Response to comments from Reviewer 1:

This study investigates the impacts of the meteorological factors and isoprene emissions on two severe ozone pollution episodes in North China Plain (NCP) by combining ground-level observations and WRF-CMAQ simulations. The effects of hot, dry and stagnant weather conditions, as well as varied factors influencing isoprene emissions, such as land cover change, high vapor pressure deficit (VPD), and urban landscape, are examined to explain the causes of ozone pollution episodes. Results show that urbanization and land cover change made significant contributions to the enhancement of MDA8 ozone in the past decades. Such effects have not been considered in most of previous studies. The topic of this study well fits the scope of ACP, however, some revisions are required before the acceptance.

RE: We thank the reviewer for the constructive comments to help us further improve the manuscript. Please see the detailed responses to your comments below.

1. GENERAL The main flaw of this study is the confusion of time scales. Land cover change and urbanization usually occurs at annual to decadal time scale. However, this study focuses on ozone episodes which happen in several days. It's not scientifically reasonable to estimate the effects of land cover or urban landscape on ozone extremes. Instead, it would be helpful to examine whether land surface changes contribute to an overall enhancement of ozone concentrations during 2010s compared to 2000s. It is clear that ozone pollution level increased fast from 2014 to 2017 (Fig. 4). However, land cover or urban landscape should not change significantly within these 4 years. As a result, the main causes of more pollution episodes are related to anthropogenic emissions, atmospheric transport, and/or weather conditions, instead of land cover change.

RE: We agree that the changes of land cover and urbanization is on the scale of annual to decade. We also agree that the main causes of pollution episodes are related to anthropogenic emissions, atmospheric transport, and/or weather conditions, instead of land cover change. This is exactly what we have described in the main manuscript. For instance, in section 3.3 (Lines 366-370), we stated that "Zooming into the two ozone episodic events (June 14-21, June 26-July 3), the mean MDA8 values of case 4 are 98.02 ppbv, 108.89 ppbv, 95.75 ppbv, and 98.98 ppbv for NCP, Beijing, Hebei and Tianjin, respectively, during the heat wave periods (June 14-21, 2017; June 26-July 3, 2017), whereas the MDA8 ozone value for the case (case 1) without biogenic emission are 87.15 ppbv, 93.06 ppbv, 84.78 ppbv and 89.65 ppbv for the corresponding region." This indicates that the contributions, i.e., anthropogenic emissions, transport, etc, except biogenic emissions accounted for the majority (more than 80%) of ozone formation.

Although the absolute contribution of biogenic emissions is smaller than the anthropogenic emissions, the substantial ozone increment is important in both understanding the mechanisms of high ozone events and ozone pollution control. To make this clear, we added a statement in the revised manuscript, "the underestimation of biogenic emission due to changes in land cover may be exaggerated in years with high temperatures and high VPD" (Lines 290). To further address the reviewer's concern regarding the effect of land cover over the recent years, we conducted another sets of simulations (the same as cases 2-4 discussed above) during June 8th to mid-July in 2016, similar period as 2017. The mean MDA8 ozone concentrations over NCP during this entire period in 2017 for case 2 is 79.03 ppbv, and statistical significant enhancement (1.34 ppbv) was achieved in case 3. In comparison to case 3, the land cover change in case 4 shows statistical significant increase as well (1.13 ppbv). Similarly, looking at the entire period in 2016 (June 8th – July 4th), statistical significant, and even higher in relative to 2016, increase was achieved in case 3 (1.55 ppbv) compared to case 2 (90.11 ppbv), and case 4 (1.23 ppbv) compared to case 3. We have added the discussions from Line 405-414 in the revised manuscript.

2. In addition, most of the isoprene observations used by this study were obtained more than ten years ago, making it difficult to evaluate the impacts of land cover change and urban landscape on BVOC emissions, which are the major merits of this study. From this aspect, these changes made limited contributions to the high ozone episodes in 2017, especially compared to the year 2014 (NOT 2003). And the title of the paper is inaccurate.

RE: We thank the reviewer for putting forward the concern of isoprene observations. In fact, we realized the importance of isoprene evaluation as well as the data limitation during our study period. Therefore, we have tried our best to search the isoprene observations from the previous literature. As far as we know, we have added the observational data available to us. Indeed, we did used the most recent observations (Fig. 7B) in 2016 and 2017. It clearly displays the cases without urban landscape substantially underestimated the isoprene emissions. We believe the evidence is clear and also agree that more isoprene observations and evaluations are needed in future.

As we addressed in the previous response, we emphasize the underestimation of biogenic emission due to changes in land cover may be exaggerated in years with high temperatures and high VPD. From the point of view, we consider the title is fine.

