# Influence of convection on the upper tropospheric $O_3$ and $NO_x$ budget in southeastern China

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Abstract. Thunderstorms can significantly influence the air composition via strong updraft and lightning nitrogen oxides  $(LNO_x)$ . In this study, the ozonesondes and TROPOMI nitrogen dioxide  $(NO_2)$  observations for two cases are combined with model to investigate the effects of typical strong convection on vertical redistribution of air pollutants in Nanjing, southeastern China. The ozonesonde observations show higher  $O_3$  and water vapor mixing ratios in the upper troposphere (UT) after

- 5 convection, indicating the strong updraft transporting lower-level airmass into the UT, and the possible downward O<sub>3</sub>-rich air near the top of UT over the convective period. During the whole convection life cycle, the UT O<sub>3</sub> production is driven by the chemistry (> 87 %5–10 times the magnitude of dynamic contribution) and reduced by the LNO<sub>x</sub> (-40 %). Sensitivity tests demonstrate that neglecting LNO<sub>x</sub> in standard TROPOMI NO<sub>2</sub> products causes overestimated air mass factors over fresh lightning regions and the opposite for outflow and aged lightning areas. Therefore, a new high-resolution retrieval algorithm
- 10 is applied to estimate the  $LNO_x$  production efficiency. Our work shows the demand for high-resolution modeling and satellite observations on  $LNO_x$  emissions of both active and dissipated convection, especially small-scale storms.

### 1 Introduction

Convection can transport the surface pollutants and moisture from the planetary boundary layer to the upper troposphere (UT) in a short time, where the gaseous pollutants have a longer lifetime due to the slower reaction rates, except for photolysis, in

15 the colder environment (Dickerson et al., 1987). As trace gases remain for more than one week in the UT, they are distributed by the upper level winds around the globe (Ridley et al., 2004). Meanwhile, the vertical profiles of trace gases are reshaped by the updraft and downdraft on timescales of hours (Barth et al., 2019)(Huntrieser et al., 2016; Barth et al., 2019). On a global scale, the chemical reactions of transported ozone  $(O_3)$  and its precursors can increase the amount of UT  $O_3$  (Lawrence et al., 2003; Murray, 2016).

- Nitrogen oxides (NO<sub>x</sub>), volatile organic compounds (VOC), and hydrogen oxide radicals (HO<sub>x</sub>) are the three main O<sub>3</sub> precursors, which can be uplifted or produced by thunderstorms (Bozem et al., 2017). Lightning produced nitrogen oxides (LNO<sub>x</sub>) is the dominant natural source of UT NO<sub>x</sub>, contributing as much as 35–45 % of global free-tropospheric ozone (Allen et al., 2010; Liaskos et al., 2015). Several lightning parameterizations have been developed for chemistry transport and climate models to evaluate the relationship between LNO<sub>x</sub> and other trace gases (Murray et al., 2012; Gordillo-Vázquez et al., 2019; Luhar et al., 2
- <sup>25</sup> Therefore, the precise estimation of  $LNO_x$  is crucial for the global  $O_3$  trend and feedback between lightning and climate change (Finney et al., 2016; Chen et al., 2021). Globally, the  $LNO_x$  is estimated as 2–8 Tg N yr<sup>-1</sup>, which is substantially less well quantified than the flash rate (Schumann and Huntrieser, 2007).

The large uncertainty of  $LNO_x$  estimation might be reduced by using cloud-resolving chemistry models in combination with satellite and aircraft observations. It is beneficial to take advantage of sonde and satellite observations for exploring the

- 30 convection effects, especially for the estimation of the  $LNO_x$  production efficiency (PE). Recently, many studies have used different satellites to quantify the  $LNO_x$  PEs (Beirle et al., 2009; Pickering et al., 2016; Zhang et al., 2020). Two main methods have been proposed to distinguish  $LNO_x$  from the  $NO_2$  background pollution: 1) subtracting the weighted temporal average  $NO_2$  of areas with few flashes before the satellite passing time (Pickering et al., 2016; Bucsela et al., 2019; Allen et al., 2019; Lapierre et al., 2020) and 2) directly using customized lightning air mass factors (AMFs) for each convection event (Beirle
- et al., 2009; Zhang et al., 2020). Recently, Allen et al. (2021) proves the potential of deriving  $LNO_x$  PEs by the geostationary lightning instruments (e.g., Lightning Mapping Imager (LMI; Yang et al., 2017), Geostationary Lightning Mapper (GLM; Rudlosky et al., 2019)), and NO<sub>2</sub> observations such as Tropospheric Emissions: Monitoring of Pollution (TEMPO; Chance et al., 2019).

Furthermore, aircraft observations and chemical models indicate that the transport from the stratosphere to the troposphere

- 40 can also increase the UT O<sub>3</sub> besides the chemical production from  $LNO_x$  (Pan et al., 2014). As revealed in the modeled Deep Convective Clouds and Chemistry 2012 Studies (DC3) and mesoscale convective system simulations, the compensation of subsidence and differential advection beneath the convective core can lead to the anvil wrapping effects (Phoenix et al., 2020) (Huntrieser et al., 2016; Phoenix et al., 2020). The different mechanisms of stratosphere–troposphere exchange and the effects on the tropospheric chemistry have been discussed in Holton et al. (1995) and Stohl (2003).
- 45 At present, most aircraft observations and model simulations of convection effects focus on the tropics or the United States (Vaughan et al., 2008; Barth et al., 2019). Little is known about the role of convection in southeastern China (Murray, 2016; Guo et al., 2017), where thunderstorm and lightning have increased significantly by urbanization during recent decades (Yang and Li, 2014)(Yang and Li, 2014; Pérez-Invernón et al., 2021). This is likely due to the increasing aerosol concentration, which can invigorate storms in a moist and convectively unstable environment (Koren et al., 2008; Rosenfeld et al., 2008; Tao et al., 2012)
- 50 . In this study, we combine ground observations and model simulations to investigate the origin of higher UT  $O_3$  and water vapor mixing ratio (Qv) after convection, and we try to distinguish the contributions of physical processes, chemical reactions and LNO<sub>x</sub>. For the first time, the TROPOMI (TROPOspheric Monitoring Instrument )-(TROPOMI; Veefkind et al., 2012)

 $NO_2$  observations are used to identify  $LNO_x$  PEs in southeastern China. Section 2 describes the used datasets with a brief introduction of the cloud-resolved chemistry model and the  $LNO_x$  retrieval method. Section 3 evaluates the model simulations

and Sect. 4 analyzes the physical and chemical effects of convection are analyzed in Sect. 4 and 5. We apply a new a priori NO<sub>2</sub> profiles into the retrieval algorithm to explore the sensitivity of AMFs to  $LNO_x$  in Sect. 6. Conclusions are summarized in Sect. 7.

