Dear Editor and Reviewer:

Thank you very much for your insightful comments concerning our 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)" (MS No.: gmd-2021-201). Those comments are all very valuable and helpful for revising and improving our manuscript. We have studied comments carefully and have made revision. The point by point responses are as following:

Responds to the referee's comments:

Referee #2:

Comment 1: P. 2, L. 58: Does "research area of observations" means coverage areas of field observations?

Response: Thank you for your comments. Yes. The "limited research area of observations" in line 58 means that the research area covered by field observation activities is always limited.

Comment 2: 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.

Response: Thank you for your valuable comments. We have separated this paragraph into three paragraphs in the latest manuscript.

Comment 3: P. 2, L. 63-64: Klich and Fuelberg (2014) is not global modeling study. Please add the appropriate reference.

Response: Thank you for your comments. We have changed this reference and have added the appropriate references in the latest manuscript as the blue text below:

"Many global chemical transport models have been used to study the distribution of air pollutants and estimate their intercontinental transport (Wai et al., 2016; Yang et al., 2017)."

Comment 4: P. 5, L. 134: What does "sensitivity analysis" mean?

Response: Thank you for your comments. As described in line 65, sensitivity analysis, "it mainly refers to the reduction in emissions in the source region by a certain extent, such as 15% or 20%, or even directly returning to zero." Because for primary pollutants, such as BC, their S-R relationships are nearly linear and are different from secondary pollutants, such as O_3. The neglect of nonlinear error in sensitivity analysis has much less impact on the results of S-R relationships of primary pollutants. To better understand the "sensitivity analysis" method here, we have revised it in the latest manuscript as the blue text below:

", so emission sensitivity analysis can provide an accurate S-R assessment."

Comment 5: P. 7, L. 192: Why do authors choose the year 2018 for simulation, even though emission for 2015 were used?