3. Page 3, line 69. The North China Plain should be defined with specific latitudes and longitudes or detailed descriptions.

RE: The defined latitudes and longitudes of the North China Plain have been added.

4. Page 8, line 194. The author could consider remove the word "medium" because the medium ozone pollution events are not analyzed.

RE: This has been corrected in the revised manuscript.

5. Page8, line 200. Fig. 1 is in the text rather than supporting information.

RE: "In the supporting information" has been deleted.

6. Page 8, lines 219-220. The sentence "Please note that…" is unnecessary and should be removed.

RE: This sentence has been deleted in the revised manuscript.

7. Page 11, Fig.4. The coordinate and scale for the daily total precipitation (yellow bars) are not shown.

RE: The coordinate of the daily precipitation is the same as Tmax in the right side, and it is marked in the coordinate title (yellow).

8. Page 11. Fig. 4. It would be better that the time series of VPD are exhibited, because the article discusses the effect of VPD on isoprene emissions in Sect.3.3.

RE: Thanks for the suggestion. We have added the VPD time series in Fig. S4 in the supporting information.

9. Figure 4: In addition to the two episodes examined in 2017, the similar hot, dry and stagnant weather condition shown several times during 2014-2017, such as 16th – 22th July in 2014, 6th–14th July in 2015 and 8th–16th July in 2017. Why these favorable conditions do not result in ozone episodes?

RE: We did notice the other episodes with relatively similar meteorological conditions, however, a few differences exist. For instance, albeit of the high temperature during July 16-22 2014, there was continuous rainfall over NCP, weakening the solar radiation and subsequent ozone formation. During July 6-14 2015, there is a relatively short period with high temperature (i.e., July 11 to 13), however, rainfall occurs from July 14, breaking the heat event. Although temperature during July 8th to 20th is relatively high, wind velocity during July 8th and 9th is too high to favor the ozone accumulation. In the following two days, the slow wind with high surface temperature provide appropriate photochemical reaction conditions which are conductive to ozone accumulation. The increased precipitation with reduced solar radiation from July 14th make the high ozone events unlikely occur.

10. Page 15, Fig.7A. The difference between case4 and case5 is 15% increase in isoprene emissions in Beijing, but why the simulated isoprene concentration is much higher in case5 compared with it in case4?

RE: Please be aware that the observations of isoprene concentrations are located in the urban area of Beijing. Although the absolute magnitude of urban isoprene emission is not too large (accounting for 15%), the location of the emissions plays a much larger role. As we know, most of the isoprene emissions from the forest in Beijing is located in the rural area, which is relatively far from the urban area. Considering the short lifetime of isoprene (about 2 hours), it may not be very efficient for these isoprene emissions to transport to the urban areas. Instead, the isoprene directly emitted from the urban areas may contribute to the isoprene concentration very efficiently. Therefore, the 15% increase in isoprene emission contributes to the substantial increase of urban isoprene concentration.

11. Page 16, Fig.8. The abbreviations of NMB, NME, MFB and MFE should be explained in the figure capture. The same issue could be considered in text line 343.

RE: These abbreviations have been added in the revised manuscript.

Response to comments from Reviewer 2:

The manuscript presents a modeling analysis to quantify the contribution of isoprene emissions under heat wave conditions in North China Plain on high ozone periods in summer 2017. It reaches two important conclusions that have not been considered in previous modeling analysis of ozone pollution during heatwaves in China: first isoprene emissions not only respond to high temperatures but also to water stress often associated with heat waves; second the recent increase in broadleaf trees and urban green spaces in China has led to increases in baseline isoprene emissions making the first point more important under heat waves. The analysis is solid and writing is clear in most parts. I recommend publication after my following comments are addressed.

RE: We thank the reviewer for the comprehensive comments to help to improve the manuscript. Please see the detailed responses to your comments below.

Main comments: My main concern is the modeling sensitivity analysis was conducted only for summer 2017 but the first part of the manuscript focuses on the difference of summer 2017 from the preceding years (2014-2016). While the sensitivity tests on isoprene emissions lead to better ozone simulation during high-ozone episodes in summer 2017, it is not clear how the same model settings affect ozone under more 'normal' conditions. This is important to know as it can show readers whether the changes in isoprene emissions in the model should be applied more broadly or only applied for certain conditions such as heatwaves. I suggest the authors pick a low ozone year and show how the same VPD and land type change schemes would affect ozone simulation in the model.

RE: We agree that it is important to do the sensitivity tests in particular over the normal year and high-ozone year. Therefore, besides the sensitivity in original manuscript, we conducted four sets of sensitivity studies for biogenic emissions over the summer of 2016, which is similar as what has been done for the summer of 2017 in the main manuscript. The specific cases include case 2 with biogenic emission using the land cover of 2003, and cases 3, 4 and 5 are the same as case 2 except for the inclusion of the VPD effect, both VPD and land cover of 2016, and VPD and land cover of 2016 combined with the effect of urban green spaces, respectively.

