July 8, 2021

Dear editor Xavier Querol and referees,

We appreciate for your work on our revised manuscript entitled "A comparative study to reveal the influence of typhoon on the transport, production and accumulation of O_3 in the Pearl River Delta, China". Hereby we present the responses to the further comments by referees. The revised version with and without marks will also be submitted.

We hope you would find the responses and revisions are adequate. If there is any question, please let us know.

Best regards,

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Response to Referee #2

Comment:
The Authors revised the original manuscript answering all the Reviewers' questions and comments adding the required missing information and clarifying the some discussed issues. The effects of typhoons on the transport, production and accumulation of O_3 are now convincingly described and discussed by the thorough analysis of episodes occurred in the Pearl River Delta, China.

Some of the revised sentences are now very long, complex and difficult to read. They should be revised for a better readability of the text. A final revision of the text and English form is therefore needed.

Response:
Thanks for your positive comments and valuable suggestions to help us improve the manuscript. The responses to the comments (in blue) and corresponding revisions (in red) are presented as follows (line numbers are those in the revised manuscript with author's changes).

Comment:
1. (Lines 123-125, Method) The content of these sentences is clear but their form could be revised and improved for better readability.

Response:
According to the suggestions of Referee #2 and #4, we revised the sentence into (in lines 126-127):
"Typhoons tend to result in more severe O_3 pollution in the PRD, as indicated by generally higher O_3 MDA1 and MDA8 values with the influence of typhoons than these without typhoons."

Comment:
2. (Lines 160-162, Method) Please check the English use of "set as".
It would be better specify that the trajectory endpoint height has been chosen to be representative of the PBL.

Response:
(1) After checking English grammar materials, "be set to + number" is more suitable than "be set as + number" to be used here.
(2) We agree. The trajectory height was chosen to better indicate the effect of long-range transport on O_3 pollution within the PBL.
Based on these suggestions, the sentence was revised into (in lines 164-165):
"Its height was set to 500 m above the ground to indicate the effect of long-range transport on O_3 pollution within the PBL (Park et al., 2007)."

Comment:
3. (Lines 175-179, Method) This sentence is very long, including a long description in brackets. It would be better readable if divided into separated sentences.

Response:
To make it brief, understandable and readable, we revised the sentence into (in lines 178-185):

"October 2015 and July 2016 featured with the most severe O_3 pollution under typhoon influence among all representative months in autumn and summer, respectively (Table S3). Therefore, O_3 pollution in these two months were modelled and used in further comparisons. Note that the modelling period in autumn was adjusted to 11 October–10 November 2015 to involve more O_3 pollution days (3–5 November 2015)."

Comment:
4. (Lines 235-241, Comparison of meteorological conditions) This sentence is very long and hardly readable. Brackets could be removed and the sentence included could be separated from the previous one.

Response:
We revised the sentence into (in lines 247-251):
"Near-surface parameters from routine monitoring datasets and the ERA-Interim re-analysis were used in the comparison. The parameters include air temperature, RH, wind speeds, cloud covers, PBL height and net surface solar radiation. Since there was no rainfall on most O_3 pollution days (indicated by the weather in Guangzhou (Table S4)), precipitation was not considered in the comparisons. For consistency, the parameters of ERA-Interim were extracted at the same time (14:00 LT) and the locations of sites (Fig. S1a) as those in routine monitoring."

Comment:
5. (Lines 359-363, Comparison of meteorological conditions) The description included in this sentence is complex and not very clear. It should be rephrased and better explained.

Response:
We revised the sentence into (in lines 370-375):
"This favoured the accumulation of locally sourced O_3. Based on the comparisons in this and previous section, typhoons did not provide more favourable conditions for O_3 production and accumulation simultaneously in the PRD in both autumn and summer, thus potentially resulting in reduced contributions in percentage of local emissions to O_3 pollution here."

Comment:
6. (Lines 503-505, Discussion and conclusions) It is not clear why the approach proposed by the Authors should be changed into that proposed by Thunis et al. (2019, 2020) for air quality management purposes. Should it be considered more reliable for a comprehensive air quality management?

