

Response to short comments (SCs) from the scientific community:

Thanks very much for your comments. Our response to your comments are listed as follows. There is an extensive discussion among the authors regarding how to revise the content. So the response is delayed, and we are sorry for this.

The authors may wish to refer to recent work by Schroeder et al. (2017) doi:

10.1002/2017JD026781, who did a thorough analysis of the utility of column HCHO/NO₂ ratios to indicate surface O₃. Schroeder et al. found that column HCHO/NO₂ ratios are not effective for determining surface O₃ sensitivity, as the "transition" range is certainly larger than the range of 1-2 described in Duncan et al (2010), and larger than the 1.3-2.8 used in this work.

Response: Thanks very much for sharing your recent work, in which a wonderful and thorough analysis of the utility of column HCHO/NO₂ ratios to indicate surface O₃ is presented. We have referred to your work and made a brief introduction of your finding in the revised version. What we gained from your work is that the column HCHO/NO₂ ratios can be used to indicate surface O₃ sensitivity but special cares should be taken or, in other words, whether the column HCHO/NO₂ ratios can be used to indicate surface O₃ sensitivity depends on conditions. This is because many additional sources of uncertainty (regional variability, seasonal variability, variable free tropospheric contributions, retrieval uncertainty, air pollution levels and meteorological conditions) may cause transition threshold vary both geographically and temporally, and thus the results from one region are not likely to be applicable globally.

Tropospheric ozone is not an emission pollutant, but produced by photochemical oxidation of CO, NO_x, and VOCs under certain meteorological condition. This process is complicated and thus shows regional representativeness. We quite follows your idea, and actually, the transition thresholds either estimated by you or Sillman (1995) or Martin et al. (2004a) or Duncan et al. (2010) were not straightly applied in this work. Instead, after referring to previous work by you, Sillman (1995), Martin et al. (2004a) and Duncan et al. (2010), we use a compromise (conservative) way to estimate the transition threshold applicable in Hefei, China. That is, we iteratively

altered the HCHO/NO₂ ratio threshold and judged whether the correlations/sensitivities of tropospheric O₃ to HCHO and NO₂ changed abruptly. Specifically, in order to find out the VOC-limited threshold, we first fit tropospheric O₃ to tropospheric HCHO lie in HCHO/NO₂ < 2 (a start point), and write down the correlation and slope, then we alter the threshold by a step size of 0.1 and perform the fit again, i.e., only fit the data pairs that lies in HCHO/NO₂ < 1.9, and so on. Finally, we compared all correlations and slopes, and regarded the ratio that has a sudden change as the VOC-limited threshold. The method to find the NO_x-limited threshold is straightforward. It is reasonable based on a fact that ozone production is more sensitive to VOCs if it is VOCs-limited and is more sensitive to NO_x if it is NO_x limited, which is also a common sense among you work, Sillman (1995), Martin et al. (2004a), and Duncan et al. (2010). Thus, there should be a turnover point near the threshold. This technique has been used by Su et al. (2017, DOI:10.1038/s41598-017-17646-x) to investigate the O₃-NO_x-VOCs sensitivities during the 2016 G20 conference, and can indeed reflect the overall O₃ production conditions in one region.

Furthermore, we have chosen the same upper limits for all gases and they are not equivalent to the real tropopause heights, but are about 3 km lower than the mean value which, derived from the NCEP database, is 15.1 km with a standard deviation (1 σ) of 1.1 km for Hefei. This manner not only ensured the accuracies of tropospheric O₃, CO, and HCHO retrievals, but also minimized the influence of transport from stratosphere, i.e., the so called STE process (stratosphere-troposphere exchange). On the other hand, we only selected the retrievals at the noon (around 13:35) for the O₃-NO_x-VOCs sensitivities, which can minimized the uncertainty due to temporal variability.

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