraction convention. Above (c) and (d), it says "2020–2019" but the caption says, "differences between 2019 and 2020", which implies "2019–2020". It might be clearest to just write out "2020 minus 2019". Finally, please state in the caption whether these statistics are averaged over the entire period. Similar comments also apply to Fig. 8.

    The inset in the lower right corner of each panel is South China Sea, which belongs to China. Done as suggested.

35. Line 315: I don't think "observations" is the correct word. Is "analyses" more accurate? Please also see line 363.

    Corrected.

36. Line 320: Please remove "slightly"; it's too subjective.

    Corrected.

37. Line 321: Please remove "Remarkably", which is also subjective, and furthermore, the differences don't seem "remarkable".

    Corrected.

38. Line 333: Please remove "slightly". Also, it seems that this behavior was only evident in Fig. 9a, so please clarify the region you are discussing.

    Corrected.

39. Fig. 9 caption: Are these statistics averaged/aggregated over the entire period and over all sites or grid points? Please clarify.

    Yes, the statistics are averaged over the entire period and over all grid points. The statement has been revised.

40. Lines 352-355: I found this chunk troublesome. The explanation you offered didn't make sense to me, and I'm not sure all your statements are accurate. Please clarify or omit.

    Thank you for your suggestion. The explanation has been deleted.

41. Throughout, including figure captions: "Skill" should be "accuracy". Skill

is "accuracy relative to a baseline", and all of the metrics you are showing are measures of accuracy, not skill. I believe every instance of "skill" needs to be changed to "accuracy".

Corrected.

42. Fig. 10 caption: Please clarify whether these statistics are averaged/aggregated over the entire time period and all sites. Same comment for Fig. 11.

    The statistics are averaged over all sites in China. Corrected.

43. Lines 369-370: Please omit "compared with the Ctrl_2016 experiment".

    Corrected.

44. Lines 373-374: Please clarify what you mean by the "background field". Do you mean the field at the every start of the period (0000 UTC 17 January 2020)?

    No, the background field means the background emission. For Emi_2019 and Emi_2020 experiments (Table R1), the 4DVAR cycling period is 6 hours. For Ctrl_2016, DA_2019 and DA_2020 experiments (Table R2), 24 h forecasts were performed every 0000 UTC from 17 January 2020 to 7 February 2020. The statement was revised.

45. Line 390: Can you point to a figure for this key result about the decrease of optimized emissions? Also, did you ever state these values in the results section (sorry if I missed it)?

    The mean optimized emission and increment SO_2 emission field was shown in Fig. in the manuscript. And as your advice, the performance of the 4DVAR system at 0000 UTC 17 January 2019 had been added in the revised manuscript. Please see our **Response 2b** to previous major comment 2b.

46. Figs. 1, 4, 5, 8, 9: Please add annotations (e.g., "a", "b") to all these figures.

    Corrected.

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