

Geosci. Model Dev. Discuss., referee comment RC2
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Comment on gmd-2021-201

Anonymous Referee #2

Referee comment on "High-resolution modeling of the distribution of surface air pollutants and their intercontinental transport by a global tropospheric atmospheric chemistry source-receptor model (GNAQPMS-SM)" by Qian Ye et al., Geosci. Model Dev. Discuss., <https://doi.org/10.5194/gmd-2021-201-RC2>, 2021

General comments:

The manuscript "High-resolution modeling the distribution of surface air pollutants and their intercontinental transport by a global tropospheric atmospheric chemistry source-receptor model (GNAQPMS-SM)" written by Qian Ye, et al. describes a global source-receptor model coupled with the GNAQPMS-SM global chemical transport model. The manuscript contains the model evaluation and assessment of source-receptor relationships of O₃, PM_{2.5}, BC, and non-sea-salt-sulfate. The topic of the manuscript is certainly within the scope of GMD. Overall, the manuscript is well written and easy to follow. I would like to consider the publication of the manuscript from GMD, while I have several comments below which should be addressed before publication.

Authors emphasize advantages of S-R calculation in their models (i.e., tagged tracer method) over sensitivity simulation analyses with perturbed emissions on the aspects of errors introduced from non-linear chemistry and computational resources in abstract, introduction, and summary. I generally agree them, whereas I miss more detailed analysis and discussion of effects of differences in S-R calculation methods. For example, the HTAP-like sensitivity simulations using GNAQPMS-SM in which emissions are perturbed for a few key regions would help isolate the effects of S-R calculation methods from the effects of the other model representations (e.g., chemical and transport processes, horizontal resolution, emission inventories, etc.).

Also, authors compare their results of S-R relationship assessment for BC and sulfate aerosols with the HTAP report. Why do not authors compare S-R relationships of surface ozone with the HTAP report? I suppose that an impact of errors introduced from non-linear chemistry on ozone S-R relationship is larger than that on aerosol S-R relationships.

Specific comments

P. 2, L. 58: Does "research area of observations" means coverage areas of field observations?

P.2-3, third paragraph in Introduction section:

This paragraph describes background information on S-R calculation methods and model resolution impacts on model performance and is mix of distinct topics. For readability, these topics need to be separated in individual paragraph.

P. 2, L. 63-64: Klich and Fuelberg (2014) is not global modeling study. Please add the appropriate reference.

P. 5, L. 134: What does "sensitivity analysis" mean?

P. 7, L. 192: Why do authors choose the year 2018 for simulation, even though emission for 2015 were used?

P. 7, Section 2.4: Please clarify treatments of soil NO_x emissions and volcanic SO₂ emissions in the simulation.

P. 11, L. 283-284: "In the model, stratospheric O₃ is constrained by relaxation towards zonally and monthly averaged values from ozone climatologies from Logan (1999)." Is it consistent with the description in section 2.3: "After calculation of tropospheric height, the monthly stratospheric ozone above the troposphere is taken from the climatic mean output from MOZART v2.4 (Horowitz et al., 2003)." (P. 7, L. 194-195)?

P. 11, L. 286: "The coarse vertical resolution..." What vertical resolutions do the GNAQPMS have in the UTLS regions? Are these errors introduced by model physical processes or tracer advection scheme?

P. 11, L. 290-292: "Note that the model overestimation in the upper troposphere does not affect our analysis of surface O₃ and its origins because O₃ in the upper troposphere has little effect on the surface O₃ concentration." Is it confirmed by authors' tagged tracer calculation?

P. 13, Section 3.1.3: There are no explanation how authors compare the model results with TROPOMI tropospheric NO₂ data (e.g., spatial and temporal sampling from model outputs, application of averaging kernels).

P. 13, Section 3.1.3: TROPOMI low biases (Verhoelst et al., 2021) may also be possible

reason for model positive biases.

P. 14, L. 330-331: "since there is higher biomass and fossil fuel burning for heating during DJF" Do seasonal cycles of precipitation affect seasonal cycles of simulated PM2.5 concentrations?

P. 16, L. 383: What is the definition of "SNA"?

P. 17, L. 389: "heterogeneous chemistry on aerosol surfaces" Please add the references which support this speculation.

P. 17, L. 401: "a trough in DJF" Is this trough over eastern China also influenced by NOx titration?

P. 18, Figure 8: What is a possible reason why model PM2.5 biases are larger in MAM than in other seasons?

P. 19, Figure 9: This figure shows "Validation of GNAQPMS simulations against EMEP observations."

P. 23, L. 467: The title of section 4 is confusing. Section 3 also contains results of this study.

P. 23, L. 472: How do biogenic secondary organic aerosols account for?

P. 27, L. 537: ", while the contribution of the top boundary is small in other receptor regions and ..." Is it correct? In Figure 14, "Boundary" have second or third most contributions in most receptor regions.

P. 28, Section 4.2: Why do authors evaluate S-R relationships of BC and sulfate in the PBL only in this section?

P. 33, L. 673: Vertical resolution is also important for resolving intercontinental transport plumes (Eastham et al., 2017). Do authors plan to investigate the sensitivity of S-R

relationships to different vertical resolutions?

Reference

Eastham, S. D. and Jacob, D. J.: Limits on the ability of global Eulerian models to resolve intercontinental transport of chemical plumes, *Atmos. Chem. Phys.*, 17, 2543–2553, <https://doi.org/10.5194/acp-17-2543-2017>, 2017.

Verhoelst, T., Compernelle, S., Pinardi, G., Lambert, J.-C., Eskes, H. J., Eichmann, K.-U., Fjæraa, A. M., Granville, J., Niemeijer, S., Cede, A., Tiefengraber, M., Hendrick, F., Pazmiño, A., Bais, A., Bazureau, A., Boersma, K. F., Bogner, K., Dehn, A., Donner, S., Elokhov, A., Gebetsberger, M., Goutail, F., Grutter de la Mora, M., Gruzdev, A., Gratsea, M., Hansen, G. H., Irie, H., Jepsen, N., Kanaya, Y., Karagkiozidis, D., Kivi, R., Kreher, K., Levelt, P. F., Liu, C., Müller, M., Navarro Comas, M., Piters, A. J. M., Pommereau, J.-P., Portafaix, T., Prados-Roman, C., Puertedura, O., Querel, R., Remmers, J., Richter, A., Rimmer, J., Rivera Cárdenas, C., Saavedra de Miguel, L., Sinyakov, V. P., Stremme, W., Strong, K., Van Roozendaal, M., Veefkind, J. P., Wagner, T., Wittrock, F., Yela González, M., and Zehner, C.: Ground-based validation of the Copernicus Sentinel-5P TROPOMI NO₂ measurements with the NDACC ZSL-DOAS, MAX-DOAS and Pandonia global networks, *Atmos. Meas. Tech.*, 14, 481–510, <https://doi.org/10.5194/amt-14-481-2021>, 2021.