Response: Thank you for your valuable comments. We choose the year 2018 for simulation because we have more observation data in China in 2018 and China have completed the toughest-ever clean air policy, where BC, NO_x, primary $PM_{2.5}$, SO_2 emissions have all significantly decreased due to the implementation of the policy. Zhang et al. (2019) reported that from 2015 to 2017 SO_2 emissions in China decreased by 35%, while NO_x and NMVOC emissions changed slightly with a decrease lower than 10%. However, this significant SO_2 change is not reflected in most emission inventories, which should lead to uncertainties on the S-R relationships, e.g., it could be a possible reason for the overestimation of SO_2 in China and may have an impact on our analysis of nss-sulphate transport from China to the downwind regions in our study. As for using different years of emission inventories in this simulation, the official inventories made by countries or regions have at least a 1-year time lag (Janssens-Maenhout et al., 2015), and the global emission inventories, e.g., EDGAR has a nearly 5-year time lag and is currently updated to year 2015. In order to understand the possible influence on inconsistent inventory years on the results, we compared the 1-year change rate of pollutants emissions in three regions (NAM, EUR, China) as shown in Fig. 1. The 1-year change rate of BC in NAM ranges around $\pm 10\%$ and in EUR ranges within $\pm 5\%$. BC emissions in China have changed within $\pm 10\%$ since 2010. The 1-year change rate of primary $PM_{2.5}$ in three regions are all within $\pm 10\%$. The 1-year change rate of NO_x and SO_2 in three regions ranges within $\pm 5\%$ and $\pm 10\%$ since 2010, respectively. All change rates are within the region and pollutant emission uncertainties (95% confidence interval) reported by Crippa et al. (2019). For example, the SO_2 emission uncertainty in China, USA, Canada is 12%, 31%, 53%, respectively. NAM refers to USA and Canada. Moreover, from the perspective of the 3-year change rate, global total emissions changed a little within $\pm 5\%$ between 2015 and 2012. We also computed the annual BC, SO_2, CO emissions in year 2017, 2018, 2019 from the Community Emissions Data System (CEDS) for Historical Emissions, their 1-year change rate of BC, SO_2 and CO ranges from -1.6% to -0.6%. It can be seen that there is no significant change of emission inventories between 2015 and 2018, and we have more observation data in China in 2018. Therefore, we chose the emission inventory 2015 to simulate the year 2018.
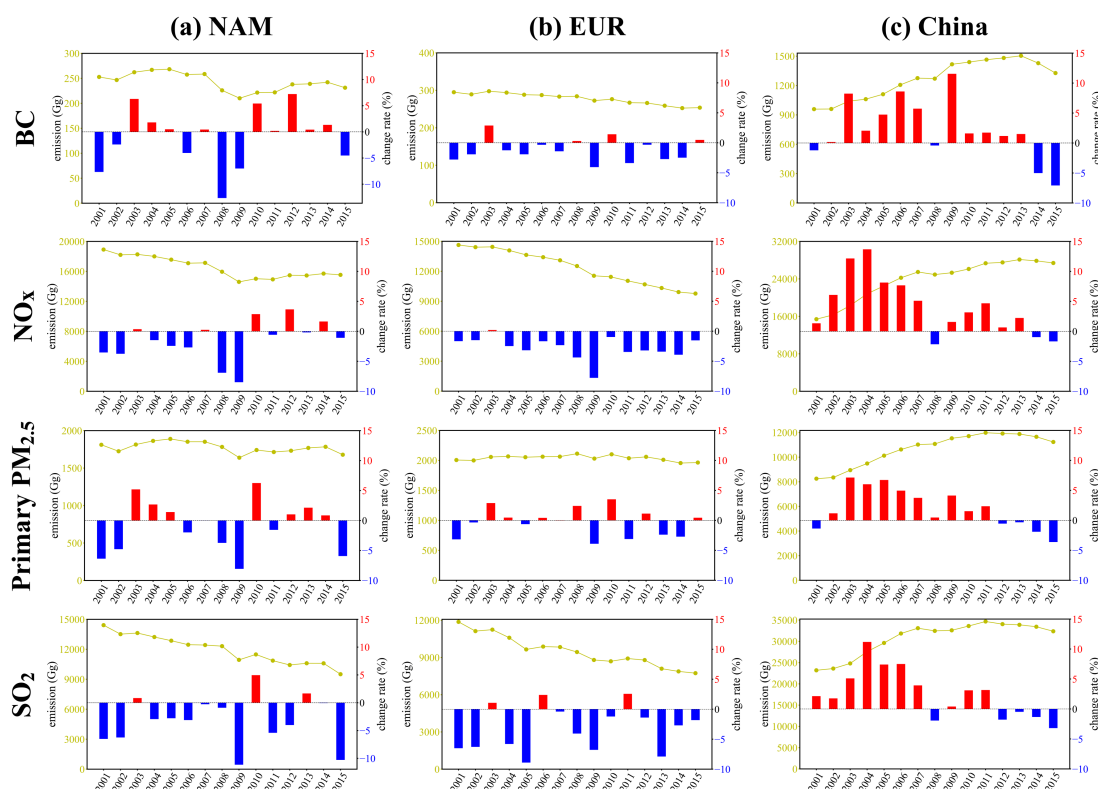
Figure. 1 Annual emissions and 1-year change rate of BC, NO$_x$, primary PM$_{2.5}$ and SO$_2$ in (a) NAM, (b) EUR, and (c) China from 2001 to 2015.

Comment 6: P. 7, Section 2.4: Please clarify treatments of soil NO$_x$ emissions and volcanic SO$_2$ emissions in the simulation.

Response: Thank you for your valuable comments. In our model, the soil NO$_x$ emissions are from Global hourly emissions for soil NO$_x$ (Hudman et al., 2012; data available at http://ftp.as.harvard.edu/gcgrid/data/ExtData/HEMCO/OFFLINE_SOILNOX/v2019-01/), and the volcanic SO$_2$ emissions are from Carn et al. (2015) (data available at http://ftp.as.harvard.edu/gcgrid/data/ExtData/HEMCO/VOLCANO/v2019-08/). We have added it in the latest manuscript.