Response:
In this study, we identified the sources of all O_3 in the PRD with or without the influence of typhoons, and suggesting that typhoons lead to higher (lower) contributions of non-local (local) emissions for O_3. Therefore, reducing emissions outside the PRD is likely to be more efficient to reduce O_3 in the PRD in the typhoon-induced scenario. For air management, the method we chose is not "dynamic" according to Thunis et al. (2019) and Thunis et al. (2020), that is, it is hard to know the response of O_3 pollution in the PRD to the fractional reduction of emissions in different regions. The efficiency of combined fractional local and non-local emission controls to reduce O_3 levels is unknown. Therefore, further studies are required to find the most effective strategies to alleviate O_3 pollution in

the PRD. In order to explain more clearly, we revised the sentence in lines 514-517 into:
"For air quality management, it is suggested to comprehensively evaluate the response of O_3 levels in the PRD to fractional local and non-local emission reductions so as to find the most effective strategies to alleviate O_3 pollution in different scenarios (Thunis et al., 2019; Thunis et al., 2020)."

Reference

Park, S. K., O'Neill, M. S., Stunder, B. J., Vokonas, P. S., Sparrow, D., Koutrakis, P., and Schwartz, J.: Source location of air pollution and cardiac autonomic function: trajectory cluster analysis for exposure assessment, J. Expo. Sci. Env. Epid., 17(5), 488-497, https://doi.org/10.1038/sj.jes.7500552, 2007.

Thunis, P., Clappier, A., Pirovano, G.: Source apportionment to support air quality management practices, A fitness-for-purpose guide (V 3.1), EUR30263, Publications Office of the European Union, ISBN 978-92-76-19744-7, doi:10.2760/47145, JRC120764, 2020.

Thunis, P., Clappier, A., Tarrason, L., Cuvelier, C., Monteiro, A., Pisoni, E., Wesseling, J., Belis, C. A., Pirovano, G., Janssen, S., Guerreiro, C., and Peduzzi, E.: Source apportionment to support air quality planning: Strengths and weaknesses of existing approaches, Environ. Int., 130, 104825, https://doi.org/10.1016/j.envint.2019.05.019, 2019.

Response to Referee #3

Comment:
Thanks for the detailed replies to the previous comments. Some revisions have been made and the manuscript is better in presenting the results and implications. Overall, the analyses and discussions are quite comprehensive. I think this manuscript is of good quality and almost ready to be published. The followings are a few comments.

Response:
Thanks for your positive comments and valuable suggestions to help us improve the manuscript. The responses to the comments (in blue) and corresponding revisions (in red) are presented as follows (line numbers are those in the revised manuscript with author's changes).

Comment:
1. From Table 1, the O_3 level on close typhoon-induced days during summer is much higher than that in other O_3 pollution days. What would be the possible reason for this? Usually, when typhoon approaches, the PRD region is under the influence of the peripheral subsidence (as in Figure S8), which facilitates the O_3 production and accumulation within the PRD region. To further justify that only typhoon-induced days were analyzed in this study, the author should consider adding a summary of the different effects on O_3 under these scenarios.

Response:
(1) We examined the meteorological conditions of the close typhoon-induced days in Sect. 3.5 of the manuscript. According to the conclusion of our analyses, favorable O_3 transport condition (strong northerly winds) and unfavorable O_3 production and accumulation conditions can be found on the close typhoon-induced days of summer. Since O_3 level is higher in north China than in south China in summer (Liu et al., 2020), enhanced transport is the reason that higher O_3 concentration in the PRD can be found in this scenario. We also calculated O_3 MDA8 concentrations in each summer scenario in Nanchang, a city to the north of the PRD with the distance of ~800 km. When northerly winds prevail, O_3 in Nanchang is likely to be transported for about 1–2 days to the PRD. As shown in Table R1, highest O_3 MDA8 in Nanchang can be found on one or two days before close typhoon-induced days in the PRD, which is also a sign of enhanced influence of O_3 transport to the south.

