Response to Referee #1:

This paper provides a thorough analysis of the impact of meteorological variability on observed ozone changes across China from 2013 to 2017. The analysis is sound, for the most part, but there are a few inaccuracies that need to be addressed, as described below. Once these items are addressed I think the paper would be acceptable for publication in ACP.

Response: We thank the referee for providing a thoughtful and detailed review of our paper. The referee's comments have helped to improve this manuscript. Below, we provide a point-by-point response to the referee's comments and summarize the changes that have been made in the revised manuscript.

Major comments:

[Comment]: 1. The panels in Figure 1 are entirely too small and need to be increased by at least a factor of two, and rearranged on the page so that they fit. I had to enlarge the images on my computer to 400% and even then they were difficult to understand as the resolution was poor. Each panel has an inset in the lower right corner, which doesn't seem to provide any information. These insets are distracting and should be removed. Likewise, the panels in Figures 3, 5 and 7 are also too small. For these figures you can expand the size of each panel by about 15% if you place the color bars underneath the panel, and move the labels on the left of the panels to positions above the panels. You can also delete the latitude and longitude labels, which aren't necessary. Then if you allow the panels to fill the full width of the page you should be able to make them significantly larger.

Response: Thanks for this comment. Regarding Figure 1, we have redrawn each panel with larger circles indicating the observational values. The composition of Figure 1 has been rearranged to enlarge the images to make them clearer. Following the referee's suggestion, we have removed the inset in each panel and placed the color bar underneath the panel in Figures 1, 3, 5, and 7. The labels on the left in Figures 3 and 5 have been changed from horizontal to vertical to save the horizontal space. We keep the latitude and longitude labels because we think this geographic information is important for readers to distinguish China's midlatitude areas (25°N to 40°N), southern (south of 25°N) and northern China (north of 40°N), which are mentioned in our manuscript. Similar changes in enlarging the panels have been made in Figures 2 and 6. Lastly, we have provided all figures with a higher resolution in the revised manuscript.

[Comment]: 2. It would be helpful to place these 2013-2017 surface ozone changes in China within the context of broader trends across Asia, as well as long-term trends in the region of China. For example, Gaudel et al. use IAGOS observations to show that ozone in the lower and mid-troposphere has increased above China, India and Southeast Asia since 1994. Xu et al. show the long-term positive trend at Mt Waliguan, and Sun et al. show the positive trend at Mt. Tai. Wang et al. show the increase of ozone at Hok Tsui when transport is from the South China Sea. And Ziemke et al. show satellite retrievals that demonstrate a board increase of tropospheric column ozone across Asia and the tropics.

Gaudel, A, et al. 2018. Tropospheric Ozone Assessment Report: Present- day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation. Elem Sci Anth, 6: 39. DOI: https://doi.org/10.1525/elementa.291

Sun, L, Xue, L, et al. 2016. Significant increase of summertime ozone at Mount Tai in Central Eastern China. Atmos. Chem. Phys. 16: 10637–10650. DOI: https://doi.org/10.5194/acp-16-10637-2016

Wang, T., Dai, J., Lam, K. S., Poon, C. N., and Brasseur, G. P. (2019), Twen- tyâA Ř˘five years of lower tropospheric ozone observations in subtropical East Asia: The influence of emissions and weather patterns, Geophysical Research Letters, 46, https://doi.org/10.1029/2019GL084459

Xu, W, Lin, W, Xu, X, Tang, J, Huang, J, Wu, H and Zhang, X. 2016. Long-term trends of surface ozone and its influencing factors at the Mt Waliguan GAW station, China– Part 1: Overall trends and characteristics. Atmos. Chem. Phys. 16: 6191–6205. DOI: https://doi.org/10.5194/acp-16-6191-2016

Ziemke, J. R., Oman, L. D., Strode, S. A., Douglass, A. R., Olsen, M. A., McPeters, R. D., Bhartia, P. K., Froidevaux,

L., Labow, G. J., Witte, J. C., Thompson, A. M., Haffner, D. P., Kramarova, N. A., Frith, S. M., Huang, L.-K., Jaross, G. R., Seftor, C. J., Deland, M. T., and Taylor, S. L.: Trends in global tropospheric ozone inferred from a composite record of TOMS/OMI/MLS/OMPS satellite measurements and the MERRA- 2 GMI simulation, Atmos. Chem. Phys., 19, 3257-3269, https://doi.org/10.5194/acp- 19-3257-2019, 2019.

Response: Our study intended to look into the effect of meteorology and emission changes on the recent surface ozone increase in China. We thank the referee for suggesting the above papers on troposphere ozone. We have made some changes in the first paragraph of the Introduction section in the revised manuscript.

Revision in the main text:

1) Line 30-33:

"With rapid urbanization and economic development, the ozone concentrations in the troposphere have increased in the past decades over most regions of Asia, including China (Gaudel et al., 2018; Sun et al., 2016; Wang et al., 2019c; Xu et al., 2016; Ziemke et al., 2019), and ground-level ozone pollution has become a major concern in China's urban and industrial regions (Wang et al., 2017; Verstraeten et al., 2015)."

[Comment]: 3. Section 3.5 What is meant by "potential velocity"? Do you mean potential vorticity? Potential vorticity has long been used as in indicator of stratospheric intrusions into the upper and mid-troposphere, where it works very well, but it just doesn't work for the lower troposphere or the surface because the signal decays by the time the intrusion reaches the lower troposphere (if it ever reaches the lower troposphere). Linking an increase of ozone at the surface to an increase of PV in the upper troposphere is just speculation. How do you know the ozone reaching the surface is from the stratosphere? Couldn't it just be ozone from the mid-troposphere? (as shown by the IAGOS profiles in Gaudel et al. 2018, there is plenty of ozone in the mid-troposphere above China during the summer months) To provide a convincing argument that there was an increase of stratospheric ozone reaching the surface you will have to implement a conserved stratospheric ozone tracer in both MOZART and in CMAQ to see if there really is an increase of this tracer at the surface (see the papers by Meiyun Lin at NOAA GFDL, or papers by Andreas Stohl using the FLEXPART model). If you can't run a tracer all you can say is that there is likely an increase of ozone transport from the free troposphere to the surface, but you don't have any way of knowing if the ozone is from the mid-troposphere or if it's from the stratosphere.

Response: Thanks for this valuable comment. What we mean is potential vorticity (PV) rather than "potential velocity". We have corrected this problem throughout the manuscript. We agree with the referee that it is unsuitable to link the ozone increase at the surface in the low-lying regions of eastern China to an increase in PV in the upper troposphere directly. However, for the Qinghai-Tibetan Plateau of western China with terrain heights > 3 km, PV analysis works well as demonstrated by many previous studies. Based on the PV results, we think that transport from the upper troposphere can explain in part the wind-induced increase in surface ozone in the western highlands. For eastern China (including southern region), we notice that the wind speeds decreased from 2013 to 2017, which would help the accumulation of ozone and ozone precursors and therefore increase ozone concentrations. Referee #2 also suggested that the ozone increases due to wind field changes could be attributed to the changes in horizontal transport. By examining the wind directions in the two years, we did not find evidence for change in vertical transport from the free troposphere to the surface or change in horizontal transport within the model domain. We have revised this section and clarified the possible reasons for the increasing ozone concentrations in terms of the changes in wind speed, horizontal transport, and vertical transport. In Figure 6, we replaced the panels about PV with those on wind speed.

Revision in the main text:

1) Line 22-23:

"The results show that the wind field change made a significant contribution to the increase in surface ozone over many parts of China."

2) Line 291-299:

"Notable increases in MDA8 O_3 in western and eastern China due to the change in wind fields were identified, which contributed significantly to the meteorology-induced increasing ozone (Fig. 3h). In the Qinghai-Tibetan Plateau of

western China whose terrain heights are greater than 3 km, the significant increase in the MDA8 O_3 mixing ratio (3 to 9 ppbv) due to wind change from 2013 to 2017 can be attributed in part to the enhanced downward transport from the upper troposphere as indicated by the increase in the potential vorticity (PV) (Fig. S6). In eastern China, the increase in O_3 level can be explained by the decrease in the wind speeds (Fig. 6h), which helps the accumulation of O_3 and its precursors and then increases ozone concentrations. There is no strong evidence for the change in the vertical transport from the free troposphere to the surface in eastern China and the horizontal transport from other regions within the modeling domain between these two years, according to the wind data (Fig. S7)."

3) Line 360-361:
"The results show that the changes in the wind fields made a significant contribution to the increase in surface ozone levels over many parts of China."

4) Line 382-384:
"The increase in MDA8 O_3 in Qinghai-Tibetan Plateau from 2013 to 2017 was ascribed to enhanced downward transport from the upper troposphere."

5) Line 646-647 (the caption of Figure 6):
"wind speed at a height of 10 m (Wind)"

[Comment]: 4. Line 304 It would be helpful to treat humidity in a consistent manner throughout the paper. In Table 1 you report values of relative humidity, while in Figure 5 your show specific humidity. Why show both types of humidity? From an ozone chemistry perspective specific humidity is most important because it scales with water vapor concentration. Relative humidity isn't useful for understanding ozone photochemistry due to its non-linear relationship to water vapor concentration.

Response: We agree that the specific humidity is more useful for understanding ozone formation than the relative humidity. However, in regular weather observing networks, ambient humidity is generally measured and reported as relative humidity rather than specific humidity. Therefore, we compared the simulated relative humidity with the observed values to evaluate the modeling results of humidity. When investigating the impact of humidity on ozone, we changed to specific humidity because it is simulated and reported by the meteorological model. We have clarified this point in the Methods section of the revised manuscript.

Revision in the main text:

1) Line 150-152:
"Here we used specific humidity rather than relative humidity because the specific humidity, which scaled with water vapor concentrations and was simulated by the model, was more useful for understanding the ozone formation chemistry."

[Comment]: 5. Line 322 This claim that precipitation can remove ozone is incorrect. The modeling study by Meleux et al. vaguely implies that precipitation removes ozone, but they don't give any mechanism or explanation, and this claim goes against the long established fact that ozone has very low solubility in water (Wesely et al., 1981). I can't think of any experimental studies that have shown that rain removes ozone from the air, although some studies have shown that chemicals in water (such as the ocean) can react with ozone if air bubbles are mixed into the ocean, or lakes (see the review by Monks et al., 2015, Atmos. Chem. Phys., 15, 8889–8973, 2015, www.atmos-chem- phys.net/15/8889/2015/ doi:10.5194/acp-15-8889-2015)

Wesely, M. L., Cook, D. R., and Williams, R. M.: Field measurement of small ozone fluxes to snow, wet bare soil, and lake water, Bound.-Lay. Meteorol., 20, 459–471, doi:10.1007/bf00122295, 1981.

Response: We agree that ozone is not water-soluble, and the amount of it removed by precipitation is limited. However, ozone precursors, such as NO_2, have relatively high solubility in water and can be removed by precipitation (Seinfeld and Pandis, 2006), which then decreases the ozone concentration (Shan et al., 2008). We have corrected this claim to

"Although precipitation can decrease ozone concentrations via the scavenging of ozone precursors (Seinfeld and Pandis, 2006; Shan et al., 2008) …" in Line 326-327 in the revised manuscript.

Reference:
Seinfeld, J. H., and Pandis, S. N.: Atmospheric Chemistry and Physics-from Air Pollution to Climate Change, John Wiley & Sons, New Jersey, 2006.
Shan, W., Yin, Y., Zhang, J., and Ding, Y.: Observational study of surface ozone at an urban site in East China, Atmos Res, 89, 252-261, https://doi.org/10.1016/j.atmosres.2008.02.014, 2008.

