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 helpful comments to improve 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 #3>

Comment: A timely and accurate emission is important for atmospheric chemistry simulation and pollution control. It is challenging and difficult to estimate the emission by using the "top-down" approach of 4DVAR. To my knowledge this is the first time when the 4DVAR system is development for optimizing SO2 emission and applied to investigate SO2 emission changes during the COVID-19 lockdown. The results shows that there is a significant decrease of SO2 emission between 2019 and 2020 due to the COVID-19 lockdown. It is reasonable and helpful for the improvement of atmospheric chemistry forecast. I suggest publishing this paper after the following points are addressed.

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

Comment 1: In the introduction, I suggest the author add some descriptions of emission optimization with the EnKF method.

Response 1: Thanks for your suggestion. For the EnKF method, many studies estimated SO_2 emissions by assimilating surface and satellite observational data in recent years, such as Dai et al (2021), Chen et al. (2019), Koukouli et al. (2018) and so on. Dai et al. (2021) developed a four-dimensional regional ensemble transform Kalman filter and showed that the SO_2 emissions over China in November 2016 decreased 49.4% in comparison to the 2010 background emission due to the implementation of emission control policies (Zheng et al., 2018).

Above literature review has been added in the introduction and discussion of revised manuscript.

Comment 2: In Fig. 2, how does the author classify the assimilating stations and verifying station?

Response 2: There are 1933 national control measurement stations in China in January 2020. The stations were gridded into the model grid ($27 \times 27\ km^2$). If there were more than 2 stations located in the same grid, one station was randomly selected to verify the improvement in using optimized emissions, and the remaining stations were used for assimilation. In this study, 508 stations were selected for verifying, while the remaining 1425 stations were used to assimilate.

This statement has been added in the revised manuscript.

Comment 3: In Fig. 3, there is not a box of observation in the flow chart. In addition, the variable of output is only SO2 emission. It should be added the initial SO2 concentration, since both the SO_2 concentration and the emission are the state variables in this study.

Response 3: Thanks for your suggestion. Figure 3 has been revised in the manuscript. The box of SO_2 observation (input) and SO_2 concentration (output) has been added.

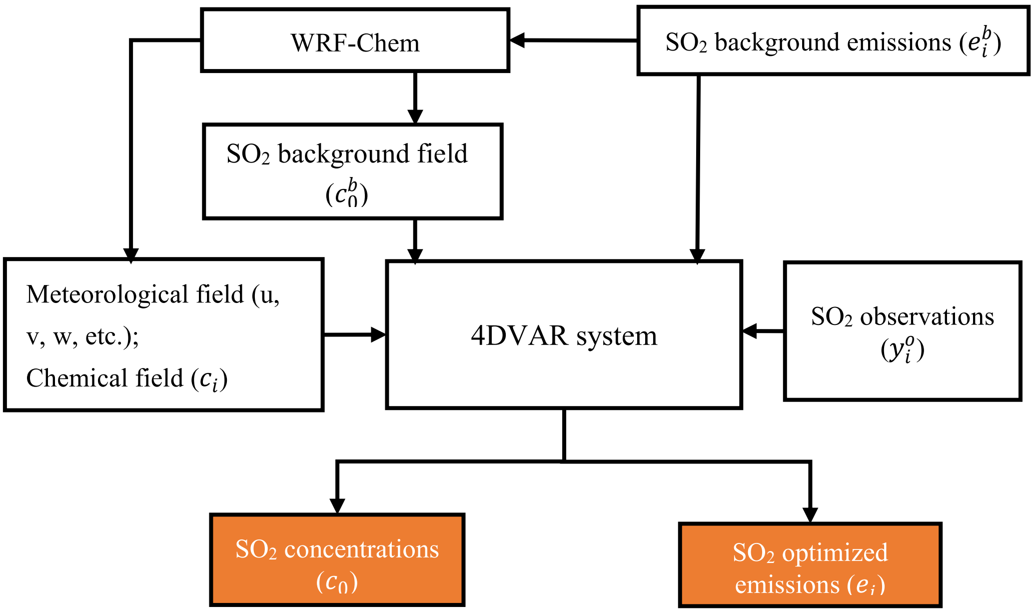

Figure R1: Flow chart of the SO_2 emissions optimization procedure in a single time step of i. The orange boxes represent the SO_2 optimized emissions and SO_2 concentrations of output. The c_0^b, c_0, e_i^b e_i *and* y_i^o are the mathematical symbols from Eq. (1).

