Dear editor and all reviewers:

We thank the editor and all reviewers for their contribution to the improvement of the ACP manuscript. Responses to reviewers on "Contrasting chemical environments in summertime for atmospheric ozone across major Chinese industrial regions: the effectiveness of emission control strategies" by Zhenze Liu et al. are given below. For clarity, the reviewer comments are given in bold, followed by our responses and modified text in our revised manuscript are given in quotes, italics and blue.

Response to Reviewer 1:

1. **In this study the UKCA chemistry-climate model is enhanced by incorporating reactive VOC tracers into the UKCA gas-phase chemistry scheme in order to better represent urban and regional-scale O_3 photochemistry, and then applied to quantify the differences in chemical environment for surface O_3 for six major industrial regions across China in summer 2016. This study is well organized and clearly written on an interesting topic – how to effectively control tropospheric ozone pollution in China. Here are some specific comments.**

We thank the reviewer for their positive comments here, and address specific concerns below.

2. **Six cities are chosen because they are located in the heavily populated regions with high emissions, but their climate is different, for example, Chongqing is often cloudy and foggy, and especially hot and muggy in summer. The amount of sunshine in Chongqing is poor, and the only months when the sunshine hours exceed 40% (but they still remain below 50%) are July and August; while Beijing has strange weather—extreme hot and cold temperatures, and high humidity to no humidity. It has glorious sunshine year-round and is hot and rainy in summer, especially in July and August. In the study impacts of meteorological conditions on local O_3 production rates are not discussed, and model results are not evaluated against observed meteorological parameters.**

We thank the reviewer for this comment and have investigated meteorological conditions further. We agree that meteorological conditions are important for O_3 formation, and many studies have focused on this aspect specifically (Gong and Liao, 2019; Liu and Wang, 2020; Shi et al., 2020). We have added additional text to introduce the importance of meteorology to O_3 formation in the introductory section of the paper:

Page 2, line 47:
"Meteorological processes also affect O_3 formation through temperature, humidity, clouds, precipitation and biogenic emissions, and a number of papers have studied meteorological impacts on O_3 over China (Gong and Liao, 2019; Liu and Wang, 2020; Shi et al., 2020). However, emission controls are the primary strategies used to reduce

O_3 pollution and we focus on these for this study, as their effectiveness for different regions has not been fully investigated."

We emphasise that the model is nudged to ECMWF ERA-interim meteorological reanalysis data, hence the meteorology over these broad regions is generally simulated well. We show a meteorological evaluation for temperature and humidity for the regions we consider in Table 1. Typically, high temperature and low humidity benefit O_3 production in China, as outlined in the studies listed above. Observed daily mean data have been obtained from the national meteorological data center http://data.cma.cn/.

Table 1: Comparison of daily mean of observed and simulated surface temperature (°C) and relative humidity (%) for six regions of China during June, July, August, 2016.

	Temperature (°C)				
	Obs.	Sim.	Bias	RMSE	r
Beijing	24.0	21.7	-2.3	2.7	0.8
Shijiazhuang	26.3	24.3	-2.0	2.8	0.7
Shanghai	27.6	25.6	-2.0	2.6	0.9
Nanjing	27.8	26.9	-0.9	2.0	0.9
Guangzhou	28.3	27.3	-1.0	1.8	0.5
Chongqing	29.4	27.5	-1.9	3.0	0.8
	Relative Humidity (%)				
	Obs.	Sim.	Bias	RMSE	r
Beijing	67.0	79.6	12.6	14.6	0.8
Shijiazhuang	72.1	80.8	8.7	13.2	0.8
Shanghai	80.4	79.8	-0.6	5.8	0.8
Nanjing	77.3	77.9	0.6	5.5	0.9
Guangzhou	85.5	82.8	-2.7	6.0	0.6
Chongqing	68.2	81.4	13.2	16.5	0.5

From Table 1, we can see that the model is biased low for simulated temperature, although this is partly due to use of temperature in the lowest model layer rather than at 2 m altitude. However, the model successfully captures regional differences in temperature – the highest in Chongqing and the lowest in BTH regions e.g. Beijing and Shijiazhuang. Relative humidity in Shanghai, Nanjing, Guangzhou is relatively well simulated; the highest relative humidity for Guangzhou is also captured by the model. The model biases for Beijing and Chongqing are larger, but this is likely to be due to their proximity to mountainous areas influencing the respective model grid boxes. Higher humidity promotes O_3 destruction hence may lead to lower surface O_3 levels, and this could partly explain negative model biases in daily mean surface O_3 levels for Beijing. However, we note that temperature and humidity are not the main influences on surface O_3 concentrations in Chongqing, and topography is a more important factor as discussed in the paper. Given our use of reliable ECMWF ERA-interim meteorological reanalysis data and the uncertainties associated with meteorological

evaluation, we choose not to include this discussion in the revised manuscript. For clarity we have added the following text to section 3:

Page 11, line 231:
"Given our use of reliable meteorological reanalysis data, we note that meteorology is not the main influence on the model biases. We therefore investigate O_3 chemical environments in different regions to explore regional differences below."