Minor comments:

[Comment]: 6. Line 45 Well, it's not the relative humidity value that is important, but rather the number of water vapor molecules that are available. It would be best to replace relative humidity with water vapor.

Response: Thanks for this valuable suggestion. We agree with the referee that it is the water vapor that is important, rather than the relative humidity value. We have replaced "relative humidity" with "water vapor" in Line 46 in the revised manuscript.

[Comment]: 7. Line 48 Change "Cloud has" to "Clouds have"

Response: Thanks for this suggestion. We have changed "Cloud has" into "Clouds have" in Line 49 in the revised manuscript.

[Comment]: 8. Line 49 I'm not sure what you mean by "cleaning efficiency". Please use another term.

Response: Thanks for this comment. We have replaced "cleaning efficiency" with "scavenging of oxidants". This sentence has been changed into "Clouds have also been shown to decrease ozone concentrations via aqueous-phase chemistry and photochemistry, which enhances scavenging of oxidants and reduces the oxidative capacity of the troposphere" in Line 49-50 in the revised manuscript.

[Comment]: 9. Line 50 How does the wet removal process increase ozone? Ozone is not water soluble. Is something else being removed by precipitation, which would otherwise destroy ozone?

Response: This is related to our response to comment 5, the ozone precursors (e.g., NO_2) rather than ozone can be removed by precipitation, which then decreases the ozone concentration. We have corrected the original statement to "precipitation decreases the ozone concentration via the wet removal of ozone precursors (Seinfeld and Pandis, 2006; Shan et al., 2008)" in Line 51-52 in the revised manuscript.

[Comment]: 10. Line 65 If you are going to report ozone values in units of ppb, rather than in micrograms per cubic meter, you cannot use the term "concentration". Instead, please use mixing ratio.

Response: Thanks for the valuable comment. We have changed "concentration" to "mixing ratio" in Line 66 in the revised manuscript. We also carefully went through the document and corrected similar issues throughout the revised manuscript.

[Comment]: 11. Line 81 implications (plural) observational data Line 82 . . .based on the observations.

Response: Thanks for pointing out these typos. We have changed "implication" into "implications" in Line 82, changed

"observation data " into "observational data" in Line 82, and changed "based on the observation data" into "based on the observations" in Line 84 in the revised manuscript.

[Comment]: 12. Line 104 Would sound better as: The equations for these statistical parameters can be found in Fan et al. (2013).

Response: Thanks for this suggestion. We have changed "The calculation equations of these statistical parameters can be found in Fan et al. (2013)" into "The equations for these statistical parameters can be found in Fan et al. (2013)" in Line 104-105 in the revised manuscript.

[Comment]: 13. Line 114 . . .which is a few grids cells smaller. . .

Response: Here, it means the CMAQ modeling domain is a few grids cells smaller. To make it clearer, we have changed this sentence "The CMAQ modeling domain covers all the land area of China and the surrounding regions, which is a few grids smaller than the WRF modeling domain to reduce the effect of the meteorological boundary from the WRF model" into "The CMAQ modeling domain, which is a few grids smaller than the WRF modeling domain to reduce the effect of the meteorological boundary from the WRF model, covers all the land areas of China and the surrounding regions" in Line 114-116 in the revised manuscript.

[Comment]: 14. Line 157-158 Please see how I modified the following sentence to improve the English. The authors can make similar changes throughout the document. Original: "Like the temperature, the simulated relative humidity was also slightly under-predicted and had a high correlation coefficient with the observation." Corrected: "Like temperature, the simulated relative humidity values were also slightly under-predicted and had a high correlation coefficient with the observations."

Response: Thanks for this valuable suggestion. We have changed this sentence into "Like temperature, the simulated relative humidity values were also slightly under-predicted and had a high correlation coefficient with the observations" in Line 161-162 in the revised manuscript. Also, we carefully went through this manuscript and made similar changes.

[Comment]: 15. Line 164 . . . conditions on ozone levels. Line 205 . . .emissions on ozone changes. . . Line 217 . . .could be comparable to or. . .

Response: Thanks for these valuable suggestions. We have changed "the ozone level" to "ozone levels" in Line 168-169, changed "to ozone changes" to "on ozone changes" in Line 212, and changed "comparable with" into "comparable to" in Line 225 in the revised manuscript.

[Comment]: 16. Line 312 I don't understand what is being said here: "Possible reasons for the ozone increase with the increase in the PBL height include low primary pollutant concentrations with the development of PBL" Are you saying that fresh emissions of NO can destroy ozone close to the surface in an urban environment, but if you have deep vertical mixing you can spread the NO vertically, which then limits ozone destruction at the surface?

Response: Thanks for this valuable comment. That is what we want to express. To make it clearer, we have changed this statement in the revised manuscript.

Revision in the main text:

1) Line 315-318:
   "Possible reasons for the ozone increase with the increase in the PBL height include lower NO concentration at the urban surface due to the deep vertical mixing, which then limits ozone destruction and increases ozone

concentrations (He et al., 2017), and more downward transport of ozone from the free troposphere where ozone is higher than the near-surface (Sun et al., 2009)."

[Comment]: 17. Line 317 What is cleaning efficiency?

Response: Similar to our response to comment 8, we have replaced "cleaning efficiency" with "scavenging of oxidants" in Line 321 in the revised manuscript.

[Comment]: 18. Line 365 Change "decreased" to "decrease"

Response: Thanks for pointing out this typo. We have changed "decreased" into "decrease" in Line 369 in the revised manuscript.

[Comment]: 19. Lines 383-386 These last two sentences should be revised. The first sentence is rather long and cumbersome and can be shortened as shown below. The second sentence is making a recommendation to policy-makers (by using the word "should") and does not belong in a scientific paper. However, it's perfectly fine to state how your results might be useful to policy-makers, by replacing "should" with "could" as shown below. "It is therefore necessary to consider meteorological variability when assessing the effectiveness of emission control policies on changes in the levels of ozone (and other air pollutants) in different cities and/or regions of China. Such an approach could be useful for the development of future air pollution mitigation policies."

Response: Thanks for these valuable suggestions. We have replaced these last two sentences with "It is therefore necessary to consider meteorological variability when assessing the effectiveness of emission control policies on changes in the levels of ozone (and other air pollutants) in different cities and/or regions of China. Such an approach could be useful for the development of future air pollution mitigation policies" in Line 387-389 in the revised manuscript.

[Comment]: 20. Figure 2, caption The second sentence is difficult to understand. The following change would help: In panel (b) only environmental monitoring sites (493) with data available in all 5 years are presented.

Response: Thanks for this valuable suggestion. We have changed the sentence "As for the observation, only environmental monitoring sites (493) with data available in all 5 years are presented" to "In panel (b), only environmental monitoring sites (493) with data available in all 5 years are presented" in Line 623 in the revised manuscript.

Response to Referee #2:

This manuscript presented a comprehensive modeling analysis on the surface ozone trends over China during 2013-2017. Significant ozone increases have been observed in China over this period in spite of the strong emission control actions implemented. Better understanding the drivers of these trends is of great scientific importance. The authors have conducted an ensemble of numerical simulations using the WRF-CMAQ air quality model to interpret these surface ozone trends, in particular, quantifying the role of meteorology in this manuscript. The results showed that the model had some success in reproducing the Chinese ozone increase trends, supporting the use of it to assess contributions from changes in anthropogenic emissions vs. changes in meteorology. The results further emphasized the importance of interannual variations in meteorology affecting the recent surface ozone trends in China.

This is an important study, representing a great step to understand the drivers of interannual changes in summertime surface ozone pollution in China. The manuscript is well organized and written, and the methodology and results sound solid. I recommend publish after the following comments been addressed.

Response: We thank the referee for providing a thoughtful review of our paper and the recognition of our work. The referee's comments have helped to improve this manuscript. Below, we provide a point-by-point response to the referee's comments and summarize the changes that have been made in the revised manuscript.

Specific comments:

[Comment]: 1. Page 5, Line 120-125:

Some recent studies have suggested that the ozone increases in China since 2013 were largely driven by the concurrent decreases in PM2.5 levels and the resulting changes in heterogenous HO_2 uptake by aerosol surfaces (Li et al., 2019a, 2019b). Since the model applied in this study reproduced the observed ozone increases, did the results support the important role of heterogenous reactions? Although the authors may discuss this issue in the second paper, I suggest put some sentences in this paper in the context of these recent findings.

Reference:

Li, K., et al., Anthropogenic drivers of 2013-2017 trends in summer surface ozone in China, P Natl Acad Sci USA, 116, 422-427, 2019.

Li, K. et al., A two-pollutant strategy for improving ozone and particulate air quality in China, Nature Geoscience, 12, 906-910, 2019.

Response: Thanks for pointing out this and suggesting the two recent papers. We have discussed the impact of heterogeneous reactions on the ozone changes in the second paper (Liu and Wang, 2020), which also supported the significant role of heterogeneous reactions in the increasing urban ozone concentration across China. Following the referee's suggestion, we have revised the manuscript and placed the incorporation of comprehensive heterogeneous chemistry into the CMAQ model within the context of these recent findings.

Revision in the main text:

1) Line 121-124:

"The original CMAQ model includes the heterogeneous reactions of only NO_2, NO_3, and N_2O_5 on aerosol surfaces. Recent studies (Li et al., 2019a; Li et al., 2019b) have suggested that the heterogeneous reactions on aerosol surfaces, mainly the uptake of HO_2, played a significant role in the increasing O_3 concentrations in China from 2013 to 2017. To better simulate the effects of aerosol on ozone via heterogeneous reactions, …"

Reference:

Liu, Y., and Wang, T.: Worsening urban ozone pollution in China from 2013 to 2017 – Part 2: The effects of emission changes and implications for multi-pollutant control, Atmos. Chem. Phys. Discuss., 2020, 1-27, 10.5194/acp-2020-53, 2020.

[Comment]: 2. Page 8, Figure 4:

The year 2013 seems to be a special year with particularly low ozone values, for example, as can be seen from Figure 4a) over Beijing (the BTH region). If the 2013 data point was removed from the linear trend calculation, then no trend was observed for Beijing. This is also the case for Guangzhou (Figure 4c) and the long-range transport ozone influences (Figure 7). Can you comment on this?

Response: Thanks for this comment. For some regions, such as Beijing and Guangzhou, the year 2013 does seem to be a special year with particularly low ozone concentration. But this is not the case in other regions such as Shanghai and Chengdu, in which the ozone concentrations in 2013 were higher than those in 2014. Figure 3a-d also shows that the changes in MDA8 O_3 during 2014-2017 relative to 2013 were increases or decreases, depending on regions and years. Such characteristics are attributed to the complex and varying roles of meteorology in ozone changes, which are highlighted in this paper.

As for the impact of long-range transport on ozone changes, we think the year 2013 is also not a special year with the lowest influence from the long-range ozone transport compared with other years. We remove the 2013 data and plot Figure R1 below to show the changes in the simulated MDA8 O_3 due to variations in long-range transport in 2015, 2016, and 2017 relative to 2014. Similar increases in ozone are still found, which supports the increasing contribution from long-range transport to ozone in these years. We have added this statement to the revised manuscript.

Revision in the main text:

1) Line 231-232:

   "As shown in Fig. 4, the changes in observed MDA8 O_3 varied in cities and years, which were generally captured by the model"

2) Line 346-348:

   "Increases in ozone levels were also found if we compared the changes in MDA8 O_3 due to variations in chemical boundary conditions relative to 2014."

