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Interactive comment

Interactive comment on "Revealing the sulfur dioxide emission reductions in China by assimilating surface observations in WRF-Chem" by Tie Dai et al.

Anonymous Referee #1

Received and published: 14 January 2021

This manuscript developed a new emission inversion system based on 4D-LETKF and WRF-Chem to update the SO2 emission by assimilating the ground-based hourly SO2 observations. The inverted SO2 emission over China in November 2016 is well in agreement with the "bottom-up" estimation, indicating that the newly developed emission inversion system can efficiently update the SO2 emissions based on the routine surface SO2 observations. Their investigation is interesting and valuable. The manuscript is well written and structured. I recommend publication after addressing the following concerns. Line 60: There are more recent research papers of ensemble-based assimilations to estimate the emission. Feng, S., Jiang, F*., Wang, H., Wang, H., Ju, W., Shen, Y., Zheng, Y., Wu, Z. & Ding, A

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(2020). NOx Emission Changes over China during the COVID-19 Epidemic Inferred from Surface NO2 Observations. Geophysical Research Letters, 47, e2020GL090080. https://doi.org/10.1029/2020GL090080 Feng, S., Jiang, F*., Wu, Z., Wang, H., Ju, W., & Wang, H. (2020). CO Emissions Inferred From Surface CO Observations Over China in December 2013 and 2017. Journal of Geophysical Research-Atmospheres, 125(7). https://doi.org/10.1029/2019JD031808 Chu, K., Z. Peng, Z. Liu, L. Lei, X. Kou, Y. Zhang, B. Xin and J. Tian: Evaluating the impact of emissions regulations on the emissions reduction during the 2015 China Victory Day Parade with an ensemble square root filter. J. Geophys. Res.-Atmos, 2018, doi:10.1002/2017JD027631 Line 76: Other two papers are also about the inverted SO2 emissions Peng, Z., Lei, L., Liu, Z., Liu, H., Chu, K., & Kou, X. (2020). Impact of assimilating meteorological observations on source emissions estimate and chemical simulations. Geophysical Research Letters, 47, e2020GL089030. https://doi.org/10.1029/2020GL089030 Peng, Z., Lei, L., Liu, Z., Sun, J., Ding, A., Ban, J., et al. (2018). The impact of multiaARspecies surface chemical observation assimilation on air quality forecasts in China. Atmospheric Chemistry and Physics, 18(23), 17,387-17,404. https://doi.org/10.5194/acpâĂŘ18âĂŘ17387âĂŘ2018)

Line 142: How do you decide the locations of the super-observations? Line 143: How do you decide the assimilated and independent verification observation sites? Line 180: How does the emission model forecast the emissions $E_{(t_{(n+1)})}$ for 12 hours? How the temporal and spatial distribution of the ensemble spread of the emissions $E_{(t_{(n+1)})}$ franged? Could you please show time series of hourly ensemble spread of the emissions $E_{(t_{(n+1)})}$ from 00:00 UTC 8 November to 00:00 UTC 18 November 2016 and their spatial distributions at typical time. Please discuss the forecast model first since the DA depends on the details of the forecast model. Line 181-183: Please write a bit more about the generation of the initial prior ensemble of SO2 emissions. And also a bit more about the spatial distribution of the ensemble spread of the prior emissions $E_{(t_0)}^-$. Line 197-200: The SO2 concentrations are updated "by recalculation of the WRF-Chem ensemble with the optimized emissions": so the uncertainties of

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the forecast SO2 concentrations could still be large. This will influence the assimilation results. Please discuss a bit more about them. Line 225-230: The spatial correlations among the grid points of the forecast emissions are not clear, so are the spatial correlations among the initial prior ensemble of SO2 emissions. Figure 3: Which data are used to obtain the averaged SO2 emissions? Could you please show the difference between the analysis and MEIC2016, or the ratio?

Line 250: Are the initial and lateral boundary chemical fields perturbed?

Line 272: Could you please show time series of hourly SO2 emissions of the prior, the forecast and the analysis of the assimilation experiments from 00:00 UTC 8 November to 00:00 UTC 18 November 2016, not only the mean spatial distribution in Figure 3. These will make the reader to understand a priori value and the adjustment SO2 emissions easily. Figure 6 and 7: I guess the SO2 concentrations are obtained from the DA experiments. But I am not sure if they are the updated results by recalculation the WRF-Chem ensemble with the optimized emissions. Could you please show the difference between the updated concentrations and the original? L391: Could you please show the diurnal variations of the inverted SO2 emissions of the DA experiments?

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Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-1259, 2020.