3. **Figure 3 shows that the model overestimates O_3 systematically in Chongqing, and observed low concentrations (<35 ppb) are not reproduced. As statistical indicators are limited and independent in this study, high correlation coefficient r does not indicate that the modeled concentrations in Chongqing are acceptable, because seasonal averaged diurnal variations in O_3 over high emission areas are quite easy to be simulated. Beside the explanation in lines 165-169, would the meteorological fields be attributable?**

Chongqing is not simulated as well as other regions as we note in the text. The overestimation of surface O_3 levels for Chongqing is likely linked to the complex topography as discussed in section 3. Surface O_3 in Chongqing is representative of higher surface altitudes characteristic of the region, leading to a systematic high bias compared with observations, and a corresponding low bias for NO_2 concentrations. Our results suggest that the region of Chongqing represents a NO_x limited regime, hence the underestimated NO_2 levels is not the cause to the overpredicted O_3 levels. This is consistent with satellite-based assessments of ozone sensitivity in China for 2016 (Wang et al., 2021). It hence suggests that complex transport patterns associated with the topography and the higher altitude of the region are the most important factors influencing O_3 levels rather than meteorology as discussed above. We have added the following text to section 3:

Page 11, line 236:
"This underestimation may lead to overestimated O_3 concentrations in a VOC-limited regime and underestimated O_3 in a NO_x-limited regime. While underestimated NO_x concentrations may reflect underestimated NO_x emissions, it is more likely to arise from the effects of dilution on NO_x."

4. **From Figure 4, we can also find that NO_2 in Chongqing is not reproduced well, and daytime NO_2 is underestimated at all sites. How would the underestimation of daytime NO_2 impact on O_3 production sensitivity?**

We agree that the underestimation of daytime NO_2 concentrations will influence O_3 sensitivity. However, higher NO_x levels lead to a more VOC limited regime so all regions except Chongqing would still be in VOC limited regimes if NO_x levels were increased. From the O_3 isopleth plot in Fig 8, we can see that increasing NO_x levels would leave Chongqing in the transition regime or still in a NO_x limited regime.

However, to address this concern we have toned down our statement that 'Chongqing is NO_x limited' to accommodate the local underestimation of NO_2 concentrations. Text added in section 6 and 7 in the revised manuscript for clarity are as follows:

Page 17, line 348:
"We note that our conclusion of NO_x limitation in Chongqing may be sensitive to our underestimation of NO_2 levels (section 3), and that with higher NO_2 Chongqing may shift from the NO_x limited regime to the VOC limited regime. However, satellite observation based studies have also identified this region as one that is largely NO_x limited, in contrast to the heavily populated coastal regions (Wang et al., 2021)."

Page 19, line 397:
"From the O_3 isopleth for Chongqing, we can see that even if NO_x emissions were increased by 40 %, Chongqing would still be in NO_x limited or transition regimes. This suggests that Chongqing is still far away from a VOC limited regime."

5. Bye the way, O_3 and NO_2 from the surface monitoring networks of China are usually recorded in the unit of μg/m3, how are they converted to ppb?

The units of $\mu g/m^3$ are converted to ppb based on temperature and pressure as follows:

$\mu g/m^3$ = ppb * molecular weight/molecular volume (litres)

where molecular volume = 22.41 * (temperature/273.15) * (1013/pressure)
T = absolute temperature (K)
P = atmospheric pressure (hPa)
Chemical species in units of mixing ratio (kg/kg) are output by the model and then converted to ppb. This unit conversion is a standard procedure and so we do not include a description of the approach in the paper.

6. Tropospheric O_3 concentrations are functions of the chain lengths of NO_x and Hox radical catalytic cycles, and ozone production rates depend on not only NO_x and VOCs concentrations, but also actinic flux and temperature. The conclusion of O_3 production across all regions except Chongqing being VOC limited needs to studies for different weather conditions.

