Responses to Reviewers' comments

To the esteemed Editor and Reviewers,

We would like to thank the reviewers for the time and efforts in reviewing our manuscript. We have revised the manuscript according to the reviewers' detailed comments, which we sincerely hope the correction will meet with the high publishing standard of the journal. Please find the point-to-point responses to the reviewers' comments as follows:

Reviewers' comments are in black.

Author's responses are in blue color.

Changes in the manuscript are in red color.

Sincerely,

Weihua Chen

On behalf of the authors

Response to Reviewer #1:

Referee #1 comment:

Wu et al. provides a novel framework for capturing the essence of nocturnal ozone increases (NOI), which is an important area of research often neglected in analysis of the ozone budget. They break the causes of NOI down into clearly discernable phenomena and present evidence that the majority of NOI events in the Pearl River Delta (PRD) are caused by Low Level Jets (LLJ). Further, they present a detailed case study of a LLJ NOI event, as well as a convective storm (Conv) NOI event.

The framework is intriguing and presents a valuable contribution to the literature, however, some modifications should be made prior to publication. In particular, the authors need to more clearly define their methodology and make a stronger case for using the K index as a proxy for convective storm events.

Response:

We thank the reviewer for the positive comments. We have revised our paper according to your comments as follow:

(1) We have reorganized the **Data and methods** section and provided clearer description regarding methodology used in this study. Please refer to our detailed responses to comments #1, #2, #3, and #5.

(2) We have introduced an additional indicator (cloud-top temperature, CTT) to prove the applicability of K index. Please refer to our detailed responses to comments #5.

General Comments:

1. The definition of NOI first appears in lines 118-120, where values increase by at least 10 μg m^{-3}. However, this comes from a 2020 reference and not every preceding study in the introduction that mentions NOI contains findings that are consistent with this strict definition (e.g. Caputi et al. 2019). It would be helpful if the authors clarify that (I assume) this is the definition they employ specifically in their study (e.g. "for our analysis, we define NOI as …"). Additionally, the definition needs to be clearer. For example, I am left unsure whether "values increasing by at least 10 μg m^{-3}" mean increasing from the daytime minimum, the previous hour's value, or what exactly.

Response:

According to the reviewer's comments, we have provided a specific definition of NOI used in this study in Lines 155-158:

'For our analysis, we define a nocturnal O_3 increase (NOI) event as O_3 concentrations peaked at night (from 21:00 LT to 06:00 LT the next day), with an increase in levels of at least 10 μg m^{-3} compared to the previous hour and a decrease of less than 10 μg m^{-3} in the next hour. The corresponding nighttime peak concentration of O_3 is referred to as the nocturnal O_3 peak (NOP) (Zhu et al., 2020).'

2. Some aspects of the methodology need clarification. In Section 3.1, are the statistics (e.g. 53 +/- 16 d yr^{-1}) from an aggregate of the air quality monitoring stations, and are the error values and error bars in Figures 3 and 4 calculated by a pooled standard deviation? The authors then discuss the proportions of events attributable to LLJ vs. Conv, does this come from the ERA5 data? If so, are the causes of an individual NOI event (LLJ, Conv, LLJ+Conv, Other) determined by an instantaneous snapshot of the meteorological conditions over the air quality station, or a regional average for a given night? Please connect the dots between the different methods discussed (e.g. CMAQ, IPR, air quality stations, meteorological stations, ERA5) and where specifically they employed in the results.

Response:

(1) The statistic of average annual frequency of NOI events (53 ± 16 d yr^{-1}) is an average value of the 16 air quality monitoring stations. We have clarified it in Lines 161-162:

'In addition, the regional values of NOI and NOP from the 16 air quality monitoring sites were averaged.'

(2) The error values and error bars in Figures 3 and 4 indicate the range of deviations for the different station. We have clarified it in Line 317, Line 344 and Line 373:

'… The error bars indicate the range of deviations for the 16 air quality sites.'

(3) The proportions of NOI events attributable to LLJs vs. Conv are calculated based on the ERA5 data. We have clarified it in Lines 164-165:

'Low-level jets (LLJs) and convective storms (Conv) are defined in this study based on the above site-specific ERA5 reanalysis dataset.'

(4) The causes of an individual NOI event are determined by an instantaneous snapshot of the meteorological conditions over the air quality station instead of a regional average for a given night. We have clarified it in Lines 141-143 and Lines 164-165:

'Since the ERA5 reanalysis dataset was gridded, the nearest-neighbour interpolation method is used to obtain site-specific meteorological variables at the 16 air quality monitoring sites.' (Lines 141-143)

'Low-level jets (LLJs) and convective storms (Conv) are defined in this study based on the above site-specific ERA5 reanalysis dataset.' (Lines 164-165)

(5) We have added Table 1 to summary the dataset used in this study and their purpose as follow:

Table 1. Summary of the dataset used in this study

Product	Period	Sites	Temporal resolution	Spatial resolution	Purpose
Observed O_3 data	2006-2019	16 sites	1 h	-	Spatiotemporal analysis of NOI and NOP, model performance
Observed vertical O_3 data	2019	Dongguan superstation	12 min	-	Analysis of an NOI event caused by Conv
Observed meteorological data	2017.09.08-2017.09.15	9 sites	1 h	-	Model performance
Observed Cloud-top Temperature (CTT) data	2019	Gridded data	1 h	0.1°	Indicator of the occurrence of convection
ERA5 reanalysis dataset	2006-2019	Gridded data	1 h	0.25°	Definition of LLJs and Conv

We have also clarified the purpose of these datasets in the revised manuscript as follows:

'The observed hourly O_3 data were used for subsequent NOI and NOP analyses, and evaluation of O_3 simulations.' (Lines 122-123)

