

Overview

The submitted manuscript deals with ozone vertical distribution in the lower troposphere over the Chinese megacities of Beijing and Hong Kong. I think that it is an interesting study giving a further insight into the complex meteorological and chemical phenomena associated with the ozone vertical distribution, especially using the Mixing-layer-height-referenced ozone values. I find that the manuscript is generally scientifically sound and well-presented, and, in my opinion, it deserves publication after the recommendations listed below are considered.

Reply: So glad to receive your positive and constructive comments. We have carefully considered your suggestions and comments, and made corresponding modifications and explanations.

General comments

The separation of the profiles based on Mixing-layer-height-referenced ozone into groups that are mainly influenced from the free troposphere (FTO₃-dominated) or the boundary layer (MLO₃-dominated) over the two examined Chinese megacities is very interesting. Since it is known that ozone pollution might affect large geographical areas through transport and photochemistry in combination to the fact that ozone is regarded as a priority pollutant in China, I think that it would be useful to examine the synoptic meteorological characteristics associated with each group of vertical ozone profiles, especially those influenced from the free troposphere and the boundary layer respectively. So, I would suggest plotting the corresponding composite charts of some meteorological parameters (geopotential height, vector wind, omega, specific humidity) in the boundary layer – free troposphere (850-700 hPa) of the highest ozone profiles (e.g. 10th percentile) which are either FTO₃-dominated or MLO₃-dominated respectively at least for summer, based for example on the categorization of profiles presented on Fig. 5 (e.g. see papers Kalabokas et al, ACP, 2013 and Kalabokas et al., Tellus, 2015, dealing with vertical ozone profiles in the Mediterranean).

Reply: Thanks for your suggestion regarding the examination of synoptic meteorological characteristics associated with different ozone profile patterns. Just as you mentioned, ozone pollution in China is usually a regional problem accompanied by regional-scale transport and photochemistry. In the revised manuscript, we added a description of large-scale meteorology (including geopotential height, vector wind at 850 hPa) associated with different ozone profile patterns (Fig. 8 in the revised manuscript).

You also suggest that attention needs to be paid to those profiles influenced from the free troposphere and the boundary layer respectively. However, we found that the distinct ozone profile patterns are mainly shaped by boundary layer ozone production (weak production or strong production), rather than vertical motions. In summer Beijing or autumn Hong Kong, the three ozone profile patterns are characterized by significant difference in MLO₃ concentrations but similarity in FTO₃ concentrations (Fig. 7 in the

revised manuscript). The similar FTO_3 concentrations among the different patterns indicate that downward transport of O_3 -rich air masses from upper level is not a decisive factor for the formation of different ozone profile patterns. Meanwhile, we examined the vertical velocity at 850 hPa in different ozone profile patterns. No significant differences are found for the different patterns, indicating a limited role of downward transport. So, we did not further add the composite charts of meteorological parameters in the boundary layer – free troposphere (850-700 hPa) of the highest ozone profiles (e.g. 10th percentile) as presented in references (Kalabokas et al, ACP, 2013 and Kalabokas et al., Tellus, 2015). We believe that the composite charts for each of different ozone profile patterns (Fig. 8 in the revised manuscript) are enough to describe the synoptic controls. In the revised manuscript, more attentions were paid to the examination of photochemistry-related processes, including the favorable local meteorology and regional precursor transport.

