Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., author comment AC4
https://doi.org/10.5194/acp-2020-1302-AC4, 2021

Reply on RC3

Peng Wang et al.

Author comment on "Unexpected enhancement of ozone exposure and health risks during National Day in China" by Peng Wang et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-1302-AC4, 2021

Comments Response

Journal: Atmospheric Chemistry and Physics

Manuscript ID: acp-2020-1302

Title: "Unexpected enhancement of ozone exposure and health risks during National Day in China"

Dear Referee #3,

We appreciate your comments to help improve the manuscript and tried our best to address your comments. The detailed responses and related changes are shown in below. Our response is in blue and the modifications in the manuscript are in red. All figures are included in the attached PDF file.

General:

The paper investigates the causes of increase in surface ozone concentration in China during the Chinese National Day Holidays (CNDH) in 2018. The authors used CMAQ model to simulation O3 production during three periods of pre, during, and after (CNDH). The result shows that the increased O3 values during CNDH are due to increase in precursor emissions and also regional transport. The impact of enhanced O3 during CNDH on public health and mortality rate in major cities in China.

The paper is well-written and fits to scope of ACP. However, it needs some clarifications on the changes in the anthropogenic emissions in the three periods. If no changes were made to the anthropogenic emission inventory to reflected the changes in emissions due to the national holiday how are you attributing the changes in O3 to this event? The relative contribution of biogenic vs. anthropogenic emissions needs to be discussed further in the paper. It may play an important role in the variation in O3 concentrations and it is totally dismissed. Please see the comment sections for the details.

Response: Thanks for the recognition of our study. We've tried our best to address these comments to improve our research. Below is the response to each comment.

General comments:

What does regional transport mean in the scale of your study? All the paper on regional transport in China that are cited in the introduction discuss one region in China and the impact of transport from a region to another hence "regional transport". Specifically, I am referring to P3, L61 where you stated the rapid increase of O3 throughout China is attributed partly to regional transport. What does this mean if the transport is between subregions in your domain?

Response: In our study, the regional transport of O_3 means the O_3 concentrations are from other regions (including one region and multiple subregions) and transported to the target region. In our model, we could track O_3 from one or multiple subregions (provinces in our study) by tagging its precursors [*Wang et al.*, 2019; *Wang et al.*, 2020] and quantify their contributions to the target region (in our study is Guangdong province). In the sentence, we inclined to emphasize the elevated O_3 during the CNDH is partly due to regional transport. For example, we tagged the O_3 percussor emissions in Beijing and other regions. Then, they formed O_3 that was transported to Shanghai under the impact of northerly wind. This is what we define as regional transport in our study.

Section 2.1: Please note in the main text that October emission is industry and residential sectors are higher than September emissions. The monthly variation in emission inventory (between September and October) can play a role in variations in O3 concentrations and it is not discussed in the paper.

Response: Thanks for the comment. We've added more related discussion in the revision.

Changes in manuscript: (line 89-90) The higher emissions rates were found during October from the residential and industrial sectors, while they kept the identical levels from transportation and power sectors. (line 195-196) These increasing O_3 precursors emissions are mainly from the residential and transportation sectors (Table S1), indicating their important roles in the elevated O_3 during the CNDH.

Section 2.1: I am not familiar with MEIC inventory, does it have a diurnal or monthly variation? Please provide more information.

Response: The detailed information of MEIC inventory could be found at http://meicmodel.org/?page_id=560. The MEIC inventory provides both annual and monthly variation inventories, and you can download this emission inventory at either temporal-resolutions. The MEIC emission did not provide the diurnal variation. In our study, we used the annual MEIC emissions and re-distributed them to monthly/daily resolution according to [*Zhang et al.*, 2007] and [*Streets et al.*, 2003]. We've added more descriptions of MEIC inventory at the revision.

Changes in manuscript: (line 86-89) The annual MEIC emission inventory was applied in this study and the monthly profile of the anthropogenic emissions was based on *Zhang et al.* [2007] and *Streets et al.* [2003] as shown in Table S1 to represent the emissions changes between September and October.

Section 2.1: This is my main questions to the authors: Is the anthropogenic emission different during CNHD? If no then how are you attributing changes in emission as one the reasons for enhanced O3. If yes then please provide more information about the changes.

Response: As shown in Table S1 and Figure S4 (now Figure 4 in the revision), the

anthropogenic emissions are different in September and October, and these changes had already been applied in the air quality model as the inputs. The higher anthropogenic emission rates, including VOCs and NO_x were distributed in October, leading to elevated O_3 during the CNDH.