'In this study, based on the above observed hourly O_3 data at the 16 air quality monitoring sites, NOI events are identified at each site, yet only one NOI event is recorded per night, regardless of how many NOI events occur in a single night.' (Lines 158-161)

'The vertical distribution of O_3 concentrations observed at the Dongguan superstation (23.02° N, 113.79° E) in 2019 is also used to investigate the impact of Conv on a particular NOI event.' (Lines 124-125)

'The observed meteorological data were used to evaluate the performance of the model.' (Line 131)

'To investigate the impacts of meteorological processes on NOI events, the ERA5 reanalysis dataset (https://cds.climate.copernicus.eu/cdsapp#!/home, last accessed on February 10, 2022) provided by the European Centre for Medium-Range Weather Forecasts (ECMWF) is used in this study.' (Lines 133-135)

'Low-level jets (LLJs) and convective storms (Conv) are defined in this study based on the above site-specific ERA5 reanalysis dataset.' (Lines 164-165)

'The observed cloud-top temperature (CTT) data for 2019 obtained from the Fengyun-2G satellite (http://satellite.nsmc.org.cn/, last accessed on August 31, 2022) are used to indicate the occurrence of convection.' (Lines 144-146)

'Due to the lack of observed vertical profiles of wind speed, the WRF-CMAQ model is employed to investigate the effects of LLJs on a selected NOI event.' (Lines 198-199)

3. In Figure 3 (and all of the accompanying analysis), the authors take data from 2006 – 2019 and break it into two halves, with a breakpoint at 2012 for (a) and (b) and 2016 for (c) and (d). It would be nice to have some physical justification for applying a discontinuity in the linear trend analysis at these specific years. For example, did any local policies on emissions change in 2012 or 2016? If there was no specific justification in mind, the authors should clearly state this section of their research as exploratory and at least speculate on a physical cause, otherwise, this could be seen as "p-hacking". Also, please state the statistical method used for calculating the p-values of the linear trends.

Response:

Thanks for your suggestion.

(1) The breakpoint at 2012 for NOI and LLJs is more likely related to the change of urbanization. We have clarified it in Lines 275-284:

'Both the frequency of NOI and LLJs present increasing trends before 2012 and decreasing trends thereafter, which was related to urbanization. Previous studies have shown that urbanization has large effects on the frequency of LLJs by changing surface conditions (roughness and soil moisture) and further affecting the turbulence and geostrophic wind speed (McCorcle, 1988; Fast and McCorcle, 1990; Kallistratova, 2008; Nikolic et al., 2019; Ziemann et al., 2019). Kallistratova (2008) and Nikolic et al. (2019) pointed out that negative correlation was found between urban areas and the frequency of LLJs. During 1987-2017, the urban areas in the PRD region grew at an average rate of 8.82% yr^{-1} (Yang et al., 2019a) and reached maximum urban land expansion growth rate of 6.66% during 2010-2015 (Zhang et al., 2021). Therefore, the trends for the frequency of NOI and LLJs were quite different during these two periods (2006-2011 and 2012-2019).'

(2) As the reviewer said, the breakpoint at 2016 for NOP and MDA8 O_3 was more likely related to the change of precursor emission. We have clarified it in Lines 299-311:

'The variations of NOP and MDA8 O_3 during the two periods (2006-2015 and 2016-2019) are more likely related to the change in precursor emissions. The continuous increase in the emissions of anthropogenic VOCs and NO_x resulted in the gradual increase of O_3 concentrations between 2006 and 2012 (Ma et al., 2016; Li et al., 2017; Zhong et al., 2018; Liao et al., 2021). However, since the implementation of Air Pollution Prevention and Control Action Plan (APPCAP) in 2013, NO_x emissions was dramatically decreased by 21% in 2017 compared to 2013 (Feng et al., 2019; Yang et al., 2019b). The weakening of NO titration caused by the dramatic decrease in NO_x emissions and the continuously increasing VOCs emissions due to the lack of controls became important drivers of the sharp rise in O_3 since 2015 (Li et al., 2019; Mousavinezhad et al., 2021; Li et al., 2022). Furthermore, the decreasing $PM_{2.5}$ levels and the increasing atmospheric oxidizing capacity in the PRD region in recent years have also been considered as important contributors to accelerated O_3 growth during 2016-2019 (Gong et al., 2018; Li et al., 2019; Han et al., 2019). Consequently, NOP and MDA8 O_3 present slower increase rate before 2015 and higher increase rate thereafter.'

(3) We used Mann-Kendall test method to calculate the p-value of the linear trends. We have clarified it in Lines 190-196:

'In this study, the nonparametric Mann-Kendall (M-K) test (Mann, 1945) is used to determine the statistical significance (p values) associated with the annual trends of NOI, NOP, MDA8 O_3, LLJs and Conv, etc. A significance level of $p < 0.05$ was used to test the significance of the inter-annual trend. The magnitude of a given trend is calculated by the nonparametric Theil-Sen (T-S) estimator (Sen, 1968). The advantage of the M-K test and the T-S estimator is that they do not require prior assumptions of the statistical distribution for the data and are resistant to outliers. The M-K test and the T-S estimator have been widely used in previous O_3 trend studies (Wang, et al., 2019; Lu et al., 2020; Li et al., 2022).'

4. I appreciate that the authors recognize the controversy of whether NOI increases or decreases the following days ozone concentration in lines 426-429. In lines 241-243, the authors state that "NOP is significantly positively correlated with MDA8 O_3 … implies that daytime O_3 concentration levels potentially affect NOP". To further strengthen this discussion on the relationship between daytime and nighttime ozone, I would suggest the authors look at the correlations between: 1) the afternoon MDA8 and the following night's NOP, and 2) the NOP and the following afternoons MDA8, and explicitly report the results from both. This will help elucidate the arrows of causality between daytime and nighttime ozone concentrations in the PRD.