Table R1 O_3 MDA8 concentrations ($\mu g/m^3$) in the PRD and Nanchang on the summer days of different scenarios

Scenarios	the PRD (max value among nine PRD cities)	Nanchang (one day before the days of each scenario)	Nanchang (two days before the days of each scenario)
Typhoon-induced	205.4	100.8	97.9
Close typhoon-induced	225.7	128.1	126.9
No-typhoon	187.8	124.1	117.9

(2) The "typhoon-induced" scenario in this study includes ~60% of O_3 pollution days under typhoon influence in each season. Therefore, through the comparisons between the typhoon-induced and no-typhoon scenarios, we can summarize the general causes of O_3 pollution in the PRD under typhoon influence, that is, O_3 transport is enhanced but favorable O_3 production and accumulation conditions

cannot be met simultaneously. The summarization of the above general typhoon influences is presented in Fig. 11,

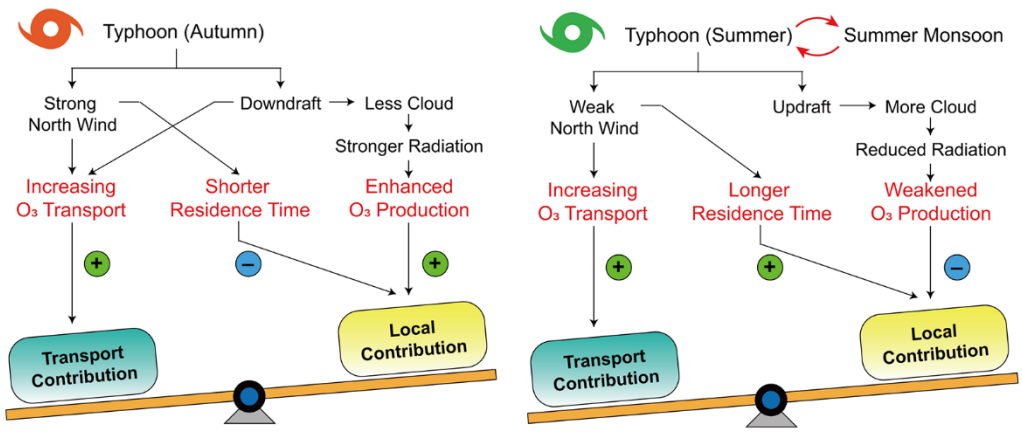

Figure 11. The summary of the causes of O_3 pollution in the PRD under typhoon influence in autumn and summer.

as well as the detailed discussions in Sect. 5 (in lines 463-481):

"In this study, we revealed the different impacts of typhoons on O_3 transport, production and accumulation in the PRD (as summarised in Fig. 11) through systematic comparisons of meteorological conditions, the contributions of various O_3 processes and sources in the typhoon-induced and no-typhoon scenarios. We found that typhoons tended to promote O_3 transport towards the PRD, but failed to provide more favourable O_3 production and accumulation conditions simultaneously, which limited the contribution of local emissions to O_3 pollution. Furthermore, there were also differences between the influence of typhoons on O_3 pollution in autumn and summer. More favourable transport conditions occurred in the typhoon-induced scenario in autumn, which was characterised by higher wind speeds and the increased influence of downdrafts. In summer, the mixed types of air masses in the typhoon-induced scenario were likely to bring more O_3 into the PRD than the clean marine air masses in the no-typhoon scenario, also suggesting enhanced O_3 transport under the influence of typhoons. Generally, typhoons led to cloudless conditions, stronger solar radiation, and thus more rapid O_3 production in autumn, but shorter APRTs (5–10 hours) suggest that locally sourced O_3 was hard to accumulate within the PRD. As a result, the contributions in percentage of local emissions to O_3 pollution decreased (slightly by ~5% for the polluted regions of the PRD in October 2015). In contrast, in summer, intensified updrafts associated with typhoons strengthened cloud formation, weakened solar radiation, and thus restrained local O_3 production. Longer APRTs (> 20 hour) under typhoon influence were far from sufficient to maintain high contributions of local emissions for O_3 pollution (which decreased by ~20% for the polluted regions of the PRD in July 2016). However, due to the variations of wind fields under different scenarios, the changes of local and transport contributions for O_3 led by typhoons were different in the southwest part of the PRD, that is, higher contribution from emissions within the PRD and reduced transport contribution occurred in the typhoon-induced scenarios in both seasons."

Close typhoon-induced scenario, including ~20% of O_3 pollution days in each season, is also considered in this study, and results show that O_3 transport is further enhanced but O_3 production and accumulation conditions both turns unfavorable in this scenario. The summary of the influence of close typhoon has been presented in Sect. 5 of the manuscript, in lines 481-483:

"As for the close typhoon-induced scenario, O_3 transport was further strengthened, but meteorological conditions in the PRD became less favourable for both the production and accumulation of O_3."