Changes in manuscript: (line 86-89) The annual MEIC emission inventory was applied in this study and the monthly profile of the anthropogenic emissions was based on *Zhang et al.* [2007] and *Streets et al.* [2003] as shown in Table S1 to represent the emissions changes between September and October. The higher emissions rates were found during October from the residential and industrial sectors, while they kept the identical levels from transportation and power sectors. (line 195-196) These increasing O_3 precursors emissions are mainly from the residential and transportation sectors (Table S1), indicating their essential roles in the elevated O_3 during the CNDH.

Section 2.1: PRE-CNDH and CNDH periods have 6 days and AFT-CNDH is 23days. Is there a specific reason for this? This makes your statistical comparisons (for example in fig 1) not fair because you are including more days in one of the periods compared to others.

Response: We included all the rest days in the AFT-CNDH to better understand the elevated O_3 in the PRD region. In this study, we found that the prominent increase of O_3 occurred in the PRD and previous studies also reported that peak O_3 always occurred in October in the PRD [*Shen et al.*, 2015; *Zhang et al.*, 2008]. In addition, as shown in Figure 2 and Figure 3, the O_3 concentrations all dropped throughout China during the AFT-CNDH. Including the 23 days in the AFT-CNDH would not change our conclusion that O_3 concentration decreased during the CNDH. Moreover, this 'unfair' approach is widely applied to analyze the changes of O_3 [*Chen et al.*, 2020] as well as other major pollutants such as ammonium [*Wu et al.*, 2019].

Figure 1. Can you add model values to plots a and b to show if model captured the variation in the MDA8 O3?

Response: The main purpose for the Figure 1 is to show the O_3 variation from the observation instead of the model performance. The model performance was comprehensively demonstrated in Figure 2 and Table S5 by comparing the prediction and observation data. The dots in Figure 2 covered all cities in Figure 1a. From Figure 2 and Table S5, we clearly showed that our model prediction was able to catch the O_3 variations and spatial distributions (Figure 2) and meet the evaluation stands (Table S5). Hence, we suggested no further revision of Figure 1.

Figure 1. Can you show on one of the maps where each the regions in plot (a) are? Why AFT-CNDH in east China is so much lower than PRE-CNDH?

Response: We've already added Figure S3 in the supplemental information (also shown below Figure R3-1) to illustrate the locations of these regions. From Figure S5, we can find the temperature dropped significantly from the CNDH to AFT-CNDH in east China from 20 to 15 °C, which leads to the lower O_3 level [*Bloomer et al.*, 2009]. In addition, the lower temperature also decreased the biogenic emissions, which further prohibit the O_3 formation. Simultaneously, the lower wind speed slowed down the regional transport of O_3 from the North China Plain to east China. All these factors comprehensively led to the lower O_3 level during the AFT-CNDH in east China.

P6 – section 3.2: Having a discussion on changes in O3 production regime in valuable. However, I suggest starting this section by discussing the differences between emissions. This way you can better distinguish between uncertainties in emissions and in the uncertainties in simulation of O3 production process.

Response: Thanks for the comments. We have re-arranged the order in the paragraph to discuss the difference in emissions followed by the changes in the sensitivity regime.

Changes in manuscript: (line 186-200) From Figure 4, the anthropogenic O_3 precursor emissions (NO_x and VOCs) increase throughout China. Increasing NO_x emissions are observed in South China, especially in Guangxi and Guangdong, with a relative increase of up to 100% during CNDH. Considering O_3 sensitivity regimes (determined by Eq. (1)), no noticeable differences are observed between PRE-CNDH and CNDH (Fig. S4). During CNDH, the VOC-limited regions are mainly in the NCP and YRD accompanied by high O_3_VOC. In South China, O_3 formation is under a transition regime in most regions, and NO_x-limited areas are in Fujian and part of Guangdong and Guangxi where have rising NO_x emissions. This is corresponding to an increasing in O_3 in these regions (Fig. 2 and Fig. 4). Simultaneously, higher anthropogenic VOC emissions are also observed during CNDH in South China, leading to elevated O_3 in the transition regime when VOCs and NO_x jointly controlled O_3 formation. These increasing O_3 precursors emissions are mainly from the residential and transportation sectors (Table S1), indicating their important roles in the elevated O_3 during the CNDH. In contrast, during AFT-CNDH, more areas turn into a transition regime in South China. The decreases in biogenic VOCs (BVOCs, compared to CNDH) (Fig. 4) due to temperature (Fig. S5) decrease MDA8 O_3 for regions in transition regime during AFT-CNDH. Accordingly, changes in O_3 highly depend on its precursor (NO_x and VOCs) emissions and the sensitivity regime.