Response:

Thanks for pointing out this critical issue, which is indeed a good point. According to the reviewer's comments, we have analyzed the correlation between the afternoon MDA8 O_3 and the following night's NOP (Fig. 12a), and the correlation between NOP and the following afternoon MDA8 O_3 (Fig. 12b), respectively. The results show that MDA8 O_3 was positively correlated with NOP, suggesting that daytime MDA8 O_3 and nighttime NOP affected each other. We have provided more discussion in Lines

496 - 501:

'… To further explore the relationship between the daytime MDA8 O_3 and nighttime NOP in the PRD region, we display the correlation between the MDA8 O_3 and the following night's NOP (shorthand MDA8-NOP) (Fig. 12a) and the NOP and the following MDA8 O_3 (shorthand NOP-MDA8) (Fig. 12b), respectively. The results show that MDA8 O_3 was positively correlated with NOP with a correlation coefficient of 0.63 ($p<0.01$) and 0.56 ($p<0.01$) for MDA8-NOP and NOP-MDA8, respectively, suggesting an interplay between daytime O_3 and NOP in the PRD region.'

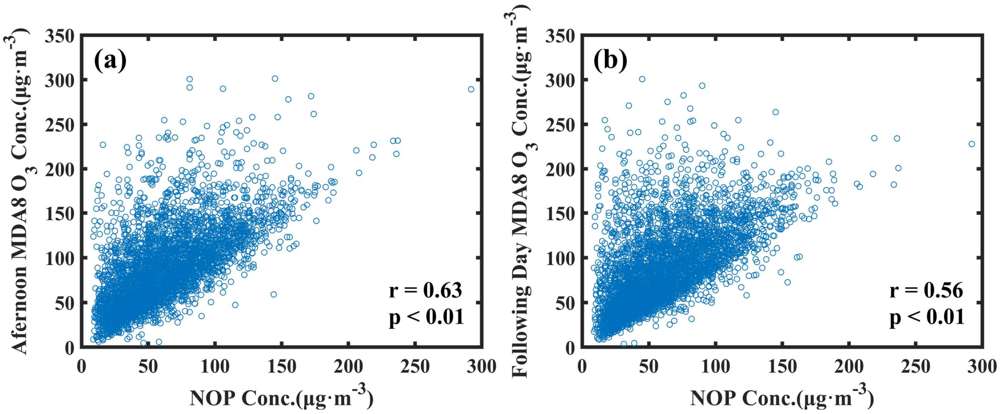

Figure 12. Correlation between (a) the afternoon MDA8 O_3 concentration and the following night's NOP concentration and (b) the NOP concentration and the following afternoon MDA8 O_3 concentration

5. As for my most significant concern, the authors use a K index (KI) > 30 as an indication of whether convective storms are occurring in the PRD on a given night. While I understand the need to make approximations when using large datasets, deep convection can occur when KI<30 and KI>30 does not guarantee the presence of deep convection, so there needs to be additional justification that KI>30 is a valid metric for what the authors are trying to capture. For example, the authors might look at a random subset of 10 nights where KI < 30 and 10 nights where KI > 30, and qualitatively compare the radar and/or satellite imagery in the PRD. Alternatively, they could look at the relationship between KI and peak vertical velocity in the ERA5 for the PRD at night, and show that KI=30 is a good cutoff for their purposes.

Response:

Thanks for pointing out this critical issue.

Firstly, apart from KI, cloud-top temperature (CTT) value was further introduced as an indicator of the occurrence of convective system. A lower CTT value suggest that the probability of convection event is higher. According to the work of Ai et al. (2016), CTT lower than -35 ℃ indicate the occurrence of convection. In addition, we have randomly selected 10 nights with KI > 30 ℃ (Table S3 and Figure S1) and 10 nights with KI < 30 ℃ (Table S4 and Figure S2) and calculated their corresponded CTT values. For the cases with KI > 30 ℃, 10 out of 10 cases were with CTT lower than -35 ℃. And the spatial distribution of CTT shows that the CTT exhibits a distinct circular lower value area over the selected sites, indicating the occurrence of convective system.

For the cases with KI < 30 ℃, 6 out of 10 cases were with CTT higher that -35 ℃, while the rest of 4 cases with no CTT data due to cloudless weather. And the spatial distribution of CTT does not show the features of a convective system, suggesting that convective processes have not been observed for the selected 10 cases with KI < 30 ℃.

The above results suggested that the KI > 30 ℃ criterion is a valid metric to capture the occurrence of convection. We have provided more information in Lines 174-185:

'Cloud-top temperature (CTT) was also introduced as an indicator of the occurrence of convective systems and further used to evaluate the applicability of KI. The lower the CTT, the higher the probability of convection event. According to the work of Ai et al. (2016), CTT lower than -35 ℃ indicates the occurrence of convection. We randomly selected 10 nights with KI > 30 ℃ (Table S3) and 10 nights with KI < 30 ℃ (Table S4) and examined the corresponding CTT values. In the cases with KI > 30 ℃, the CTT values were lower than -35 ℃ in 10 out of 10 nights (Table S3). And the spatial distribution of CTT showed that they had a distinct circular area with lower value over the selected sites, indicating the occurrence of convective systems (Fig. S1). For the cases with KI < 30 ℃, 6 out of 10 nights were with CTT higher than -35 ℃, while the rest 4 nights had no CTT data due to cloudless weather (Table S4). The spatial distribution of CTT did not show the features of a convective system (Fig. S2), suggesting that convection was not observed for the selected 10 cases with KI < 30 ℃. The above results suggest that the KI > 30 ℃ criterion is a valid metric to capture the occurrence of convection.'