To address the reviewer's comments on how VPD and land cover changes affect ozone under normal year, we conducted another sets of simulations (the same as cases 2-4 discussed above) during June 8th to mid-July in 2016, similar period as 2017. The mean MDA8 ozone concentrations over NCP during this entire period in 2017 for case 2 is 79.03 ppbv, and statistical significant enhancement (1.34 ppbv) was achieved in case 3. In comparison to case 3, the land cover change in case 4 shows statistical significant increase as well (1.13 ppbv). As expected, looking at the entire period in 2016 (June 8–July 4), statistical significant, and even higher in relative to 2016, increase was achieved in case 3 (1.55 ppbv) compared to case 2 (90.11 ppbv), and case 4 (1.23 ppbv) compared to case 3. Therefore, the land cover and VPD may be applied in either episodic events or normal conditions. We have added the discussions from Line 405 to 414 in the revised manuscript.

Second, the VPD effect on isoprene emissions from Zhang and Wang (2016) was derived from one case study in the southeast US based on a modeling sensitivity analysis. It is not a process-level algorithm as MEGAN and whether the VPD scheme would work for the time period and spatial domain outside the study of Zhang and Wang (2016) has not been investigated. This should be stated in the manuscript. The authors may not be aware that a new water stress effect on isoprene emissions is now included in the newest MEGAN3; see Jiang et al. (2018). This literature should be referenced in the manuscript and discussed.

RE:

The uncertainty of Zhang and Wang (2016) has been addressed in the revised manuscript.

Regarding Jiang et al. (2018), we have illustrated the new approach and the potential impact on ozone formation in the revised manuscript, which has been shown below as well.

In the latest version MEGAN 3 (Jiang et al., 2018), a new approach was developed to quantify the drought effect on the isoprene emissions based on both photosynthesis and water stress, yielding a general reduction of monthly mean isoprene emission across the globe, including northern China. The impact of changes in isoprene emissions, based on the new method, on ozone formation deserves further evaluation in future.

Technical issues: Line 41: add ozone after MDA8.

RE: This has been corrected in the revised manuscript.

Figure 1: explain what H, B, and T in the figure stands for.

RE: They have been added in the revised manuscript.

Line 195: How was the interpolation done? Simply averaging over a 0.5x0.5degree grid? As this is a comparison of observations between different years, I do not understand why spatial interpolation is necessary. Is it because the site numbers differ by year?

RE:

The reviewer is correct and a simple averaging was used for the interpolation. The reason to interpolate was due to the differences in number of sites among different years.

Figure 3: how is the regression relationship calculated for each summer? For each day, do you average over all the sites over NCP, average over the 0.5x0.5 degree grid, or treat individual sites as separate data points? Will the slope and correlation slope change with those different representations of data?

RE: The meteorological observations from CMA and ozone observations from China National Environmental Monitoring Centre were used to calculate their regression relationship. However, the sites numbers and locations between meteorological and ozone observations were different. Therefore, to pair up the air quality and meteorological stations, for each air quality station, the closest meteorological station was selected. After that, daily meteorology and ozone data were averaged regionally over the NCP.

The meteorological sites were uniform distribution in general, whereas most air quality sites were located in urban area (shown in Fig.1). In this case, if there is no pairing up, in another word, direct regional mean was operated on meteorology and ozone observations separately, the comparison then loses the consistency. We also tested this method (Fig. R3), yielding much lower correlation coefficient. Thus, the pairing up method was used in the correlation analysis.