Comment 4: Why the author firstly optimized the SO2 emission of 2019 from the emission of 2016. Did the author directly optimize the emission of 2020 from the emission of 2016?

Response 4: The goal of this study is to optimize the SO_2 emissions in January 2020 by the 4DVAR method and evaluate the influence of COVID-19 on SO_2 emissions. And the difference between 2019 and 2020 emissions during the same period reflected the influence of COVID-19 lockdown. However, since there were no 2019 emissions, we only first generated 2019 optimized emissions using the 4DVAR system, and the MEIC_2016 was set as background emission. Then the 2020 emissions were optimized, and 2019 optimized emissions were set as background emissions.

Comment 5: The author did not show the increment field of SO_2 concentration. I suggest the author add it. The scatter point of SO_2 concentration between observation and assimilation also should be illustrated.

Response 5: Thanks for your suggestion. The increment of SO_2 concentration and emissions at 0000 UTC 17 January 2019 have been added in the revised manuscript to estimate the 4DVAR system's efficacy that assimilated real observations.

Figure R2 shows the model simulated and observed SO_2 concentrations at 0000 UTC 17 January 2019. The MEIC_2016 was the background emission, and the hourly surface SO_2 observations during 0000–0600 UTC was assimilated. The observed SO_2 concentrations (Fig. R2(a)) showed that the most polluted area was located in North China Plain and Northeast China, and the observed SO_2 concentrations in southern China were generally lower than 20 μg m^{-3}. Compared with the observations, the SO_2 background concentrations were overestimated in Southern China, especially in Central China, Sichuan Basin, and Pearl River Delta. In addition, the SO_2 concentrations were underestimated in Northern China, Western China, and Southeast China. The increment of SO_2 concentrations showed the same change trends with the difference of observation and background field (Fig. R2(c)), reflecting the improvement of SO_2 concentration analysis field. Compared with the background field, the mean bias in analysis field improved from -2.8 to 1.8 μg m^{-3}, and the RMSE decreased from 23.1 to 11.8 μg m^{-3}. The CORR of analysis field increased from 0.2 to 0.8, suggesting the accuracy of SO_2 analysis field were improved using 4DVAR method.

Figure R2(e) is the background emission from MEIC 2016 at 0000 UTC and Fig. R2(f) shows the increment of SO_2 emissions at 0000 UTC 17 January 2019. The SO_2 background emission (Fig. R2(e)) showed that there were high emissions in South China, especially in Central China, Sichuan Basin and Pearl River Delta. The increment of SO_2 emissions (Fig. R2(f)) decreased the emissions in these regions. Zheng et al (2018) also found the emissions decreased in Southern China due to the implementation of emission reduction policies in China. The positive increment of SO_2 emissions (Fig. R2(f)) was lower than 1 mol km^{-2} h^{-1} in Southeast China, but the increment of SO_2 concentrations (Fig. R2(c)) was generally more than 10 μg m^{-3}, indicating the difference between background field and observation in Southeast

China was caused by the uncertainty of initial concentration field, not the emissions.