Table S3. Site-specific values of KI and CTT for the randomly selected cases with KI > 30 °C

Time (LT)	Site	KI (°C)	CTT (°C)	Figure
2019/04/11 22:00	WQS	33	-49	Figure S1 (a)
2019/04/16 00:00	XP	33	-45	Figure S1 (b)
2019/05/26 00:00	JJJ	32	-62	Figure S1 (c)
2019/06/25 23:00	DH	34	-51	Figure S1 (d)
2019/07/02 22:00	NCYL	35	-54	Figure S1 (e)
2019/07/21 21:00	NCYL	32	-47	Figure S1 (f)
2019/08/08 22:00	LY	39	-70	Figure S1 (g)
2019/08/24 23:00	LY	31	-68	Figure S1 (h)
2019/09/14 23:00	HJC	31	-48	Figure S1 (i)
2019/10/07 00:00	TJ	37	-68	Figure S1 (j)

Table S4. Site-specific values of KI and CTT for the randomly selected cases with KI < 30 °C

Time (LT)	Site	KI (°C)	CTT (°C)	Figure
2019/04/12 22:00	WQS	29	12	Figure S2 (a)
2019/04/17 00:00	XP	11	Cloudless	Figure S2 (b)
2019/05/03 00:00	JJJ	29	2	Figure S2 (c)
2019/06/27 23:00	DH	25	Cloudless	Figure S2 (d)
2019/07/04 22:00	NCYL	26	-3	Figure S2 (e)
2019/07/25 21:00	NCYL	27	Cloudless	Figure S2 (f)
2019/08/04 22:00	LY	26	-20	Figure S2 (g)
2019/08/20 23:00	DH	29	-6	Figure S2 (h)
2019/09/15 23:00	HJC	28	Cloudless	Figure S2 (i)
2019/10/08 00:00	TJ	26	19	Figure S2 (j)

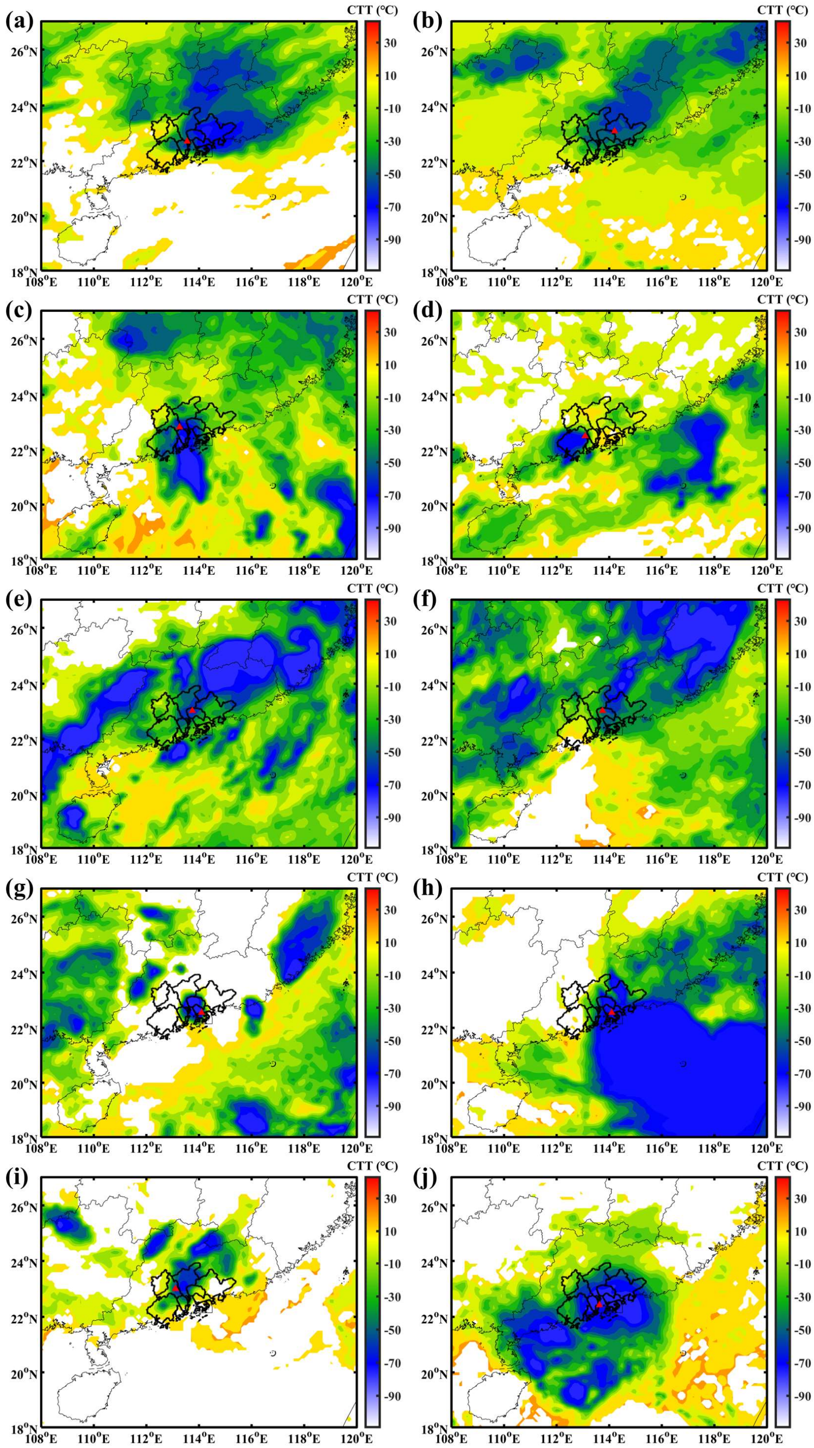

Figure S1. Spatial distribution of CTT for the randomly selected cases with KI > 30 °C. (a) to (j) refer to Table S3.

