



Comment on “Transport of substantial stratospheric ozone to the surface by a dying typhoon and shallow convection” by Chen et al. (2022)

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Abstract. Chen et al. (2022) analyzed the 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. The analysis seems to be valid in the view-point of atmospheric physics. This comment revisits the NOE phenomenon on the basis of the China National
15 Environmental Monitoring Center (CNEMC) network data ever used in Chen et al. (2022), together with the CNEMC data from Zibo (ZB), and O₃, NO_x, PAN and VOCs data from the Zibo supersite operated by China Research Academy of Environmental Sciences (CRAES). We found (a) O_x (O₃+NO₂) levels during the NOE period approaching to those of O₃ during 14:00-17:00 LT; (b) the relationship between O₃ and PAN (peroxyacetic nitric anhydride) consistent with dominance of chemical and physical processes within the boundary layer, and (c) estimated photochemical ages of air mass being
20 shorter than one day and showing no drastic increases during the NOE. We argue that the NOE was not caused by typhoon-induced stratospheric intrusion but originated from fresh photochemical production in the lower troposphere.

1 Introduction

Chen et al. (2022) reported a phenomenon of rapid nocturnal ozone (O₃) enhancement (NOE) occurred at the surface level during the night of 31 July 2021 in six cities in the North China Plain (NCP, 34-40 °N, 114-121 °E). Prior to the NOE, the
25 NCP was impacted by Typhoon In-fa, which was largely weakened by 30 July 2021. The mesoscale convective systems (MCSs) formed and passed through the NCP at night on 31 July 2021. Chen et al. (2022) concluded that the NOE phenomenon resulted from "the direct stratospheric intrusion to reach the surface" and was "induced by the multi-scale interactions between the dying Typhoon In-fa and local MCSs". The study suggested that the dying Typhoon In-fa induced stratospheric troposphere transport (STT) of O₃ followed by downdrafts of shallow convections, which resulted in "transport
30 of substantial stratospheric ozone to the surface". The relatively high O₃-low water vapor and CO (HOLWCO)



concentrations observed at some sites in the NCP and the relative variations of water vapor, O₃ profiles respectively from radiosonde data and the AIRS satellite product were used to support the conclusions.

STT processes were triggered by the large-scale circulation or synoptic-scale dynamical processes (Holton et al., 1995). A global study (Škerlak et al., 2014) shows that STT displays strong regional distribution and seasonal variations, and the NCP is not a hot region particularly in summer. STT can be an important source of tropospheric O₃, particularly in regions where the photochemistry is weak (Lelieveld and Dentner, 2000). However, tropospheric O₃ originates dominantly from photochemistry within the troposphere and photochemically produced O₃ (PPO) is the more important O₃ source not only in the middle-low troposphere (Lelieveld and Dentener, 2000; Logan, 1985) but also in the upper troposphere (Chameides, 1978; Liu et al., 1983; Jaeglé et al., 1998). Anthropogenic and natural O₃ precursors convectively transported from the surface or lower troposphere and lightning produced NO_x may involve PPO in the upper troposphere. Precursors emitted near the surface contribute largely to PPO in the surface and boundary layer, which can be transported upwards through the warm conveyor belt (Bethan et al., 1998; Cooper et al., 2002), spread in the free atmosphere and delivered over a long range by atmospheric circulations (Parrish et al., 1998). PPO in the surface boundary layer is mainly removed by NO_x titration reactions and dry deposition. The NCP is a hot region of PPO from the surface level up to 2.5 km in the summer (e.g., Ding et al., 2008) and has demonstrated rapid long-term increases in surface O₃ levels (Ma et al., 2016; Lu et al., 2018; Lyu et al., 2023).