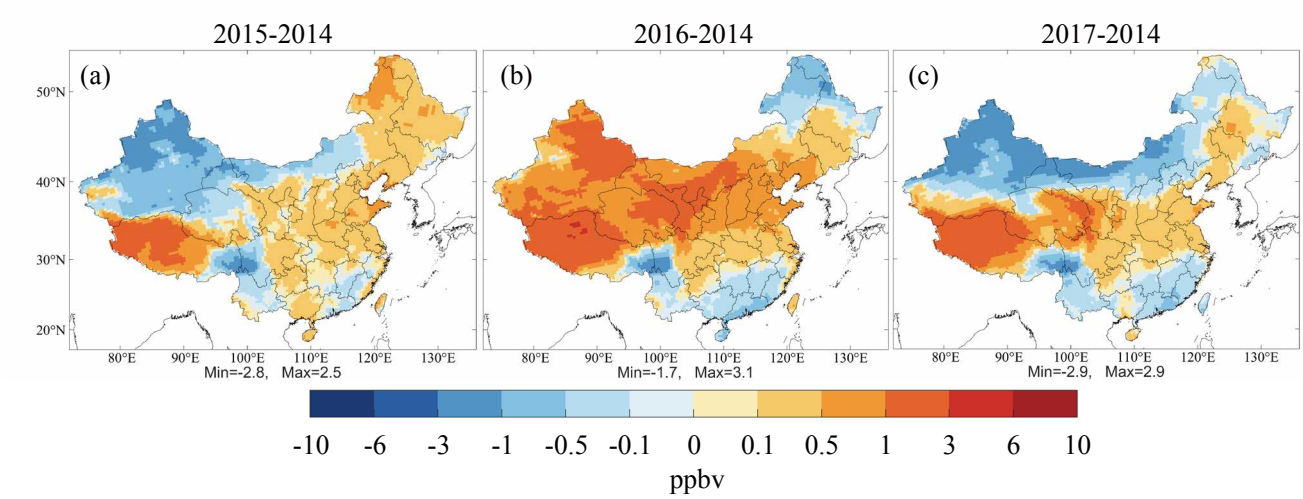

Figure R1 Changes in the simulated summer surface MDA8 O_3 mixing ratios due to variations in long-range transport over China in 2015, 2016, and 2017 relative to 2014.

[Comment]: 3. Page 10, first paragraph of section 3.5:

Figure 6 showed that the 2013-2017 changes in wind significantly increased surface ozone over most regions of China, and the authors attributed the ozone increases to enhanced transport from the lower stratosphere. This is not clear to me. It may explain some of the surface ozone increases in the northern and central China as argued by the PV changes, but how about the southern China where I think stratospheric ozone influences would be low at surface? It is not clear that enhanced ozone transport from the lower stratosphere could lead to 6-10 ppbv surface ozone increases in the southern

China. I wonder whether changes in horizontal winds still contribute there, e.g., changes in the wind speed and the summer Asian monsoon. Please clarify.

Response: Thanks for this comment. Referee #1 raised a similar concern. We think that the PV analysis works well for Qinghai-Tibetan Plateau of western China with terrain heights > 3km. But for other regions (eastern and southern), surface ozone increases are due to a decrease in wind speeds, which would help the accumulation of ozone and ozone precursors in these regions and increase ozone concentrations. By examining the wind directions in the two years, we did not find evidence for change in vertical transport from the free troposphere to the surface or change in horizontal transport within the model domain. We have revised this section and clarified the possible reasons for the increasing ozone concentrations in terms of the changes in wind speed, horizontal transport, and vertical transport. In Figure 6, we replaced the panels about PV with those about wind speed.

Revision in the main text:

6) Line 22-23:
"The results show that the wind field change made a significant contribution to the increase in surface ozone over many parts of China."
7) Line 291-299:
"Notable increases in MDA8 O_3 in western and eastern China due to the change in wind fields were identified, which contributed significantly to the meteorology-induced increasing ozone (Fig. 3h). In the Qinghai-Tibetan Plateau of western China whose terrain heights are greater than 3 km, the significant increase in the MDA8 O_3 mixing ratio (3 to 9 ppbv) due to wind change from 2013 to 2017 can be attributed in part to the enhanced downward transport from the upper troposphere as indicated by the increase in the potential vorticity (PV) (Fig. S6). In eastern China, the increase in O_3 level can be explained by the decrease in the wind speeds (Fig. 6h), which helps the accumulation of O_3 and its precursors and then increases ozone concentrations. There is no strong evidence for the change in the vertical transport from the free troposphere to the surface in eastern China and the horizontal transport from other regions within the modeling domain between these two years, according to the wind data (Fig. S7)."
8) Line 360-361:
"The results show that the changes in the wind fields made a significant contribution to the increase in surface ozone levels over many parts of China."
9) Line 382-384:
"The increase in MDA8 O_3 in Qinghai-Tibetan Plateau from 2013 to 2017 was ascribed to enhanced downward transport from the upper troposphere."
10) Line 646-647 (the caption of Figure 6):
"wind speed at a height of 10 m (Wind)"

[Comment]: 4. Page 10, Line 301-302:

The statement "we found that the impact of temperature via the change in the chemical reaction rate was more significant than that via the change in biogenic emissions from 2013 to 2017" need to be more quantitative. It is difficult to read from Figure 5 and Figure 6 (the color bars are too small). It might depend on regions. I suggest compare their values averaged over the key regions and over China.

Response: Thanks for this suggestion. In the revised manuscript, we have enlarged the panels in Figures 5 and 6 and provided high-resolution figures, which can help to compare the impacts of changes in temperature and biogenic emissions on MDA8 O_3. Figure 5o and 6c are put together in Figure R2 below. We also compared their values averaged in four typical cities, namely Beijing, Shanghai, Guangzhou, and Chengdu, in Figure R3 (also being provided in the supplementary material as Figure S8). We noted that the impact of temperature and biogenic emissions on ozone could be different in some regions (e.g., opposite impacts in Beijing), which can be explained by the transport of emitted pollutants. However, the increases (decreases) in MDA8 O_3 due to changes in temperature were generally higher than those due to changes in biogenic emissions, which supported the statement in our manuscript.

Revision in the main text:

1) Line 304-306:

"However, comparing Fig. 5o to Fig. 6c, we found that the impact of temperature via the change in the chemical reaction rates was generally more significant than that via the change in biogenic emissions from 2013 to 2017 (also see Fig. S8 for the quantitative comparisons in different cities)."

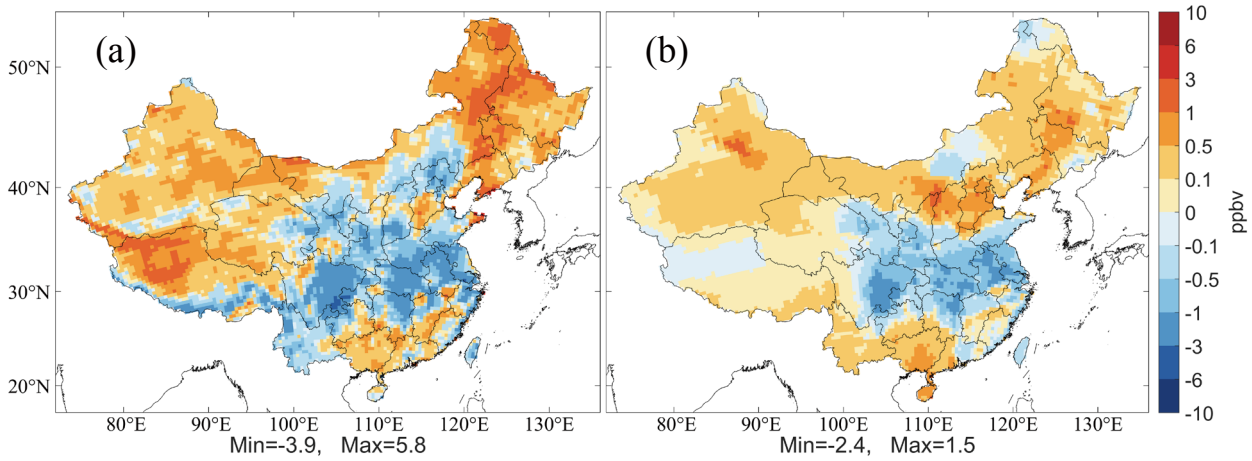

Figure R2 Changes in the simulated summer surface MDA8 O_3 mixing ratios due to the changes in (a) temperature and (b) biogenic emissions in 2017 relative to 2013.

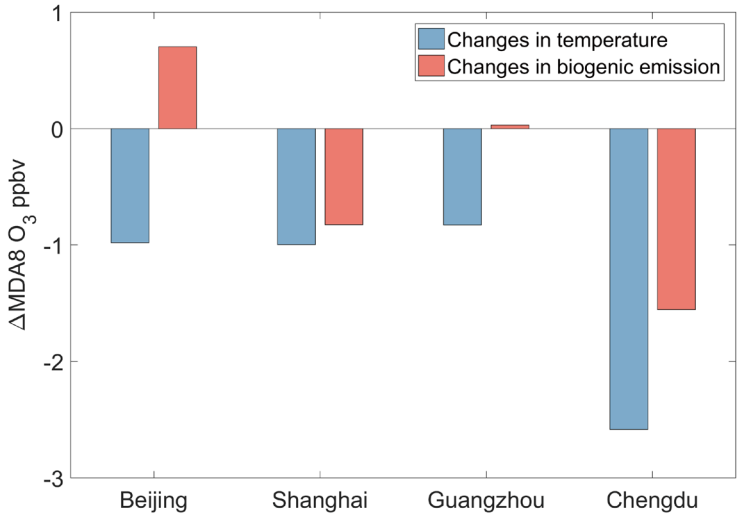

Figure R3 (Figure S8) Changes in the simulated summer surface MDA8 O_3 mixing ratios due to the changes in temperature and biogenic emissions in 2017 relative to 2013 in Beijing, Shanghai, Guangzhou, and Chengdu.

[Comment]: 5. Page 12, Line 342-345:

The statements here seem to imply that transport of PAN led to the long-range transport of ozone influences. How about transport of ozone itself? Which one is the main pathway? One way to quantify and to separate the influences is to conduct a simulation fixing PAN in the 2013 chemical boundary conditions, yet I do not want to push the authors to do more model simulations. Can you explain the issue with present analyses and results?

Response: Thanks for pointing out this issue. The original statement does seem to indicate that the transport of PAN is

the only pathway affecting the long-range transport of ozone, which needs to be corrected. Previous observation and modeling studies (West et al., 2009; Wild et al., 2004) have suggested that ozone and its precursors, namely NO_x (or its carrier PAN), VOCs and CO, can be transported a long distance and then affect the ozone concentration in remote regions. We have corrected this statement in the revised manuscript.

We also think that investigating the pathway of long-range O_3 transport is an interesting and important topic and agree that one way to address this issue is to conduct many simulations fixing different pollutants individually in the chemical boundary conditions and compare the simulation results with each other. As the present study focuses on the impact of meteorological parameters, we prefer to address which chemical contributed most during long-range transport in a future study.

Revision in the main text:

1) Line 347-348:

"Ozone and its precursors can be transported a long distance and then affect surface O_3 in remote regions (West et al., 2009; Wild et al., 2004)."