Comment 7: P. 11, L. 283-284: "In the model, stratospheric O$_3$ 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)?

Response: Thank you for your valuable comments. These two descriptions are consistent. The Horowitz et al. (2003) study where we referred the treatment method also used original values from

ozone climatologies from Logan (1999) in stratospheric O_3. Therefore, two references we both added in our manuscript.

Comment 8: 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?

Response: Thank you for your comments. In the vertical direction, we divide 20 layers. The bottom layer is about 50 m and the top layer is about 20 km. About 6 layers are located above 6 km. As you said, these errors may introduce by model physical processes. The inadequate resolution of the tropopause and excessive cross-tropopause transport of O_3 in the model could result in the overestimation of O_3. The top boundary of O_3 in our model are referred from Horowitz et al. (2003). The vertical direction is divided into 34 layers extending up to 4hPa in Horowitz et al. (2003), and the model they used tends to overestimate O_3 in the vicinity of the tropopause by 25% or more. Similar reasons have also been pointed out that inadequate resolution of tropopause location and excessive production of O_3 in the UTLS regions are responsible for the overestimation. Above 15 km, we only divide 3 layers and it's relatively coarse. In the horizontal direction, our resolution is 0.5°, which is sufficient that Eastham and Jacob (2017) has reported that 1° is sufficient. Wei et al. (2019) computed the global budgets and proved global mass conservation of model which used the same advection scheme with us, so the advection scheme should not have much influence on our overestimation.

Comment 9: P. 11, L. 290-292: "Note that the model overestimation in the upper troposphere does not affect our analysis of surface O_3 and its origins because O_3 in the upper troposphere has little effect on the surface O_3 concentration." Is it confirmed by authors' tagged tracer calculation?

Response: Thank you for your comments. This statement is imprecise and mainly focuses on the mid-latitude continent in the Northern Hemisphere. Compared with the contribution of photochemical reactions, stratosphere is not the major contributor in the main continents in the Northern Hemisphere. We select 4 stations, Resolute, Hilo, Hohenpeissenberg and Sapporo. Resolute and Hilo are located in NAM; Hohenpeissenberg is located in EUR; Sapporo is located in EA. The transport from stratosphere can reach a peak of around 10 ppb to surface concentration in DJF, with relative contribution lower than 20%. Their seasonal contributions of top boundary are compared with Sudo and Akimoto (2007) results as shown in Fig. 2. Sudo and Akimoto (2007) simulation captures the variation of O_3 in the upper troposphere and stratosphere well. Although our results overestimate the contribution, it's still within an acceptable range, with R value 0.67, NMB value 33% and FAC2 value 72.9%. We also capture the seasonal variation of stratosphere

contribution. However, we have to admit that according to the results of S-R relationships, the Southern Hemisphere is greatly affected by stratospheric transport, and this overestimation will inevitably overestimate the transport from stratosphere. We have deleted this sentence in the latest manuscript.
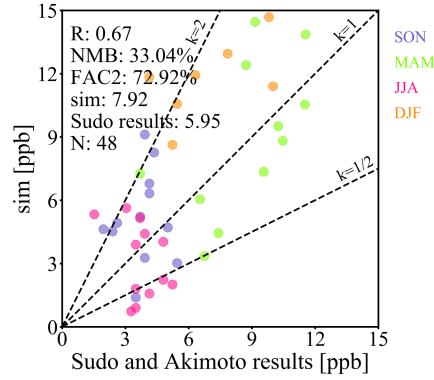
Figure 2. Scatterplots of simulated (sim) and Sudo and Akimoto (2007) seasonal stratosphere contributions to O$_3$ at 4 stations. The black dotted lines are the 2:1, 1:1, and 1:2 reference lines from left to right. Different seasons are plotted with circles in different colors.

Comment 10: P. 13, Section 3.1.3: There are no explanation how authors compare the model results with TROPOMI tropospheric NO$_2$ data (e.g., spatial and temporal sampling from model outputs, application of averaging kernels).

