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

Response to Anonymous Referee #1

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We thank the reviewer for his/her positive comments and very careful reading of our article. The individual corrections suggested are addressed below. The reviewer's comments will be shown in **red**, our response in **blue**, and changes made to the paper are shown in **black** block quotes. Unless otherwise indicated, page and line numbers correspond to the original manuscript. Figures, tables, or equations referenced as "Rn" are numbered within this response; if these are used in the changes to the paper, they will be replaced with the proper number in the revised version.

1) Line 18: Huntrieser et al. (2016) measured ozone gradients in thunderstorms. Please cite and explain their results here. Mention again in lines 36-40.

Huntrieser, H., Lichtenstern, M., Scheibe, M., Aufmhoff, H., Schlager, H., Pucik, T., ... & Barth, M. C. (2016). On the origin of pronounced O3 gradients in the thunderstorm outflow region during DC3. Journal of Geophysical Research: Atmospheres, 121(11), 6600-6637.

Thanks for the reference and we have added it. Because the conclusion of Huntrieser et al. (2016) is similar to Phoenix et al. (2020), we combined them in the same sentence.

> Meanwhile, the vertical profiles of trace gases are reshaped by the updraft and downdraft on timescales of hours (**Huntrieser et al., 2016; Barth et al., 2019**).
>
> ... As revealed in the **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 (**Huntrieser et al., 2016; Phoenix et al., 2020**).

Lines 19 - 24: Murray et al. (2012) and Gordillo-Vázquez et al. (2019) used global models to study the relationship between LNOx and other trace gases. Please cite here.

Murray, L. T., Jacob, D. J., Logan, J. A., Hudman, R. C., & Koshak, W. J. (2012). Optimized regional and interannual variability of lightning in a global chemical transport model constrained by LIS/OTD satellite data. Journal of Geophysical Research: Atmospheres, 117(D20).

Gordilloâ Vázquez, F. J., Pérezâ Invernón, F. J., Huntrieser, H., & Smith, A. K. (2019). Comparison of six lightning parameterizations in CAM5 and the impact on global atmospheric chemistry. Earth and Space Science, 6(12), 2317-2346.

Added.

> Several lightning parameterizations have been developed for chemistry transport and climate models to evaluate the relationship between LNO_x and other trace gases (Murray et al., 2012; Gordillo-Vázquez et al., 2019; Luhar et al., 2021; Pérez-Invernón et al., 2021).

Lines 43-44: Please give a brief explanation on why thunderstorms and lightning have increased due to urbanization. Is it because of the effect of aerosols on cloud electrification [Tao et al. (2012), Pérez-Invernón et al. (2021)]?

Tao, W. K., Chen, J. P., Li, Z., Wang, C., & Zhang, C. (2012). Impact of aerosols on convective clouds and precipitation. Reviews of Geophysics, 50(2).

Pérez-Invernón, F. J., Huntrieser, H., Gordillo-Vázquez, F. J., & Soler, S. (2021). Influence of the COVID-19 lockdown on lightning activity in the Po Valley. Atmospheric Research, 263, 105808.

Thanks for the suggestion. Yang and Li (2014) (cited in the manuscript) suggests a large impact of aerosols on thunderstorm activities. We have added it.

> 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; 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**).

Line 46: Although described in other part of the manuscript, it could be useful for the reader giving a reference for TROPOMI here, as it is the first time it appears.

Added.

> For the first time, the TROPOspheric Monitoring Instrument (**TROPOMI; Veefkind et al., 2012**) NO_2 observations are used to identify LNO_x PEs in southeastern China.

Line 74: intro-cloud -> intra-cloud

Fixed.

> ... while ENTLN and WWLLN detect both **intra-cloud** (IC) and CG flashes with specific detection frequency ...

Section 2.2: The main weakness of the manuscript is the poor estimation on the DE of the employed lightning systems. A reliable estimation on the total number of lightning flashes is needed to calculate the LNOx. Would it be possible combining the lightning data from the used lightning dataset with lightning data from ISS-LIS in order to evaluate the combinience of using the 3:1 ratio? ISS-LIS reports IC+CG lightning. The DE of ISS-LIS is well established and quite constant all around the world [Blakeslee et al. (2020)].

Blakeslee, R. J., Lang, T. J., Koshak, W. J., Buechler, D., Gatlin, P., Mach, D. M., ... & Christian, H. (2020). Three years of the lightning imaging sensor onboard the international space station: Expanded global coverage and enhanced applications. Journal of Geophysical Research: Atmospheres,125(16), e2020JD032918.