Comment:
2. In terms of calculating the APRTs, is the Kriging method used for plotting Figure 7? If not, which field or target grid is obtained?

Response:
Yes, the Kriging method was used to plot the distribution of APRTs in Fig. 7. We mentioned that in the Method section, in lines 174-175:
"APRTs in each point were averaged, and these averaged APRT values in all points were interpolated using the Kriging method to obtain field results for the further comparisons."

Comment:
3. In section 3.2, "southwest winds in the no-typhoon scenario in summer" (line 292 in the version with track changes). The prevailing wind direction on no-typhoon days needs to be clarified. In Figure S5d, it seems that southeast wind dominates. Also in Figure 3, the air masses are mainly from the south and east of PRD. However, in Figure 8d, the PRD region is mainly controlled by the southwest wind.

Response:
Thanks for pointing out that. South or southeast winds may prevail on no-typhoon days in summer in the PRD, thus "southerly wind" is more suitable to summarize the prevailing wind direction in this scenario. We revised the sentence in lines 296-297 into:
"… and southerly winds in the no-typhoon scenario in summer"
and the sentence in lines 458-459 into:
"… the site was located in the upwind regions under the prevailing of southerly winds,"

Comment:
4. In line 321-322 in the version with track changes, more evidence is needed to demonstrate the direct vertical transport of O_3. Downdrafts may not necessarily enhance the downward transport of O_3. In addition, the O_3 concentrations over the PRD region at different levels did not show much difference from Figure 5 (e, i). What were the average O_3 concentrations in the whole PRD region at different levels?

Response:
This study showed that downdrafts, high O_3 and low humidity occur in the same regions at multiple heights, thus suggesting the high possibility of downwards O_3 transport (Roux et al., 2020; Wang et al., 2020). We agree that downdrafts may not necessarily enhance the downward transport of O_3 when O_3 concentrations decrease with increase in altitude. Therefore, we calculated the mean O_3 mixing ratios in ERA-Interim in the area with the longitude of 21.5°N–24.5°N and the latitude of 111.5°E–115.5°E, which embraces the whole PRD. At 850 hPa, 700 hPa and 500 hPa, the value is 45.5 ppb, 48.5 ppb, and 46.5 ppb, respectively, on the typhoon-induced days of autumn as shown in Fig. 5 (a, e, i). O_3 levels at higher altitudes (700 hPa, 500 hPa) are slightly higher than those at 850 hPa, thus downwards O_3 transport potentially contribute to more severe O_3 pollution near the ground. More studies are required in future to quantify these contributions in different scenarios.

Reference

Liu, R., Ma, Z., Liu, Y., Shao, Y., Zhao, W., and Bi, J.: Spatiotemporal distributions of surface ozone levels in China from 2005 to 2017: A machine learning approach, Environ. Int., 142, 105823, https://doi.org/10.1016/j.envint.2020.105823, 2020.

Roux, F., Clark, H., Wang, K.-Y., Rohs, S., Sauvage, B., and Nédélec, P.: The influence of typhoons on atmospheric composition deduced from IAGOS measurements over Taipei, Atmos. Chem. Phys., 20, 3945–3963, https://doi.org/10.5194/acp-20-3945-2020, 2020.

Wang, H., Wang, W., Huang, X., and Ding A.: Impacts of stratosphere-to-troposphere-transport on summertime surface ozone over eastern China, Sci. Bull., 65, 276–279, https://doi.org/10.1016/j.scib.2019.11.017, 2020.

Response to Referee #4

Comment:
This manuscript systematically studied the difference between the typhoon-induced and no-typhoon O_3 pollution in the PRD area of China. It elucidated the influence of typhoon on O_3 transport, production and accumulation and found the seasonal difference of such influence. The revised version of the manuscript would be a good fit to ACP and publishable if minor comments below are addressed.

Response:
Thanks for your positive comments and valuable suggestions to help us improve the manuscript. The responses to the comments (in blue) and corresponding revisions (in red) are presented as follows (line numbers are those in the revised manuscript with author's changes).