Reference:

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Worsening urban ozone pollution in China from 2013 to 2017 – Part 1: The complex and varying roles of meteorology

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Abstract. China has suffered from increasing levels of ozone pollution in urban areas despite the implementation of various stringent emission reduction measures since 2013. In this study, we conducted numerical experiments with an up-to-date regional chemical transport model to assess the contribution of the changes in meteorological conditions and anthropogenic emissions to the summer ozone level from 2013 to 2017 in various regions of China. The model can faithfully reproduce the observed meteorological parameters and air pollutant concentrations and capture the increasing trend in the surface maximum daily 8-hour average (MDA8) ozone (O_3) from 2013 to 2017. The emission control measures implemented by the government induced a decrease in MDA8 O_3 levels in rural areas but an increase in urban areas. The meteorological influence on the ozone trend varied by region and by year and could be comparable to or even more significant than the impact of changes in anthropogenic emissions. Meteorological conditions can modulate the ozone concentration via direct (e.g., increasing reaction rates at higher temperatures) and indirect (e.g., increasing biogenic emissions at higher temperatures) effects. As an essential source of volatile organic compounds that contributes to ozone formation, the variation in biogenic emissions during summer varied across regions and was mainly affected by temperature. China's midlatitude areas (25°N to 40°N) experienced a significant decrease in MDA8 O_3 due to a decline in biogenic emissions, especially for the Yangtze River Delta and Sichuan Basin regions in 2014 and 2015. In contrast, in northern (north of 40°N) and southern (south of 25°N) China, higher temperatures after 2013 led to an increase in MDA8 O_3 via an increase in biogenic emissions. We also assessed the individual effects of changes in temperature, specific humidity, wind field, planetary boundary layer height, clouds, and precipitation on ozone levels from 2013 to 2017. The results show that the wind field change made a significant contribution to the increase in surface ozone over many parts of China. The long-range transport of ozone and its precursors from outside the modeling domain also contributed to the increase in MDA8 O_3 in China, especially on the Qinghai-Tibetan Plateau (an increase of 1 to 4 ppbv). Our study represents the most comprehensive and up-to-date analysis of the impact of changes in meteorology on ozone across China and highlights the importance of considering meteorological variations when assessing the effectiveness of emission control on changes in the ozone levels in recent years.

1 Introduction

Elevated concentrations of ozone (O_3) on the earth's surface are harmful to human health and terrestrial vegetation (Lefohn et

al., 2018; Lelieveld et al., 2015; Fleming et al., 2018). With rapid urbanization and economic development, the ozone concentrations in the troposphere have increased in the past decades over most regions of Asia, including China (Gaudel et al., 2018; Sun et al., 2016; Wang et al., 2019c; Xu et al., 2016; Ziemke et al., 2019), and ground-level ozone pollution has become a major concern in China's urban and industrial regions (Wang et al., 2017; Verstraeten et al., 2015). In 2013, the Chinese government launched the Air Pollution Prevention and Control Action Plan to reduce anthropogenic emissions. The Chinese Ministry of Ecology and Environment reported that the observed concentrations of primary pollutants had decreased significantly since these strict control measures (http://www.mee.gov.cn). However, the ozone concentrations in major urban areas of China have continued to increase, and the magnitude and frequency of high-ozone events are much greater in China than in cities in Japan, South Korea, Europe, and the United States (Lu et al., 2018). The ozone problem has become a new challenge to air quality management in China. A comprehensive understanding of the causes of the increase in surface ozone levels in China is necessary to develop a comprehensive whole-air improvement strategy.

Ground-level ozone is produced in situ by chemical reactions from ozone precursors, NO_x, volatile organic compounds (VOCs), and carbon monoxide (CO) or transported from outside the region and from higher altitudes (Atkinson, 2000; Roelofs and Lelieveld, 1997; Akimoto et al., 2015). Meteorological conditions affect the surface ozone concentrations directly via changes in chemical reaction rates, dilution, wet and dry removal, and transport flux or indirectly via changes in natural emissions (Lu et al., 2019b; Lin et al., 2008). As for the direct effects, an increase in temperature can enhance ozone formation by altering the chemical reaction rates (Lee et al., 2014; Fu et al., 2015), and an increase in water vapor can lead to a decrease in ozone concentrations in the lower troposphere (Kalabokas et al., 2015; He et al., 2017). An increase in the planetary boundary layer (PBL) height can decrease ozone levels via dilution of primary pollutants into a larger volume of air (Sanchez-Ccoyllo et al., 2006). Clouds have also been shown to decrease ozone concentrations via aqueous-phase chemistry and photochemistry, which enhances scavenging of oxidants and reduces the oxidative capacity of the troposphere (Lelieveld and Crutzen, 1990), and precipitation decreases the ozone concentration via the wet removal of ozone precursors (Seinfeld and Pandis, 2006; Shan et al., 2008). Wind fields can significantly affect ozone by transporting ozone and ozone precursors in and out of the region of interest (Lu et al., 2019a; Sanchez-Ccoyllo et al., 2006). As for the indirect effects, an increase in temperature can enhance the biogenic emissions of VOCs and thus affect ozone production (Tarvainen et al., 2005; Guenther et al., 2006; Im et al., 2011).

Several studies have used statistical analysis or numerical modeling to assess the effects of meteorological variations on the recent urban ozone trend in China. Using a convergent cross-mapping method to overcome the interactions between various factors, Chen et al. (2019) quantified the influence of individual meteorological factors on the O_3 concentration in Beijing from 2006 to 2016. The results indicated that temperature was the critical meteorological driver of the summer ozone concentrations in Beijing. Cheng et al. (2019) applied the Kolmogorov-Zurbenko filtering method to the ozone variations in Beijing from 2006 to 2017, and the results suggested that the relative contribution of meteorological conditions to long-term variation in ozone was only 2% to 3%, but short-term ozone concentrations were affected significantly by variations in meteorological

conditions. Yin et al. (2019) also used the Kolmogorov-Zurbenko approach to analyze the ozone data for Guangzhou from 2014 to 2018 and showed that four meteorological factors, including temperature, relative humidity, etc., accounted for 76% of the variability in the baseline ozone level. Wang et al. (2019b) used a chemistry transport model (Community Multiscale Air Quality modeling system; CMAQ) to investigate the response of summer ozone concentrations to changes in meteorological conditions from 2013 to 2015, and showed that the maximum daily 8-hour average (MDA8) O_3 mixing ratio decreased by 5 to 10 ppb in most cities due to changes in meteorological conditions and biogenic emissions in the latter two years, except for some cities in eastern, south-central, and southwestern China in which the ozone mixing ratio increased by less than 10 ppb. Lu et al. (2019a) used the GEOS-Chem model to explore changes in source attributions contributing to ozone changes over China in 2016 and 2017 and suggested that the increases in ozone in 2017 relative to 2016 were mainly caused by higher background ozone driven by hotter and drier weather conditions.

Despite these studies, a more comprehensive understanding of the role of the meteorological conditions in the recent ozone changes is still warranted. Previous analyses with a statistical method have been limited to a few cities (i.e., Beijing and Guangzhou). China has a vast territory with a wide range of climates, so the meteorological conditions in various parts of China may have experienced different changes in recent years. Previous chemical transport modeling studies examined either the meteorological impact for 2 or 3 years (no more), the combined (not individual) effects of meteorological parameters, or the combined (not separate) effect of biogenic emission and meteorology changes.

The objective of our study is to investigate the effects of changes in meteorological conditions and anthropogenic emissions on summer surface ozone increases over China from 2013 to 2017 using an up-to-date regional chemical transport model driven by the interannual meteorological conditions and anthropogenic emissions over the 5 years. This paper (part 1) assesses the role of meteorological conditions, and a companion paper (Part 2; (Liu and Wang, 2020)) focuses on the role of anthropogenic emissions and implications for multi-pollutant control. Section 2 introduces the observational data, the model used, and experiment settings. In Section 3, we first evaluate the simulated meteorological factors and pollutant concentrations based on the observations. Subsequently, we separate the changes in the MDA8 O_3 due to the variations in meteorological conditions and anthropogenic emissions by conducting numerical sensitivity experiments and explore their contributions to the ozone changes during the 5 years. Considering the importance of biogenic emissions to ozone production, we estimate the meteorology-driven biogenic emissions over China from 2013 to 2017 and assess their impacts on the variations in ozone. Lastly, the effects of changes in individual meteorological factors are examined, and the role of long-range transport is assessed. Section 4 summarizes the conclusions.

2 Methods

2.1 Measurement data

We used observational data to evaluate the meteorological parameters and air pollutant concentrations simulated by the Weather Research and Forecasting (WRF)-CMAQ model. The daily meteorological observations were obtained from the National Meteorological Information Center (http://data.cma.cn), including the daily average temperature at a height of 2 m, relative humidity at a height of 2 m, wind speed at a height of 10 m, and surface pressure at ~700 ground weather stations in China. The observed concentrations of air pollutants were obtained from the China National Environmental Monitoring Center (http://106.37.208.233:20035/), including SO_2, NO_2, CO, MDA8 O_3, and $PM_{2.5}$. In 2013, there were 493 environmental monitoring stations in 74 major cities, mostly in urban areas. As a result, only these stations have continuous 5-year observations of pollutants from 2013 to 2017. With the increasing recognition of the air pollution problem in China, more monitoring stations have been built since 2013, and the total number exceeded 1500 in 2017. We applied data quality control to the observed pollutant concentrations to remove unreliable outliers following the approach used in previous studies (Lu et al., 2018; Song et al., 2017). The locations of environmental monitoring stations are presented in Fig. S1.

To evaluate the model performance, we calculated some statistical parameters, including the mean observation, mean simulation, mean bias, mean absolute gross error, root mean square error, index of agreement, and correlation coefficient. The equations for these statistical parameters can be found in Fan et al. (2013).

2.2 Model setting

The CMAQ modeling system (Byun and Schere, 2006) was developed by the United States Environmental Protection Agency (US EPA) to approach air quality as a whole by including state-of-the-science capabilities to model multiple air quality issues, including tropospheric ozone, fine particles, toxins, acid deposition, and visibility degradation. This study used the latest CMAQ model (version 5.2.1), an offline chemical transport model without considering the effects of air pollutants on meteorological fields. The meteorological inputs are driven by the WRF model. Table S1 shows the settings of the physical parameterization schemes for the WRF model. The meteorological initial and boundary conditions were provided by NCEP/NCAR FNL reanalysis data with a horizontal resolution of 1°. Fig. S1 shows the modeling domains for the WRF and CMAQ model with a horizontal resolution of 36 km. The model has 23 vertical layers and reaches 50 hPa. The CMAQ modeling domain, which is a few grids smaller than the WRF modeling domain to reduce the effect of the meteorological boundary from the WRF model, covers all the land areas of China and the surrounding regions. The boundary conditions of chemical species for CMAQ were derived from the modeling results of the global chemistry transport model, Model for Ozone and Related Chemical Tracers, version 4 (MOZART-4) (http://www.acom.ucar.edu/wrf-chem/mozart.shtml) (Emmons et al., 2010). We used SAPRC07TIC (Carter, 2010; Hutzell et al., 2012; Xie et al., 2013; Lin et al., 2013) as the gas-phase chemical mechanism and AERO6i (Murphy et al., 2017; Pye et al., 2017) as the aerosol mechanism in the CMAQ model.

The original CMAQ model includes the heterogeneous reactions of only NO_2, NO_3, and N_2O_5 on aerosol surfaces. Recent studies (Li et al., 2019a; Li et al., 2019b) have suggested that the heterogeneous reactions on aerosol surfaces, mainly the uptake of HO_2, played a significant role in the increasing O_3 concentrations in China from 2013 to 2017. To better simulate the effects of aerosol on ozone via heterogeneous reactions, we updated the heterogeneous reaction rates of NO_2 and NO_3 on the aerosol surface and incorporated more heterogeneous reactions into the CMAQ model, including the uptake of HO_2, O_3, OH, and H_2O_2. The detailed heterogeneous reactions in the updated CMAQ model are listed in Table S2. We select the "best guess" uptake coefficients of these gases, which have been widely used in previous chemical transport model studies (Jacob, 2000; Zhu et al., 2010; Zhang and Carmichael, 1999; Fu et al., 2019; Liao et al., 2004). These improvements help the CMAQ model better simulate ozone and other pollutants, and their influence and that of aerosol on the ozone concentration via various heterogeneous reactions are evaluated in the companion paper (Part 2; (Liu and Wang, 2020)).