#### 2 Datasets

#### 2.1 Ozonesonde Datadata

- 60 Five ozonesondes were launched from the Nanjing National Reference Climatological Station (31.93° N, 118.90° E) on 25 July 2019 and 01 September 2020, both days experienced strong convection. Both pre-convection and during-convection/post-convection campaigns were designed to investigate the convection effects. The convection and ozonesonde trajectories are illustrated in Fig. 1a and b.
- Three Institute of Atmospheric Physics (IAP) ozonesondes had been launched near the airmass convection that developed on 25 July 2019. The IAP ozonesonde uses an electrochemical concentration cell (ECC). The complete parameters and performance are described in Zhang et al. (2014). Its average bias is less than 0.3 mPa from the surface up to 2.5 km, close to zero below 9 km, and less than 0.5 mPa between 9 km and 18 km. The first IAP ozonesonde was launched at 05:35 UTC on a sunny day (23 July) and the other two were at 05:10 UTC (pre-convection) and 06:35 UTC (post-convection) on 25 July. Because of water leakage, the pre-convection one lost signal just a few seconds after the release, and instead the ozonesonde launched on
- 70 23 July is chosen. Although the time interval is two days, the largest relative difference of forecast  $O_3$  profiles above 10 km is usually smaller than 25 % (Fig. S2), therefore the daily variation cannot explain the observed difference of more than >65 %.

The two Vaisala ECC ozonesondes were launched successfully at 23:45 UTC 31 August (pre-convection) and 06:10 UTC 01 September (during-convection), respectively, following the standard manual to ensure the precision is better than 5 % and the accuracy is within  $\pm(5-10)$  % below 30 km (Smit et al., 2007). The captured squall line was developing from the convergence

75 of cold air and typhoon Maysak's outer region circulation. Note that the during-convection ozonesonde entered directly into the cloud, providing a unique opportunity of exploring the ozone affected by the convective clouds.

#### 2.2 Lightning Datadata

Three lightning datasets were used in this study: the China National Lightning Detection Network (CNLDN; Yang et al., 2015), the Earth Networks Total Lightning Network (ENTLN; Marchand et al., 2019), and the World Wide Lightning Location

80 Network (WWLLN; Rodger et al., 2006). The detection efficiency (DE) of cloud-to-ground (CG) flashes is about 90 % for the CNLDN data in Jiangsu province (Li et al., 2017a) while ENTLN and WWLLN detect both intro-cloud intra-cloud (IC) and CG flashes with specific detection frequency (1 Hz–12 MHz for ENTLN and 3–30 kHz for WWLLN). To increase the lightning data coverage in our study, the CG flashes of three datasets are combined.



**Figure 1.** (a, b) The convection detected by the FY-4A Advanced Geostationary Radiation Imager (AGRI) visible channel (0.65  $\mu$ m) field at the time when the post-convection/during-convection ozonesondes reached around 10 km. The pre-convection ozonesonde trajectories are colored blue while others are in red. The white star symbol stands for the observation station and the thin yellow lines are the TROPOMI swath pixels. (c) and (d) are the observed O<sub>3</sub> (red) and Qv (blue) profiles in the pre-convection (dashed) and post-convection/during-convection (solid) periods. The initial (dashed) and simulated post-convection or during-convection (solid) O<sub>3</sub> profiles are in green. The dark gray shading is the 50 % confidence interval while the light one is the 90 % confidence interval. The gray lines are the lapse rate tropopauses.

In the ENTLN data, groups of pulses are classified as a flash if they are within 700 ms and 10 km. Both strokes and lightning flashes composed of one or more strokes are included in the preprocessed data obtained from the ENTLN. The detailed processing algorithm of the WWLLN is given by Rodger et al. (2004). The WWLLN strokes and pulses are combined with ENTLN into one dataset (ENGLN) within 10 km and 0.7 s as mentioned in Virts and Goodman (2020). Then, the ENGLN data are combined with the CNLDN dataset within. To increase the lightning data coverage in our study, the CG flashes of ENGLN and CNLDN datasets are combined using spatial and temporal clustering criteria of 10 km and 0.5 s (Zhao et al., 2020). The

- 90 Although CG detection efficiency for ENTLN and WWLLN of ENGLN is not known for this region due to a lack of validation data. However, merging these three datasets should provide a sufficiently high CG flash detection efficiency for this analysis. Because the IC DE of all these lightning data is low in China, we conservatively used use the merged CG data with a constant ratio (IC/CG ratio of 3:1) of IC and CG based on Wu et al. (2016) and Bandholnopparat et al. (2020). IC data will become more accurate if more Chinese total lightning networks, such as Beijing Lightning Network (BLNET: Srivastava et al., 2017), 95 are available.

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#### 2.3 TROPOMI Datadata

On 13 October 2017, the TROPOMI on Sentinel-5 Precursor satellite was launched successfully (Veefkind et al., 2012). For the current study, we used the Royal Netherlands Meteorological Institute (KNMI) standard product v2.1-test as input to our  $LNO_{x}$  retrieval algorithm. A spike removal is included in the product to better deal with the detector saturation and blooming effects, which enables more valid data over the bright clouds generated by convection (Ludewig et al., 2020; van Geffen et al., 2021). Each pixel includes a slant column density quality flag (no2 scd flag=0, see Appendix A) that can be used to get the data without known retrieval errors (Allen et al., 2021; van Geffen et al., 2021).

The official NO<sub>2</sub> columns are retrieved using the near-ultraviolet and visible (UV-VIS, 405–465 nm) spectrometer backscattered solar radiation measurements on the TROPOMI (van Geffen et al., 2015). The retrieval consists of three main procedures for each measured Level-1b spectrum:

1) Total NO<sub>2</sub> slant column density (SCD) is determined by the DOAS method.

2) The stratospheric and tropospheric SCDs are separated by data assimilation of slant columns in the Tracer Model, version 5, tailored for the application of satellite retrievals (TM5-MP; Williams et al., 2017).

3) The stratospheric and tropospheric  $NO_2$  vertical column density (VCD) are obtained via the air mass factor (AMF) look-up tables (Lorente et al., 2017). 110

We replaced the tropospheric AMF (AMF<sub>trop</sub>) with a new AMF called AMF for  $LNO_x$  (AMF<sub>LNOx</sub>) to derive the tropospheric  $LNO_x$  vertical column density (VCD<sub>LNOx</sub>). The concept of AMF<sub>LNOx</sub> inherits from the tropospheric AMF (AMF<sub>trop</sub>) derived by a function of several parameters (solar zenith angle, viewing zenith angle, relative azimuth angle, surface albedo, surface pressure, cloud fraction, cloud height, and a priori trace gas profile)and. Briefly, the numerator is the modeled tropospheric