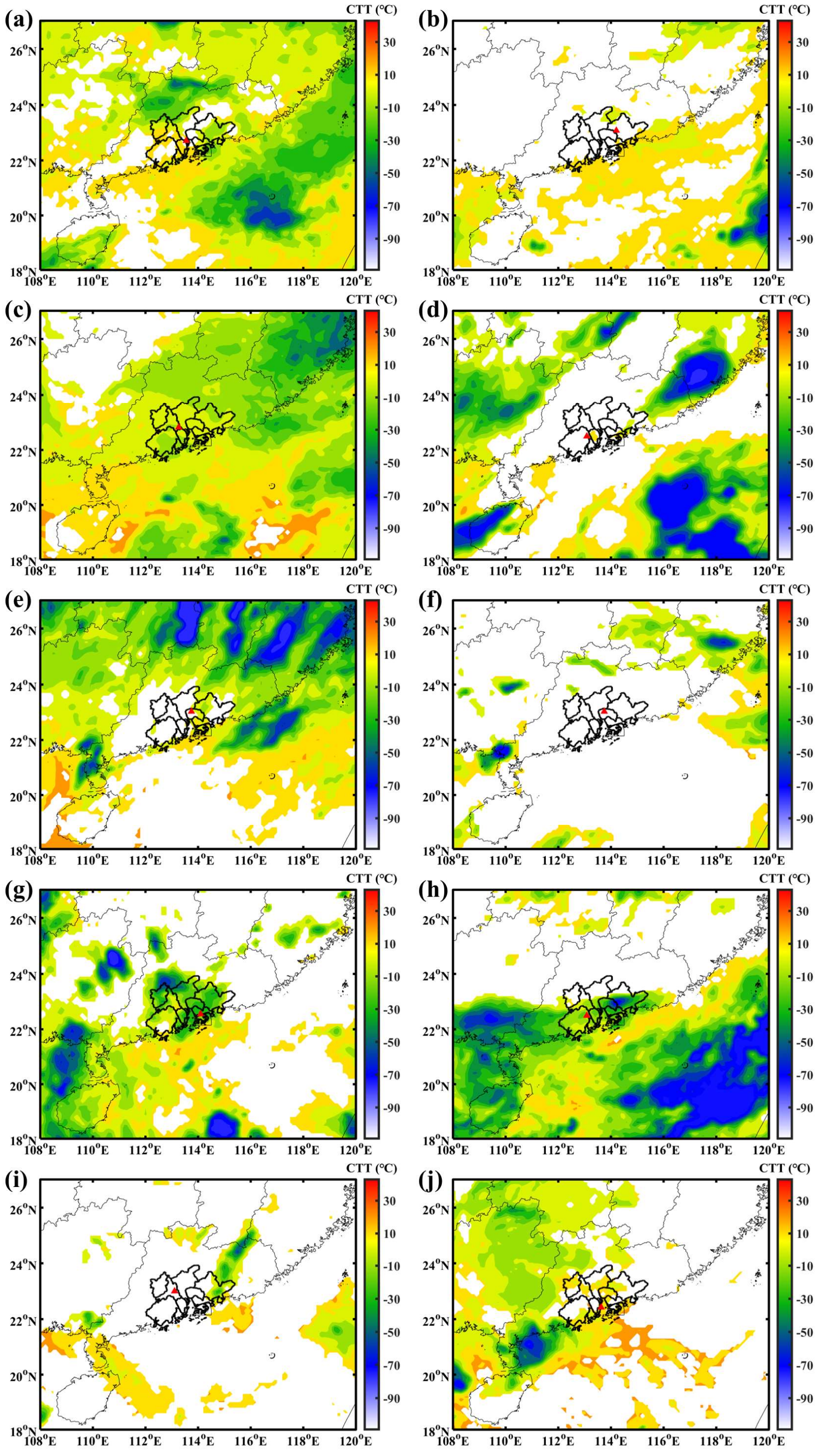

Figure S2. Spatial distribution of CTT for the randomly selected cases with KI < 30 °C. (a) to (j) refer to Table S4.

6. On a related note to (5), the case study of a Conv event presented in section 3.6 could use some additional supporting evidence and data. Figure 9b shows updrafts of only up to 5 cm s^{-1}, which are at least an order of magnitude lower than what would be expected in convective showers and

thunderstorms. While some light precipitation is indicated in Figure 9a, it would be better to also see a radar and/or satellite image of the alleged convective storms that night.

Response:

Thanks for pointing out this critical issue.

(1) We agree with the reviewer that the vertical wind velocity was relatively low, which has also been found in previous studies (Ploeger et al., 2021). Although the vertical wind velocity was relatively low, the vertical velocity results show continuous updraft trends at 1-3 km altitude during the afternoon (Fig. 8b), which still can indicated the happening of convection.

(2) According to the reviewer's comments, we used additional data (cloud-top temperature, CTT) to provide evidence for the occurrence of convection. The spatial distribution of CCT (Fig. 9) showed that the CTT was -66 °C over Dongguan, which was lower than the criteria (-35 °C) for the happening of convection. The CTT results further suggested a high possibility of the happening of convection process.