Typhoons are tropical cyclones formed over the western North Pacific regions, which have well-organized structures of updrafts and downdrafts over hundreds and thousands of kilometres (Ahrens and Henson, 2016). A large-scale tropical cyclone with well self-organized character is able to induce dynamical processes and form an outflow layer in the upper troposphere and lower stratosphere (UTLS) and cause strong downdrafts in the periphery of the cyclone (Merrill, 1988; Ahrens and Henson, 2016). The strong air subsidence in the periphery of a typhoon can theoretically lead to the STT of O₃. It was suggested that the observed enhancement of O₃ in the middle troposphere over the Indian Ocean was caused by the STT through ageostrophic process linked to the strong tropical cyclone Marlene, which occurred in April 1995 (Baray et al., 1999). This suggestion is supported by a modeling study (de Bellevue et al., 2007). However, the idea that O₃ increases in the mid- and upper-troposphere are directly from the STT processes induced by typhoons is less supported by in-situ aircraft-borne observations by Cario et al. (2008) and the literature reviewed therein. Especially, the comparative studies on the Supertyphoon Mireille (1991) during the Pacific Exploratory Mission (PEM)-West A campaign (Newell et al., 1996a; Preston et al., 2019) and the Hurricanes Floyd (1999) and Georges (1998) in the Atlantic Ocean during their phases of intensification and weakening (Carsey and Willoughy, 2005) provided little evidence of the STT of O₃. On the other hand, the analysis of ERA5 PV (potential vorticity) and air mass with HOLWCO observed below 12 km by the In-service Aircraft for a Global Observing System (IGOS) indicated the occurrence of STT induced by typhoons (Roux et al., 2020; Z. Chen et al., 2021). However, it should be noted that atmospheric large-scale subsidence over East Asia can also be induced by the strong summer subtropical high. Photochemically aged pollution air masses may also show HOLWCO features as observed in the PEM-West A campaign (Newell et al. 1996b; Stoller et al., 1999).