115  $NO_2$  slant column density (SCD<sub>tropNO<sub>2</sub></sub>) and the denominator is the modeled VCD (VCD<sub>NO<sub>2</sub></sub> or VCD<sub>LNO<sub>2</sub></sub>). In detail, these two AMFs can be calculated as:

$$AMF_{\underline{\text{LNO}_x}} \underset{\sim}{\text{Trop}} = \frac{(1 - f_{effNO_2}) \int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p) \, dp + f_{effNO_2} \int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p) NO_2(p) \, dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p) \, dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p) \, dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p) \, dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p) \, dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p) \, dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p) \, dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p) \, dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p) \, dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p) \, dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p) \, dp}{\int_{p_{surf}}^{p_{tp}} MD_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} W_{clear}(p) \, dp}{\int_{p_{surf}}^{p_{tp}} MD_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} W_{clear}(p) \, dp}{\int_{p_{surf}}^{p_{tp}} MD_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} W_{clear}(p) \, dp}{\int_{p_{surf}}^{p_{tp}} MD_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} W_{clear}(p) \, dp}{\int_{p_{surf}}^{p_{tp}} MD_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} W_{clear}(p) \, dp}{\int_{p_{surf}}^{p_{tp}} MD_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} W_{clear}(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} MD_x(p) \, dp} \frac{(1 - f_r) \int_{p_{surf}}^{p$$

$$AMF_{LNO_x} = \frac{(1 - f_r) \int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p) \, dp + f_r \int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p) NO_2(p) \, dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p) \, dp}$$
(2)

(1)

where  $p_{surf}$  is the surface pressure,  $p_{tp}$  is the tropopause pressure,  $p_{cloud}$  is the cloud optical pressure,  $f_{effNO_2}$  is the 120 effective cloud  $f_x$  is the cloud radiance fraction in the NO<sub>2</sub> window,  $w_{clear}$  and  $w_{cloudy}$  are respectively the pressure-dependent scattering weights from the lookup table (Lorente et al., 2017) for clear and cloudy parts, and  $NO_2(p)$  is the NO<sub>2</sub> vertical profile simulated by WRF-Chem. Besides,  $LNO_x(p)$  is the LNO<sub>x</sub> vertical profile calculated by the difference of vertical profile between WRF-Chem simulations with and without lightning. All other parameters in the KNMI v2.1-test product, including the total SCD, stratospheric SCD, total VCD, stratospheric VCD, surface albedo, and scattering weights, remain 125 unchanged.

In comparison with this study, Pickering et al. (2016), Allen et al. (2019), Bucsela et al. (2019), and Allen et al. (2021) defined another AMF<sub>LNO<sub>x</sub></sub> to convert SCD<sub>tropNO<sub>2</sub></sub> to the tropospheric NO<sub>x</sub> vertical column density (VCD<sub>NO<sub>x</sub></sub>). Then, their VCD<sub>LNO<sub>x</sub></sub> can be calculated as the VCD<sub>NO<sub>x</sub></sub> subtracted by a tropospheric NO<sub>x</sub> background. Because our AMF<sub>LNO<sub>x</sub></sub> converts the SCD<sub>tropNO<sub>2</sub></sub> to VCD<sub>LNO<sub>x</sub></sub> directly, the additional estimation of background NO<sub>2</sub> is not needed for calculating LNO<sub>x</sub> PE in Sect. 6.2.

#### 130 2.4 Model Simulations

This study uses Weather Research and Forecasting model with chemistry (WRF-Chem) version 4.1.4. The initial and boundary conditions of meteorological parameters are provided by the hourly ECMWF atmospheric reanalysis (ERA5) data (Hersbach et al., 2020). Simulations are performed with one-way nesting with 75 vertical levels and a 50 hPa model top. The domain settings are illustrated in Fig. S1. The microphysical processes are computed with the WRF Single-Moment 6-class scheme

- 135 (WSM6; Hong and Lim, 2006), while the shortwave and longwave radiation is calculated by the Rapid Radiative Transfer Model for GCMs scheme (RRTMG; Iacono et al., 2008). The land surface processes are simulated by the Noah scheme (Koren et al., 1999). However, we use different planetary boundary layer (PBL) parameterizations to simulate the convection. Specifically, the 2019 case uses the Yonsei University scheme (YSU; Hong et al., 2006), while the Quasi-Normal Scale Elimination (QNSE; Sukoriansky et al., 2005) is applied to the 2020 case.
- 140 The chemical initial and boundary conditions are defined using the output from the Whole Atmosphere Community Climate Model (WACCM, https://www.acom.ucar.edu/waccm/, last access: March 22, 2022). The initial O<sub>3</sub> profile of the 2020 case is replaced by the O<sub>3</sub> profile from the ozonesonde. Anthropogenic emissions are driven by the 2016 Multi-resolution Emission Inventory for China (MEIC) version 1.3 (http://www.meicmodel.org/, last access: March 22, 2022). The Model of Emissions of Gases and Aerosol from Nature (MEGAN; Guenther et al., 2006) is used for biogenic emissions. The chemical mechanism is
- 145 the Model for Ozone and Related chemical Tracers (MOZART) gas phase chemistry and Goddard Chemistry Aerosol Radiation and Transport aerosols (GOCART) for aerosols (Pfister et al., 2011). The photolysis rates are adjusted by the presence of aerosols and clouds using the new TUV photolysis option with the scaled cloud optical depth (cloud\_fraction<sup>1.5</sup>). Note that

the bimodal profile modified from the standard Ott et al. (2010) profile is employed as the vertical distribution of LNO in WRF-Chem (Laughner and Cohen, 2017), while the  $LNO_x$  parameterization is activated as 500 mol NO per flash (Zhu et al.,

2019). The resulting lightning nitrogen monoxide (LNO) and lightning nitrogen dioxide (LNO<sub>2</sub>) profiles are defined as the

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difference of vertical profiles between simulations with and without lightning.

To simulate the convection and  $LNO_x$  realistically, a lightning data assimilation (LDA) technique is applied to WRF-Chem. The details of the LDA technique can be found in Fierro et al. (2012) and Li et al. (2017b) and are briefly illustrated here. The water vapor mass mixing ratio is increased at constant temperature layers in columns where flashes occur:

# 155 $Q_v = AQ_{\text{sat}} + BQ_{\text{sat}} \tanh(CX) \left[1 - \tanh\left(DQ_q^{\alpha}\right)\right]$ (3)

where Q<sub>sat</sub> is the water vapor saturation mixing ratio (g kg<sup>-1</sup>), Q<sub>g</sub> is the graupel mixing ratio (g kg<sup>-1</sup>) and X is the flash rate. In our simulations, the layer between 263.15 K and 290.15 K is chosen to let the convection root in the PBL quickly as the Q<sub>v</sub> in the lower troposphere is the deeper layer (Marchand and Fuelberg, 2014; Finney et al., 2016; Li et al., 2017b). Parameter settings follow Li et al. (2017b): A = 0.94, B = 0.2, C = 0.001 and D = 0.25 and α = 2.2. The resampled total lightning flashes data are read through the Auxiliary Input Stream of WRF every 10 minutes. For example, if the beginning time of LDA is 05:00 UTC with a time step of 10 min, all flashes in a specific grid between 05:00 and 05:10 UTC are summed as the contribution during this period. At the next time step, the flashes are classified as the next new group. Therefore, the flash count is the flash rate density (units: flashes 10 min<sup>-1</sup> dx km<sup>-1</sup> dy km<sup>-1</sup>), where dx and dy are the resolutions of model grids in the x and y direction, respectively. This leads to flash rate densities that are the same through all nested domains as done in Fierro et al. (2012) and Li et al. (2017b), and these are used as the flash counts in the WRF-Chem directly following the method employed in CMAQ (Kang et al., 2019b, a, 2020)(Kang et al., 2019a, b, 2020).