Comment:
1. Line 91: "We mainly used the ERA-Interim re-analysis product in the analyses due to its more available parameters and high spatial coverage". Regarding "more available parameters", does it compare to observations or to other reanalysis datasets such as GDAS? Why "mention spatial coverage" here if only ERA-Interim extractions at the monitoring locations are used.

Response:
(1) ERA-Interim has more available parameters than other re-analysis datasets. Take three re-analysis datasets (FNL, GDAS, ERA-Interim) used in this study for example. As shown in Table R2, ERA-Interim has higher temporal/spatial resolutions and more parameter types than FNL and GDAS.

Table R2 The temporal/spatial resolution and parameters of the FNL, GDAS, and ERA-Interim re-analysis datasets

Parameter	FNL	GDAS	ERA-Interim
Temporal Resolution	6 h	6 h	3 h/6 h
Horizontal Resolution	1°	1°	0.25°
Vertical Layers	31	23	37
Air Temperature	√	√	√
Humidity (Relative or Absolute)	√	√	√
Planetary Boundary Layer Height	√	√	√
Horizontal Wind (u and v)	√	√	√
Vertical Wind (w)	√	√	√
Cloud Cover		√	√
Cloud Water Content	√		√
Solar Radiation			√
O_3 Mixing Ratio	√		√

In order to express clearly, we revised the sentence into (in lines 90-91):
"We mainly used the ERA-Interim re-analysis product in the comparisons due to its more available parameters than other re-analysis datasets and high spatial coverage…"

(2) In the comparison of meteorological parameters within the PRD, ERA-Interim was extracted at the monitoring locations to keep the comparison consistent. Besides, ERA-Interim on a larger scale

(both horizontally and vertically) was also utilized to compare O_3 transport, production and accumulation conditions, thus "high spatial coverage" is mentioned here.

Comment:
2. Line 97:"near-surface parameters from the forecast fields". What does it mean "forecast fields" if these fields came from the ERA-Interim re-analysis?

Response:
According to the documentation of ERA-Interim (https://confluence.ecmwf.int/display/CKB/ERA-Interim%3A+documentation; last assess: June 2021), there is two types of data, that is, analyses and forecast data, in ERA-Interim re-analysis. Analyses data was generated using observations, whereas forecasts data was generated using models and can provide more parameters with higher temporal and spatial coverage. According to this comment, we think that "fields" can be confusing and "data" is better. Thus we revised the sentence in line 94 into:
"(1) near-surface parameters from the analyses data …",
and the sentence in line 98 into:
"(2) near-surface parameters from the forecast data …".

Comment:
3. Line 124: "Higher O_3 MDA1 and MDA8 values can be generally found with the appearance of typhoons in comparison with days without typhoons in July, whereas these values are similar in October, further indicating the important role of typhoons in O_3 pollution in the PRD." Better to just delete ", whereas these values are similar in October", and replace it with "and October".

Response:
We agree that the expression needs to be brief and clear. The sentence is revised as (in lines 126-127):
"Typhoons tend to result in more severe O_3 pollution in the PRD, as indicated by generally higher O_3 MDA1 and MDA8 values with the influence of typhoons than these without typhoons."

Comment:
4. Line 158: "Hysplit model (Stein et al., 2015) with the Global Data Assimilation System (GDAS) and Line 190: "The Weather Research and Forecasting (WRF) model (version 3.2) provided the meteorological fields used as inputs." Why not use WRF outputs in Hysplit for consistency? Was GDAS or ERA-Interim used to drive WRF? What's the reason to use two different reanalysis datasets in the same study, considering the potential inconsistency between them?

Response:
In order to keep consistency, ideally, the meteorology inputs should be the same one. In this study, three datasets were used, namely, ERA-Interim used in the comparisons, FNL to drive WRF, GDAS to drive the Hysplit. The reasons of the above selections are as follows:

(1) ERA-Interim: According to Table R2 and our response to the first comment of Referee #4, ERA-Interim has more parameters with high temporal/spatial resolution and spatial coverage than other re-analysis, thus was used in the comparisons. Besides, we also used the observations within the PRD in the comparisons. Similar values as well as the same comparison results between scenarios using two datasets suggest the validity of this selection.

(2) FNL: FNL was normally used to drive the WRF. The statistics in Table S5 (of the Supplement) indicate the overall consistency between the WRF-modelling results and observations. Therefore, the selection of FNL as WRF input is acceptable for this study.