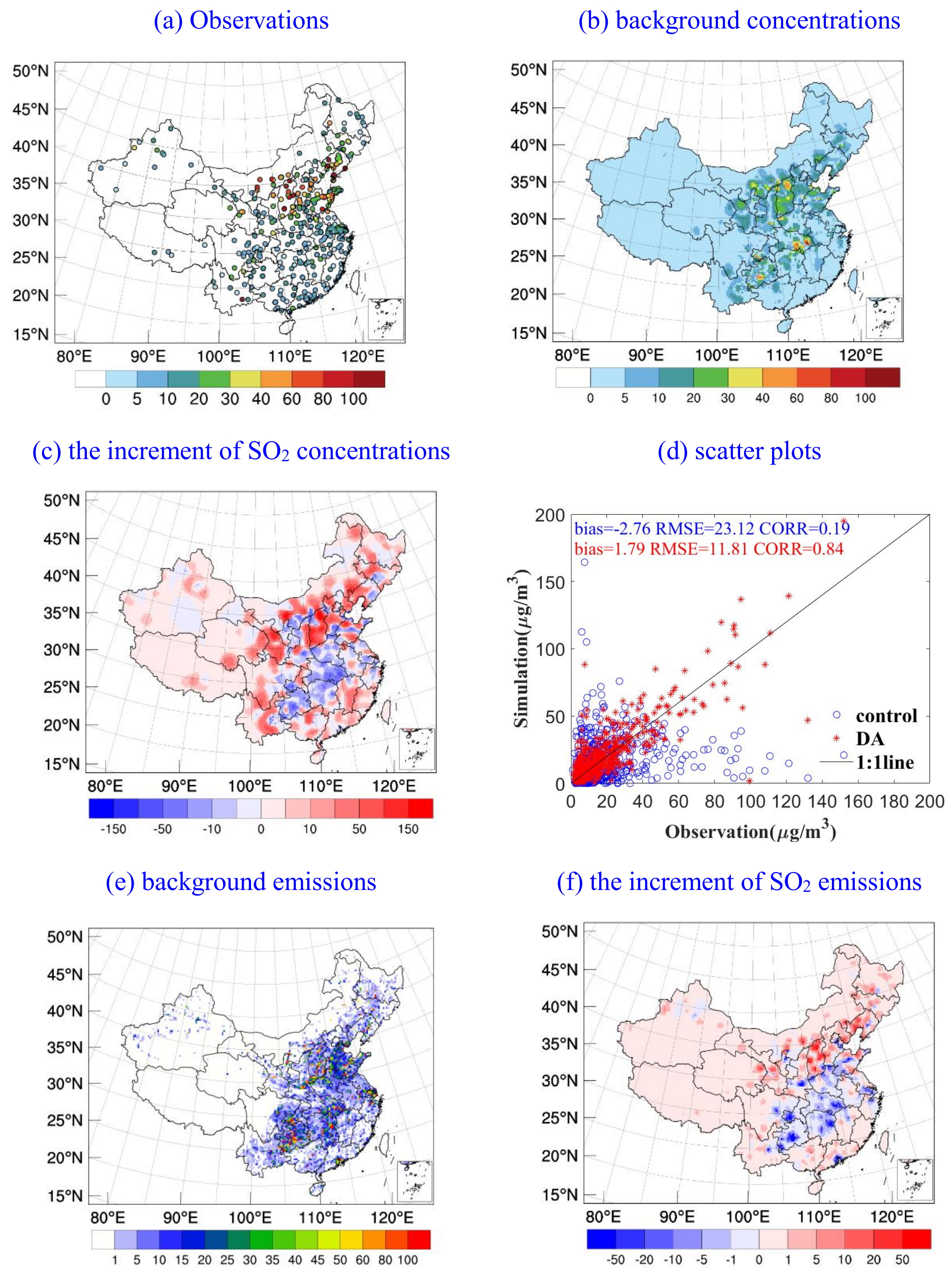

Figure R2: The simulated and observed SO_2 concentrations at 0000 UTC 17 January 2019. (a) Observations, (b) background concentrations, (c) the increment of SO_2 concentrations, (d) scatter plots, (e) background emission, and (f) the increment of SO_2 emission. Units: $\mu g\ m^{-3}$ for (a), (b), (c), and (d), and mol $km^{-2}\ h^{-1}$ for (e) and (f) .

Comment 6: L166-168： The variables such as *Lturb* and *Ldry* in the description and Eq. (9)-(13) are inconsistent. Please clarify.

Response 6: Sorry for misleading. The statement had been deleted.

Comment 7: L172: K_z should be $\overline{K_z}$ in Eq. (10).

Response 7: Corrected.