#### 3 Model Evaluation

Compared with the radar observation, the simulated initiations of convection are ahead by 60 minutes and 30 minutes for the 25 July 2019 and 01 September 2020 cases, respectively (Fig. 2 and Fig. 3). The lightning assimilations are applied with the same time step forwards. For comparisons below, we choose the matched stages instead of the same time.

The airmass convection on 25 July 2019 was initialized as isolated cells. The WRF-Chem reproduces the position and intensity of isolated convections at the initial stage (Fig. 2a and 2d). At 05:40 UTC, the cells were presented with the northeast-southwest orientation and the column maximum radar reflectivity (CRF) reached 60 dBZ (Fig. 2b), which is stronger than the simulated convection (CRF = 55 dBZ, Fig. 2e). The vertical cross section of simulated radar reflectivity across the core of

175 cells is compared with the observation (Fig. S3). Although there were much missing data caused by the long distance between convection and radar, the horizontal and vertical structures of isolated cells are roughly shown without artificial interpolation. While the simulated 45 dBZ contour extends to 12 km, the observed one only reaches 10 km because of reduced data quality above 10 km.

The squall line on 25 July 1 September, 2020 was born in the north, strengthened, and moved towards the observation site (Fig. 3). The strongest convective stage with a CRF of 60 dBZ was at 05:50 UTC which is the TROPOMI overpass time (Fig. 180 3b and 3e). Although the highest level reached was lower than that of 2019 case, the reflectivity in the lower troposphere (2–8 km) was larger and broader (Fig. S4). Note that the simulated dissipated cells deviate from the radar observation and this leads to the region for the ozonesonde comparison moving to the west of station (Fig. 3c and 3f).



Figure 2. Observed radar composite reflectivity at (a) 04:20 UTC, (b) 05:40 UTC, and (c) 06:50 UTC. (d-e) WRF-Chem simulated composite reflectivity one hour before the radar observation times. The AB solid lines in (b) and (e) are cross section lines for Fig. S3. The CD solid line in (e) is the cross section line for Fig. 4b. The black rectangle is the region for the comparison with ozonesonde.

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The measured  $O_3$  and Qv profiles at different convection stages are shown in Fig. 1c and d. Generally, the observed UT  $O_3$ and Qv are higher with convection, while the largest enhancements are between 10 km and 16 km. However, the 2020 case showed a larger increase in the lower troposphere (2–8 km, LT). Additionally, a two-valley shape of  $O_3$  profile exists in both cases but at different levels: 2/8 km for the 2019 airmass and 4/10 km for the 2020 squall line. Although the WRF-Chem model tends to underestimate the  $O_3$  concentration in the LT and UT for the 2019 and 2020 cases, respectively, it can reproduce the detailed  $O_3$  structures and provide the opportunity to analyze the mechanisms of convection.

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Three possible sources can explain the enhancements of  $O_3$  in the UT: convective transport, chemical production, and  $O_3$  directly produced by lightning. Only the first two factors are discussed in detail in Sect. 4, as lightning  $O_3$  is beyond the scope of this study and still uncertain as shown by limited observations and model simulations (Morris et al., 2010) (Morris et al., 2010; Ripoll et al., 2014).



Figure 3. Same as Fig. 2 but for the case on 01 September 2020. The simulation time is 30 minutes ahead of each radar observation.

#### 4 Convection Impactsimpacts

195 To evaluate the higher UT O<sub>3</sub> concentration after the convection, the mean vertical profiles of O<sub>3</sub> in the atmospheric regions passed by the ozonesondes are illustrated at three stages of convection: initiation, development, and dissipation (Fig. 4a and d). The UT O<sub>3</sub> increases continuously during the whole cycle of the 2020 case, but for the 2019 case, it declines at the developing stage because of the uplift of O<sub>3</sub>-poor air while later it starts rising again. This phenomenon can be explained by the O<sub>3</sub> vertical cross sections at the developing stages (Fig. 4b and e). For the 2019 case, the cells of low O<sub>3</sub> concentration reach 16 km by the updraft, and then the high O<sub>3</sub> air wrapped behind the convection moves into the region. However, the observed increasing O<sub>3</sub> of the 2020 case is mostly from the vertically transported background O<sub>3</sub>.

To determine the processes causing the differences between the two cases, we analyzed the outputs of mean integrated physical rates (IPR) from 10 to 14 km during the convective period (Fig. 4c and f). Generally, the opposite trend of the horizontal advection (advh) and vertical advection (advz) governs the net decrease in UT  $O_3$  production rate of the 2019 case.

Note that the advz contribution is negative between 10 and 11.5 km and positive between 11.5 and 13.8 km. This is due to the uplifted O<sub>3</sub>-poor air and downward O<sub>3</sub>-rich air caused by the stronger updraft compared with the 2020 case. As indicated by the higher  $Q_v$  after convection (Fig. 1c) and tropopause height (Fig. 4b), the updraft of the 2020 case is not strong enough to wrap the stratospheric O<sub>3</sub> like the mesoscale convective system (Phoenix et al., 2020). While the dynamic processes play an important role in the O<sub>3</sub> production, the positive chemistry contribution cannot be neglected in both cases and leads to the



**Figure 4.** (a, d) The mean  $O_3$  profiles in the regions passed by the ozonesondes at three stages: initiation (green), development (orange), and dissipation (blue). (b, e) Vertical  $O_3$  distribution within the convective periods along the line crossing the convective core (Fig. 2e and Fig. 3f). The blue dashed lines stand for the boundaries of regions passed by the ozonesondes, and the lapse rate tropopause is shown as the white dots. The cloud boundaries (the sum of cloud liquid water mixing ratio  $[q_{cloud}]$  and ice mixing ratio  $[q_{ice}] \ge 0.01$  g/kg) are shown in white lines. (c, f) The vertical distributions of the  $O_3$  net production rate (net) and tendency due to horizontal advection (advh), vertical (advz) advection, and chemistry (chem) during the convective periods.

210 net increase in UT O<sub>3</sub> during the convective period of 2020 case. Specifically, the dynamic transport (chemical production) occupies 114 % (-14 %) of the total UT O<sub>3</sub> decrease during the convective period of the 2019 case, the chemical production (dynamic transport) is responsible for 249 % (-149 %) of the total O<sub>3</sub> increase for the 2020 case (Table 1). During the life cycle of both cases , the chemistry tendency occupies more than 87 % of the chemistry increases O<sub>3</sub> production in the UT regions passed by the ozonesondesin both cases and the magnitude of the effect is 5–10 times that of dynamic effects. This demonstrates the dominant chemistry role in the overall effects of convection.