2.3 Emission

For anthropogenic emissions, we used the multi-resolution emission inventory for China (MEIC) for 2013 to 2017 (http://www.meicmodel.org/), which was developed by Tsinghua University and has been evaluated by satellite data and ground observations (Zheng et al., 2018). International shipping emissions in 2010 were obtained from the Hemispheric Transport Atmospheric Pollution emissions version 2.0 dataset (Janssens-Maenhout et al., 2015). Biogenic emissions from 2013 to 2017 were calculated from the Model of Emissions of Gas and Aerosols from Nature (MEGAN) (Guenther et al., 2006) driven by the interannual summer meteorological inputs from the WRF model.

2.4 Experiment setting

The model simulations were conducted for the summers (June, July, and August) from 2013 to 2017, driven by interannual meteorology and anthropogenic emissions, namely, the base simulations. The shipping emissions remained unchanged in the 5-year simulation due to a lack of data for recent years. To investigate the causes of the increasing surface ozone levels in China, we conducted four sets of modeling experiments based on the simulation of 2013. The first was designed to evaluate the effects of changes in meteorological conditions and anthropogenic emissions (Table S3). We derived the effects of meteorological variation by comparing the simulated ozone concentrations in different years but with the same anthropogenic emissions and chemical boundary conditions as those from 2013. The effects of changes in anthropogenic emissions were derived by comparing the simulated ozone values in 2013 but with anthropogenic emissions from different years. The second set was designed to evaluate the effects of variations in biogenic emissions driven by meteorological conditions (Table S4), which were derived by comparing the simulated ozone values in 2013 but with biogenic emissions from different years. The third set was designed to evaluate the contributions of the individual meteorological parameters to the ozone change from 2013 to 2017 (Table S5), including temperature, specific humidity, wind field, PBL height, clouds, and precipitation. Here we used

specific humidity rather than relative humidity because the specific humidity, which scaled with water vapor concentrations and was simulated by the model, was more useful for understanding the ozone formation chemistry. The fourth set (Table S6) was designed to evaluate the contribution of long-range transport from outside the CMAQ modeling domain (Fig S1) by comparing the simulated ozone levels in 2013 with those with chemical boundary conditions from MOZART from different years.

3 Results

3.1 Model evaluation

Table 1 shows the evaluation results for temperature, relative humidity, wind speed, and surface pressure. The results for all weather stations in China were averaged. The simulated temperatures at the height of 2 m were slightly underestimated with biases of less than 0.6 °C in 5 years. The high correlation coefficients (over 0.82) indicate that the WRF model can capture variations in temperature. Like temperature, the simulated relative humidity values were also slightly under-predicted and had a high correlation coefficient with the observations. The simulated wind speeds at the height of 10 m were slightly overestimated by about 0.5 m s^{-1} due to the underestimation of the effects of urban topography in the WRF model and were often found in other WRF modeling studies (Fan et al., 2015; Hu et al., 2016). The WRF model faithfully reproduces surface pressures for 5 years with low biases and high correlation coefficients. The WRF model could also capture the temporal variations in meteorological parameters. For example, the simulated temperature at the height of 2 m decreased from 2013 to 2015 and then increased from 2015 to 2017, which is consistent with the observations. The good performance of the WRF model gives us the confidence to use the simulations to study the effects of variations in meteorological conditions on ozone levels.

Table 2 presents the evaluation results for air pollutant concentrations in China. Generally, the CMAQ model has excellent performance on the simulated pollutant concentrations with low biases, high index of agreement, and high correlation coefficients. The simulated NO_2 mixing ratio was slightly underestimated for these 5 years in general, which can be explained in part by the fact that the NO_2 concentrations in the national network were measured using the catalytic conversion method, which overestimates NO_2, especially during periods with active photochemistry and at locations away from primary emission sources (Xu et al., 2013; Zhang et al., 2017; Fu et al., 2019). The simulated CO mixing ratio is underestimated significantly by the CMAQ model, which might be due to the missing sources of CO such as biomass burning. The CMAQ model predicts a slightly higher MDA8 O_3 mixing ratio, which could be explained by the artificial mixing of ozone precursors in modeling grids leading to higher ozone production efficiency and positive ozone biases, especially for models with coarser resolutions (Young et al., 2018; Chen et al., 2018; Yu et al., 2016). However, the overall CMAQ model performance is acceptable and can support further investigation of the drivers of increasing ozone levels in China.

3.2 Rate of change in ozone due to meteorology and anthropogenic emission

Fig. 1 shows the spatial distribution of the summer surface MDA8 O_3 level over land areas of China in summer from 2013 to 2017. The CMAQ model can faithfully capture the spatiotemporal variations in the observed MDA8 O_3 level. Both the simulations and the observations exhibit elevated concentration in midlatitude areas, including the North China Plain (NCP), Yangtze River Delta (YRD), Sichuan Basin (SCB), and large areas in central and western China. The O_3 levels in southern China are lower than those in northern China, but they are relatively high in the Pearl River Delta (PRD) region.

We applied the linear regression method to obtain rates of change in the simulated and observed interannual MDA8 O_3, which are shown in Fig. 2. In general, the observed MDA8 O_3 mixing ratios present an increasing trend from 2013 to 2017 in many of the 493 monitoring stations (most located in urban areas of 74 major cities), while the trend result at some of these sites has relatively low confidence level as indicated by large p values (Fig. S2). The model captured 57% of the observed rate of increase averaged for the 493 sites (Fig S3). With the aid of the model simulations, the characteristics of the changes in MDA8 O_3 levels were revealed for all areas, including those with no monitoring stations. Both observations and model simulations show that NCP, YRD, SCB, northeastern China, and some areas in western China experienced increasing levels of ozone pollution. Interestingly, the model results revealed that MDA8 O_3 levels were decreasing in large parts of rural areas that could not be covered by the current monitoring stations, such as northwestern China and southern China.

We separated the change rates of simulated MDA8 O_3 into that due to variations in meteorological conditions and changes in anthropogenic emissions (also see Fig. 2). Here, the impact of biogenic emission variation is included in the effects of meteorological variation because it is affected by meteorology. The result shows that the change rates of ozone over China were more affected by meteorological changes than by emission changes in terms of spatial distribution. The regions with an increasing or decreasing trend of ozone were generally consistent with the contributions from variations in meteorology except for some regions whose ozone trends were dominated by anthropogenic emission changes. The changes in anthropogenic emission have resulted in ozone increases in NCP, YRD, PRD, SCB, and other scattered megacities but decreases in rural regions. This discrepancy can be explained by the different ozone formation regimes in urban (VOCs-limited) and rural (NO_x-limited) areas (Li et al., 2019a; Wang et al., 2019a). A recent study reported the observations of surface ozone during 1994-2018 at a coastal site in southern China and revealed no significant changes in the ozone levels in the outflow of air masses from China mainland during the recent years (Wang et al., 2019c). These results suggest that nationwide NO_x emission reductions may have decreased ozone production over large regions despite causing ozone increase in urban areas. The impact of anthropogenic emission changes on ozone levels in recent years remains a challenging and momentous topic and will be assessed in the companion paper (Part 2; (Liu and Wang, 2020)). The present paper focuses on the effects of meteorological conditions.

3.3 Impact of meteorological conditions and anthropogenic emissions relative to 2013

We next quantified the impact of meteorological conditions and anthropogenic emissions on ozone changes from 2013 to 2017 relative to 2013 (Fig. 3). The changes in MDA8 O_3 from the base simulation varied spatially and yearly, mainly as a result of meteorological conditions. The variation in the MDA8 O_3 mixing ratio due to meteorological changes ranged from -12.7 to 15.3 ppbv over China from 2014 to 2017 relative to 2013. The emission-induced MDA8 O_3 changes in each year exhibited similar spatial patterns, which were consistent with those of the change rates due to emission changes (Fig. 2d). The impact of emission changes on the MDA8 O_3 concentrations became increasingly significant as anthropogenic emissions were further reduced. Our results differ from those by Wang et al. (2019b), who suggested a less important role of meteorology in the variation of ozone from 2013 to 2015. The discrepancy could be due to the difference in the chemical mechanisms and method used for quantifying the effects of emission changes. They calculated the changes in ozone levels due to emission variations by subtracting simulated changes due to meteorological conditions variations from total observed changes, and we calculated by comparing the simulated difference between the simulations in 2013 driven by anthropogenic emissions from different years.

We further found that in some specific regions and years, the changes in MDA8 O_3 due to meteorological variation could be comparable to or greater than those due to emission changes, which highlights the significant role of meteorological conditions in ozone variations. As a result, we selected four megacities in different regions of China to further examine the impact of changes in meteorological conditions and anthropogenic emissions on ozone levels (Fig. 4)—Beijing, Shanghai, Guangzhou, and Chengdu—which are principal cities in the Beijing-Tianjin-Hebei (BTH) region in the north, the YRD in the east, the PRD in the south, and the SCB in the southwestern part of China, respectively (see Fig. S1 for their locations). The numbers of monitoring sites used to obtain the average values for these four cities were 12, 9, 11, and 8, respectively. Most of these sites are situated inside the city, and thus the average results represent the conditions in urban areas. As shown in Fig. 4, the changes in observed MDA8 O_3 varied in cities and years, which were generally captured by the model (except for the changes of MDA8 O_3 in Beijing in 2014 and 2015, which were underestimated, likely due to underestimation of anthropogenic emissions in Beijing and its surrounding regions in those two years). The contributions of anthropogenic emissions to MDA8 O_3 exhibited an almost linear increasing trend in the four cities from 2013 to 2017, whereas the contribution of meteorology could be positive or negative, depending on the region and year.

In Beijing (the BTH region), the variations in meteorological conditions had little effect on the MDA8 O_3 changes from 2014 to 2017 relative to 2013. The increase in ozone was driven primarily by the changes in anthropogenic emissions. This characteristic can also be identified in the larger BTH region, as shown in Fig. 3. In Shanghai (the YRD region), the effects of meteorology were comparable with those of anthropogenic emissions in terms of the absolute values of the contribution to MDA8 O_3 changes. From 2014 to 2016, the meteorology was unfavorable for ozone formation, which masks the ozone increase

due to emission changes. However, meteorological conditions became a positive driver in 2017, leading to a drastic increase in the total MDA8 O_3 mixing ratio (over 10 ppb). In Guangzhou (the PRD region), the role of meteorological conditions was opposite that in Shanghai. The weather changes were conducive to ozone formation from 2014 to 2016 compared with 2013, contributing to a large increase in MDA8 O_3 by over 10 ppbv; in 2017, however, the impact of changes in meteorological conditions on ozone levels decreased substantially, leading to a moderate increase in MDA8 O_3 in that year compared with 2013. In Chengdu (the SCB region), the impact of meteorological conditions on ozone variation was limited in these years, and the ozone level was mainly affected by emission changes, similar to the situation in Beijing. Our result is similar to those by Wang et al. (2019b) for Shanghai and Guangzhou from 2013 to 2015, both indicating meteorological variations unfavorable for ozone formation in Shanghai and favorable in Guangzhou. On the other hand, our simulations differ from theirs that showed a considerable negative contribution of the meteorology variations to ozone levels in Beijing and Chengdu. Our study and that of Chen et al. (2019) suggest a weak role of meteorology variation in the summer ozone trend in Beijing. In addition to these four regions, we found a significant impact of meteorology on the ozone change in western China, especially the Qinghai-Tibetan Plateau (Fig. 3). The meteorological variations contributed to considerable increases in MDA8 O_3 in 2014-2017 in this region relative to 2013.