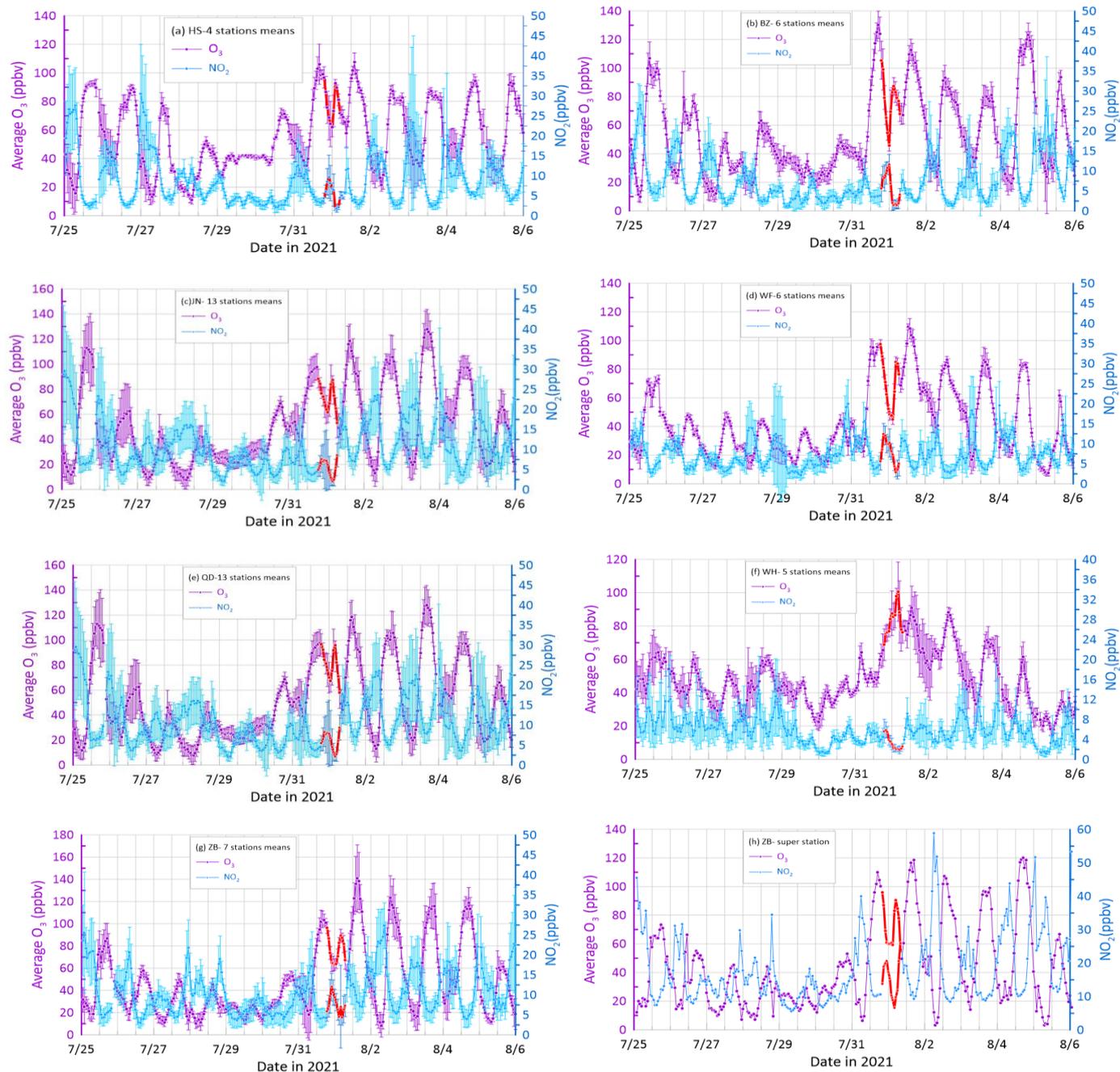


65 2 Data

Surface O₃, NO_x, and meteorology data collected in the cities Hengshui (HS), Binzhou (BZ), Jinan (JN), Weifang (WF), Qingdao (QD), Weihai (WH) and the newly added Zibo (ZB) were from the China National Environmental Monitoring Center (CNEMC) network (<https://quotsoft.net/air>). The geographical location of ZB is shown in Figure S1. In addition, hourly averages of surface O₃, PAN, NO_x, and VOCs were obtained from a supersite in ZB, operated by China Research Academy of Environmental Sciences (CRAES). Ambient O₃ and NO_x at the supersite were monitored using a Model 49i ozone analyzer and a Model 42i NO/NO₂/NO_x analyzer (both from Thermo Fischer Scientific), respectively. The analyzers were calibrated weekly. Quasi-continuous measurement of PAN was made using a gas chromatograph coupled with an electron capture detector (GC-ECD) (ZC-PANs, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences). The GC-ECD system was calibrated seasonally using PAN inline produced from CH₃COCH₃+NO reactions under UV irradiation (J. Chen et al., 2021). Samples of VOCs were taken hourly and analyzed using a coupled gas chromatograph-mass spectrometry (GC-MS) system (5800-GM, Thermo Fischer Scientific), which was calibrated monthly using standard gas mixture from Linda, containing 116 species including hydrocarbons, oxygenated and halogenated hydrocarbons.

3 Verification of results in Chen et al. (2022)

The hourly averages of surface O₃ and NO_x in the six cities (HS, BZ, JN, WF, QD, and WH) listed in Chen et al., (2022) as well as those from ZB and the ZB supersite are shown in Figure 1. The data of O₃ and NO_x from 18:00 LT on 31 July to 06:00 LT on 1 August 2021 are highlighted in red lines. Although PPO was not obvious during 29-30 July due to the weather conditions, the diurnal variations of O₃ and NO_x on most days displayed typical features being controlled by the PPO process, with O₃ maxima and correspondingly NO_x minima during 14:00~17:00 and rapid nighttime O₃ decreases due to substantial NO_x titration reactions. As reported by Chen et al. (2022), a clear NOE was observed during the night of 31 July in HS, BZ, JN, WF, and QD. Our data from ZB (Figures 1g and 1h) also confirm the occurrence of this NOE. However, it is noteworthy that NOE events occurred not only during the night of 31 July but also during some other nights in these cities with O₃ enhancement of 5-20 ppbv. The frequency of NOE was highest in WH, in details: O₃ increased from 24±8 ppbv at 22:00 LT on 27 July to 46±21 ppbv at 01:00 LT on 28 July; from 23±5 ppbv at 23:00 LT on 29 July to 40±4 ppbv at 04:00 LT on 30 July; from 43±2 ppbv at 00:00 LT to 57±3 ppbv at 04:00 LT on 31 July; from 54±19 ppbv at 00:00 LT to 66±5 ppbv at 02:00 LT on 2 August; from 42±17 ppbv at 00:00 LT to 48±5 ppbv at 03:00 LT on 3 August; from 20±4 ppbv at 02:00 LT to 25±2 ppbv at 03:00 LT on 5 August. Other NOE events occurred, for example, on 4 August in HS, on 28 July and 5 August in BZ, on 26 July and 4 August in JN, on 4 August in WF and QD.