#### 5 Impact of lightning $NO_x$ on the $O_3$ profile

Furthermore, the IPR outputs including LNO emission are compared with these excluding LNO to explore the effects of  $LNO_x$  on  $O_3$  (Table 1). The  $LNO_x$  reduces the net  $O_3$  production by 25 % and 40 % during the convective period and life cycle of the 2019 case, respectively. This effect, induced by the The decreased chemistry contribution ris less significant ( $<\leq 1\%$ ) for the 2020 case which has a smaller lightning density near the station. Consistent with Ott et al. (2007), the net loss of ozone

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due to lightning is less than 3 ppbv at all height levels, thus the comparison with ozonesonde is not affected by  $LNO_x$  (Fig. 1). However, Note that the  $LNO_x$  can certainly enhance the downwind ozone production in on the scale of days (Pickering et al., 1996; DeCaria et al., 2005). Therefore, it is necessary to estimate the  $LNO_x$  production efficiency accurately PE accurately (discussed later in Sect. 6.2).

Period	Time	LNO (mol/flash)	$advh + advz^*$	chem*	net*
Life Cycle	2019-07-25	0	-3.3 (-24.6 %)	16.7 (124.6 %)	13.4
	(03:20-05:40)	500	-2.3 (-28.8 %)	10.3 (128.8 %)	8.0
	2020-09-01	0	3.4 (9.6 %)	32.0 (90.4 %)	35.4
	(04:20-06:40)	500	4.4 (12.1 %)	31.9 (87.8 %)	36.3
Convective Period	2019-07-25	0	-19.6 (140.0 %)	5.6 (-40.0 %)	-14.0
	(04:20-05:00)	500	-20.0 (114.3 %)	2.5 (-14.3 % )	-17.5
	2020-09-01	0	-9.7 (-131.1 %)	17.1 (231.1 % )	7.4
	(05:40-06:20)	500	-10.1 (-148.5 %)	16.9 (248.5 % )	6.8

Table 1. Process Analysis Table analysis table for the Mean mean O<sub>3</sub> Integrated Tendencies integrated tendencies (10-14 km).

\*The unit is  $10^{10}$  molec. cm<sup>-3</sup>. The percentage is the proportion of each part in the net O<sub>3</sub> change.

Additionally, the convection was divided into three regions by TROPOMI data: fresh lightning, downwind of fresh lightning, and aged lightning (See Sect. 6.1 and Fig. 8 for details). Firstly, the difference of O<sub>3</sub> (ΔO<sub>3</sub>) profiles is obtained with different LNO PE assumption (Fig. 5a–c). In contrast with the net loss of ozone (< 4 ppbv) over all height levels in Ott et al. (2007), the ΔO<sub>3</sub> is mostly positive (< 1 ppbv) between 2 km and 5 km and negative (> -3 ppbv) between 5 km and 12 km in our cases. The higher PE (700 mol/flash) slightly reduces the O<sub>3</sub> concentration by less than 1 ppbv at all levels compared with the
default PE (500 mol/flash) and it even leads to negative ΔO<sub>3</sub> between 2 km and 5 km downwind of fresh lightning (Fig. 5b).

The maximum  $O_3$  loss is between 8 km and 10 km due to the peak of  $LNO_x$  (up to 2.6 ppby) introduced in the model.

Then, the integrated reaction rate (IRR) is applied to determine the chemistry mechanism and the effect of LNO on the  $O_3$  variation for two layers where the  $\Delta O_3$  is opposite: 800 hPa–500 hPa ( $\Delta O_3 > 0$ ) and 500 hPa–200 hPa ( $\Delta O_3 < 0$ ). The tropospheric  $O_3$  is mainly controlled by five reaction rate terms (Pickering et al., 1990; Bozem et al., 2014):

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$$\frac{d}{dt}[O_3] \equiv k_1[NO][HO_2] + \sum_i k_i[NO][R_iO_2]$$

$$-k_{3}[H_{2}O][O(^{1}D)] - k_{4}[HO_{2}][O_{3}] - k_{5}[OH][O_{3}]$$
(4)

where  $k_i$  is the rate coefficient of the reaction between peroxy radicals ( $R_iO_2$ ) and NO. The time series of each contribution to  $O_3$  production are illustrated in Fig. 5d–i. Overall, the time series of IRR are more variable for the 2019 case due to the stronger activity as clarified in Sect. 4. The total net chemistry production of  $O_3$  keeps positive for both layers. In detail, the reaction between NO and HO<sub>2</sub> always dominates the production while the oxidation of NO by RO<sub>2</sub> is about 40 %–60 % of that production. The dominant loss of  $O_3$  is the photolysis (described by the reaction of  $O(^1D)$  and  $H_2O$ ) while the reaction between  $O_3$  and OH is comparable during the convective period. The lowest contribution to  $O_3$  loss,  $O_3 + HO_2 \rightarrow OH + 2O_2$ , is reduced during the convection because of the production of LNO, which captures the HO<sub>2</sub> reacting with  $O_3$ . Note that although the increase of total IRR induced by  $LNO_x$  can reach  $1.36 \times 10^7$  molec. cm<sup>-3</sup> s<sup>-1</sup> and  $2.60 \times 10^6$  molec. cm<sup>-3</sup> s<sup>-1</sup> in the low layer and high layer over these three regions, respectively, the net  $O_3$  production actually decreases in the high layers (Fig. 5a–c) due to the combination of dynamic transport and chemical production related to  $LNO_x$ .

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**Figure 5.** (a–c) Changes in  $O_3$  profiles due to  $LNO_x$  at TROPOMI overpass time in three regions (fresh lightning region, downwind of fresh lightning, and aged lightning area) as defined in Fig. 7. (d–f) Time series of the mean integrated reaction rate (IRR) between 800 hPa and 500 hPa. The legend shows detailed species and reactions. The Total IRR is the  $O_3$  loss IRR subtracted from the  $O_3$  production IRR (red and orange lines). The solid line shows the IRR with LNO (500 mol/flash) while the dashed line is without LNO. (g–i) Same as (d–f) but between 500 hPa and 200 hPa.

#### 6 TROPOMI products over the convection

#### 6.1 Relation of between lightning and TROPOMI products

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As the LNO<sub>x</sub> production estimation from TROPOMI depends upon the tropospheric NO<sub>2</sub> slant column density (SCD<sub>tropNO<sub>2</sub></sub>), we compare the SCD<sub>tropNO<sub>2</sub></sub> distributions with the observed lightning flashes (Fig. 6a–d). Although the SCD<sub>tropNO<sub>2</sub></sub> over the most active pixels is not valid due to the detector saturation and blooming effect, the nearby or outflow regions still have useful data. While the flashes occurred less than 30 minutes before the TROPOMI overpass time in the 2019 case, the 2020 case had both fresh and aged LNO<sub>2</sub>, generating the stripe of high SCD<sub>tropNO<sub>2</sub></sub> surrounded by low values in (Fig. 6bd).