Yes, that would be a better method for total lightning networks. Because the CLDN only detects CG flashes and the IC DE of ENGLN is low in China, we have decided to use the constant IC:CG ratio from Wu et al. (2016) and Bandholnopparat et al. (2020). If there are more total lightning networks deployed in China, we will use that for future studies with a better DE estimation. The uncertainty of LNO_x PE estimation caused by DE is evaluated by changing the ratio to 2:1 and 4:1.

> Although CG detection efficiency of ENGLN is not known for this region due to a lack of validation data, 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 use the merged CG data with a constant IC/CG ratio of 3:1 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), are available**.

Equation (1): I think a more detailed explanation on eq. (1) is needed.

Added.

We replaced the tropospheric AMF ($\mathrm{AMF_{trop}}$) with a new AMF called AMF for LNO_x ($\mathrm{AMF_{LNO_x}}$) to derive the tropospheric LNO_x vertical column density ($\mathrm{VCD_{LNO_x}}$). The concept of $\mathrm{AMF_{LNO_x}}$ inherits from the $\mathrm{AMF_{trop}}$ 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). **Briefly, the numerator is the modeled tropospheric $\mathbf{NO_2}$ slant column density ($\mathbf{SCD_{tropNO_2}}$) and the denominator is the modeled VCD ($\mathbf{VCD_{NO_2}}$ or $\mathbf{VCD_{LNO_x}}$). In detail, these two AMFs can be calculated as**:

$$AMF_{\text{Trop}} = \frac{(1 - f_{effNO_2}) \int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p)\, dp + f_{effNO_2} \int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p) NO_2(p)\, dp}{\int_{p_{surf}}^{p_{tp}} NO_2(p)\, dp} \tag{R1}$$

$$AMF_{\text{LNO}_x} = \frac{(1 - f_{effNO_2}) \int_{p_{surf}}^{p_{tp}} w_{clear}(p) NO_2(p)\, dp + f_{effNO_2} \int_{p_{cloud}}^{p_{tp}} w_{cloudy}(p) NO_2(p)\, dp}{\int_{p_{surf}}^{p_{tp}} LNO_x(p)\, dp} \tag{R2}$$

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 effective cloud fraction in the $\mathrm{NO_2}$ 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 $\mathrm{NO_2}$ vertical profile simulated by WRF-Chem. Besides, $LNO_x(p)$ is the LNO_x vertical profile calculated by the difference of vertical profile between WRF-Chem simulations with and without lightning.

Lines 174–176: Ripoll et al. (2014) developed a detailed chemistry model for lightning channel including ozone. It would be beneficial for the manuscript citing it at this point.

Ripoll, J. F., Zinn, J., Jeffery, C. A., & Colestock, P. L. (2014). On the dynamics of hot air plasmas related to lightning discharges: 1. Gas dynamics. Journal of Geophysical Research: Atmospheres, 119(15), 9196-9217.

Thanks for your suggestion. It is a valuable model. Added.

Only the first two factors are discussed in detail in Sect. 4, as lightning $\mathrm{O_3}$ is beyond the scope of this study and still uncertain as shown by limited observations and model simulations **(Morris et al., 2010; Ripoll et al., 2014)**.

Line 246: There is still a significant uncertainty in the lifetime of NO2 in or near the field of convection [e.g., Beirle et al. (2010), Pickering et al. 2016 ...]. According to literature, the lifetime can vary between 3 h and 2 days. Although it is mentioned below, please mention here.

Added.

> Since the lifetime of NO_2 is $\sim$ 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_x estimation.

Lines 257-263: Comparing the sources of uncertainty in the text is hard. A Table for the uncertainties could be useful.

Yes, it can make it easier to read. Added (Table R1).

Table R1 . Uncertainties for the estimation of LNO_x production efficiency.

Type	**Uncertainty (%)**
LNO_x lifetime	27 %
Lightning detection efficiency	27 %
NO/NO_2 ratio	30 %
LNO profile	26 %
Stratospheric vertical column	7 %
Others	10 %
Net	56 %

Lines 276-279: Mention explicitly if TROPOMI is useful or not here.

Fixed.

> The WRF-Chem results are incorporated into the retrieval algorithm to explore how LNO_x affects the official TROPOMI products and **prove that** TROPOMI is useful for LNO_x studies over small-scale convection regions.

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