Response: Thank you for your comments. The horizontal resolution of our simulation is 0.5°×0.5° and the resolution of TROPOMI data is 0.125°×0.125°. The column of NO$_2$ (DU) is computed as follows:

$$DU = \sum_{i=1}^{n} \frac{plev_i \times conc_i \times 100 \times 6.02 \times 10^{23}}{8.34 \times T_i \times 10^{13}} \times dz_i$$

where $plev_i$, $conc_i$, T_i, dz_i are pressure, concentration of NO$_2$, temperature and height of the ith layer, respectively.

The Tropospheric Monitoring Instrument (TROPOMI) was loaded onboard the Sentinel-5 satellite for fine gas monitoring and the TROPOMI NO$_2$ data retrieval is described in the product Algorithm Theoretical Basis Document (van Geffen et al., 2020). The monthly tropospheric NO$_2$ column concentration data from the Royal Netherlands Meteorological Research Institute (KNMI) used in our paper has measurement loss in some areas. We compared the NO$_2$ columns through one-by-one correspondence between the simulation time and the TROPOMI data observed time, the model grid cell and TROPOMI data grid cell. The simulation results (DU) when drawing, are filled with model results if the grid cell misses measurement in the TROPOMI data. In order to be consistent, in the latest manuscript we have eliminated the grid cells in our simulation which is missing in the TROPOMI data as shown in Fig. 3 below.

(a) Simulation   **(b) TROPOMI**

0.1 0.2 0.3 0.4 0.5   1   3   5   7   10   15
10^{15} molecules/cm^2
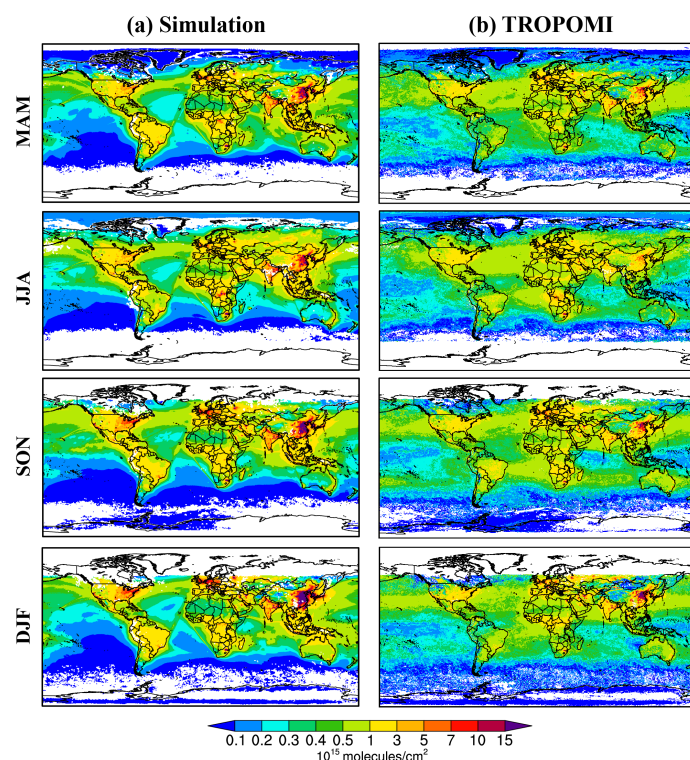
Figure 3. Spatial distributions of seasonal mean NO$_2$ columns from (a) GNAQPMS averaged in 2018 and (b) the TROPOMI data.

Comment 11: P. 13, Section 3.1.3: TROPOMI low biases (Verhoelst et al., 2021) may also be possible reason for model positive biases.

Response: Thank you for your valuable suggestions. We have added this possible reason in the latest manuscript as the blue text below:

"A negative bias for TROPOMI satellite data compared to the ground-based measurements (Verhoelst et al., 2021) may also be a reason for our model positive biases."

Comment 12: 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 PM$_{2.5}$ concentrations?