95 **Figure 1: Time series of hourly averages of O_3 (purple) and NO_x (blue) with one standard deviation in several NCP cities between 25 July and 5 August 2021. Data from 18:00 LT on 31 July to 06:00 LT on 1 August are highlighted in red. The multisite data from HS (a), BZ (b), JN (c), WF (d), and ZB (g) are available at <https://quotsoft.net/air> (last access: 15 April 2023; X. L. Wang, 2020). Data from the ZB supersite(h) is provided by the Chinese Academy of Environmental Sciences (CRAES).**



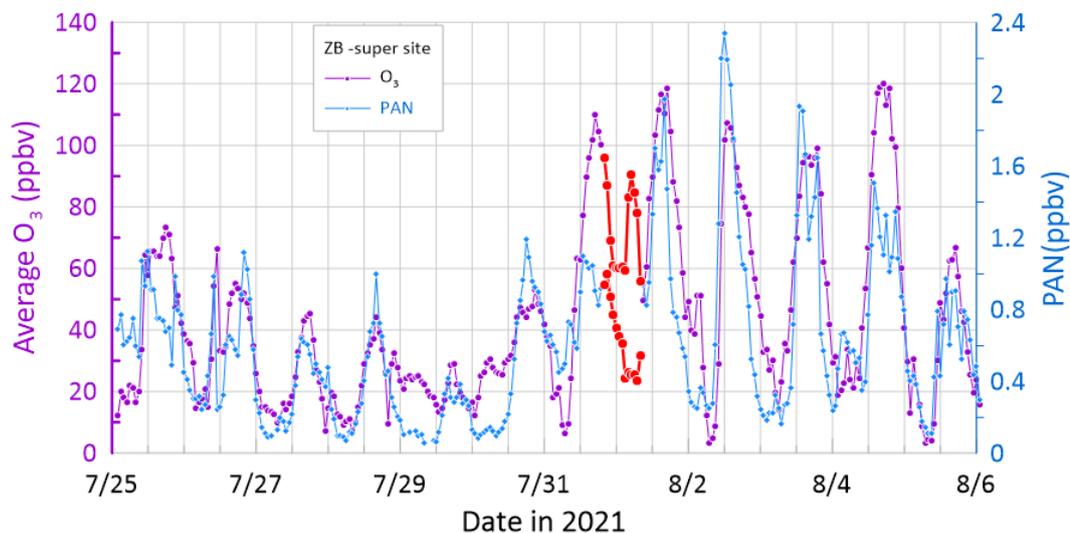
Following the method of He et al. (2022), we make comparison of O_3 averages during 14:00-17:00 LT on 31 July in the
100 above cities with the respective O_x (O_3+NO_2) averages during the periods of the maximum NOE between 31 July and 1
August (Table 1). Such comparison facilitates the judgement whether or not the NOE was caused by downward mixing of air
in the residual layer into the nocturnal boundary layer. It can be seen that, except for WH, the O_x averages approach to or
obviously lower than the respective daytime O_3 averages during 14:00-17:00 LT. In the mega-cities QD and JN, the average
levels of O_x during the maximum NOE were nearly the same as those of daytime O_3 , while O_x in the other cities (excluding
105 WH) was at least a few ppbv lower than daytime O_3 .

