Response to Community Comments#1

The impact of stratospheric intrusion on O_3 pollution is an important topic of research, and this paper presents very interesting analysis on this topic. The paper is well organized and for the most part, methodologically sound. More in-depth analysis, as suggested below, is required before this manuscript can be accepted. A minor revision is suggested with a few reasons.

Response: Thanks for the positive feedback. Please see our point-by-point responses below.

1. L24, 29, 31: ozone has been defined as O_3 on lines 16. Please check similar issues for other abbreviation terms.

Response: Thank for the suggestion. We have corrected the ozone with abbreviations in L24, L29, L31, L100, L117, L220, L223, L229, L231, L232, L239, L273, L410, L538, L539, L544, L547 in the revised manuscript.

2. L42: I disagree with the author's viewpoint that "Stratospheric intrusions and photochemical production are two major contributors to tropospheric ozone". Regional transport can include emissions from urban areas, industrial zones, or other regions that may contain ozone precursors or other compounds that affect ozone formation and degradation. Therefore, considering the impact of regional transport on tropospheric ozone is crucial.

Response: Thank you for the comment. We agree with the reviewer that regional transport of ozone and ozone precursors is crucial to tropospheric ozone enhancement. The sources for ozone regional transport can be classified into stratosphere and troposphere. The ozone from the stratosphere intrusion and tropospheric photochemical ozone reaction can be further transported from local to other regions. The regional transport of ozone precursors will finally impact the local ozone through photochemical ozone production. Regional transport can be considered as the dynamic driver that contributes based on two major sources, i.e. stratospheric intrusions and photochemical production. To address the discussion of ozone contributors more comprehensively, we mentioned the influence of regional transport in the Part 4 Discussion and Conclusion in the manuscript P20L559-564: “The dynamical and chemical drivers for such long-term tropospheric changes deserve further analysis in the future. Here, we propose several mechanisms based on related research that could potentially contribute to observed tropospheric O_3 enhancements in East Asia. Regional transport is one important contributor to tropospheric O_3 enhancement. Compared with the other two Japanese sites, Naha, to the east of China, is susceptible to regional transport of air pollution from China. The prevailing westerly winds bring O_3-enriched air from eastern China to Naha, resulting in a substantial increase of O_3 from the middle to upper troposphere”.

3. L168-169: Is a horizontal resolution of 2.8°× 2.8° too coarse?

Response: Thank the reviewer for the comment. In the study, we used the global chemistry-climate model EMAC to simulate the 40-year changes of tropospheric ozone. The model considered the interaction of chemistry and dynamic processes between the surface and the middle atmosphere. It is always good to have simulation results with fine time resolution. However, it would be computationally expensive to run such a long-term hindcast simulation with finer spatial resolution on a global range. The current spatial resolution for the simulation is the

trade-off between performance and computational capacity. According to the evaluation results in 3.2.1, the EMAC model reasonably simulates the spatial and temporal variations in tropospheric ozone as compared to the ozone observations at the four sounding sites.

4. L185-186：What is the loss rate of O_3S tracer? Please provide a calculation method.

Response: Thank you for the question. The stratospheric ozone tracer O_3S is destroyed in the troposphere as O_3 (Roelofs and Lelieveld, 1997; Jöckel et al., 2006). The total chemical loss rate of O_3S in the troposphere is given by:

$$O_3 + OH \rightarrow O_2 + HO_2 \quad (1)$$

$$O_3 + HO_2 \rightarrow 2O_2 + OH \quad (2)$$

$$O_3 + h\nu \rightarrow O(1D) + O_2 \quad (3)$$

$$O(1D) + H_2O \rightarrow 2OH \quad (4)$$

$$L_{O_{3s}} = k_1[OH] + k_2[HO_2] + \frac{J_3 k_4 [H_2O]}{k_2[M] + k_4[H_2O]}$$

In which $\boldsymbol{k}$ and $\boldsymbol{J}$ values represent the reaction rates and photodissociation coefficients, respectively. The square brackets indicate concentrations. The dry deposition of O_3S is calculated the same as O_3.