Having a figure that shows the differences between NOx and VOC emissions (in different periods within your simulation) as one the main figures will be very helpful.

Response: Thanks for the suggestion. We've moved the original Figure S4 (the emissions differences figure) to the main text (now Figure 4 in the revision).

P8 – Discussion on changes in transportation emission.

I think making these changes in transportation sector emission and running another simulation can reflect the actual changes that occur in the emissions during the national holiday. Without considering these changes the conclusion seems weak and incomplete to me. I'm not suggesting to add a real time vehicle emission inventory. You can simply increase the emission from transportation sector by factor of 2.2 during the national holiday and study the impact on O3 values.

Response: Thanks for the comment. We conducted the test case (adjustment case, ADJ case) by increasing the MEIC transportation emissions by a factor of 2.2 during the CNDH (Figure R3-2). From Figure R3-2, the O_3 increased in the ADJ case in vast areas of China, especially in the YRD and PRD regions (up to ~10 ppb). The higher emission rates in transportation lead to higher O_3 levels during the CNDH. However, the ADJ case may overestimate the impacts from the travel activities during the CNDH on O_3. The adjustment factor (2.2 in the ADJ case) should only be applied in the vehicle emissions, while we used it in the entire transportation sector that includes non-road sources. The MEIC emission inventory only provides the total transportation emissions instead of the more explicit sectors. Consequently, we did not make changes in the revision since we prefer more accurate results to be shown in the manuscript. In the following study, we'll continue developing real-time vehicle emissions and have a more comprehensive understanding of the holiday impacts of O_3.

Specific comments:

P4 – L83. It is not clear to me if or how much the anthropogenic emission has changed on CNDH days. Also having September and October months in the simulation, probably biogenic emission changes as well. Please be more specific about changes in emissions

during the simulation period.

Response: The anthropogenic and biogenic emissions have different temporal resolutions. The temporal resolution for anthropogenic emissions is monthly and hourly for biogenic emissions. For anthropogenic emissions, although we used hourly emissions in the air quality model, we just allocated the annual amount to different months and used the same hourly profile for each day (difference between workdays and weekends). As a result, we discussed the monthly difference (September and October) to investigate the elevated O_3 issue, and the anthropogenic emissions remained the same from CNDH to AFT-CNDH. For the biogenic emissions, we used the MEGAN model [*Guenther et al.*, 2012] to generate. The MEGAN model requires the hourly meteorology data as inputs to reflect the hourly differences each day.

The anthropogenic emissions, including NO_x and VOCs, increased from September (PRE-CNDH) to October (CNDH and AFT-CNDH). The discussion of biogenic emissions from CNDH to AFT-CNDH illustrates its role in decreasing O_3 in the AFT-CNDH. We also investigated the difference in biogenic emissions between PRE-CNDH and CNDH (Figure R3-3). The biogenic emissions have slightly decreased during the CNDH in the PRD due to the lower temperature. However, the increasing anthropogenic emissions and regional transport enhanced the O_3 concentrations during the CNDH. We also modified Figure S4 (now Figure 4 in the revision also Figure R3-4 in the response) to show these emission differences better.

Changes in the manuscript: (line 86-90) The annual MEIC emission inventory was applied in this study and the monthly profile of the anthropogenic emissions was based on *Zhang et al.* [2007] and *Streets et al.* [2003] as shown in Table S1 to represent the emissions changes between September and October. The higher emissions rates were found during October from the residential and industrial sectors, while they kept the identical levels from transportation and power sectors. (line 227-229) The enhanced regional transport and the increasing anthropogenic emissions synergistically lead to the rising O_3 during the CNDH, offsetting the impacts from the lower BVOC emissions (Fig. 4).

P4 – L100-107. I suggest briefly explain which benchmarks you used for meteorology and O3 performance in this paragraph.

Response: Thanks for the comment. We've added the related references in the revision.

Changes in the manuscript: (line 107-109) All the statistics results of the WRF model are satisfied with the benchmarks [*Emery et al.*, 2001] except for the GE of temperature (T2) and wind speed (WD) went beyond the benchmark by 25% and 46%, respectively (Table S4). (line 112-114) The model performance of O_3 was within the criteria [*EPA*, 2005] with a slight underestimation compared to observations, demonstrating our simulation is capable of the O_3 study in China (Table S5).