Table 1: Averages of surface O_3 during 14:00-17:00 LT and O_x during the periods of the maximum NOE in the night from 31 July to 1 August 2021.

Sites	NOE time	Mean $O_3 \pm \text{std}$ (ppbv)	Mean $O_x \pm \text{std}$ (ppbv)
HS	01:00-02:00	101.7 \pm 3.5	93.1 \pm 2.2
BZ	01:00-03:00	124.1 \pm 5.4	94.6 \pm 2.3
JN	02:00-03:00	96.1 \pm 1.6	96.6 \pm 2.2
WF	04:00-05:00	92.5 \pm 4.8	88 \pm 0.3
QD	02:00-03:00	96.1 \pm 1.6	96.6 \pm 2.2
WH	02:00-03:00	59.2 \pm 7.1	101.3 \pm 1.9
ZB	02:00-05:00	102.6 \pm 1.4	90.9 \pm 4.2
ZB-Supersite	02:00-05:00	104.2 \pm 4.3	93.8 \pm 3.0

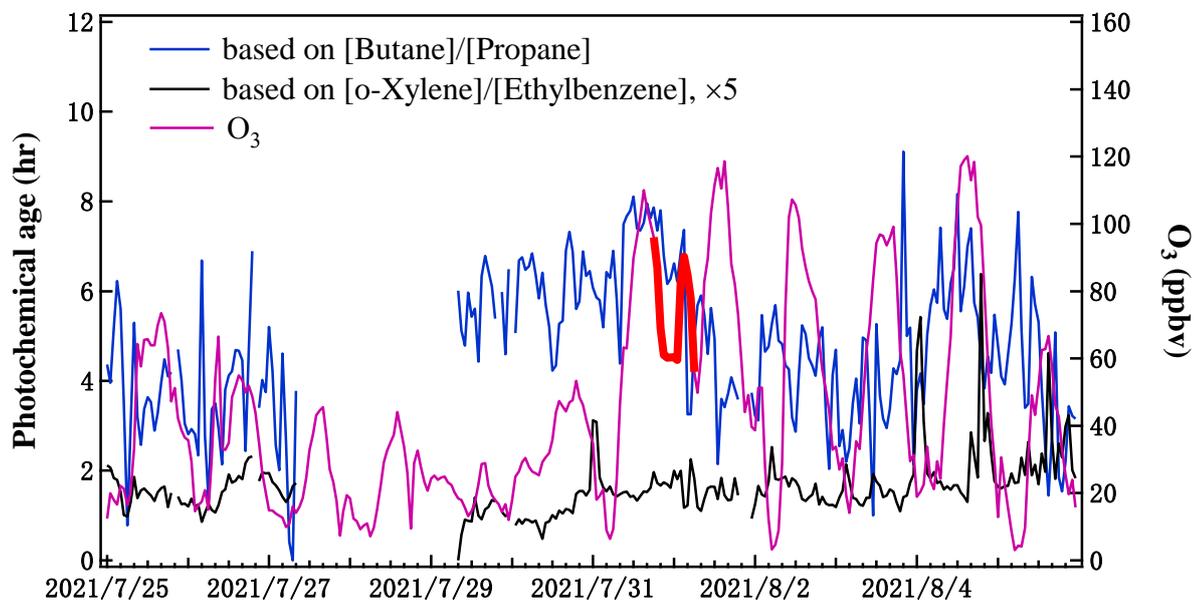
WH is a relatively smaller city in the tip of the Shandong Peninsula. The higher O_x concentration for WH in Table 1 was
110 probably related to the influence of the diurnal alternations of land-sea breezes. During the daytime (particularly afternoon),
when sea breeze dominates, PPO is significantly diluted by cleaner air from the marine boundary layer. The daily maximum
of O_3 in WH is generally observed between 11:00 LT and 13:00 LT, rather than between 14:00 LT and 17:00 LT. If the O_3
average for WH in Table 1 were replaced with that from 11:00 LT to 13:00 LT on 1 August (80 \pm 2.0 ppbv), then the
difference between O_x and O_3 would be reduced to only 13.00 ppbv. At night, when the land breeze dominates, the near
115 surface level of WH is usually controlled by divergence, which induces downdraft from the residual layer, transports
daytime PPO residing in the residual layer to the surface, and resulted in the NOE. This is the main reason of highly frequent
NOE emerging in WH.