**Figure 6.** Events on 25 July 2019 (left) and 07 September 2020 (right). (a, b) The tropospheric NO<sub>2</sub> slant column density (SCD<sub>tropNO<sub>2</sub></sub>, filled color) and cloud optical pressure (line color). (c, d) The <u>effective</u> cloud <u>radiance</u> fraction in the NO<sub>2</sub> window ( $f_{effNO<sub>2</sub>}$ ) and flashes whose color depends on the occurring time relative to the TROPOMI overpass time. (e, f) The distribution of the LNO<sub>x</sub> vertical column densities (VCD<sub>LNO<sub>x</sub></sub>). The solid white rectangle is used for the summation of flashes while the dashed one is for VCD<sub>LNO<sub>x</sub></sub>. The solid white black border is Jiangsu province. These white grid cells stand for missing TROPOMI data (no2\_scd\_flag  $\neq$  0), as defined in Appendix A.

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Specifically, the SCD<sub>tropNO<sub>2</sub></sub> of the convective pixels ( $f_{effNO_2,L} \ge 0.7$ ) is smaller than that in other regions. This is opposite to previous studies of large-scale convective systems with high flash density (Beirle et al., 2009). Four factors could lead to this unexpected result: the cloud top heights, flash counts, flash occurring time, and background NO<sub>2</sub>. Either the inadequate flash or weak convection could lead to a smaller SCD<sub>tropNO<sub>2</sub></sub> over pixels with  $f_{effNO_2,L} \approx 1$  because the TROPOMI can only see the

LNO<sub>2</sub> above the clouds. In other words, the polluted NO<sub>2</sub> below the broken or thinner clouds is partially exposed if  $f_{effNO_2}$  r

< 1. The sensitivity tests of the WRF-Chem a priori SCD<sub>tropNO2</sub> also can help explain this phenomenon clearly (Fig. S5). The

260 pixels of high SCD<sub>ttopNO<sub>2</sub></sub> with low cloud fraction belong to background NO<sub>2</sub> pollution (Fig. S5a and e), but the SCD<sub>ttopNO<sub>2</sub></sub> increased by UT LNO<sub>2</sub> is still visible compared with the lower SCD<sub>ttopNO<sub>2</sub></sub> without LNO<sub>2</sub> (Fig. S5b–d and f–h).

An LNO PE upper limit of 700 mol NO/flash (Ott et al., 2010) is applied to WRF-Chem for investigating the importance of  $LNO_x$  for the  $AMF_{trop}$  and  $AMF_{LNO_x}$  calculations. The changes in the retrieved AMFs are examined by replacing profiles in three tropospheric layers independently: middle troposphere (MT, 800 hPa to 400 hPa), upper troposphere (UT, 400 hPa to 150

- 265 hPa), and troposphere (surface to tropopause). Unless otherwise specified, the changes of AMFs are obtained by increasing LNO<sub>x</sub> in this Section Engine 7 shows that the AMFs-AMF changes are mostly controlled by the LNO<sub>x</sub> in the UT layer where the detection sensitivity is high (Laughner and Cohen, 2017)(Beirle et al., 2009; Laughner and Cohen, 2017) and the LNO<sub>x</sub> production reaches the peak (Fig. S6). While the AMF<sub>LNO<sub>x</sub></sub> always decreases decreases by 5 %-40 % for both cases, the changes of AMF<sub>trop</sub> ( $\Delta$ AMF<sub>trop</sub>) are regionally specific and can be classified by the lightning activity: fresh lightning (MT
- 270  $\Delta AMF_{trop} < -20\%$ , downwind of fresh lightning (MT  $\Delta AMF_{trop} > 20\%$ ), and aged lightning (UT  $\Delta AMF_{trop} > 20\%$ ). Figure 8a illustrates the relationship between the cloud optical pressure and  $f_{effNO_2}$  p<sub>cloud</sub> and  $f_r$  over these three regions. The fresh lightning pixels own clouds clouds are higher than 400 hPa and  $f_{effNO_2}$  larger than 0.6( $p_{cloud} < 400$  hPa) and  $f_r$  is larger than 0.8 over fresh lightning pixels, but both aged lightning and downwind of fresh lightning areas have clouds lower than 400 hPa. This coincides with the mean cloud pressures in Fig. S6 and explains why UT  $\Delta AMF_{trop} > 20\%$  exits in Fig. 7b<sub>i</sub> and b<sub>ii</sub>, and 275 indicates <sub>iii</sub>, indicating the possibility of estimating LNO<sub>x</sub> estimations over the aged lightning regions (Sect. 6.2).
- As defined in Appendix AB,  $\text{SCD}_{\text{tropNO}_2}^{\text{LNO}_x}/\text{SCD}_{\text{tropNO}_2}^{\text{noLNO}_x}$  and  $\text{VCD}_{\text{tropNO}_2}^{\text{LNO}_x}/\text{VCD}_{\text{tropNO}_2}^{\text{noLNO}_x}$  can be used to determine which parameter controls  $\Delta \text{AMF}_{\text{trop}}$ : enhanced a priori  $\text{SCD}_{\text{tropNO}_2}$  or a priori  $\text{VCD}_{\text{tropNO}_2}$  (Fig. 8b–d). Briefly, the dominant one belongs to the larger ratio. First, if the LNO<sub>2</sub> is included in the tropospheric layer of a priori NO<sub>2</sub> profiles (Fig. 7c<sub>i</sub>), the AMF<sub>trop</sub> decreases over most fresh lightning pixels because of the increased a priori  $\text{VCD}_{\text{tropNO}_2}$  (Fig. 8b). The situation is opposite for the
- 280 downwind of the fresh lightning region. There the AMF<sub>trop</sub> is larger no matter which layer is chosen (Fig.  $7a_i-c_i$ ), because the LNO<sub>2</sub> was convected, reached above the cloud top, and led to the larger a priori SCD<sub>tropNO2</sub> (Fig. 8c). Interestingly, for the 2020 case, the AMF<sub>trop</sub> of aged lightning pixels increases more than 50 % for the UT layer (Fig.  $7b_{iii}$ ). It demonstrates the important role of advected UT LNO<sub>2</sub> and that the cloud exists as a barrier, causing the difference between a priori SCD<sub>tropNO2</sub> and a priori VCD<sub>tropNO2</sub> (Fig. 8d). Although the difference is smaller than that of the other two regions due to the LNO<sub>2</sub>
- 285 lifetime, it is still useful for retrieving the LNO<sub>x</sub>. Besides, considering the region-specific LNO<sub>x</sub> effects on AMFs, we need to include the representation of LNO<sub>2</sub> in the TROPOMI NO<sub>2</sub> retrievals better, especially outflow regions. The comparisons (Fig. S6) between the TROPOMI standard NO<sub>2</sub> profiles from TM5-MP and WRF-Chem also illustrate the importance of LNO<sub>x</sub>, resolved convection transport, and emissions. Aircraft observations of NO and NO<sub>2</sub> will be useful to determine the exact roles (Laughner and Cohen, 2017).