5. L191-193: This looks pretty strange. Considering that the tropopause height decreases with increasing latitude, the author defines 200 hPa as the upper troposphere for Hong Kong and NAHA, while defining 400 hPa as the upper troposphere for Tsukuba and Sapporo. However, the author defines both mid-level and lower-level troposphere as 500 hPa and 850 hPa for all four sites.

Response: Thank you for the comments. We agree with the reviewer that choosing the same model pressure layers for the middle and low troposphere for four sites at different latitudes can introduce uncertainties. Due to the uneven distribution of pressure layers, it's hard to accurately classify the range of upper, middle, and lower troposphere as our definition for normalized height in observations. Hence, we choose the same specific layer to simply and coherently represent the middle and lower layers in the troposphere for a better comparison with the observed ozone trends. To avoid the uncertainties induced by the layer selection in model results, we add more comparison and analysis based on the time-height cross sections of modeled ozone as well as ozone tracer, as shown in Figures 8, 9, 10, 11.

6. *Could you provide the data source of the tropopause height in Figure 2?*

Response: The tropopause height in Figure 2 is calculated by ozonesonde observations based on the World Meteorological Organization lapse rate tropopause definition. The detailed information about calculation is at P4L145-150 in the manuscript:" Due to the latitudinal differences and the seasonal variations in tropopause height across the four O_3-sounding observation sites, it is inappropriate to apply a specific height as the tropopause height. We thus employ the World Meteorological Organization lapse rate tropopause definition to calculate the tropopause height (hereafter called Z_t) for each site and O_3 profile. The Z_t is defined as the level at which the lapse rate decreases to 2 K km^{-1} or less, provided that the average lapse rate between this level and all higher levels within 2 km does not exceed 2 K km^{-1} (WMO, 1957)." We modified the title of Figure 2 and Figure 4 as "Black

dash lines indicate the tropopause heights calculated by observations according to the WMO lapse rate tropopause definition" in the revised manuscript.

7. Again, ozone has been defined!

Response: Thank you for the suggestion. We have double-checked and corrected the ozone with O_3 in the revised manuscript.

8. The uncertainty of stratospheric O_3 tagging method needs to be discussed.

Response: Thank the reviewer for the suggestion. The uncertainties of the stratospheric O_3 tagging method including: Model Uncertainties: Model uncertainties can arise from inaccuracies in representing atmospheric dynamics, parameterizations of physical and chemical processes, and spatial or temporal resolution; Chemical Kinetics Uncertainties: Chemical reactions involving O_3 can have uncertainties in their rate constants and reaction mechanisms. These uncertainties can propagate through the modeling process and affect the accuracy of ozone tagging results; Vertical Transport Uncertainties: The vertical transport of tagged O_3 in the atmosphere, particularly between the troposphere and stratosphere, is complex and can be subject to uncertainties in the representation of processes such as convection, mixing, and advection; Sensitivity to Initial Conditions: The results of ozone tagging simulations can be sensitive to the initial conditions used in the atmospheric models. Uncertainties in the initial state of the atmosphere, including ozone concentrations and meteorological parameters, can influence the simulated ozone distribution and its attribution to different sources. As for our EMAC hincast simulation, it considers the interaction of chemistry and dynamic processes between the surface and the middle atmosphere (Jöckel et al., 2016), with the specific dynamics nudging by Newtonian relaxation towards ECMWF ERA-5 reanalysis meteorological data (Hersbach et al., 2020). So the uncertainties mentioned above such as Chemical Kinetics Uncertainties, Sensitivity to Initial Conditions, and Vertical Transport Uncertainties are maximum optimized.

Since there is no such observation available for the evaluation of the O_3 tagging results. We can only infer the performance of tagged O_3 by the overall simulated O_3. In general, the EMAC model reasonably simulates the spatial and temporal variations in tropospheric O_3 as compared to the O_3 observations at the four sounding sites. Consistency between the model and observations suggests that the O_3 tagging results remain valuable despite uncertainties related to the model uncertainties. Moreover, the model can effectively be used to investigate the stratospheric ozone contributions to the tropospheric O_3 changes.

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