Response: Thank you for your comments. The removal of pollutants from the atmosphere can occur in one of two ways: by wet deposition or dry deposition. Wet deposition is the process of washing out pollutants from the air by precipitation or cloud droplets. Seasonal cycles of precipitation will affect wet deposition, and then affect seasonal cycles of simulated PM$_{2.5}$ concentration. The simulated precipitation field is shown in Fig. 4. The PM$_{2.5}$ concentration in major PM$_{2.5}$ hotspots India, East Asia, and Central Africa reaches a peak during DJF and a trough in JJA. Similarly, the precipitation reaches a maximum in JJA and a minimum in DJF in these regions, which is

corresponding to the wet deposition peak in JJA. Moreover, the Planetary Boundary Layer (PBL) plays an important role by exchanging heat and moisture between the surface and free atmosphere. A high concentration of $PM_{2.5}$ will enhance the stability of PBL, which in turn decreases the PBL height. Lower PBL height is associated with weaker turbulence mixing, leading to favorable conditions for the accumulation of air pollutants in the shallow PBL and this positive feedback mechanism will strengthen in DJF (Petaja et al., 2016; Yin et al., 2019).
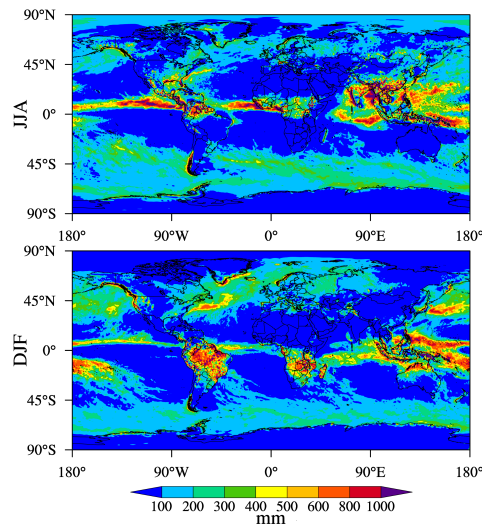
Figure 4. Simulated seasonal precipitation in JJA and DJF.

We have updated these reasons in the latest manuscript as the blue text below:

", and the lowest values are during JJA and SON since there is higher biomass and fossil fuel burning for heating, less precipitation and strengthened positive feedback between aerosols and PBL (Petaja et al., 2016) during DJF."

Comment 13: P. 16, L. 383: What is the definition of "SNA"?

Response: Thanks for your comments. As described at line 250, SNA "refers to the sum of sulphate, nitrate and ammonium".

Comment 14: P. 17, L. 389: "heterogeneous chemistry on aerosol surfaces" Please add the references which support this speculation.

Response: Thank you for your comments. We have added the appropriate references to support as the blue text below:

"heterogeneous chemistry on aerosol surfaces (Bauer and Koch, 2005; Andreae and Crutzen, 1997)"

Comment 15: P. 17, L. 401: "a trough in DJF" Is this trough over eastern China also influenced by NO_x titration?

Response: Thank you for your valuable comments. We compared the seasonal spatial distribution of NO and NO_2 as shown in Fig. 5. Surface NO and NO_2 over eastern China reaches a maximum in DJF and SON and a minimum in MAM and JJA, which is opposite from the surface O_3 concentration. The O_3 trough in DJF at urbanized eastern China could be due to the enhanced O_3 titration effect by the increase of NO_x. We have added this possible reason in the latest manuscript as blue text below:

", and this seasonality is determined by abundant photochemical reactions in JJA and the East Asian monsoon. The enhanced titration of O_3 by the increased NO_x in DJF could also be a possible reason."
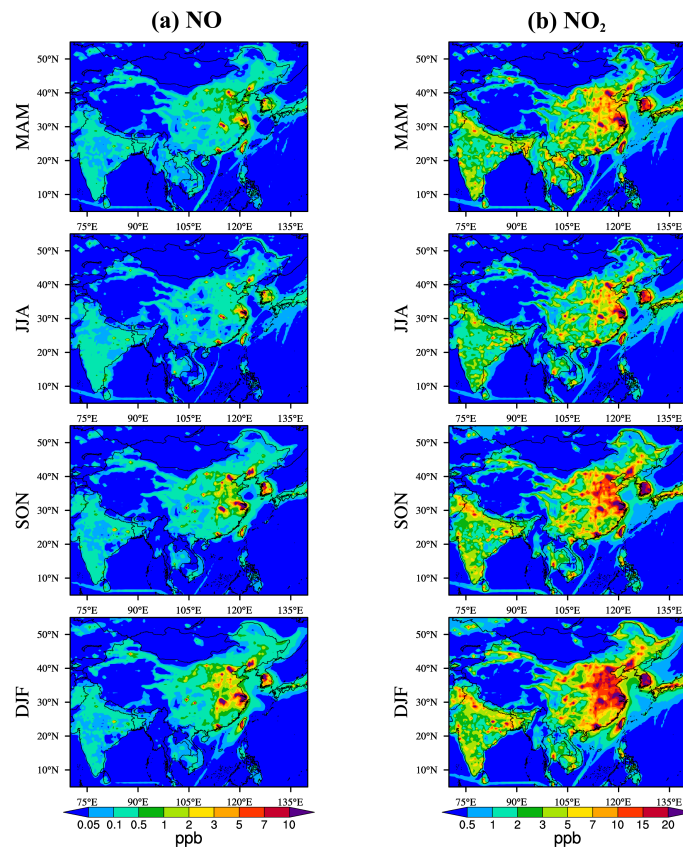
Figure 5. Spatial distributions of seasonal mean (a) NO and (b) NO_2 surface concentrations.

Comment 16: P. 18, Figure 8: What is a possible reason why model $PM_{2.5}$ biases are larger in MAM than in other seasons?

Response: Thank you for your comments. Seasonally, 72.85% of all dust storms occurred in the spring (MAM), 16.39% in winter (DJF), 8.71% in summer (JJA), and 2.05% in autumn (SON) in northern China (Deng et al., 2013). Many studies have stated that the mechanism for the outbreak and development of dust storms are not yet fully understood and the techniques for local dust storm in China forecasting require further improvement (Huang et al., 2006; Sun et al., 2006; Wang et al.,

2019). Therefore, the larger PM$_{2.5}$ biases in MAM may be due to the spring dust storms over northern China, which is difficult to simulate, e.g., several stations underestimated and circled by the red box in Fig. 6. We have added this reason in the latest manuscript as the blue text below:

"The model shows good spatial correlations with observations and little seasonal bias, with R values ranging from 0.91-0.92 and NMB values within $\pm$15%. The frequent spring dust storms in northern China are an important reason for the larger PM$_{2.5}$ biases in MAM."
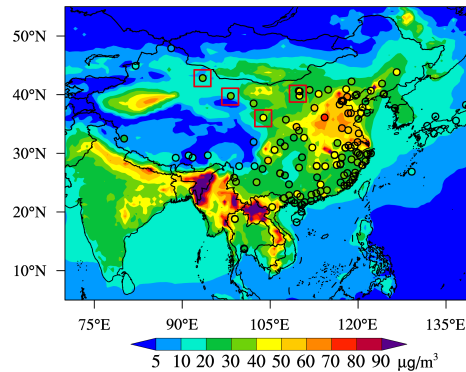
Figure 6. Spatial distribution of simulated (shaded) surface PM$_{2.5}$ concentrations compared with observations (solid circles) in MAM in East Asia.

Comment 17: P. 19, Figure 9: This figure shows "Validation of GNAQPMS simulations against EMEP observations."

Response: Thank you for your comments. We have revised this sentence in the latest manuscript as the blue text below:

"Figure 9 shows the validation of GNAQPMS simulations against EMEP observations."

Comment 18: P. 23, L. 467: The title of section 4 is confusing. Section 3 also contains results of this study.

Response: Thank you for your comments. We have changed the title of section 4 to "Results of S-R relationships" in the latest manuscript.

Comment 19: P. 23, L. 472: How do biogenic secondary organic aerosols account for?

Response: Thank you for your comments. Biogenic secondary organic aerosols are considered to be from natural emission sources.