**Figure 7.** The percent differences of AMFs by replacing the a priori NO<sub>2</sub> profiles at three layers: middle troposphere (left), upper troposphere (middle), and troposphere (right).  $\Delta$ AMF<sub>trop</sub> is the comparison of the AMF<sub>trop</sub> with 500 mol NO per flash and relative to 0 mol NO per flash.  $\Delta$ AMF<sub>LNO<sub>x</sub></sub> is the comparison of the AMF<sub>LNO<sub>x</sub></sub> with 700 mol NO per flash and relative to 500 mol NO per flash. Three regions are annotated: fresh lightning (blue), downwind of fresh lightning (red), and aged lightning (green). Because of the quite large AMF<sub>LNO<sub>x</sub></sub> values in pixels with little lightning,  $\Delta$ AMF<sub>LNO<sub>x</sub></sub> is shown over pixels where 0 < AMF<sub>LNO<sub>x</sub></sub> < 10.

## 290 6.2 Estimations of LNO<sub>x</sub>

Satellite observations with fresh convection are usually used to estimate the  $LNO_x$  PE. However, it is difficult to apply the same method to regions with small convection like the 2019 case, because of the pixel saturation of TROPOMI and limited coverage of convective area (Fig. 6ec). Instead, we focus on dissipated convection which is the southern part of the 2020 case, while the



**Figure 8.** (a) The relationship between cloud pressure and  $f_{effNO_2}$  cloud radiance fraction for three regions defined in Fig. 7: fresh lightning region, downwind of fresh lightning, and aged lightning area. (b–d) The a priori  $SCD_{tropNO_2}^{LNO_x}/SCD_{tropNO_2}^{noLNO_x}$  and a priori  $VCD_{tropNO_2}^{LNO_x}/VCD_{tropNO_2}^{noLNO_x}$  of pixels in these three regions. The LNO<sub>x</sub> superscript indicates that the a priori variable is calculated with LNO<sub>x</sub> (500 mol NO per flash) and the noLNO<sub>x</sub> superscript is without LNO<sub>x</sub>.

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northern part has multiple missing values regardless of the adequate flashes and high cloud fractions. As shown in Fig. 6f9b–c, the time differences between flashes and TROPOMI overpass time are longer than 30 minutes but shorter than 3 hours. Since the lifetime of NO<sub>2</sub> is ~ 3 hours in or near the field of convection (Nault et al., 2016) and ranges from 2 to 12 h depending on the convective location, these pixels can still be used for the LNO<sub>x</sub> estimation. Equation (5) is applied to determine the mean LNO<sub>x</sub> PE (mol/flash):

$$PE_{LNO_x} = \sum_{p} V_i A_i / \sum_{N} F_j e^{-(t_0 - t_j)/\tau}$$
(5)

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where *p* stands for pixels affected by  $\text{LNO}_x$ ,  $V_i \text{ (mol/m}^2)$  is the  $\text{LNO}_x$  vertical column densities ( $\text{VCD}_{\text{LNO}_x} = \text{SCD}_{\text{tropNO}_2}$ /  $\text{AMF}_{\text{LNO}_x}$ ) over pixel *i* with an area called  $A_i \text{ (m}^2)$ , *N* is the total number of flashes contributing to  $\text{VCD}_{\text{LNO}_x}$ , and the exponential component considers the lifetime of  $\text{NO}_x$  for each flash ( $F_j$ ). Specifically,  $t_0$  is the time of TROPOMI overpass,  $t_j$ is the time of the lightning flash, and  $\tau$  represents the  $\text{NO}_x$  3-hour lifetime near convection. As the dissipated convection produced enough lightning and the UT winds within the storm were blowing from the west-northwest to east-southeast (Fig. 9a), the pattern of  $VCD_{LNO_x}$  can still be clearly identified (dashed rectangle in Fig. 6f). Additionally, we need to enlarge the selected region for flashes, because the UT winds within the storm were blowing from the west-northwest to east-southeast(). Fortunately, there is a low  $VCD_{LNO_x}$  strip separating the northern and southern convection. With the manual selection of the correct region careful selection, the  $LNO_x$  PE is estimated as 88.60 mol  $NO_x$  per flashand it will be 90 mol  $NO_x$ . Although there are a few lightning flashes related to the  $VCD_{LNO_x}$  is outside of the region selection, it only affects the  $LNO_x$ per flash, applying the linear interpolation over the few invalid pixelsPE by  $\approx 2$  mol, which is within the uncertainty discussed







Following Allen et al. (2019) and Zhang et al. (2020), the uncertainty of LNO<sub>x</sub> is determined by LNO<sub>2</sub> lifetime, background NO<sub>2</sub>, *x* lifetime, lightning DE, NO/NO<sub>2</sub> ratio, LNO profile, and other sources (summarized in Table 2). The lifetime (τ) of NO<sub>2</sub> is replaced by 2 and 6 hours to evaluate the uncertainty as 27 % while another uncertainty is also 27 % related to lightning DE by
changing the ratios ratio of IC to CG to 2:1 and 4:1. The uncertainty caused by modeled NO/NO<sub>2</sub> ratios is assumed to be 30 % based on Allen et al. (2019) and the uncertainty related to LNO profile is 29-26 % by using the a priori NO<sub>2</sub> profile with 330 and 700 mol NO per flash. The uncertainty associated with the stratospheric vertical column amount, f<sub>ettNO2</sub>, and is considered as 7 % by applying a bias of ± 10<sup>14</sup> molec. cm<sup>2</sup> (van Geffen et al., 2021). The uncertainty caused by other possible error sources including systematic errors in slant columnsis considered , optical cloud pressure, and NO<sub>2</sub> redistributed by convection is difficult to quantify and assumed as 10 % (Allen et al., 2021) following Allen et al. (2021). Assuming no correlation between errors, the total uncertainty (58-56 %) is estimated as the square root of the sum of the squares of all individual uncertainties. As a result, the LNO<sub>x</sub> PE is 88.60 ± 50.33 mol NO<sub>x</sub> per flash. Even though the LNO<sub>x</sub> per flash) over the continental United States

(Zhang et al., 2020) - and at the lower end of  $120 \pm 65 \text{ mol NO}_x$  per flash obtained in Allen et al. (2021). Thus, more studies over China are necessary for the estimation of region-dependent LNO<sub>x</sub> PEs.

Type	Uncertainty (%)
$\underline{\text{LNO}_x \text{ lifetime}}$	<u>27 %</u>
Lightning detection efficiency	27 %
NO/NO <sub>2</sub> ratio	<u>30 %</u>
LNO profile	<u>26 %</u>
Stratospheric vertical column	<u>7 %</u>
Others	10 %
Net	<u>56 %</u>

Table 2. Uncertainties for the estimation of LNO<sub>x</sub> production efficiency.