We have provided the related description in Lines 398-404:

'The KI remains above 36 °C (Fig. 8a) and the vertical velocity show continuous updraft trends at 1-3 km altitude from 14:00 to 23:00 (Fig. 8b), indicating a high possibility of convection. Although the magnitude of vertical velocity was relatively low, it also has been found in previous studies (Ploeger et al., 2021). In addition, the spatial distribution of CTT show that the CTT value at 18:00 over Dongguan was around -70 °C (Fig. 9a), which was lower than the criterion (-35 °C) for the happening of convection process. The results of KI, the vertical velocity and the CTT indicate that the possibility of a convection process is high.'

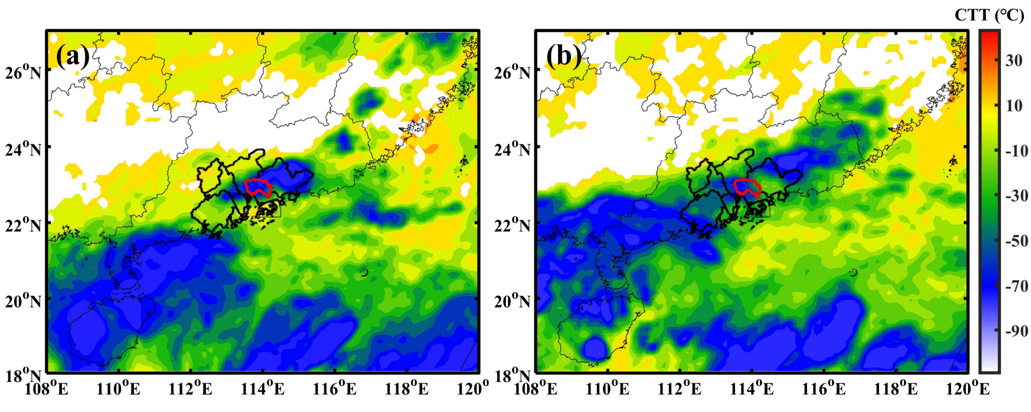

Figure 9. Spatial distribution of cloud-top temperature (CTT) at (a) 18:00 and (b) 21:00 LT on September 3, 2019.

Specific Comments:

7. Line 45: Please explicitly introduce the chemical reaction for NO titration for unfamiliar readers.

Response:

Thanks for your suggestion. We have added more descript in Lines 43-46:

'During nighttime, O_3 production ceases owing to the absence of sunlight, and dry deposition and NO titration (Eq. (1)) remove O_3 directly from the atmosphere, lead to relatively low O_3 concentrations at night (Jacob, 2000; Brown et al., 2006).

$$NO + O_3 \rightarrow NO_2 + O_2 \quad (1)$$'

8. Line 50: "around 3:00 in the morning" local time or UTC? Please specify.

Response:

It is local time. We have modified it in Line 51:

'… around 3:00 (LT) in the UK …'

9. Line 50: "118 μg m^{-3} in the UK" compared to roughly what values in the daytime?

Response:

Thank you. We compared the value 118μg m^{-3} with monthly average daytime values (69 ± 10 μg m^{-3}). We have modified the description in Lines 50-52:

'Kulkarni et al. (2015) found that NOI events were observed around 03:00 (LT) in the UK, with concentrations as high as 118 μg m^{-3}, which was much higher than the monthly average daytime O_3 concentration (69 ± 10 μg m^{-3}).'

10. Lines 51-52: "and the annual trend was found to be increasing" in terms of frequency of occurrence or intensity? Or both?

Response:

The "increasing" refers to the nocturnal O_3 concentration. We have modified it in Line 53-54:

'… and the annual trend of nocturnal O_3 concentration was found to on the increase …'

11. Lines 52-53: "High nocturnal O_3…pollution events" but in lines 426-429 you mention that this is

controversial. Better to be consistent.

Response:

We have deleted the original sentence "High nocturnal O_3…pollution events" to avoid confusion, and modified the description in Lines 490-496:

'The occurrence of NOI events is likely to impact the O_3 levels on the following day, which makes O_3 prevention more complex and challenging (Ravishankara, 2009; Sullivan et al., 2017). However, the relationship between NOI events and the following daytime O_3 pollution remains unclear and controversial. Kuang et al. (2011) and Sullivan et al. (2017) revealed that NOI events led to a higher increasing rate of O_3 and worse air quality on the following day, while Klein et al. (2019) and Caputi et al. (2019) observed lower O_3 levels during the daytime following NOI events.'

12. Line 54: We use the word "proven" in mathematics but not science. "Shown", "suggested", "provided evidence for", or anything similar could be used instead.

Response:

We have replaced "proven" with "shown" throughout the manuscript.

13. Line 63: "With an altitude of about 500 m" this is a general average cited in Stull, how well does this apply to the PRD?

Response:

Sugimoto et al. (2009) had conducted a campaign to observe boundary layer height in the PRD region by using lidar and found that the nocturnal boundary layer height is around 500 m. And Fan et al. (2022) reported a nocturnal boundary height of around 400 m in the PRD region. Therefore, the nocturnal boundary layer height was around 400-500 m in the PRD regions, which was comparable with the average value provided by Stull, (1988). We have modified the description in Lines 62-63:

'… with an altitude of 400-500m (Stull, 1988; Sugimoto et al., 2009; Fan et al., 2022).'

14. Line 69: Please clarify what is meant by "dynamic variation".

Response:

We apologized for our vague description. We have rewritten this sentence in lines 70-72:

'Dias-Junior et al. (2017) revealed that downdrafts induced by Conv play an important role in triggering

NOI events in the Amazon region of Brazil based on 1-yr observations.'

15. Line 71: Please change "Tropospheric" to "Free Tropospheric" because we are distinguishing multiple layers of the troposphere in this study (residual layer, nocturnal boundary layer, free troposphere).

Response:

Modified as suggested.

16. Line 96: "attributable to differences … urbanization". Is this because of the differences in nighttime NO emissions in urban vs. rural areas? Please state.

