

Response to reviewers' comments

We thank the reviewers for the constructive comments and suggestions, which are very positive to improve scientific content of the manuscript. We have revised the manuscript appropriately and addressed all the reviewers' comments point-by-point for consideration as below. The remarks from the reviewers are shown in black, and our responses are shown in blue color. All the page and line numbers mentioned following are refer to the revised manuscript without change tracked.

Reviewer

This paper presents a set of recent (2018) measurements of trace gases from a ground site in Shanghai to assess the factors that lead to photochemical ozone pollution in that region of China. The measurements span five months of nearly continuous measurements. They include NO_x and speciated VOCs, among other chemical measurements, together with standard meteorological data (but not including boundary layer dynamics).

The results are analyzed in the context of three different case studies of high, medium and low ozone. Several different standard metrics of photochemistry and ozone production are used to analyze the data using both observationally derived quantities as well as box modeling.

While the overall measurements and analysis are standard and do not present any novel data or analysis methods, they do represent a comprehensive analysis from a particular year and location in China, a highly polluted region that is currently undergoing a transition from recent high emissions to somewhat lower and more controlled emissions of common air pollutants. They will therefore represent a useful data point and analysis of factors that control ozone pollution in a Chinese megacity.

The manuscript is generally well written and easy to follow.

I recommend publication following attention to the minor comments and technical corrections below.

R: Thanks very much for the comments. This paper focuses on comparing atmospheric photochemistry and radical chemistry at different ozone levels in Shanghai in summer 2018. Although the traditional data and analysis methods are used, we did a comprehensive analysis of atmospheric photochemistry in specific years and locations in China. Since ozone pollution is the big challenge for the air quality during summer and the long-term observations show that the mean mixing ratio of O₃ concentration in Shanghai increased 67% from 2006 to 2015 at a growth rate of 1.1 ppbv/year (Gao et al., 2017), it is necessary to study the atmospheric photochemical behavior under different ozone levels and explore the contribution of precursor VOCs to ozone generation.

Minor comments are given below.

Line 21, Abstract: AQI is not defined here nor referenced further in the text. The wording is also not clear. 92.2% of all the days in the observation period? Or some fraction of the AQI?

R: Thanks for the suggestion. AQI, Air Quality Index, comes from 'Technical

Regulation on Ambient Air Quality Index' formulated by Ministry of Environmental Protection of China (now called Ministry of Ecology and Environment of China) to regulate the daily and real-time report on air quality index.

92.2% refers to the ratio of the days without air pollution to the total days during the observation period. And we have rewritten this sentence to 'Five months of observations from 1 May to 30 September 2018 showed that the air quality level is in lightly polluted and even worse (Ambient Air Quality Index, AQI>100) for 12 days, of which ozone is the primary pollutant for 10 days, indicating that ozone is the main challenge of air quality in Shanghai in the summer of 2018.'. Please refer to Line 19-22.

Line 34, Abstract: "Concentration ratio" should be defined. This is the summed mixing ratio of these species relative to what? Total NMVOC? Or total carbon? Also, the statement that follows implies that these four compounds could be controlled, but since HCHO is not a direct emission, it would result from control of all VOC and could not be targeted individually.

R: Thanks for the suggestion. 'Concentration ratio' means the ratio of certain NMVOC concentration to total NMVOC concentration. We have rewritten this sentence as 'The concentration ratio (~23%) of these four species to total NMVOCs is not proportional to their contribution (~55%) to OFP'. Please refer to Line 35-36.

In general, the sources of HCHO can be attributed to the primary and secondary contribution, as well as the background. The primary sources of HCHO are mainly from fossil fuels, industrial and vehicular emissions (Lui et al., 2017). Previous studies have shown that the contribution of primary source to HCHO in Summer in Wuhan, China reached $32.4 \pm 6.5\%$, primary source contributed 40% to HCHO in Houston in Summer, and the annual average contribution of primary source to HCHO were 42.52% in Shanghai in 2016 (Buzcu Guven and Olaguer, 2011; Su et al., 2019; Yang et al., 2019). This indicates that the primary source of HCHO cannot be ignored, and the controlling of the primary emission of HCHO also make sense. In addition, the secondary formation of formaldehyde is also indeed important, which means that the level of precursors of formaldehyde needs to be controlled. So we prefer to keep the current statements.

Line 73: The differences described are not all a function of metropolitan areas but also of the season in which the measurements took place. The Ren 2003 reference, for example, was in winter, one of the main reasons that HONO photolysis is listed as important. The list is also not a comprehensive literature review, which should be stated, as there are numerous similar analyses in addition to those listed here.