To further understand the source characteristic of NOE, the hourly average concentrations of surface PAN (peroxyacetic
nitric anhydride) and O_3 observed at the ZB supersite are displayed in Figure 2. It can be seen that the variations of PAN and
120 O_3 were in phase and the concentrations of both gases were well correlated ($r^2=0.60$, $p<0.0001$, $n=283$), indicating that the
variations of PAN and O_3 were driven mainly by chemical and physical processes within the boundary layer. The maximum
 O_3 from 02:00 LT to 05:00 LT on 1 August was 88 \pm 5 ppbv, matching with PAN of 0.44 \pm 0.02 ppbv, which was still
significantly higher than those lowest values in nighttime of other dates and did not show sign of significant impact of STT.



125 **Figure 2. Time series of hourly average of O₃ (purple) and PAN (blue) at the ZB supersite between 25 July and 5 August 2021. Data from 18:00 LT on 31 July to 06:00 LT on 1 August are highlighted in red.**

Photochemical ages of air masses arrived at the ZB supersite from 25 July to 5 August 2021 were estimated based on hourly VOCs (volatile organic compounds) measurements. The methodologies of the estimation are given in detail in Text S1. Figure 3 shows two sets of estimated photochemical ages based on ratios [Butane]/[Propane] and [o-Xylene]/[Ethylbenzene], respectively, together with hourly O₃ concentrations from the supersite. As can be seen in the figure, photochemical ages estimated based on [o-Xylene]/[Ethylbenzene] are much shorter than those based on [Butane]/[Propane]. Since we used the observed maximum [Butane]/[Propane] and [o-Xylene]/[Ethylbenzene] in the calculations instead of the respective initial [Butane]/[Propane] and [o-Xylene]/[Ethylbenzene], the photochemical ages were underestimated, particularly those based on measurements of o-xylene and ethylbenzene, which are much more reactive than the alkanes (see Text S1). The estimated photochemical ages can be used to check the major conclusion in Chen et al. (2022) even though they could be underestimated. Here, the actual values of photochemical ages are less important than their variations during, before and after the NOE. The photochemical ages of stratospheric air masses are usually longer than one year (Diallo et al., 2012). However, all estimated photochemical ages in Figure 3, including those for the NOE period, are much shorter than one day. More importantly, the photochemical ages for the NOE from 31 July to 1 August 2021 varied roughly in the middle of all estimated ages and showed no drastic increases, which would be expected if the surface air had contained a significant fraction of stratospheric air. Therefore, our observation-based calculations of photochemical ages do not support the conclusion made by Chen et al. (2022) about the mechanisms of the NOE. It is very likely that the NOE was not a result of typhoon-induced stratospheric intrusion, instead, it was originated from fresh photochemical production of O₃ in the lower troposphere.



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Figure 3. Variations of estimated photochemical ages of air masses arrived at the ZB supersite and the O₃ mixing ratio (purple). The O₃ data from 18:00 LT on 31 July to 06:00 LT on 1 August 2021 are highlighted in bold red line. The photochemical ages were estimated based [Butane]/[Propane] (blue) and [o-Xylene]/[Ethylbenzene] (black), respectively.