Response:

We apologized for the vague expression in the original manuscript. We have rewritten this sentence in lines 96-98:

'Tong and Leung (2012) observed a double-peak pattern of diurnal O_3 variation in Hong Kong during 1990-2005, and found that nocturnal O_3 peaks are sometimes higher than daytime maxima.'

17. Lines 103-104: "Moreover, high daytime … in the PRD region". Please add citation. Also, another motivating factor would probably be the high population in the PRD and the number of people air quality affects in this region?

Response:

We have modified this sentence in line 104-107:

'… In addition, high population densities and increasing number of people active at night in the PRD region make NOI events an important potential risk to human health (Kurt et al., 2016; Carré et al., 2017; Yang et al., 2019a; Zhang et al., 2021).'

18. Figure 1: If it wouldn't create much additional work, it may be worth shading this map with terrain instead of coloring the political regions. The fill colors for the political boundaries don't add much useful information to the plot.

Response:

We have accordingly replotted Figure 1 as follows:

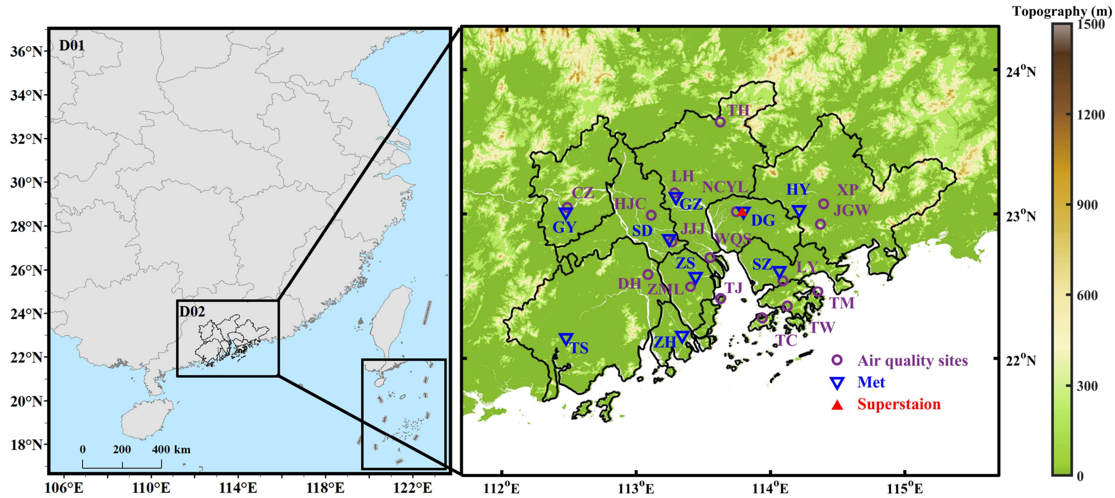

Figure 1. Model domains and locations of 16 air quality monitoring sites (purple dots), 9 meteorological stations (blue triangles), and Dongguan superstation (red triangles). The underlying figure shows the elevation of the terrain (m).

19. Line 142: Again, please avoid using the words “proof” or “proven” in a science article.

Response:

We have replaced “proven” with “shown” throughout the manuscript.

20. Line 164: Consider changing “explored” to “utilized”

Response:

We have replaced “explored” with “utilized” in the revised manuscript.

21. Lines 201-205: These statistics are cutoff ranges that the EPA considers a model acceptable or unacceptable to use, correct? Please specify the purpose of introducing these values, statistics, and ranges here – it isn’t entire clear.

Response:

The reviewer is correct. These statistics are cutoff ranges that the EPA considers a model acceptable or unacceptable to use. We introduced these statistics to evaluate the model performance by using some specific quantified index. We have modified it in Lines 245-251:

‘The evaluation protocols of the U.S. Environmental Protection Agency (EPA, 2017) are used to evaluate the performance of the meteorological parameters. The simulated results were accepted when the statistics met the criteria listed as follows: MB ≤ ±0.5 °C and IoA ≥ 0.8 for simulated T2; MB ≤ ±5% and IoA ≥ 0.6 for simulated RH; and MB ≤ ±0.5 m/s, RMSE ≤ 2.0 m/s, and IoA ≥ 0.6 for simulated WS10. The evaluation protocols of the Ministry of Environmental Protection of China (MEP, 2015) are used to evaluate the performance of O_3 and the simulated results were acceptable if the statistics met the criteria listed below: −15% < NMB < 15%, NME < 35%, and r > 0.4.'

22. Lines 219-220: In order to consider LLJ events a "downdraft", there must be an assumption that the LLJ is inducing turbulent mixing from the vertical wind shear it creates. Please state this.

Response:

We have clarified it in Lines 167-168:

'It's worth noting that the LLJs defined in this study only consider the turbulence mixing induced by their vertical wind shear.'

23. Line 264: "Below" -> please specify "below the jet"

Response:

We have replaced "below" with "below the jet" in Line 323.

'Given that LLJs can enhance turbulence below the jet …'

24. Lines 278 – 279: "it can bring clean marine air into the PRD region" I assume this is at the surface?

Response:

The reviewer is correct. We have modified this sentence in Lines 337-338:

'… This is mainly due to the typical Asian monsoon circulation, which brings clean marine air to the lower troposphere of the PRD region in summer…'

25. Line 297: what is meant exactly by O_3 from the daytime "enters" the RL between 21:00 and 03:00? Why isn't the O_3 already inside the RL from the minute the daytime boundary layer fades into the RL?

Response:

The reviewer is correct. O_3 has already stayed inside the RL instead of entering the RL. We have modified this sentence in Lines 358-359:

'As the sun sets and the daytime boundary layer fades away, the O_3 produced during daytime remains at a relatively high level in the RL during 21:00-03:00.'