R: Thanks for the suggestion. The literature review should be more comprehensive and detailed. We have introduced the HO_x sources among different places and also highlighted its change due to the observational periods/seasons, as following 'For example, ozone photolysis is the dominant OH source in Nashville (Martinez et al., 2003); HONO photolysis has a more important role in New York City (Ren et al., 2003), Paris (Michoud et al., 2012) and Santiago (Elshorbany et al., 2009), Wangdu, China

(Tan et al., 2017) and London (Whalley et al., 2016; Whalley et al., 2018); HCHO photolysis is a significant source of OH in Milan (Alicke et al., 2002); while OVOCs photolysis plays a more critical role in Mexico City (Sheehy et al., 2010), Beijing (Liu et al., 2012), London (Emmerson et al., 2007) and Hong Kong (Xue et al., 2016). However, it also should be noted that the sources of HO_x also changed with different observational seasons/periods even in the same place. The HO_x production in New York City was reported to be dominated by HONO photolysis during daytime but O₃ reactions with alkenes at night in winter (Ren et al., 2006). The main source of radicals was the reaction of O₃ and alkenes during whole day in winter, while HONO photolysis dominated the source of radicals in the morning but photolysis of carbonyls at noon of summer in Tokyo (Kanaya et al., 2007).’ Please refer to Line 74-83.

Line 89: Remove “the of”. What does the growth rate refer to? Average O₃? Maximum O₃? Number of air quality exceedances?

R: Thanks for the suggestion. The imprecise expression may lead misunderstanding. The growth rate here refers to the mean concentration of O₃. So we have rewritten this sentence into “The long-term observations show that the mean mixing ratio of O₃ at the downtown urban site in Shanghai increased 67% from 2006 to 2015 at a growth rate of 1.1 ppbv/year”. Please refer to Line 94-96.

Line 125: Define “ultra-low temperature”

R: The ultra-low temperature freezing collection device adopted electronic refrigeration, and the internal temperature of the cold trap could reach -150 °C, which can completely capture the target compound. Please refer to Line 131.

Line 141: PAN is not technically defined as an oxidant, but is co-produced with O₃.

R: Thanks for the suggestion. PAN does not belong to oxidants, it is the same photochemical product as O₃. What we want to say here is that O₃ and PAN are secondary products. The ‘oxidant formation’ has been corrected to ‘secondary products formation’. Please refer to Line 147.

Line 145-146: The model procedure is not clear. A seven-day run is constrained to data throughout, with seven days of continuous measurements? Or is the run constrained to some sort of diel average? Why does it require four days to reach a steady state? Which species require this spin up time?

R: Thanks for the suggestion, we have introduced the model procedure and describe it accurately and specifically. Please refer to Line 149-160. A seven-day run is constrained by seven-day continuous measurements, where the first four days of pre-simulation are for unmeasured, model-generated intermediate species (HO₂, RO₂, PAN, etc.) to reach steady-state concentrations during the last three days of simulation. Previous literature reported that the pre-simulation time was set to 4 day, 5 days or 9 days (Xue et al., 2014; Xue et al., 2016; Li et al., 2018). Considering the lifetime of the model-generated intermediate species and the simulation time cost, the pre-simulation time is set to four days in this study.

Line 150, AOC: The definition of AOC does not include NO_x, notably not the reaction of OH with NO₂, but also not RO₂ + NO reactions (i.e., producing organic nitrates). These are chain termination steps and so perhaps are excluded for that reason, but the exclusion would not then fit the definition that follows of defining the “removal rate of most pollutants”, since NO_x (as well as SO_x) is excluded. Some comment or caveat to this effect is warranted, even if the definition is simply following prior literature. The quantity as defined is not as commonly used as other metrics in this paper.

R: Thanks for the suggestion. Indeed the definition of AOC is to follow the previous literature, and the "removal rate of most pollutants" does not conform exactly to AOC definition. According to the definition of AOC, AOC actually determines the removal rate of VOC, CO and CH₄.

We have modified this part to ‘According to the definition of AOC, it can be calculated by the equation (E1) (Elshorbany et al., 2009; Xue et al., 2016):

$$AOC = \sum_i k_{Y_i} [Y_i] [X] \quad (E1)$$

Where Y_i are VOCs, CO, and CH₄, X are oxidants (OH, O₃, and NO₃), and k_{Y_i} is the bi-molecular rate constant for the reaction of Y_i with X. Atmospheric oxidation capacity determines the rate of Y_i removal (Prinn and Resources, 2003).’. Please refer to Line 163-166.