NO_x and VOCs are the main primary O_3 precursor species emitted from emission sources. Therefore, we construct O_3 response surfaces with respect to changing NO_x and VOC emissions, and our aim is to investigate the impacts of changing NO_x and VOC emissions on surface O_3 concentrations. We choose to represent O_3 sensitivity to NO_x and VOC emissions because it is largely driven by ambient abundances of NO_x and VOCs – reflected by VOC and NO_x limited regimes. We agree that actinic flux and temperature alter O_3 production rates through their effects on photolysis rates and chemical reaction rates, and these are fully captured in our model chemistry scheme, which includes 101 species and 348 reactions (see section 2.1). We note that O_3

sensitivity is affected by photolysis rates and temperature, but highlight that we use the meteorological conditions appropriate to each region to calculate the sensitivity. It is a good point that O_3 sensitivity could be investigated in future studies for different weather conditions, and it is also worthy investigating how O_3 sensitivity changes throughout a day. However, this study aims to provide a broad assessment of regional O_3 sensitivity with the implication of emission control strategies in the summer time, and focus on specific time periods or unusual meteorological conditions is not necessary for this.

7. **Generally model simulated O_3 production rates are quite sensitive to the chemical mechanism beside other model inputs. As stated in lines 101-105, VOCs such as alkenes and aromatics are abundant in industrial areas of China, but why is ethene not incorporated into the UKCA gas-phase chemistry scheme?**

The extended chemistry scheme for UKCA introduced in this study provides highly reactive VOC species to permit representation of more active photochemical environments. We incorporate C_3H_6, C_4H_{10} and toluene as proxies for the principal chemically reactive VOC families: alkenes, alkanes and aromatics. The tracer C_3H_6 thus implicitly includes ethene as a part of the alkene family. This approach to VOC chemistry is described in detail in Stockwell et al. (1990). We highlight that this extended chemistry scheme permits a more realistic VOC oxidation environment, and is more suitable for high emission areas than the original chemistry scheme that did not include these short-lived VOC tracers.

Response to Reviewer 2:

1. **This manuscript investigated chemical environment for surface O_3 for six major industrial regions across China in summer 2016. Detailed chemistry-climate model simulations were employed to diagnose ozone sensitivity to precursors and contrast the effectiveness of different measures to reduce surface O_3 concentrations. This manuscript is helpful to understand ozone pollution mechanism in Chinese cities, and within the scope of ACP. I think it is publishable in ACP after my following concerns are addressed.**

We thank the reviewer for their positive comments, and specific concerns are addressed below.

2. **Line 215: The gross rate of production $P(O_3)$ actually represents the production rate of O_X (O_3 + NO_2) through the reaction HO_2 (RO_2) +NO. Therefore, the net ozone production rate should include the loss term NO_2+OH (Wang et al., 2019. doi.org/10.5194/acp-19-9413-2019). In addition to OH+NO_2 and RO_2+NO_2, the loss of NOx should also include RO_2+NO and OH+HONO When calculating OPE. Please give specific quantification even though these reactions play a minor role in the loss of NOx.**

We thank the reviewer for this point. The calculation of O_3 production that the reviewer points out is based on including NO_2 in the definition of odd oxygen (O_x). We note that there are different definitions of O_x and sometimes O_3 and NO_2 are together defined as O_x. These wider definitions include O_3, $O(^1D)$, $O(^3P)$, NO_2 and various other NO_y species (NO_3, N_2O_5, HO_2NO_2, HNO_3, PAN, MPAN), see Section 4 in Horowitz et al. (2003). There is still ongoing debate about the best way to consider O_x, and recent studies have used an even wider definition (Bates and Jacob, 2020). However, we choose to take a simpler and more traditional definition of O_x including only O_3 and O species, following Kleinman et al. (1997). No approach provides a complete assessment of all odd oxygen, but we feel strongly that the simplicity and greater interpretability of this more traditional approach outweighs the benefits of including a large number of minor species, while also allowing direct comparability with earlier studies which have used the same approach. Using this approach, O_3 production is the sum of HO_2/RO_2 + NO reactions and O_3 loss is the sum of direct reactions of O_3 with OH, HO_2 and alkenes, and $O(^1D)$ + H_2O. We now state this clearly in section 5:

Page 15, line 298:
"We define the net O_3 production rate (ppb/h) as the gross rate of production of O_3, $P(O_3)$, from the reactions HO_2 + NO and RO_2 + NO minus the gross rate of loss of O_3, $L(O_3)$, from the reactions $O(^1D)$ + H_2O, O_3 + OH, O_3 + HO_2 and O_3 + VOCs."