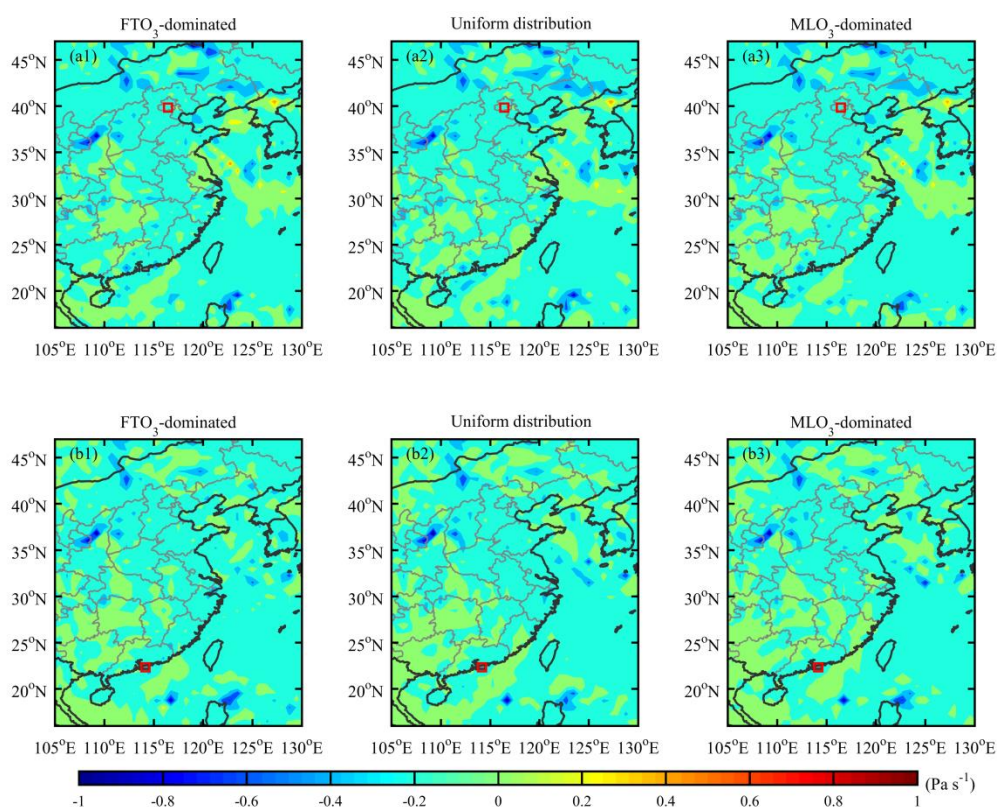


Figure S1. Compositing vertical velocity at 850 hPa according to different ozone profile patterns in (a) summer of Beijing and (b) autumn of Hong Kong. The red boxes indicate the locations of Beijing and Hong Kong.

Different from previous studies in the Mediterranean, high emissions of ozone precursors in China causes wide-range variability in MLO_3 production under the varying synoptic meteorological conditions (favorable or unfavorable). We noted that your mentioned references in the Mediterranean defined the lower troposphere as the height range of 1.5-5.0 km, excluding the atmospheric mixing layer (< 1.5 km). However, the lower troposphere defined in our study included the atmospheric mixing layer. The inclusion of

atmospheric mixing layer makes the wide-range MLO₃ variability a decisive factor for the formation of different lower-tropospheric ozone profile patterns in our study. As we mentioned in manuscript, the mixing layer top inversion is so strong to inhibit the ozone exchange between free troposphere and mixing layer. Though that vertical exchange may still works through micro-scale turbulence mixing near the mixing layer top, a complete understanding of this process is not possible given current data and knowledge.

Compared with the lower troposphere focused in this study, our previous studies had discussed the effects of vertical motions on the whole tropospheric ozone profile patterns in Hong Kong and Beijing (Liao et al., 2021; Zeng et al 2023). We found that dynamical processes (e.g., stratospheric intrusion) play a very important role in the upper troposphere. However, the stratospheric ozone intrusion had a very limited influence (or weak signal) on ozone in the atmospheric boundary layer. Recently, Chen et al. (2022) analyzed an event of rapid nocturnal O₃ enhancement (NOE) observed on 31 July 2021 at surface level in the North China Plain and proposed transport of substantial stratosphere ozone to the surface by Typhoon In-fa followed by downdraft of shallow convection as the mechanism of the NOE event. However, Zheng et al. (2023) argued that the NOE was not caused by typhoon-induced stratospheric intrusion but originated from fresh photochemical production in the lower troposphere.

Liao, Z., Ling, Z., Gao, M., Sun, J., Zhao, W., Ma, P., et al. Tropospheric ozone variability over Hong Kong based on recent 20 years (2000–2019) ozonesonde observation. *Journal of Geophysical Research: Atmospheres*, 2021,126, e2020JD033054. <https://doi.org/10.1029/2020JD033054>.

Zeng, Y., Zhang J., Li, D., Liao, Z., et al. Vertical distribution of tropospheric ozone and its sources of precursors over Beijing: Results from ~ 20 years of ozonesonde measurements based on clustering analysis. *Atmospheric Research* 284 (2023) 106610.

Chen, Z., Liu, J., Qie, X., Cheng, X., Shen, Y., Yang, M., Jiang, R., and Liu, X.: Transport of substantial stratospheric ozone to the surface by a dying typhoon and shallow convection, *Atmos. Chem. Phys.*, 22, 8221–8240, <https://doi.org/10.5194/acp-22-8221-2022>, 2022.

Zheng, X., Yang, W., Sun, Y., Geng, C., Liu, Y., and Xu, X.: Comment on “Transport of substantial stratospheric ozone to the surface by a dying typhoon and shallow convection” by Chen et al. (2022), *EGUsphere* [preprint], <https://doi.org/10.5194/egusphere-2023-2336>, 2023.

The charts might cover the geographical area between the two megacities, which is densely populated and with important air pollutant emissions. This would give a good idea of the geographical extend of the large-scale tropospheric ozone subsidence observed during summer months over Beijing or respectively to the uplifting of boundary layer air towards the free troposphere observed over Hong Kong.

Reply: Thank you for this constructive suggestion. We made the charts covering the Eastern China (Figs. 8 in the revised manuscript). However, these charts are more used to reflect large-scale transport of ozone and its precursors, rather than ozone subsidence.

I think that the above supplementary work would strengthen the results of the paper by giving a further insight of the local or regional nature of the meteorological phenomena associated with FTO₃-dominated or MLO₃-dominated ozone profiles observed over Beijing and Hong Kong. In addition to their scientific importance, these results might be useful for the development of more elaborated ozone pollution abatement strategies in the examined areas of China.

Reply: Thank you for your constructive suggestions again. The corresponding revisions according to your suggestions have substantially strengthened the results by giving a further insight of the local or regional nature of the meteorological phenomena associated with FTO₃-dominated or MLO₃-dominated ozone profiles observed over Beijing and Hong Kong. We believe that the results will be useful for the development of more elaborated ozone pollution abatement strategies in the whole eastern China.