P6 – L158. In south China…

Are you referring to model or obs values? Please clarify.

Response: It refers to the model values. Sorry for being unclear, and we've already changed it in the revision.

Changes in the manuscript: (line 167-168) In South China, the predicted MDA8 O_3 reaches ~90 ppb that is approximately 1.2 times of the Class II standard with an average increase rate of 30%.

P6 – L 159. In contrast…

What is the reason for this?

Response: The "in contrast" is used to describe the O_3 variation during the AFT-CNDH, which is quite different from that in the CNDH. We removed the "in contrast" in the revision.

Changes in manuscript: (line 167-168) The highest MDA8 O_3 drops sharply to 60 ppb in the same regions in AFT-CNDH.

P6-L160. High O3_NOx…

What is a high O3_NOx and O3_VOC level? Also in Fig 2 b and c can you use same range for O3_NOx and O3_VOC? And perhaps a better color bar? The values of O3_NOx in south China during CNDH are not readable.

Response: We used "high" in order to describe the increasing trend of O_3_NO_x and O_3_VOC during the CNDH compared to PRD-CNDH. Sorry for being unclear, and we changed the corresponding content. We also improved the range for O_3_NO_x and O_3_VOC in Figure 2 to better reflect their variations from PRE-CNDH to AFT-CNDH.

P8 – L183 – Fig S4:

This is the part that confuses me the most. Is the increase of NOx (and AVOC) emission in Oct due to the national holiday or it is for the whole month? If it's for the whole month how are you attributing it to the national holiday (only from Oct 1-7)?

Response: Sorry for being unclear. The increase of NO_x and AVOC emissions in October is for the whole month in October compared to September. So, in the emissions comparison figure (Figure 4 in the revision), we only compared anthropogenic changes at a monthly scale. We will work on the real-time emission inventory in the CNDH, trying to develop a more comprehensive understanding of O_3 holiday impacts.

The differences between e and f (if it shows biogenic VOC emissions) is a natural occurring event and not related to changes in anthropogenic activities. How much of the changes in ozone can be attributed to this? I would like to see BVOC emission maps for PRE-CNDH as well given the highest temperature occurred in PRE-CNDH. Can this justify lower O3 values in AFT-CNDH that we see in Fig 1?

Response: The change of the biogenic VOC (BVOC) emissions is a naturally occurring event, which highly depends on the meteorological parameters and land covers [*Guenther et al.*, 2012]. Since the BVOCs are regarded as an important precursor, their variations could affect the O_3 level. We showed the BVOC emissions in PRE-CNDH in Figure R3-3. On average, the BVOC emissions during the PRE-CNDH. A recent study reported that when the BVOC emissions decreased 40%, the O_3 would drop ~3-4 ppb [*Wang et al.*, 2021]. The lower BVOC emissions may decrease the O_3 concentration in the CNDH, similar to that in the AFT-CNDH.

Changes in manuscript: (line 227-229) The enhanced regional transport and the increasing anthropogenic emissions synergistically lead to the rising O_3 during the CNDH, offsetting the impacts from the lower BVOC emissions (Fig. 4).

Can you provide difference plots for AVOC and BVOS plots? Also, why different time frames are considered for BVOC plots?

Response: We've added the difference plots for AVOC and BVOC in the revised Figure 4 (also Figure R3-4). As we explained in the previous response, we used different temporal

resolutions when we generated the AVOC and BVOC emissions. For AVOC emissions (MEIC emission inventory in this study), we used the annual inventory and re-distributed it into the different months. In each month, we only have two different patterns: weekdays and weekends (the temporal profile is the same for each month). So, the AVOC emissions only reflect the monthly differences. The BVOC emissions were generated using the MEGAN model that requires the hourly meteorology parameters as inputs. So, the BVOC emissions can reflect the daily or even hourly differences.

P9 – L 221: Fig S11 and Fig S12. this is not correct.

Response: Thanks. We've corrected it in the revision.

Changes in the manuscript: (line 242-243) The O_3_NO_x shows more significant regional transport characteristics than that of the O_3_VOC (Fig. S11 and Fig. S12).

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Please also note the supplement to this comment:
https://acp.copernicus.org/preprints/acp-2020-1302/acp-2020-1302-AC4-supplement.pdf