Comment 20: 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.

Response: Thank you for pointing out our imprecise express. Sources of O_3 to the troposphere include downward transport from the stratosphere (Junge, 1962) and photochemical production from precursors (Monks et al., 2009). The smaller contribution of the top boundary is relative to the contribution of photochemical reactions. We have revised it in the latest manuscript as the blue text below:

   "The contribution of the top boundary is large in SPO due to the low tropopause of SPO and the simplified treatment of the stratosphere, while the contribution of the top boundary is smaller in Northern Hemisphere receptor regions compared with the contribution of photochemical reactions."

Comment 21: P. 28, Section 4.2: Why do authors evaluate S-R relationships of BC and sulfate in the PBL only in this section?

Response: Thank you for your careful comments. First, the S-R relationships of $PM_{2.5}$ and O_3 in the surface layer have already been briefly analyzed in section 4.1, and we hope to show the advantage that our model can quantify the contributions of multiple air pollutants from various source regions at the same time in one simulation. Secondly, BC as the representative of primary aerosol and sulphate as the representative of secondary aerosol both make a great influence on our environment and climate. They tend to play opposite roles in their radiative effects. While sulphate is characteristic of strong radiation scattering and thus leads to a cooling surface (negative radiative forcing), BC heats up the atmosphere owing to its excellent ability of absorbing solar radiation. It's important to analyze their S-R relationships. In addition, due to the limitation of manuscript length, our purpose is to introduce the added S-R module, which can help us analyze the S-R relationships of pollutants, rather than discussing the results of each tagged pollutant in detail. Therefore, we only evaluate S-R relationships of BC and sulfate in section 4.2.

Comment 22: P. 33, L. 673: Vertical resolution is also important for resolving intercontinental transport plumes (Eastham and Jacob, 2017). Do authors plan to investigate the sensitivity of S-R relationships to different vertical resolutions?

Response: Thank you for your valuable suggestions which really provide a great thinking direction. As Eastham and Jacob (2017) stated, the inadequate vertical grid resolution cannot properly resolve plume gradients. We have already divided the vertical layer into 40 layers to better analyze the S-R relationships, although this new vertical resolution is much coarser compared with a vertical resolution of 100 m in the free troposphere. About 20 layers are located above 3km and about 12 layers are located above 6 km. We will incorporate this into our future plans. It's a great question worthy of careful discussion.

Comment 23: 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.

Response: Thank you for your constructive comments. Indeed, in addition to the different methods of studying the S-R relationships, the model, horizontal resolution, emission inventories we used are also different from HTAP used, which may bring some uncertainties when we compare their differences. As you suggested, we will carry out some experiments about emission sensitivity analysis in the following manuscript.

We try to do some comparisons on the two approaches. Taken O_3 as an example, emission sensitivity approach estimates intercontinental transport of surface O_3 resulting from 20% reductions of anthropogenic O_3 precursor emissions for each source region, and HTAP chose this method. It's attributable to the precursor emitting locations. In contrast, for O_3, it's tagged by the produced locations in our model because its production relates to hundreds and thousands of reactions and its precursor relates to NO_x and hundreds of VOCs. It could lead to certain uncertainties when we tag precursor emitting locations of O_3. Li et al. (2008) compared these two methods for the contribution of high O_3 concentration regions in China and found that the difference could be as much as 30%.

Based on the average of all grid cells in the receptor region, we compute the contribution of source region EUR and NAM transport to receptor region EA and compare the results with Fiore et al. (2009) study which is cited in HTAP report. When extrapolated to a 100% source contribution, as shown in Fig. 8b, the Fiore et al. (2009) results suggest that EU and NA contribute from 0.5-2.8 ppb and 0.3-1.4 ppb to surface O_3 in EA in MAM and JJA, respectively. NAM's contribution to EA in our results are consistently lower, but the annual results are within the range given by other studies which Fiore et al. (2009) cited in their Fig. 11. There are many possible reasons for this difference. Their results are multi-model estimates and model horizontal resolution ranged from 5°×5° to 1° ×1°. The different methods are used. We have considered the amounts O_3 produced in source region