We regard to the reaction NO_2 + OH as the principal loss of NO_x but it is not a loss of O_3 because we do not include NO_2 as part of odd oxygen – this is an advantage of the

simpler definition of O_x. The NO_x loss term is defined as NO_2 + OH in Liu et al. (1987), assuming that the lifetime of NO_x is determined by the formation and deposition of HNO_3 in the daytime. However, the formation of organic nitrate ($RONO_2$) is also a minor loss of NO_2 as the reviewer points out. We therefore include this reaction in the NO_x loss term as the reviewer suggests. It also has minor effects on the calculation of OPE. To account for the reviewer's suggestion, we have redefined the NO_x loss and OPE in the paper and have updated Fig. 6. Our results in the text are unchanged. We have also introduced $RONO_2$ in the introductory section. Oxidation of HONO by OH does not remove NO_x so this reaction is not included in the NO_x loss term. The modified text is shown below.

Page 2, Line 60:
"However, at high NO_x concentrations, nitric acid (HNO_3), peroxy nitrates (RO_2NO_2) and organic nitrates ($RONO_2$) are easily formed as NO_x reacts with OH and RO_2. These species are the main sinks of radicals and NO_x, and are readily removed from the atmosphere by deposition or exported to remote areas (Horowitz et al., 1998). Therefore, increasing NO_x concentrations increase O_3 production, but also accelerate the formation of NO_x sinks, leading to less efficient O_3 formation."

Page 15, Line 308:
"The loss of NO_x, $L(NO_x)$, is principally determined by the reactions OH + NO_2, RO_2 + NO_2 and RO + NO_2, which produce HNO_3, RO_2NO_2 and $RONO_2$ respectively."

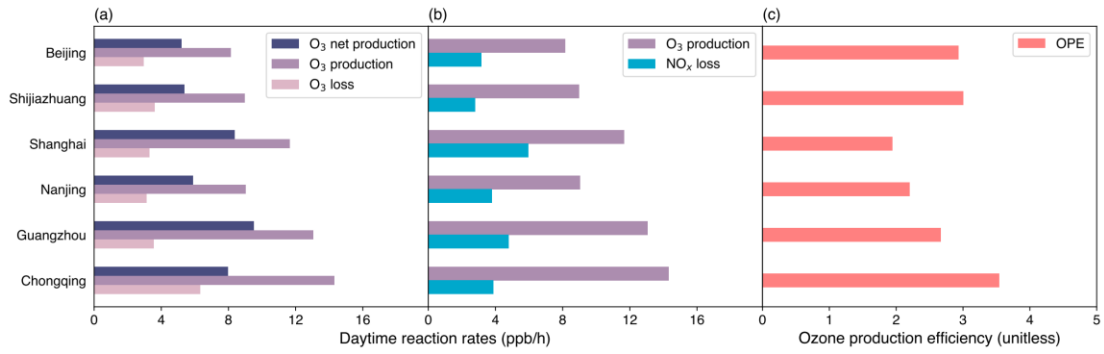

Figure 6: Simulated surface daytime **(a)** net O_3 production rates, gross O_3 production rates and gross O_3 loss rates (ppb/h) **(b)** gross O_3 production rates and NO_x loss rates (ppb/h) **(c)** OPE (unitless) for the six industrial regions in JJA, 2016, China.

3. **Figure 4 shows significant underestimation for NO_2 in daytime, but overestimation for NO_2 at nighttime. The overestimation of NO_2 at night maybe related to underestimated nighttime chemistry such as the removal of NO_3 and N_2O_5 through heterogenous uptake (Li et al., 2018; Li et al., 2019). A short discuss should be performed. Additionally, how do these underestimation and overestimation for NO2 influence your diagnosis of ozone sensitivity? For example, the underestimation of NO2 in Chongqing will lead to more NOx-**

limited, which likely misleads the actual situation.