26. Line 299: Horizontal transport to where? And by "vertical transport" of O_3 are the authors referring to dry deposition, convection, or both?

Response:

(1) The horizontal transport means the O_3 remained at the RL can be horizontal transported to the downwind area.

(2) The vertical transport includes the vertical exchange caused by LLJs, convection, and the O_3 dry deposition processes.

We have modified this sentence in Lines 360-362:

'After 03:00, the O_3 concentrations in the RL decreased due to horizontal transport to downwind area and vertical transport (e.g., LLJs, convection, O_3 dry deposition process) during 21:00-03:00, …'

27. Line 319: How are individual sites classified as either rural or urban?

Response:

HKEPD (2017) classified the individual sites into urban and rural according to the land use type and surrounding environment of the monitoring stations.

According to the comments #7 provided by the reviewer #2, the spatial distribution of the sites is more important than the type of these sites. Therefore, we only discuss the spatial difference instead of difference between urban and rural, and we have deleted the description associated with the difference between urban and rural in the revised manuscript.

28. Figure 8: Similar to general comment (3), please provide a justification for the break at 2012.

Response:

We have deleted Figure 8 and the associated description in the revised manuscript. Please refer to our detailed responses to the reviewer #2 comments #7.

29. Lines 352-353: the modeled downdraft in Figure 9b occurs *after* the observed O_3 intrusion into the nocturnal boundary layer. It may be that the timing of the model is slightly off, but this should

be acknowledged rather than stated as a clear cause an effect.

Response:

Thanks for pointed out this issue. We have acknowledged it in Lines 415-418:

'Although the modeled downdraft occurred at 22:00-23:00 (Fig. 8b) was around half an hour later than the observed O_3 intrusion into the nocturnal boundary layer (Fig. 8c) due to the model bias, the modeled results can still generally capture the occurrence of convection processes.'

30. Line 369: "meet the criteria" -> the EPA criteria specified earlier? Please clarify.

Response:

The reviewer is correct. We have clarified it in Lines 431-432:

'The results show that WS10 was reasonably well simulated, as the regional average of MB, RMSE, and IoA met the EPA criteria mentioned in section 2.6.'

31. Figure 10: The black lines (NET) is categorically different from ozone and wind because it is not a meteorological phenomena. This was a bit confusing to my eye at first because it is plotted along with ozone and wind, but in reality it relates more to the bars. Consider at least changing the circle marker to a triangle for the black NET lines.

Response:

According to the reviewers' comments, we have replotted Figure 10 as follows:

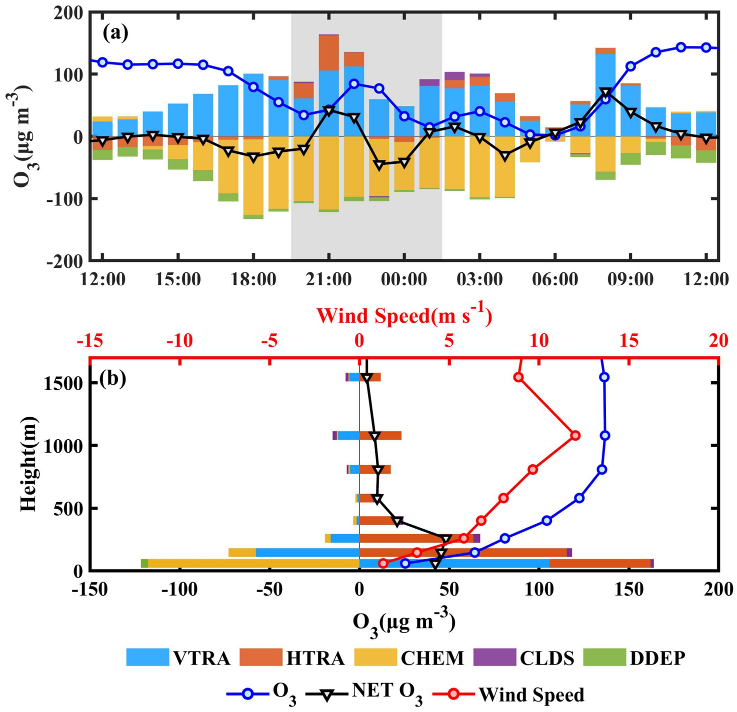

Figure 10. Contribution of individual processes to (a) hourly O_3 concentration near the surface during September 13-14, 2017 and (b) vertical O_3 concentration at 21:00 on September 13, 2017. VTRA: vertical transport, the net effect of vertical advection and diffusion; HTRA: horizontal transport, the net effect of horizontal advection and diffusion; CHEM: gas-phase chemistry; CLDS: cloud processes; DDEP: dry deposition; NET: the net change in O_3 due to all atmospheric processes.

32. Figure 10b: Would it be possible to plot the momentum flux in the model as well to get an indication of shear below the LLJ? Or no because this was not a large eddy simulation?

Response:

We apologized that we cannot plot the momentum flux since we did not conduct large eddy simulation.

33. Line 428: Caputi et al. 2019 also found lower ozone the following day when more mixing of ozone from the residual layer to nocturnal boundary layer occurred overnight.

Response:

We have modified this sentence in Lines 493-496:

'Kuang et al. (2011) and Sullivan et al. (2017) revealed that NOI events led to a higher increasing rate of O_3 and worse air quality on the following day, while Klein et al. (2019) and Caputi et al. (2019) observed lower O_3 levels during the daytime following NOI events.'

Technical Comments:

34. Line 91: “Long-tern” Long-term?

Response:

Modified as suggested.

35. Figure 6b caption: reference to blue but no blue in figure, assume orange?

Response:

Modified as suggested.

Reference:

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