which is direct transport and have neglected the quantities of O_3 produced during long range transport. For example, in our study, OCN contributed about 19.5% to surface O_3 concentration in NAM and this O_3 produced in OCN and transported to NAM. EA contributed only 3.6% to NAM which means that O_3 produced in EA transports to NAM and makes a 3.6% contribution. It's lower than realistic transport contribution because many contributions has been attributed to OCN due to the precursors could transport to OCN under the control of westerly winds, react and produce O_3 on OCN. The 19.5% contribution of OCN includes the contribution of EA's precursors transported to OCN, NAM's precursors transported to OCN, and O_3 local produced due to precursors emitted from traffic source in OCN. As shown in Fig. 8a, in receptor region NAM, although most of our results are within the range, we still underestimate the real contribution from EA to NAM. Here we propose an indirect evaluation method to roughly estimate the maximum contribution of EA transport to NAM. Considering the NO_x from EA transported to OCN, we find a location where EA's contribution to surface NO_x in OCN decreases and starts to be lower than 5%, then OCN local contributions to surface O_3 in the west of this location are all simply attributed to EA due to the precursor emitted in EA and transport to OCN. We compute the attenuation rate of O_3 transported from EA to NAM, then estimate how much O_3 that produced in OCN from precursors emitted in EA and transported to OCN can transport to NAM. The total contribution of this indirect transport and EA direct transport are considered as real contribution of EA to NAM. The new results are shown in Fig. 8a as purple dots. It can be seen that this evaluation method could influence S-R relationships a lot in DJF and MAM, and is around 1 ppb higher than the upper limit of Fiore et al. (2009). In fact, due to the traffic source in OCN and some natural emissions in OCN, the NO_x contribution standard of 5% from EA standard we select is lower, which means the results marked as purple dots are the maximum contribution from EA to NAM.
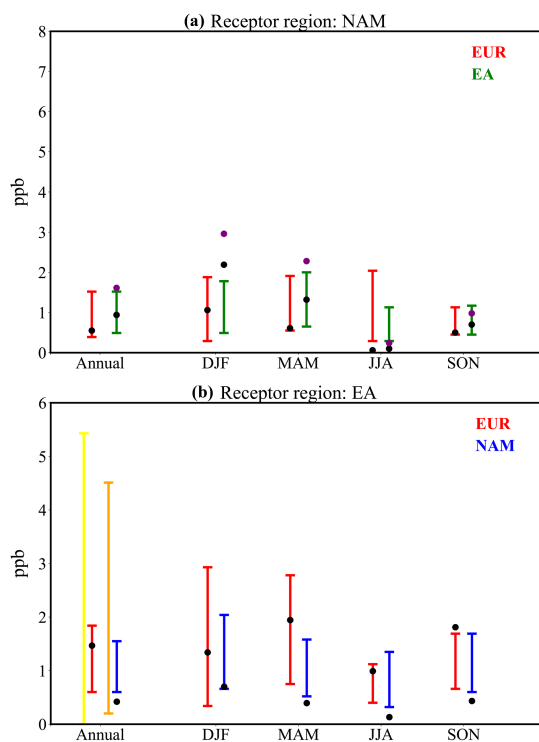
Figure 8. Annual and seasonal mean contribution to NAM and EA surface O₃ from source regions. (red vertical bars for EUR, blue vertical bars for NAM, green vertical bars for EA (Fiore et al., 2009); yellow vertical bars for EUR, orange vertical bars for NAM but from other studies which Fiore et al. (2009) cited in their Fig. 11; black dots for our results, purple dots for our new approach results). The contributions from Fiore et al. (2009) are estimated by linearly scaling the simulated surface O₃ response to the combined 20% decreases in anthropogenic emissions of NOₓ, CO, and NMVOC in the source regions to 100% decreases.

We appreciate for your positive comments and valuable suggestions to improve the quality of our manuscript.

On behalf of all the co-authors, best regards,

Qian Ye

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