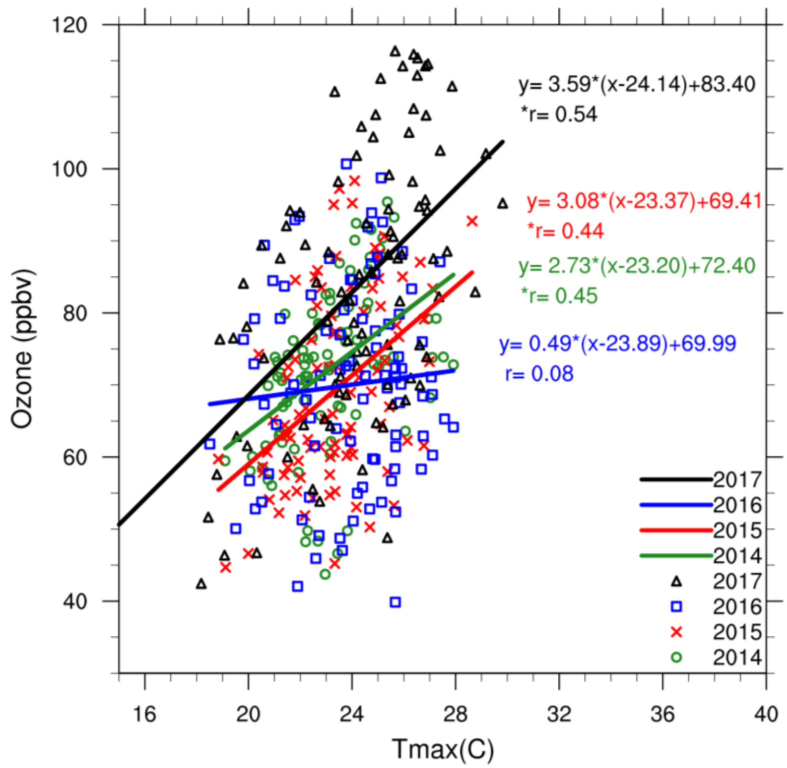

Fig. R3 The correlation between summer MDA8 ozone and daily maximum 2-meter temperature (Tmax) during 2014-2017 over NCP. Regional mean was calculated from the observational sites over NCP.

Line 228: Figure 4 shows all four years, not just 2017.

RE: This has been corrected in the revised manuscript.

Line 230-231: Did you verify these two periods were hit by heat waves? What definition of heat waves were used? Did the heat wave affect the whole NCP or some portion of it? It will make your argument stronger if there is also a spatial association that the sites affected by the heat waves had higher ozone concentrations than those sites not affected. I also suggest the authors describe the temperature in degrees for the two high-ozone periods in the text. July 6-14 2017 had higher temperature than the two periods you noted and ozone was also high. Why did you not highlight that period in the analysis?

RE: Right, during the two periods, the North China Plain was hit by heat waves. The definitions we used in this study is: for each station, three or more continuous days with daily maximum 2-meter air temperature exceeding the 95th percentile of the summers during the past thirty-year (1987-2016). Since the meteorological measurement data mostly located near the airports (blue dots in Fig. 1 of the main manuscript), the distribution is relatively sparse and not all the stations were hit by the heat waves. For example, there is a total of 58 stations, and among these stations, 22 of them were hit by heat waves during June 14-21 and June 26-July 3.

Regarding the comments of ozone concentration comparison between the heat waves and non-heat wave periods, we first match the meteorological stations with observational ozone, and then composite the ozone concentrations during heat waves and non-heat waves. The mean MDA8 during heat waves and non-heat waves are 111.62 and 107.96 ppbv, indicating higher ozone concentrations during heat waves, further implying the impact of heat waves on ozone formation.

Since the reviewer suggest to add the descriptions of temperature in degrees, we now added in section 3.2 in the revised manuscript. During the first three days of these two high ozone episodic events, the regional mean daily maximum temperature is 32.3 °C, accounting for 90th percentile relative to a thirty-year period during 1987-2016. Moreover, almost half of the stations with at least three continuous days exceeding their respective 95th percentile from 1987-2016, satisfying the definitions of heat waves.

Although temperature during July 8 to 20 is relatively high, wind velocity during July 8th and 9th is too high to favor the ozone accumulation. In the following two days, the slow wind with high surface temperature provide appropriate photochemical reaction conditions which are conductive to ozone accumulation. The increased precipitation with reduced solar radiation (figure of radiation not shown) from July 14th make the high ozone events unlikely occur.

Figure 4: What is the gray dashed line in each panel?

RE: The blue dash lines in each panel represented 31 degree Celsius, and this has been explained in the revised manuscript.

Line 268-278: the land cover change is shown for a longer period (between 2003 and 2016), not over the study period (2014-2017). I think the authors meant to say that the use of an older land cover map can lead to underestimate of isoprene emissions and such an underestimate may be exaggerated in years with high temperatures and high VPD. I suggest the authors make this point explicitly in the text.

RE: This has been clearly addressed in the revised manuscript, which is shown below as well.

Combining the point *a)* described above, the underestimation of biogenic emission due to changes in land cover may be exaggerated in years with high temperatures and high VPD.

Reference：

Jiang, X. Y., Guenther, A., Potosnak, M., Geron, C., Seco, R., Karl, T., Kim, S., Gu, L. H., and Pallardy, S.: Isoprene emission response to drought and the impact on global atmospheric chemistry (vol 183, pg 69, 2018), Atmos Environ, 185, 272-273, 2018.

Zhang, Y. Z., and Wang, Y. H.: Climate-driven ground-level ozone extreme in the fall over the Southeast United States, Proc Natl Acad Sci USA, 113, 10025-10030, 10.1073/pnas.1602563113, 2016.