4 Discussion and Conclusions

150 The NOE event presented by Chen et al. (2022) was actually one of the normal cases of NOE associated with the compensatory downdrafts induced by convection cells (Betts et al., 2002). Occurrences of NOE had been reported in the NCP during the summer (e.g., Ma et al., 2013; Jia et al., 2015). Similar phenomena were found in southern China (He et al., 2021), in the Bay of Bengal in India (Sahu and Lal, 2006) and even in Amazonia (Betts et al., 2002). The daytime PPO is transported upward to and resides in the residual layer, while surface O₃ is largely removed at night mainly by NO_x titrations, forming a large positive lower tropospheric gradient of O₃ from surface to the residual layer during the night and early morning period, as often reported in the literature (e.g., Ma et al., 2013; Jia et al., 2015; Wang et al., 2017; Tang et al., 2017; Zhao et al., 2019; Zhu et al., 2020). The positive gradient of O₃ can be strongly disturbed by nighttime convective processes or low-level jets and the compensatory downdrafts in convection systems can cause NOE events, as reported for a Amazonia site (Betts et al., 2002) and a NCP site (Jia et al., 2015) and systematically summarized in He et al. (2022).

160 Previous investigations on surface O₃ enhancement associated with passage of typhoons revealed two possible mechanisms: i) stratospheric O₃ is ultimately transported to the surface level after typhoon-induced STT (Jiang et al., 2015; Wang et al., 2020; Zhan et al., 2020; Chen et al., 2022; Meng et al., 2022); ii) formation and accumulation of O₃ as well as emissions of O₃ precursors in the boundary layer are promoted under meteorological conditions accompanying strong atmospheric subsidence in typhoon periphery (Hung and Lo, 2015; Shao et al., 2022; Wang et al., 2022). Directly before the



165 NOE event reported by Chen et al. (2022), the photochemical formation of O_3 in the NCP was obviously intensified after a few days of weakening (Figure 1). The O_3 -rich air spread within the boundary layer during the daytime of 31 July and remained in the residual layer at night. Given the favorable thermal-dynamical condition like MCSs, the PPO in the residual layer could easily be conveyed downward to the surface, leading to NOE in the surface layer, as also shown in other studies (Shu et al., 2016; Qu et al., 2021; Ouyang et al., 2022; He et al., 2022). Our analysis supports the conclusion that this NOE
170 event was caused by rapid downward transport of daytime PPO residing in the residual layer.

The STT of O_3 is often observed in the free troposphere through balloon or aircraft-based observations and air masses associated with identified STT of O_3 exhibit usually the feature of HOLWCO. However, air mass with HOLWCO feature in free troposphere do not necessarily mean that the O_3 enhancement is originated from the stratosphere (Stoller et al., 1999). For those observations made at a high mountain site (Izaña, 28°18'N, 16°30'W, 2370 m a.s.l.), even with stratospheric tracers
175 (such as 7Be), the contribution of PPO to the rise of O_3 at surface level was important and the stratosphere seemed not to be a direct source (Prospero et al., 1995; Graustein and Turekian, 1996). Although the air with HOLWCO, induced by katabatic winds, was observed at the base camp of Mount Everest (about 5000 m a.s.l.), the source of O_3 from the stratosphere was not confirmed (Zhu et al., 2006). Simultaneous observations of O_3 and PAN at the Namco (4545 m a.s.l.) in Tibet captured air masses with high O_3 and low water vapor, which were accompanied with increases of PAN, suggesting that PPO during the
180 long-range transport might be one of the major sources of elevated O_3 (Xu et al., 2018). These examples show that even at high altitude sites, HOLWCO phenomena may not be caused by STT. Therefore, more cautions should be taken when attributing HOLWCO events at low altitude sites to stratospheric impact, whether or not there was an influence from a typhoon or other synoptic system. It is suggested that if possible, each case should be verified by analyzing both physical and chemical processes before making a conclusion.

185 **Data availability.** The data used in this study are available upon request to the corresponding author (xiaobin_xu@189.cn).

Author contributions. XZ and XX designed the research. WY was responsible for the observations at the Zibo supersite. CG and YL validated the data from the supersite. XZ, YS and XX performed the data analysis. XZ and XX prepared the manuscript.

Competing interests. The contact author has declared that none of the authors has any competing interests.

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