#### 7 Conclusions

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Both the 2019 and 2020 cases saw enhanced upper tropospheric (UT)  $O_3$  concentrations according to the ozonesonde observations. As revealed by modeling using WRF-Chem, the dynamic contribution of  $O_3$  variation in the 2019 case was generated by mixing with the UT  $O_3$ -rich air due to strong updraft, while it was caused by vertical advection of high background  $O_3$ 

in the 2020 case. The detailed analysis of integrated physical rates shows that the dynamic processes represent 114 % ~ 149 % of dominate the UT O<sub>3</sub> decrease during the convective stage of both cases. However, in the convection life cycle, the chemistry reactions contribute more than 87 % contribution of chemistry reactions to the UT O<sub>3</sub> production is 5–10 times that of dynamics. Besides, the UT O<sub>3</sub> enhancement of the 2019 case decreases by 40 % if the lightning nitrogen oxides (LNO<sub>x</sub>) is included in the model, indicating the importance of the LNO<sub>x</sub>. Utilizing the outputs of integrated reaction rates improves our understanding of the chemical reactions and effects of lightning nitrogen oxides (LNO<sub>x</sub>) on O<sub>3</sub>. While the reaction of NO and HO<sub>2</sub> dominates the production of O<sub>3</sub>, the O<sub>3</sub> loss processes are suppressed because the LNO<sub>x</sub> reacts with the HO<sub>2</sub> which is

the reactant in the production. The WRF-Chem results are incorporated into the retrieval algorithm to explore how  $LNO_x$  affects the official TROPOMI products and whether prove that TROPOMI is useful for LNO<sub>x</sub> studies over evently evaluated and whether proves that TROPOMI

products and whether prove that TROPOMI is useful for  $LNO_x$  studies over small-scale convection regions. The sensitivity tests imply that air mass factors of tropospheric  $NO_2$  are smaller for fresh lightning regions and the opposite for the aged lightning pixels or downwind of fresh lightning. The air mass factors of tropospheric  $LNO_x$  always decrease with increased  $LNO_x$ . Since the  $LNO_x$  affects the variation of tropospheric  $NO_2$  in the outflow regions, better consideration of  $LNO_x$  is essential for studies of the tropical and mid-latitude regions in summer.

Because the saturation of TROPOMI pixels and blooming effects lead to the failure of detecting LNO<sub>x</sub> over active convec-345 tion, we focus on the dissipation regions where aged LNO<sub>x</sub> still exists. The production efficiency is estimated to be  $\frac{88.60}{2}$  ± 50-33 mol LNO<sub>x</sub> per flash, which is less than but still within the range of previous studies. Although the current results are limited, the technique can be implemented worldwide to region-specific  $LNO_x$  retrievals. To quantify and refine the  $LNO_x$ production estimates, TROPOMI data over both active and dissipated convection could provide valuable information.

*Code and data availability*. Data used are obtained from https://www.acom.ucar.edu/waccm/download.shtml (WACCM), http://meicmodel. 350 org/ (MEIC), and https://www.acom.ucar.edu/wrf-chem/download.shtml (MEGAN). The relevant data and analysis code are hosted at https://github.com/zxdawn/Xin ACP 2021 Convection Effect (Zhang, 2021c, d). The retrieval algorithm is available at https://github.com/ zxdawn/S5P-WRFChem (Zhang, 2021a). The modified WRF-Chem source code is available to the public at https://github.com/zxdawn/ WRF-Chem-LDA-LFR (Zhang, 2021b).

# Appendix A: Contributions to $\Delta AMF_{tron}$ Flag definitions used in this study

355 The no2 scd flag is introduced to make usage of the  $NO_2$  slant column density (SCD) data easier, by gathering information from a few variables into one flag (Table A1). This flag can thus be used for filtering, though with care as it probably does not cover all possible situation. Here " $\delta$ " refers to the SCD error (in mol/m<sup>2</sup>) and "pqf" stands for processing quality flag.

 Table A1. Definition of no2\_scd\_flag.

	Meaning
-1	no SCD value due to saturation limit exceeded, i.e. pqf=54
$\overset{0}{\sim}$	SCD with $\delta < 3.3 \times 10^{-5}$ & no error reported
$\stackrel{1}{\sim}$	SCD with $\delta < 3.3 \times 10^{-5}$ & error reported: pqf=55
2~~~	SCD with $\delta < 3.3 \times 10^{-5}$ & other error reported, e.g. pqf=41
3	SCD with $\delta \ge 3.3 \times 10^{-5}$ & no error reported
<u>4</u>	SCD with $\delta \ge 3.3 \times 10^{-5}$ & error reported: pqf=55
5	SCD with $\delta \ge 3.3 \times 10^{-5}$ & other error reported, e.g. pqf=41
FillValue	no SCD due other error (prior to the Differential Optical Absorption Spectroscopy fit)

# Appendix B: Contributions to $\Delta AMF_{trop}$

In Fig. 8b–d, the contribution of LNO<sub>x</sub> to  $\Delta AMF_{trop}$  is divided into two parts:  $SCD_{tropNO_2}^{LNO_x}/SCD_{tropNO_2}^{noLNO_x}$  and  $VCD_{tropNO_2}^{LNO_x}/VCD_{tropNO_2}^{noLNO_x}$ , where the LNO<sub>x</sub> superscript indicates that the a priori variable is calculated with LNO<sub>x</sub> (500 mol NO per flash) and the noLNO<sub>x</sub> 360 superscript is without  $LNO_x$ . The two contributions are derived by taking the logarithm of Eq. (B1) and Eq. (B2) and then subtracting them into Eq. (B3). Here, several abbreviations are defined to simplify the symbols: S is  $SCD_{tropNO_2}$ , V is  $VCD_{tropNO_2}$ , subscript 1 is with  $LNO_x$ , and 0 is without  $LNO_x$ .

$$AMF_1 = \frac{S_1}{V_1} \tag{B1}$$

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$$AMF_0 = \frac{S_0}{V_0}$$
 (B2)

$$log(AMF_{1}) - log(AMF_{0}) = log(\frac{S_{1}}{V_{1}}) - log(\frac{S_{0}}{V_{0}})$$
  
= log( $\frac{S_{1}}{S_{0}}$ ) - log( $\frac{V_{1}}{V_{0}}$ ) (B3)

Therefore, if  $\frac{S_1}{S_0}$  is larger than  $\frac{V_1}{V_0}$  (the solid line is higher than the dashed line in Fig. 8b–d), then AMF<sub>1</sub> is larger than AMF<sub>0</sub>. In other words, these two variables determine how a priori LNO<sub>x</sub> affects the retrieval of NO<sub>2</sub>.

Author contributions. YY directed the research and YY, XZ, and RvdA designed the research with feedback from the other co-authors;
 XZ, RvdA, HE, and JvG developed the retrieval algorithm; XZ modified lightning assimilation code written by YL; JLL provided guidance and supporting data on the ENTLN data; XZ, KC, XK, ZZ, JH, CH, JZ, XY, and HC participated in the field campaigns; XZ performed simulations and analysis with the help of YY, RvdA, XK, JC, CH, and RS; XZ, YY, RvdA, and JLL interpreted the data and discussed the results. XZ drafted the manuscript with comments from the co-authors; RvdA, YY, and JLL edited the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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