3.4 Impact of meteorology-driven biogenic emissions relative to 2013

Temperature is an important meteorological driver of biogenic emissions. Fig. 5 displays the temperature changes in land areas of China from 2014 to 2017 compared with 2013. The changes in the spatial distribution were similar in these four years. A decrease in temperature was found in midlatitude areas (25°N to 40°N), and an increase was found in southern (south of 25°N) and northern China (north of 40°N). As shown in Fig. S4, in midlatitude areas such as the BTH, YRD, and SCB, the temperature decreased from 2013 to 2015 and then increased from 2015 to 2017. In contrast, in southern China, such as the PRD region, the temperature increased during 2013-2014 and then slightly decreased during 2014-2017. The variation in the observed temperature is well captured by the WRF model, which enables the MEGAN model to calculate the variation of biogenic emissions driven by the realistic temperature. We present the results of biogenic isoprene emissions because isoprene is generally the most abundant biogenic VOC and has the highest ozone formation potential (Zheng et al., 2009). Large isoprene emissions were found in the southern parts of China and northeast China, which have high vegetation covers in summer (Fig. 5f). The spatial and interannual variations of biogenic isoprene emissions followed the changes in temperatures in China, leading to similar changes in MDA8 O_3 concentrations.

In midlatitude areas of China, the variations in biogenic emissions induced a decrease in the MDA8 O_3 level after 2013. The most significant decrease in the MDA8 O_3 level was found in the YRD and SCB regions, where there were high biogenic emissions and a drastic temperature decrease. In 2014 and 2015, the MDA8 O_3 mixing ratio decreased by ~5 ppbv in these two regions compared with 2013. The changes in ozone were less affected by biogenic emissions due to the lower biogenic

emissions and smaller variation of temperature in the BTH region. In southern and northern China, the increase in temperature and then biogenic emissions since 2013 led to an enhancement of the MDA8 O_3 mixing ratio by up to 1 to 2 ppbv (Fig. 5). In Guangzhou, for example, affected by temperature-dependent biogenic emissions, the MDA8 O_3 increased by 0.8 ppbv from 2013 to 2014 and then decreased slightly from 2014 to 2017 (Fig. S5).

The changes in MDA8 O_3 concentrations due to changes in biogenic emissions in Shanghai and Guangzhou (Fig. S5) generally matched the total changes in ozone levels due to variations in meteorological conditions and provided a considerable contribution to them (Fig. 4). The variations in biogenic emissions were mostly affected by temperature. In section 3.5, we also found that the changes in ozone levels caused by temperature variations via altering chemical reaction rates had an even more significant impact than via changing biogenic emissions in 2017 relative to 2013. As a result, the temperature can play an important role in the variations in ozone levels in recent years. Previous studies also demonstrated the significant role of temperature in the ozone trend in China and other regions (Hsu, 2007; Jing et al., 2014; Lee et al., 2014). However, the role of meteorology is complex and the changes in other meteorological factors can counteract this effect. In Beijing and Chengdu, for example, the changes in ozone levels due to variations in meteorology were insignificant and could not reflect those caused by variations in temperature-dependent biogenic emissions (Fig. 4).

3.5 Impact of individual meteorological parameters in 2017 relative to 2013

Fig. 6 shows the individual effects of changing temperature, humidity, wind field, PBL height, clouds, and precipitation between 2017 and 2013 on the ozone level. Of all the meteorological parameters, the change of wind fields had the most significant impact on MDA8 O_3. It led to an increase in MDA8 O_3 mixing ratio in nearly all of China, with a maximum of 9.1 ppbv (Fig. 6i). Notable increases in MDA8 O_3 in western and eastern China due to the change in wind fields were identified, which contributed significantly to the meteorology-induced increasing ozone (Fig. 3h). In the Qinghai-Tibetan Plateau of western China whose terrain heights are greater than 3 km, the significant increase in the MDA8 O_3 mixing ratio (3 to 9 ppbv) due to wind change from 2013 to 2017 can be attributed in part to the enhanced downward transport from the upper troposphere as indicated by the increase in the potential vorticity (PV) (Fig. S6). In eastern China, the increase in O_3 level can be explained by the decrease in the wind speeds (Fig. 6h), which helps the accumulation of O_3 and its precursors and then increases ozone concentrations. There is no strong evidence for the change in the vertical transport from the free troposphere to the surface in eastern China and the horizontal transport from other regions within the modeling domain between these two years, according to the wind data (Fig. S7).

In addition to wind fields, other meteorological parameters contribute to the ozone change. Because a high temperature facilitates the formation of ozone via the increase in chemical reaction rates, the changes in ozone due to temperature were consistent with the changes in temperature in terms of spatial distribution (Fig. 6b and c). The MDA8 O_3 decreased in central China and increased in other parts of China. This change in the spatial distribution was similar to that due to the changes in

biogenic emissions because they were both affected by temperature. However, comparing Fig. 5o to Fig. 6c, we found that the impact of temperature via the change in the chemical reaction rates was generally more significant than that via the change in biogenic emissions from 2013 to 2017 (also see Fig. S8 for the quantitative comparisons in different cities).

The specific humidity decreased in central China and northeast China but increased in other parts of China from 2013 to 2017 (Fig. 6e). A decrease in the specific humidity in central China led to an increase in the MDA8 O_3 concentration in localized areas, and an increase in other parts of China resulted in a decrease in the MDA8 O_3 concentration in a large area (Fig. 6f). A negative correlation between ozone concentration and humidity in various regions over China was also reported in many previous studies (Ma et al., 2019; Li et al., 2019c).

From 2013 to 2017, the PBL height increased in most parts of China, including NCP, northeast China, and northwest China (Fig. 6k). Our modeling results show that the increase in the PBL height enhanced the MDA8 O_3 concentration in most parts of China (Fig. 6l). A positive correlation between the PBL height and the ozone level in China is also revealed in the statistical results of He et al. (2017). Possible reasons for the ozone increase with the increase in the PBL height include lower NO concentration at the urban surface due to the deep vertical mixing, which then limits ozone destruction and increases ozone concentrations (He et al., 2017), and more downward transport of ozone from the free troposphere where ozone is higher than the near-surface (Sun et al., 2009).

The cloud fraction increased in southwestern China and the Qinghai-Tibetan Plateau but slightly decreased in other parts of China from 2013 to 2017 (Fig. 6n). Because clouds can decrease ozone concentrations via aqueous-phase chemistry and photochemistry to enhance scavenging of oxidants and reduce the oxidative capacity of the troposphere (Lelieveld and Crutzen, 1990), the MDA8 O_3 mixing ratio in most parts of China increased except for southwestern China and the Qinghai-Tibetan Plateau (Fig. 6o).

The change in precipitation was similar to that of the cloud fraction in terms of spatial distribution (Fig. 6q) but made an opposite contribution to ozone levels compared with clouds (Fig. 6r). A positive correlation ($p=0.95$) between ozone and precipitation was also reported by the statistical results of Li et al. (2019c). Although precipitation can decrease ozone concentrations via the scavenging of ozone precursors (Seinfeld and Pandis, 2006; Shan et al., 2008), an increase in precipitation may decrease aerosol concentrations that could increase ozone levels by altering photolysis rates and heterogeneous reactions.

The meteorological parameters that dominate ozone changes can differ in the four megacities (Fig. S9). The decrease in cloud cover was the important meteorological cause that increased MDA8 O_3 in Beijing from 2013 to 2017, the wind field change was the dominant factor that increased and decreased the MDA8 O_3 level in Shanghai and Guangzhou, respectively, and the decrease in temperature contributed primarily to the decline in MDA8 O_3 in Chengdu. However, the effect of the dominant meteorological factor on variations in the ozone level could be counteracted by the influence of other meteorological factors. For example, in Shanghai, the significant positive effect of changes in wind fields on ozone formation was offset by the

negative effects of changes in temperature and precipitation, leading to the smaller increase in the ozone level due to the overall meteorological changes in 2017 compared with 2013 (Fig. 6b). The synergistic or counteracting effects from individual meteorological factors can give rise to the complex impact of the overall meteorology on ozone variations.

3.6 Impact of long-range transport relative to 2013

The chemical boundary conditions for the CMAQ model were derived from the results of the MOZART global model, which can represent the long-range transport of ozone and its precursors from outside the CMAQ modeling domain to China and surrounding regions (Fig S1). We changed the chemical boundary conditions in 2013 to different years to investigate the role of long-range transport in ozone variations in China, and the results are shown in Fig. 7. Changes in long-range transport after 2013 increased the MDA8 O_3 mixing ratio over China except for some areas in northwestern China. Compared with a small increase in MDA8 O_3 (<1ppbv) in eastern China, the Qinghai-Tibetan Plateau encountered a significant increase in MDA8 O_3 by about 1 to 4 ppbv due to changes in long-range transport after 2013. Increases in ozone levels were also found if we compared the changes in MDA8 O_3 due to variations in chemical boundary conditions relative to 2014. Ozone and its precursors can be transported a long distance and then affect surface O_3 in remote regions (West et al., 2009; Wild et al., 2004). A previous MOZART study by Li et al. (2014) found that the transport from the emissions of all Eurasian regions except China contributed 10 to 15 ppbv to the surface O_3 mixing ratio over western China. An analysis of 10-day back trajectories at Mount Waliguan (a remote mountain site in western China) also showed that the air mass from central Asia contributed to the high O_3 levels observed at the site during summer via long-range transport in the free troposphere (Wang et al., 2006). Our study indicates a considerable increase (1 to 4 ppbv) in this long-range transport contribution since 2013.

4 Conclusions

This study explored the impact of changes in meteorological conditions and anthropogenic emissions on the recent ozone variations across China. The changes in anthropogenic emissions since 2013 increased the MDA8 O_3 levels in urban areas but decreased the ozone levels in rural areas. The meteorological impact on the ozone trend varied by region and by year and could be comparable with or even larger than the impact of changes in anthropogenic emissions. The individual effects of changes in temperature, specific humidity, wind field, planetary boundary layer height, clouds, and precipitation from 2013 to 2017 on the ozone levels were examined in this study. The results show that the changes in the wind fields made a significant contribution to the increase in surface ozone levels over many parts of China. The main findings for various regions of China are summarized as follows.

1) In Beijing (the Beijing-Tianjin-Hebei region), the contribution of meteorological changes to the variations in summer ozone was small in 2014-2017 relative to 2013, and the changes in anthropogenic emissions dominated the increase in ozone. Decreasing cloud cover was the dominant meteorological factor that contributed to the increase in MDA8 O_3.

2) In Shanghai (the Yangtze River Delta region), meteorological variation decreased ozone formation from 2014 to 2016, which masked a large increase in ozone due to changes in emissions. The meteorological conditions in 2017 became a positive driver for an increase in ozone, leading to a drastic increase in the total MDA8 O_3 mixing ratio (over 10 ppb). Changes in biogenic emissions had a significant impact on the ozone level in this region. The temperature decrease after 2013 resulted in a considerable decline in MDA8 O_3, especially in 2014 and 2015. The wind field change was the dominant factor that increased MDA8 O_3 in 2017 relative to 2013.