Line 162, OH chain length: This is one of several available definitions. The assumption in this formulation appears to be that OH + NO₂ is the major chain termination reaction. This is shown in the later analysis but not justified here. The later analysis needs to be referenced to justify this equation. In some instances, RO₂ + NO producing organic nitrates is competitive with OH + NO₂. No mention is made of this chain termination step, nor is its importance ever assessed in the context of other metrics. This chain termination reaction needs to be included in the metrics of ozone photochemistry somewhere in this paper.

R: After in-depth reading of relevant literature, we have a deeper understanding of chain length. The ratio of the rate of HO_x cycling reactions to HO_x termination is called the chain length, as in Equation 1 (Martinez et al., 2003). When the termination reaction of HO_x is dominated by the reaction of NO₂ and OH, the definition can be simplified as equation (E4). When we use the simplified equation (E4), we need to declare that the reaction between OH and NO₂ is the main termination reaction of radicals.

$$\text{Chain Length} = \frac{[OH]k_{OH-L(HO_x)}}{L(HO_x)} \quad (E2)$$

$$L(HO_x) = k_{OH+NO_2} [OH][NO_2] + 2k_{HO_2+HO_2} [HO_2]^2 + 2k_{OH+HO_2} [OH][HO_2] + 2k_{HO_2+RO_2} [HO_2][RO_2] + 2k_{RO_2+RO_2} [RO_2][RO_2] \quad (E3)$$

$$OH \text{ Chain Length} = \frac{k_{OH} [OH] - k_{OH+NO_2+M} [OH][NO_2]}{k_{OH+NO_2+M} [OH][NO_2]} \quad (E4)$$

This is one of several definitions available based on the assumption that OH + NO₂ is the main chain termination reaction, which is further discussed in Sect 3.3. Please refer to Line 180-181 and Line 376-383.

Line 191-192: Is radiation the only factor? From Figure 1, it appears that meteorology and transport could also easily have been important. The temperature, relative humidity, and distribution of wind vectors were also different between the two periods.

R: Thanks for the suggestion. After comparing other meteorological parameters, it can be seen that not only the radiation of Case 1 was higher than that of Case 2, but also the temperature, humidity and pressure of Case 1 are different from Case 2. The temperature difference between day and night in Case 1 was greater than Case 2, and the humidity and air pressure were lower than Case 2 (see Figure R1). These meteorological conditions are conducive to photochemical reactions. We have revised it. Please refer to Line 209-212.

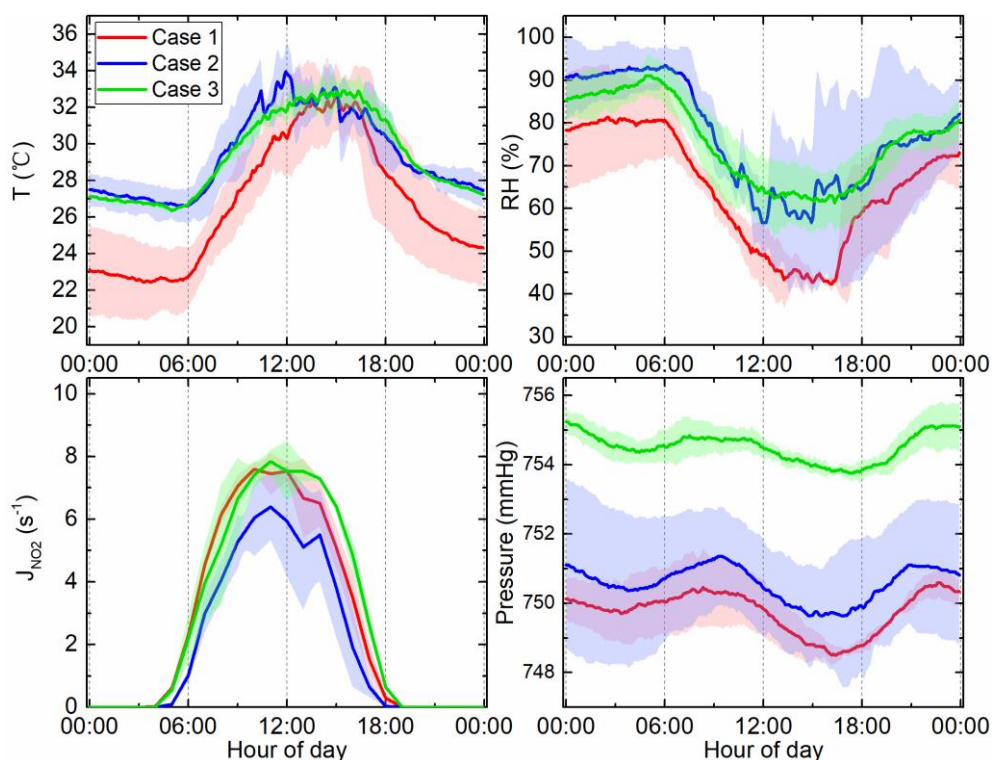


Figure R1. Mean diurnal profiles of meteorological parameters for three cases. The shaded areas denote the standard deviation.

