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Comment on acp-2021-1099

Yunting Fang (Referee)

Referee comment on "Diagnosing the stratospheric proportion in tropospheric ozone using triple oxygen isotopes as tracers" by Hao Xu et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-1099-RC2, 2022

The manuscript submitted by Hao Xu et al. reports seasonal variations in triple oxygen isotopes of tropospheric ozone for two sites in Japan, and quantified the proportion of stratosphere-troposphere transport. The authors used a multistep nitrite-coated filter-pack system, which was newly-developed, to collect air samples, for the two selected sites in Japan. They found that the two sites had a similar seasonal pattern in triple oxygen isotopic composition of tropospheric ozone in the terminal position, with averages of 37 permil in at both sites and the highest values in April. The day and night difference had also been examined for one of the two sites, and a difference of 1.4 permil was observed. Using the relationship between concentration and the triple oxygen isotopic composition for collected samples over study period, the authors identified the triple oxygen isotopic composition was 44.3 permil for stratospheric ozone and 34.8 permil for the ozone produced in troposphere, respectively. With these values, the proportion of stratosphere-troposphere transport was quantified with the simple mixing model to be 23% to 36%. The stratosphere-troposphere transport was further evidenced by a positive relationship between triple oxygen isotopic composition of tropospheric ozone and the radionuclide Beryllium-7 activities over the two study sites. The study was well designed and the manuscript was clearly written.

I have one major concern about the partition equations (equations 7 to 9) for tropospheric ozone with oxygen isotopic composition. As stated in the section 4.4, tropospheric ozone can be considered to include three components, i.e., background ozone in the troposphere, stratospheric ozone supplied to the troposphere via stratosphere-troposphere transport (STT) and the ozone produced in situ in the troposphere through photochemical reaction. However, in the equations 7 to 9, the tropospheric ozone was considered to only include the later two components. I suggest the authors to modify these equations, by first subtracting the background ozone from the troposphere ozone with results of concentration and oxygen isotopic composition in figure 4 (orange circles), then partitioning them into components with equations 7 to 9. After that, combine the results and the proportion of background ozone which is STT orignal. By so doing, it may reduce the proportion of STT and solve the discrepancy between the observation from the present study and the CHASER model.

Specific comments:

1, To check if the difference between day and night time or between two sites is statistically significant, the authors could use repeated measurement ANOVA.

2, For lines 215-218, I suggest move them into the method sections or discussion section, because they did not state any results on Beryllium-7 activities.