3) In Chengdu (the Sichuan Basin region), the impact of meteorological conditions on changes in ozone was limited from 2013 to 2017, and the ozone concentration was mainly affected by emissions, as in Beijing. However, the biogenic emissions induced by meteorological conditions were important and led to a moderate decrease in the ozone level. The drop in temperature contributed to the decrease in MDA8 O_3 in 2017 relative to 2013.

4) In Guangzhou (the Pearl River Delta region), the meteorological conditions were more conducive to ozone formation from 2014 to 2016 than in 2013, which led to a significant increase (>10 ppbv) in MDA8 O_3. In 2017, the impact of changes in meteorological conditions on ozone levels decreased substantially, and the increase in MDA8 O_3 was small and only due to the changes in emissions. The biogenic emissions driven by meteorological conditions were also important to ozone formation and increased MDA8 O_3 after 2013. The wind field change was the dominant meteorological factor that decreased MDA8 O_3 in 2017 compared with 2013.

5) In western China, the increase in ozone concentrations was mainly caused by meteorological conditions. The increase in MDA8 O_3 in Qinghai-Tibetan Plateau from 2013 to 2017 was ascribed to enhanced downward transport from the upper troposphere. The long-range transport of ozone and its precursors from outside the modeling domain also contributed to the increase in MDA8 O_3 in most parts of China, especially on the Qinghai-Tibetan Plateau.

In conclusion, our study highlights the complex but varying effects of meteorological conditions on surface ozone levels across the regions of China and for different years. It is therefore necessary to consider meteorological variability when assessing the effectiveness of emission control policies on changes in the levels of ozone (and other air pollutants) in different cities and/or regions of China. Such an approach could be useful for the development of future air pollution mitigation policies.

Author contribution

T.W. initiated the research, Y.M.L. and T.W. designed the paper framework. Y.M.L. ran the model, processed the data, and made the plots. Y.M.L. and T.W. analyzed the results and wrote the paper.

Competing interests

The authors declare that they have no conflict of interest.

Code/Data availability

The code or data used in this study are available upon request from Yiming Liu (yming.liu@polyu.edu.hk) and Tao Wang (cetwang@polyu.edu.hk).

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Table 1: Evaluation results for the meteorological factors (T2 is temperature at a height of 2 m; RH2 is relative humidity at a height of 2 m; WS10 is wind speed at a height of 10 m; PRS is surface pressure; Num is number of sites with available observation for statistics; OBS is mean observation; SIM is mean simulation; MB is mean bias; MAGE is mean absolute gross error; RMSE is root mean square error; IOA is index of agreement; r is correlation coefficient; OBS, SIM, MB, MAGE, and RMSE have the same units as given in the first column, while IOA and r have no unit).

Species	Year	Num	OBS	SIM	MB	MAGE	RMSE	IOA	r
T2	2013	692	23.9	23.3	-0.6	2.1	2.3	0.99	0.82
(ºC)	2014	690	23.1	22.8	-0.3	1.9	2.2	0.99	0.82
	2015	708	23.0	22.5	-0.5	1.9	2.2	0.99	0.83
	2016	694	23.8	23.2	-0.6	2.0	2.3	0.99	0.82
	2017	694	23.7	23.2	-0.5	2.0	2.2	0.99	0.86
RH2	2013	692	70.5	67.6	-2.9	10.1	11.9	0.99	0.72
(%)	2014	690	72.2	67.2	-5.0	10.1	11.9	0.99	0.68
	2015	708	71.2	67.6	-3.6	9.2	10.9	0.99	0.72
	2016	694	72.0	68.4	-3.6	9.2	10.9	0.99	0.71
	2017	694	71.7	67.7	-4.1	9.4	11.1	0.99	0.73
WS10	2013	692	2.1	2.8	0.7	1.0	1.2	0.93	0.53
(m/s)	2014	690	1.9	2.5	0.5	0.9	1.0	0.94	0.47
	2015	708	2.1	2.6	0.6	0.9	1.1	0.94	0.54
	2016	694	2.1	2.6	0.5	0.9	1.1	0.94	0.50
	2017	694	2.1	2.6	0.5	0.9	1.1	0.94	0.49
PRS	2013	692	922.4	906.2	-16.2	21.0	21.0	0.99	0.98
(hPa)	2014	690	924.0	907.8	-16.3	21.1	21.1	0.99	0.98
	2015	708	924.0	907.9	-16.1	21.1	21.1	0.99	0.97
	2016	694	923.4	907.5	-15.9	20.7	20.7	0.99	0.98
	2017	694	923.4	907.8	-15.6	20.5	20.6	0.99	0.98

Table 2: Evaluation results for the air pollutants in China (Num is number of sites with available observation for statistics; OBS is mean observation; SIM is mean simulation; MB is mean bias; MAGE is mean absolute gross error; RMSE is root mean square error; IOA is index of agreement; r is correlation coefficient; OBS, SIM, MB, MAGE, and RMSE have the same units as given in the first column, while IOA and r have no unit).

Species	Year	Num	OBS	SIM	MB	MAGE	RMSE	IOA	r
SO_2	2013	408	7.1	12.0	4.9	7.6	9.1	0.79	0.28
(ppbv)	2014	867	6.4	9.0	2.6	5.9	7.0	0.80	0.26
	2015	1410	5.0	5.2	0.2	4.0	4.8	0.77	0.23
	2016	1422	4.4	4.1	-0.3	3.4	4.0	0.77	0.24
	2017	1474	3.8	3.2	-0.6	2.7	3.1	0.77	0.22
NO_2	2013	430	15.1	16.6	1.4	7.0	8.3	0.91	0.41
(ppbv)	2014	843	13.9	13.8	-0.1	6.6	7.7	0.89	0.37
	2015	1411	11.3	9.9	-1.4	5.7	6.7	0.84	0.34
	2016	1420	10.9	9.5	-1.4	5.5	6.4	0.85	0.35
	2017	1480	11.3	9.5	-1.8	5.9	6.8	0.83	0.32
CO	2013	436	0.71	0.34	-0.37	0.39	0.45	0.81	0.33
(ppmv)	2014	872	0.75	0.32	-0.42	0.44	0.49	0.79	0.34
	2015	1400	0.65	0.28	-0.38	0.39	0.44	0.78	0.32
	2016	1419	0.65	0.26	-0.39	0.40	0.44	0.78	0.33
	2017	1473	0.62	0.25	-0.37	0.38	0.41	0.78	0.30
MDA8 O_3	2013	371	50.9	57.7	6.8	17.8	21.3	0.95	0.55
(ppbv)	2014	836	52.5	59.2	6.7	17.7	21.1	0.95	0.54
	2015	1361	50.4	56.4	5.9	15.3	18.3	0.96	0.55
	2016	1373	52.3	57.6	5.3	13.4	16.3	0.97	0.61
	2017	1440	56.3	58.3	1.9	13.1	16.1	0.98	0.63
$PM_{2.5}$	2013	437	44.4	42.8	-1.7	19.4	26.0	0.91	0.58
($\mu g/m^3$)	2014	869	43.8	43.6	-0.2	19.1	24.5	0.92	0.57
	2015	1401	35.3	31.6	-3.7	16.4	20.6	0.89	0.54
	2016	1411	29.7	27.0	-2.7	13.5	17.0	0.90	0.54
	2017	1462	27.8	24.5	-3.3	12.6	15.8	0.89	0.52

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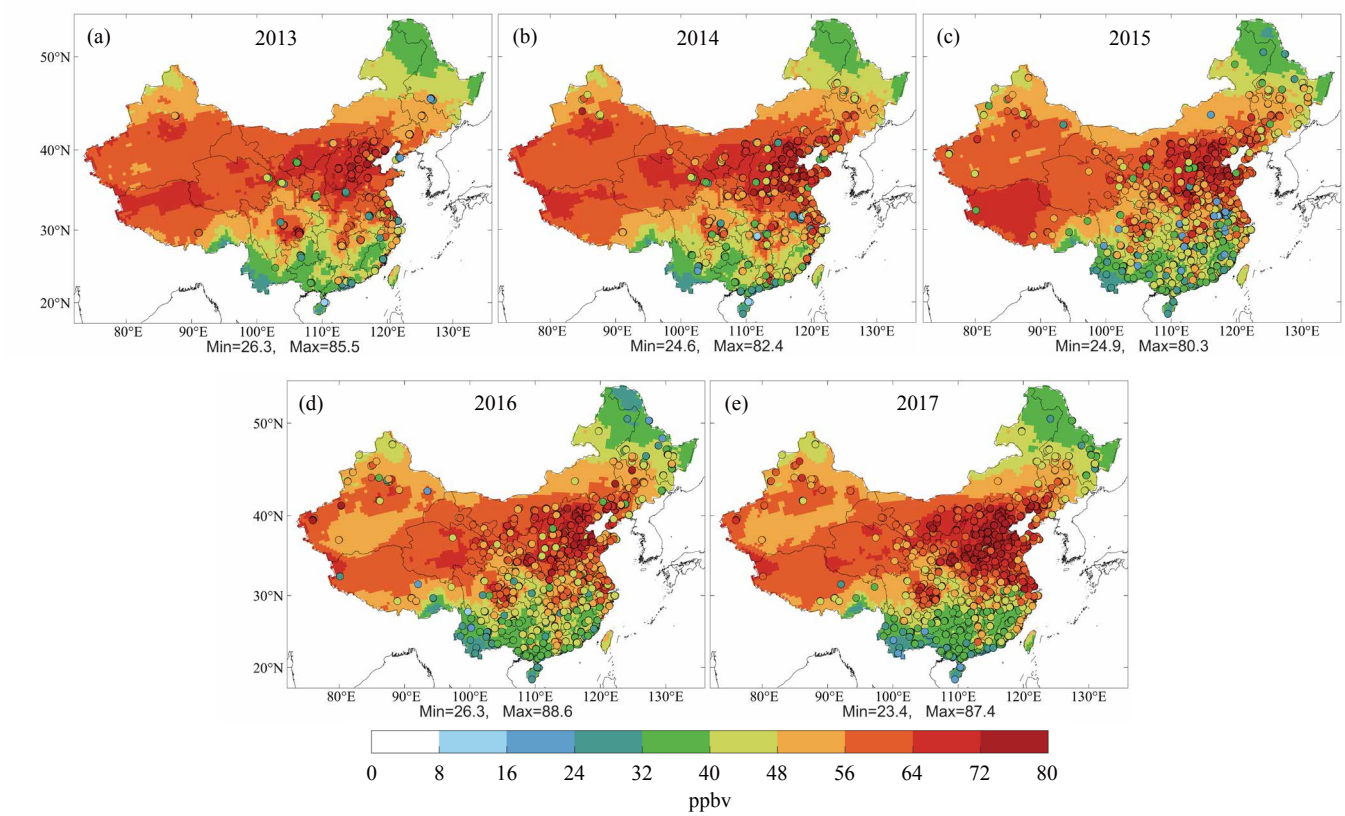

Figure 1: Spatial distribution of the simulated surface maximum daily 8-hour average (MDA8) O_3 mixing ratios across land areas of China in summer (June-August) of 2013 (a), 2014 (b), 2015 (c), 2016 (d), and 2017 (e). Circles with color are the available observed values at environmental monitoring stations in each year.