(3) GDAS: GDAS is the mostly used dataset to drive the Hysplit. In this study, WRF was only used to simulate the meteorology in two represented months (Oct. 2015, July 2016), thus the modelling results of WRF were not used to calculate the trajectories.

Comment:
5. Line 191: "… process the anthropogenic and biogenic emission files". "Emission files" better to be "emissions".

Response:
Accepted. We revised the sentence in lines 197-198 into:
"SMOKE (version 2.5) and MEGAN (version 2.10) were used to process the anthropogenic and biogenic emissions, respectively."

Comment:
6. Line 219: "… the difference between two sensitivity cases where emissions expect Ei and all of the emissions are zeroed out, respectively". Is "expect" in fact "except"?

Response:
Yes. Thanks for pointing out this mistake. We revised the sentence in lines 224-225 into:
"… (2) the difference between two sensitivity cases where emissions except E_i and all of the emissions are zeroed out, respectively (i.e., bottom-up BFM)."

Comment:
7. Line 303: "scenario in summer were overall higher than these in the corresponding no-typhoon scenario". Better to replace "these" with "those"?

Response:
Accepted. We revised the sentence in lines 313-315 into:
"… the absolute values of vertical wind speeds in the typhoon-induced scenario in summer were overall higher than those in the corresponding no-typhoon scenario."

Comment:
8. Supplement Figure S11: "Comparisons between the observational and modelling mean O_3 MDA8, daily NO_2 and NMHCs concentrations in the PRD." Better to use "observed and modeled", instead of "observational and modelling".

Response:
Accepted. We revised the first sentence of Fig. S11 note into:
"**Figure S11**. Comparisons between the observed and modelled mean O_3 MDA8, daily NO_2 and NMHCs concentrations in the PRD."

Comment:

9. Figure S11 and Table S5. The conclusions of this study that based on modeling results rely on accuracy of the simulations, especially on consistency in performance of the simulations of different seasons as well as of different scenarios that with or without typhoon influence. Is there any significant performance difference between typhoon induced and no-typhoon scenarios for simulated meteorology and air quality?

Response:

The modelling performance statistics of meteorological parameters (air temperature, relative humidity and wind speed) and pollutant concentrations (O_3-MDA8, NO_2, NMHCs) on the typhoon-induced and no-typhoon days of two represented months are shown in Table R3. For most parameters, WRF/CMAQ underestimates or overestimates them in the two scenarios of the same season, and the statistics are overall close. Therefore, there is no significant difference between typhoon-induced and no-typhoon scenarios in two months.

Table R3 The modelling performance statistics of meteorological parameters and pollutant concentrations on the typhoon-induced and no-typhoon days of two represented months. MB, mean bias; RMSE, root mean square error; NMB, normalized mean bias

Parameters	Statistics	Typhoon-induced, Oct. 2015	No-typhoon, Oct. 2015	Typhoon-induced, July 2016	No-typhoon, July 2016
Air temperature	MB (K)	-0.51	-0.24	0.52	-0.66
	RMSE (K)	2.16	1.70	2.45	1.92
Relative humidity	MB (%)	-5.33	-3.43	-6.85	-2.83
	RMSE (%)	14.45	10.58	14.31	11.27
Wind speed	MB (m/s)	0.52	0.61	0.09	0.15
	RMSE (m/s)	1.21	1.18	1.23	1.04
O_3 MDA8	MB ($\mu g/m^3$)	8.5	19.1	12.6	16.0
	NMB	0.05	0.15	0.08	0.12
NO_2	MB ($\mu g/m^3$)	-16.1	-12.3	-5.4	4.1
	NMB	-0.35	-0.28	-0.17	0.21
NMHCs	MB ($\mu g/m^3$)	-5.0	-3.7	-4.9	-5.0
	NMB	-0.20	-0.14	-0.33	-0.31

The following sentence was added to the end of Sect. 1 (Evaluation of WRF and CMAQ modelling results within the PRD) in Supplement:

"As shown in Fig. S6, there is no significant difference between the modelling performance of meteorological parameters and pollutant concentrations in the typhoon-induced and no-typhoon scenarios, ensuring the validity of comparisons using WRF and CMAQ."

Also, Table R3 was added to the Supplement as Table S6.