Line 212: Acetylene is not technically an alkene but rather an alkyne. It is much less reactive than alkenes towards OH. This is described in footnote c of table 2, but would be better also in the text. The lumping of acetylene with alkenes is not really appropriate, but if it is done, the statement that this compounds is far less reactive with alkenes toward OH needs to be explicit.

R: Acetylene is indeed not technically an alkene, but an alkyne. Considering that both acetylene and alkenes are unsaturated aliphatic hydrocarbons, which have unsaturated bonds, acetylene and other species with carbon-carbon double bonds are classified as alkenes category for the convenience of statistics. We have followed the comment and made the description clearly that the reactivity of acetylene with OH is far less than that

of alkenes with OH and need to be explicit. Please refer to Line 238-239.

Line 215, Table 2: Units are given in footnote (b) but are otherwise difficult to find. Suggest moving this description to the table caption.

R: We have followed the comments and move this description to the table caption. Please refer to Line 233-235.

Line 229-230, and 235-239: Were there NO₃ measurements to define nighttime AOC? The NO₃ measurement (by DOAS?) is not specified in the experimental techniques. Was this a calculated quantity? Was there nighttime NO at the surface level measurement site that limited NO₃?

R: We have not measured the NO₃ concentration in this study. The presented NO₃ data and its contribution to nighttime AOC were the simulated results.

Line 238: Numbers given for NO₃ do not match the figure, which always shows much larger AOC due to OH. Do these percentages refer to nighttime data only?

R: These percentages of NO₃ to AOC refer to nighttime only. The time periods that these percentages refer to should be indicated clearly in the manuscript. Please refer to Line 256-257.

Line 310: Clarify what is meant by “all within 10”. This could imply a factor of 10 difference between chain lengths, which is likely not what is intended.

R: Thanks for the suggestion. As Reviewer #1 commented on this, I have changed ‘within 10’ to ‘less than 8’. Please refer to Line 329.

Line 312: This is not “probably” due to higher NO_x, but rather simply “due to higher NO_x”, correct? The dependence should not be difficult to infer.

R: Thanks for the suggestion. As shown in Figure R2 and Table 1, the high mixing ratio of NO_x in Case 1 during the 09:00-14:00 results in a relatively larger sink of OH + NO₂. We have followed the comment and remove ‘probably’. Please refer to Line 331-332.

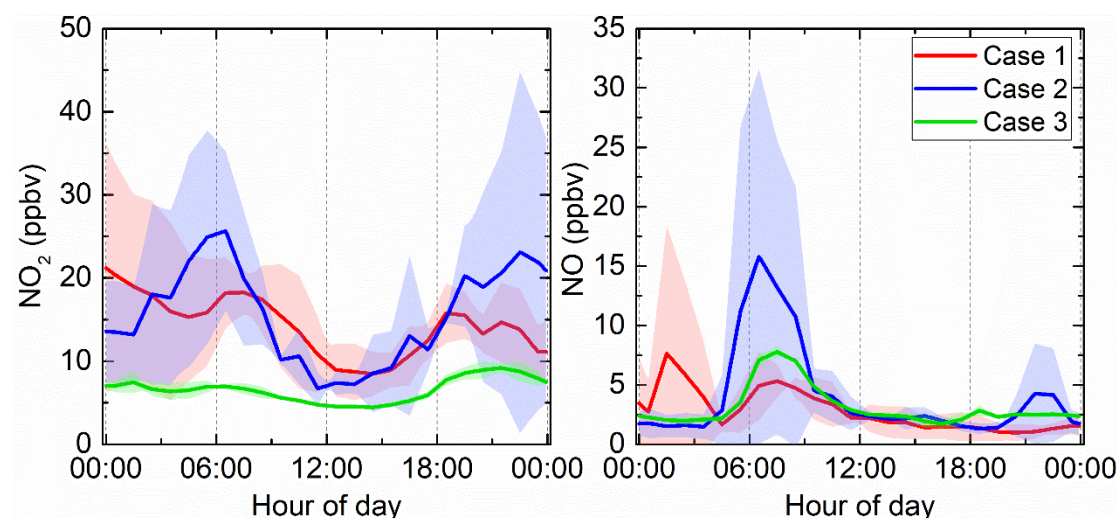


Figure R2. Mean diurnal profiles of NO₂ and NO for three cases. The shaded areas denote the

standard deviation.

