Response to the Comments of Referees

Manuscript ID: acp-2022-301

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

Author: Yiwen Hu, Zengliang Zang*, Xiaoyan Ma*, Yi Li, Yanfei Liang, Wei You, Xiaobin Pan, Zhijin Li

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 #1 >

Comment: This manuscript describes the development and application of a 4DVAR system to optimize SO2 emissions in China. An OSSE test shows improved consistency with the true emissions after optimizing emissions using this system. The framework has also been applied to estimate SO2 emissions during the COVID19 shutdown and shows a reduction of 18% compared to 2019. The topic fits the readership of ACP. I recommend publication after addressing the following comments:

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: L18, please specify the studied region in the abstract.

Response 1: Corrected.

Comment 2: L64 – 77, I would expect literature reviews on the application of 4D-Var to SO2 emission estimates in this paragraph. There are several of such studies. How are these 4D-Var estimates compared with previous EnKF SO2 estimates and your results?

Response 2: Thanks for your suggestion. For the 4DVAR method, Qu et al. (2019) estimated SO_2 emissions based on the GEOS-Chem model and its adjoint model by assimilating OMI observations and found the SO_2 emissions decreased by 48% over China from 2008 to 2016. 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 local ensemble transform Kalman filter and showed that the SO_2 emissions over China in November 2016 decreased by 49.4% in comparison to the 2010 background emission due to national pollution control policies (Zheng et al., 2018).

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

Comment 3: Eq (1), it was not clear to me whether H is an operator or a matrix from my first glance. I suggest using a different font for H.

Response 3: $\boldsymbol{H}$ is an observation operator that computes the observation estimates from the state variables and is also a vector. Additionally, the $\boldsymbol{H}$ in our 4DVAR system is Linear. Thus we use an italic bold font $\boldsymbol{H}$ to represent the operator in the revise manuscript.

Comment 4: Eq (3), the use of H delta(c) here implies that the operator is linear, but I doubt that for SO2. Could you discuss the impact of this assumption on the results?

Response 4: In this study, $\boldsymbol{H}$ plays a role of interpolation from the model grid to the observed value and is linear. And the tangent linear operators Γ, L(in Eq. (6) and (7)) were applied to calculate c_i(or δc_i) in this study. $\Gamma, and\ L$ are derived from WRF-Chem are very complex and computational demanding, thus, we simplify the CTM to focus on SO_2.

Focusing on SO_2, the governing equation for the concentration of species in WRF-Chem can be written as:

$$\frac{\partial c}{\partial t} = -u\frac{\partial c}{\partial x} - v\frac{\partial c}{\partial y} - w\frac{\partial c}{\partial z} + \frac{\partial}{\partial x}\left(K_x\frac{\partial c}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_y\frac{\partial c}{\partial y}\right) + \frac{\partial}{\partial z}\left(K_z\frac{\partial c}{\partial z}\right) - \mathbf{e}^{-\Lambda}\frac{\partial c}{\partial t} - r\frac{\partial c}{\partial t} + V_m\frac{\rho_{air}}{\rho}\frac{\Delta S}{dz}e \quad (1)$$

where c is the gas/aerosol concentration, ∂x and ∂y are the horizontal resolutions of the model, ∂z is the vertical resolution, and $u, v, and\ w$ denote the wind in $x, y, and\ z$ directions, respectively. $K_x, K_y\ and\ K_z$ are turbulent exchange coefficient in $x, y, and\ z$ directions based on K theory of turbulence. Λ is the loss rate. $\mathbf{e}$ is the base of natural logarithms equal to 0.272. r is the chemical reaction rate of the species. e denotes the emission source of the species. $V_m = 22.4\times10^{-3}\ m^3\ mol^{-1}$ is the molar volume of the gas, ρ is the air density of the actual atmosphere ($kg\ m^{-3}$), ρ_{air} is the standard air density, and ΔS is the grid area.

According to Eq. (1), the change in concentrations is approximately linear because the concentrations only relate to the physicochemical parameters, such as u, v, w, K_z, Λ, $r, \rho,$ 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. R1). The average difference in concentrations between the two experiments was 0.3 in the grid of $i = 94$, $j = 152$ during 1 hour (Fig. R1a), which was also 1% of the total average concentrations. The mean difference in concentrations over China was 0.1 (Fig.R1b). Thus, it is concluded that the error from the linearization is very small and negligible.

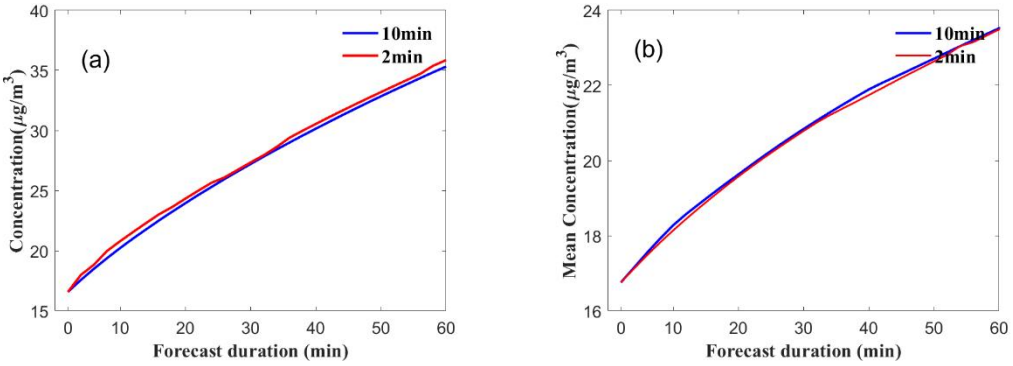

Figure R1: The forecast concentrations in the forward process by using different values in: (a) the grid of $i = 94$, $j = 152$ and (b) China.