The heterogeneous chemistry scheme in UKCA includes heterogenous uptake for NO_3 and N_2O_5 – the conversion of N_2O_5 to aqueous HNO_3. Uptake coefficients are estimated for the different aerosol types included in the GLOMAP aerosol scheme. In UKCA, the lack of nitrate aerosol in the aerosol scheme may result in the lower uptake of nitrogen (Archibald et al., 2020), particularly in regions with high NO_x emissions. Therefore, the heterogeneous removal of nitrogen may be biased, potentially leading to higher NO_2 and lower O_3 concentrations at nighttime. We have now added further text to discuss the potential impacts of heterogeneous processes in section 3 on model evaluation:

Page 7, Line 202:
"In addition, nighttime heterogeneous uptake of nitrogen on aerosols remains highly uncertain due to the complexity in estimating uptake coefficients for different aerosol composition/mixing states (Lowe et al., 2015; Tham et al., 2018). In UKCA, the lack of nitrate aerosol in the aerosol scheme may result in a lower uptake of nitrogen (Archibald et al., 2020), particularly in regions with high NO_x emissions. Therefore, there may be a bias in the heterogeneous removal of nitrogen, potentially leading to higher NO_2 and lower O_3 concentrations at nighttime."

However, we note that we focus on daytime O_3 levels and O_3 sensitivity, and therefore uncertainties in nighttime chemistry have a relatively small influence on our results.

As in our response to the reviewer 1, we now include discussion of the impacts of underestimated NO_2 levels on O_3 sensitivity. We note that higher NO_2 levels for Beijing, Shijiazhuang, Shanghai, Nanjing, Guangzhou would lead to more VOC limited chemical environments, so this would not affect our conclusions. However, it is possible that higher NO_x emissions might shift Chongqing into a transition regime (see Fig 8 in section 6). We have adjusted the statement that 'Chongqing is NO_x limited' in sections 6 and 7 as follows:

Page 17, line 348:
"We note that our conclusion of NO_x limitation in Chongqing may be sensitive to our underestimation of NO_2 levels (section 3), and that with higher NO_2 Chongqing may shift from the NO_x limited regime to the VOC limited regime. However, satellite observation based studies have also identified this region as one that is largely NO_x limited, in contrast to the heavily populated coastal regions (Wang et al., 2021)."

Page 19, line 397:
"From the O_3 isopleth for Chongqing, we can see that even if NO_x emissions were increased by 40 %, Chongqing would still be in NO_x limited or transition regimes. This suggests that Chongqing is still far away from a VOC limited regime."

4. **Figure 8. shows ozone increased from 70 ppb to over 80 ppb during 2013-2019. However, observed ozone concentrations in Beijing didn't increased significantly during the period or decreased after 2015 in spite that ozone increased over North China Plain (Lu et al., 2018. DOI: 10.1021/acs.estlett.8b00366; Tang et al., 2020. doi.org/10.1016/j.atmosres.2020.105333). This needs further explanations.**

Tang et al. (2021) suggest that the chemical environment for Beijing is now NO_x limited hence reductions of NO_x emissions lower surface O_3 levels. However, our study focuses on the regional scale situation rather than on urban measurement locations. Many studies have shown an overall increasing trend in O_3 levels on the North China Plain in recent years, as the reviewer notes. It would not be representative to use a single measurement site in the outskirts of the city to interpret the overall trend of the whole region. We have added text in the conclusion section to emphasise the aim of this study:

Page 23, line 476:
"This study hence provides a broad assessment of the O_3 sensitivities for these regions with the implication of emission control strategies."

5. **Line 270: How do you obtain VOC and NOx emissions in 2018 and 2019 given that Cheng et al (2019) just estimated emissions during 2013-2017. Please give specific description.**

We assume that NO_x and VOC emissions in the Beijing region continue to follow the same trend as they do between 2013 and 2016. We have now added further text to clarify this in section 7. We already note in the conclusions that emission inventory development is a limitation for accurately predicting O_3 trends.

Page 18, line 383:
"For context, Fig. 8a also shows the simulated daytime O_3 changes between 2013 and 2019 in the Beijing region assuming that the emission changes observed between 2013 and 2016 continue at the same rate until 2019 (Cheng et al., 2019)."

6. **Line 145: There are only 450 measurement stations in 2013, growing to 1,500 stations in 2017 and 1670 stations in 2019.**

Yes, the number of measurement sites in China increased substantially after 2013. The text in the paper hase been modified to reflect this.

Page 6, Line 177:
"450 measurement stations in China started operating in 2013, growing rapidly to 1670 stations by 2019."

7. **Line 300: "summer-mean ozone" should be "daily mean ozone".**

We replace ‘summer-mean ozone’ with ‘summer daily-mean ozone’ throughout the text in section 7.

Below are all references used in the responses. Added references in the revised manuscript are shown as track changes.

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