Line 313-315: The OH chain length is described at the beginning of the paragraph as being similar to ozone production efficiency, yet here trend in OH chain length is show to be opposite to ozone abundance. Can the authors reconcile these statements?

R: During the daytime, the greater the chain length, the greater the amount of O₃ produced per NO_x molecule converted to HNO₃. Thus, the chain length is related to ozone production efficiency (OPE), which is given by $\Delta O_3 / \Delta (NO_y - NO_x)$ (note: NO_y = NO_x + HNO₃ + NO₃ + PAN) (Wang et al., 2018). In the profile of OH chain length in Figure 5, the OH chain length in Case 3 is longer accompanied with the lowest ozone mixing ratio, meaning that per NO_x converted into HNO₃ produces more O₃ whereas the daytime NO_x mixing ratio in Case 3 is almost half that of Case 1 and 2 (see Figure R1), causing ozone mixing ratios to be lower than Case 1 and 2. In addition, we found that the OH chain length was opposite to the ozone level, and the explanation given was also due to the lower NO_x mixing ratios in previous studies (Mao et al., 2010; Ling et al., 2014).

Line 344-350: The photolysis of ClNO₂ was noted in an earlier section and should be noted here again as previous studies have shown it to be as important as HONO during the morning hours (e.g., Young, ES&T, v 46, p10965, 2012)

R: Thanks for the suggestion. We have followed the comments and stated the importance of ClNO₂ photolysis to OH sources in the morning. Please refer to Line 369-370.

Line 367: Where do the calculated MIR coefficients come from in this equation? How are they determined?

R: MIR coefficients come from Carter (2010) research, as listed in Table 1. The Maximum Incremental Reactivity (MIR) scale is determined by adjusting the input ratio of VOC to NO_x in model (built on the SAPRC atmospheric chemical mechanisms) to maximize the incremental reactivity of a base VOC mixture. Please refer to Carter (2010) research for details.

Line 412-414: Same comment as in the abstract. While three of the four NMVOC can in fact be controlled, formaldehyde is mainly the secondary oxidation product from a wide range of other compounds and cannot be controlled directly.

R: Please also refer to the responses to the minor comments #2. Many studies have reported that the primary source is a non-negligible source of formaldehyde (Buzcu Guven and Olaguer, 2011; Lui et al., 2017; Su et al., 2019; Yang et al., 2019). So we consider that the regulating on the primary sources of HCHO also can make sense.

Technical Corrections

Line 23, Abstract: The word “premise” is not properly used here.

R: Thanks for the suggestion. Now we are using ‘precondition’. Please refer to Line 23.

Line 32, Abstract: “radical” rather than “radicals”

R: We have corrected it. Please refer to Line 33.

Line 83: Replace “even more” with “increasingly”; Line 86: remove the word “around”

R: Thanks for the suggestion. The ‘even more’ has been corrected to ‘increasingly’. And we have removed the word ‘around’. Please refer to Line 89 and Line 92.

Line 104, 123, 153, 336, 409: Replace “Besides” with “Additionally”

R: We have followed the suggestions and replaced ‘Additionally’ with ‘Besides’. Please refer to Line 112, 130, 168, 358 and 447.

Line 143: “input” rather than “inputted”

R: We have corrected ‘inputted’ to ‘input’. Please refer to Line 150.

Line 170: Pollutants shown in Figure 1 are given in mixing ratio, not concentration units.

R: We have followed the suggestion and replaced ‘concentrations’ with ‘mixing ratios’ and elsewhere in the manuscript. Please refer to Line 199 and other places.

Line 223: “based” rather than “base”

R: Thanks for the suggestion. The ‘base’ has been replaced with ‘based’. Please refer to Line 242.

Line 289: Eliminate the word “besides”

R: We have been removed the ‘besides’. Please refer to Line 309.

Line 373: Figure gives mixing ratios rather than concentrations. Specify mixing ratio in text.

R: We have followed the suggestion and replaced ‘concentrations’ with ‘mixing ratios’ and elsewhere in the manuscript. Please refer to Line 411 and other places.

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