Comment 5: L239, it is not clear to me what is the objective of these experiments just based on what is described here. Please clarify.

Response 5: Thank you for your suggestion. The goal of this study is to estimate the influence of COVID-19 lockdown on SO_2 emissions. But, the newest emission inventory of MEIC is for 2016 by statistics. It is considerable inaccurate due to the emissions reduction policies. The difference between 2019 and 2020 emissions during the same period reflected the influence of COVID-19 lockdown on SO_2 emissions. Thus, the SO_2 emissions during the study period in 2019 was also optimized.

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 from17 January to 7 February, 2020

On the base of three emission inventories of 2016, 2019 and 2020, three sets of forecast experiments were performed on the emissions during COVID-19 from 17 January to7 February 2020 (Table R2) to estimate the improvement of SO_2 forecasts using optimized emissions. Three experiments were run daily with 24 h forecasts from 17 January to 7 February 2020, and all experiments used the same WRF-Chem domain settings and physiochemical parameters. The MEIC_2016 emissions were used in the Ctrl_2016 experiment. For the DA_2019 experiment, the 2019 optimized emissions were used to simulate SO_2 concentrations during the study period. For the DA_2020 experiment, the 2020 optimized emissions were applied. The SO_2 initial condition 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 on 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 3: 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

The description of experiments has been revised in the revised manuscript.

Comment 6: Fig 7 & 8, how are these emission changes compared with previous studies?

Response 6: Figure 7 in the original manuscript shows the spatial differences and variations in emission ratios between 2019 and 2020. Compared with the 2019 optimized emissions, the 2020 optimized emissions decreased over most areas of the country due to the lockdown. The averaged reducing ratios of emission were 9.2% over China. Especially, the reducing ratios were more than 40.0% in most areas of North China and Central China. Zheng et al. (2020) showed that SO_2 emissions in China decreased by 12.0% in January and February 2020 compared to that in 2019. Fan et al. (2020) also found the SO_2 concentration decreased by 20.0–50.0% around COVID-19 lockdown in the spring 2020 over China based on TROPOMI satellite data. Our results were similar with the previous studies.

Figure 8 shows the same analysis as Fig. 7, but for central China. The averaged emission value in Wuhan was 43.0 mol km^{-2} h^{-1} in 2019 and 34.0 mol km^{-2} h^{-1} in 2020, showing a reduction of 21.0%. Almost all emissions in the Hubei Province decreased by 5–10 mol km^{-2} h^{-1}. Al-qaness et al. (2021) also found a decrease of SO_2 concentrations with 15% around Wuhan. In addition, the heavy emissions with the value exceeding 20.0 mol km^{-2} h^{-1} were most located around large cities. These heavy emissions decreased more than –5 mol km^{-2} h^{-1}, and the negative ratios were >20.0%. The large reduction of SO_2 emissions were coming from industrial and domestic coal combustion and power plants decreased during the COVID-19 lockdown (Zheng et al., 2018, 2020; Bian et al., 2019; van der A et al., 2017).

These sentences has been added in the revised manuscript.

Comment 7: Fig 11, the observation is significantly smaller than the simulations, even after DA. Could you address this a bit more and discuss the implications of this? How is this compared to other studies?

Response 7: Thank you for your suggestion. Figure 11a in the original manuscript shows the observation is smaller than the simulations in Central China. Note that the

mean concentration from the simulation of DA_2020 is close to the concentration of observation, suggesting that the 2020 optimized emissions were generally consistent with the real emissions in 2020. In 4DVAR optimization process, each grid will be influenced by surrounding grids because of the advection and vertical mixing. The theory of 4DVAR method is to take a balance between the observation and background field and obtain the optimized field. Therefore, when the observation values are small and those of the background field are large, the value of the optimized field will be larger than the observation.

The discussion had been added in the revised manuscript.

Reference

Al-qaness, M. A. A., Fan, H., Ewees, A. A., Yousri, D., and Abd Elaziz, M.: Improved ANFIS model for forecasting Wuhan City Air Quality and analysis COVID-19 lockdown impacts on air quality, Environmental Research, 194, 110607, https://doi.org/10.1016/j.envres.2020.110607, 2021.

Chen, D., Liu, Z., Ban, J., and Chen, M.: The 2015 and 2016 wintertime air pollution in China: SO2 emission changes derived from a WRF-Chem/EnKF coupled data assimilation system, Atmos. Chem. Phys., 19, 8619-8650, 10.5194/acp-19-8619-2019, 2019.

Dai, T., Cheng, Y., Goto, D., Li, Y., Tang, X., Shi, G., and Nakajima, T.: Revealing the sulfur dioxide emission reductions in China by assimilating surface observations in WRF-Chem, Atmos. Chem. Phys., 21, 4357-4379, 10.5194/acp-21-4357-2021, 2021.

Qu, Z., Henze, D. K., Theys, N., Wang, J., and Wang, W.: Hybrid Mass Balance/4D-Var Joint Inversion of NOx and SO2 Emissions in East Asia, Journal of Geophysical Research: Atmospheres, 124, 8203-8224, https://doi.org/10.1029/2018JD030240, 2019.

Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng, Y., He, K., and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions, Atmospheric Chemistry and Physics, 18, 14095-14111, 10.5194/acp-18-14095-2018, 2018.

Zheng, B., Zhang, Q., Geng, G.-n., Chen, C., Shi, Q., Cui, M., Lei, Y., and He, K. J. E. S. S. D.: Changes in China's anthropogenic emissions and air quality during the COVID-19 pandemic in 2020,

2021.