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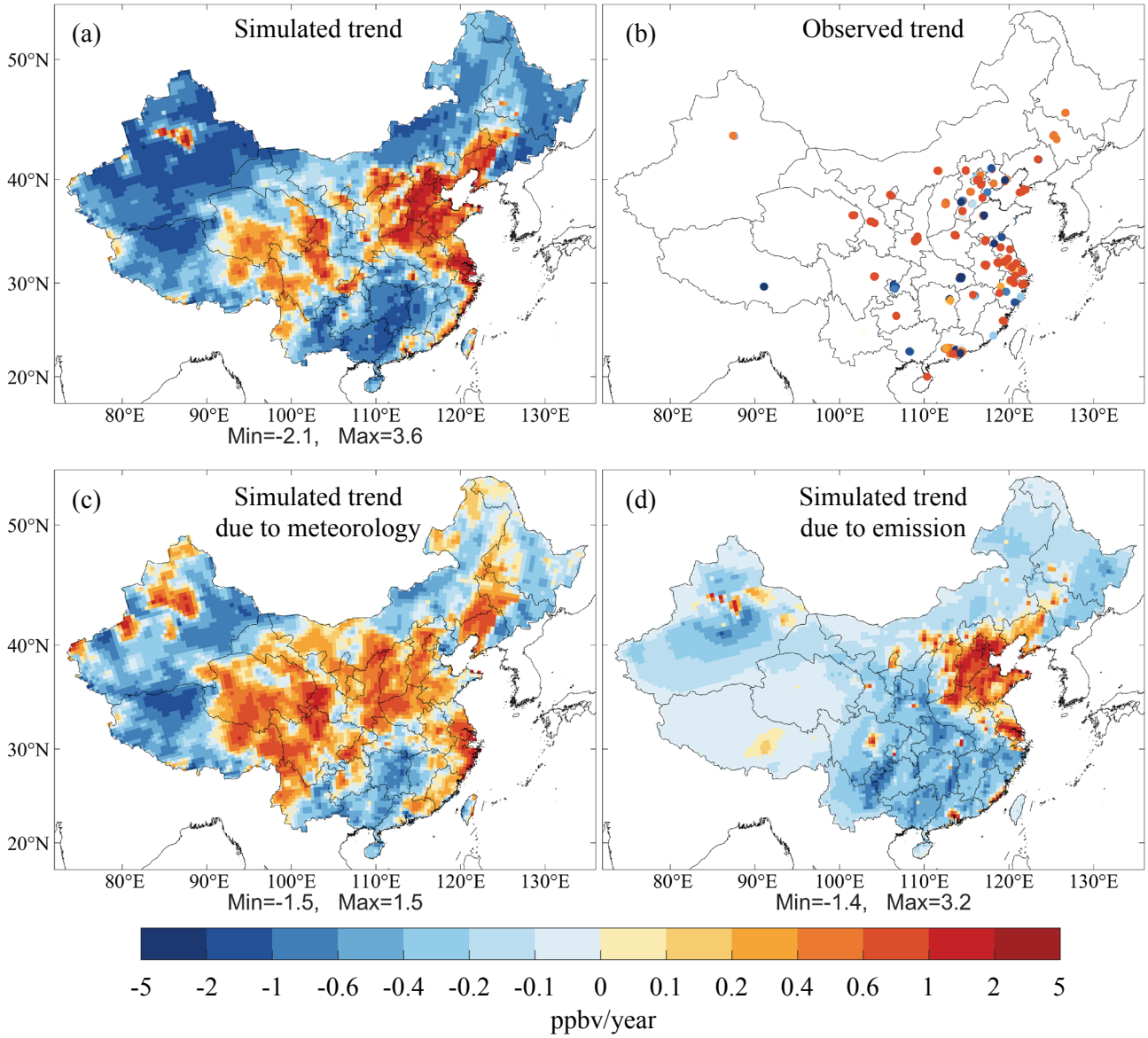

Figure 2: Rates of changes in the simulated (a) and observed (b) surface MDA8 O_3 mixing ratios over land areas of China in summer from 2013 to 2017. In panel (b), only environmental monitoring sites (493) with data available in all 5 years are presented. (c) and (d) present the rates of changes in the simulated MDA8 O_3 mixing ratios due to variations in meteorological conditions and anthropogenic emissions over land areas of China in summer from 2013 to 2017 (see methods). The corresponding p values of regression are presented in Fig. S2.

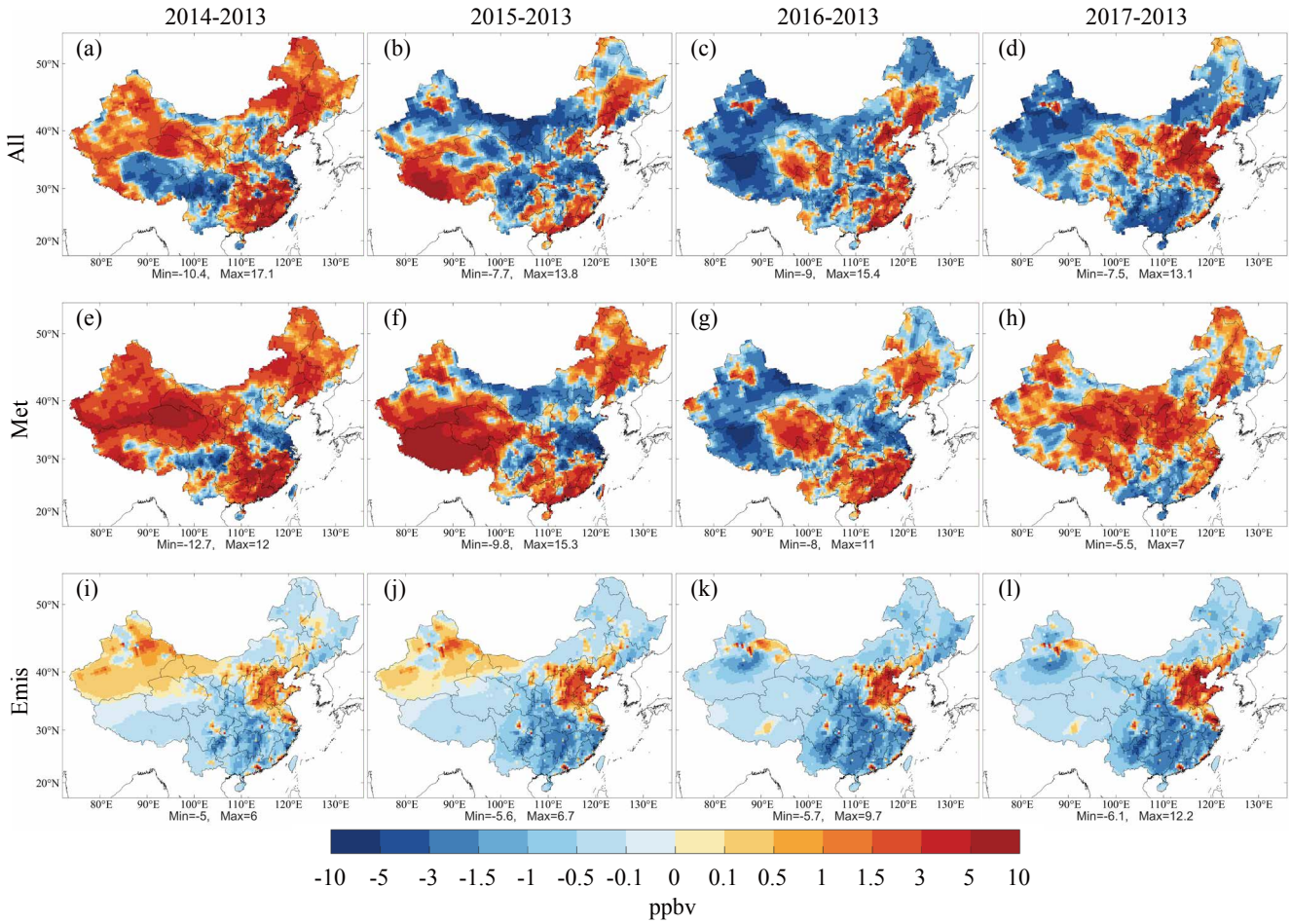

Figure 3: Changes in the simulated summer surface MDA8 O_3 mixing ratios from the base simulation (All, the top row), and those due to variations in meteorological conditions (Met, the central row) and anthropogenic emissions (Emis, the bottom row) in 2014, 2015, 2016 and 2017 relative to 2013.

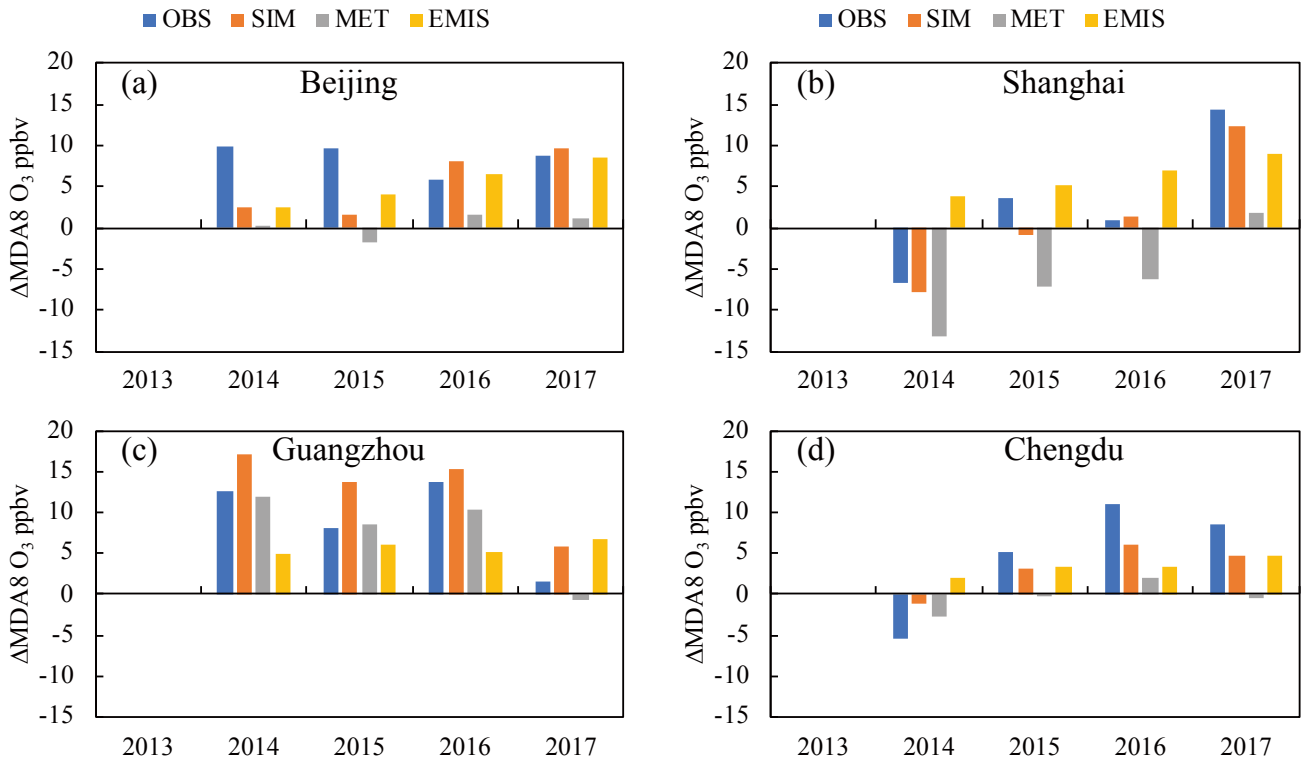

Figure 4: Interannual changes in the simulated (SIM) and observed (OBS) summer surface MDA8 O_3 mixing ratios and those due to variations in meteorological conditions (MET) and anthropogenic emissions (EMIS) in (a) Beijing, (b) Shanghai, (c) Guangzhou, and (d) Chengdu in 2013-2017 relative to 2013.

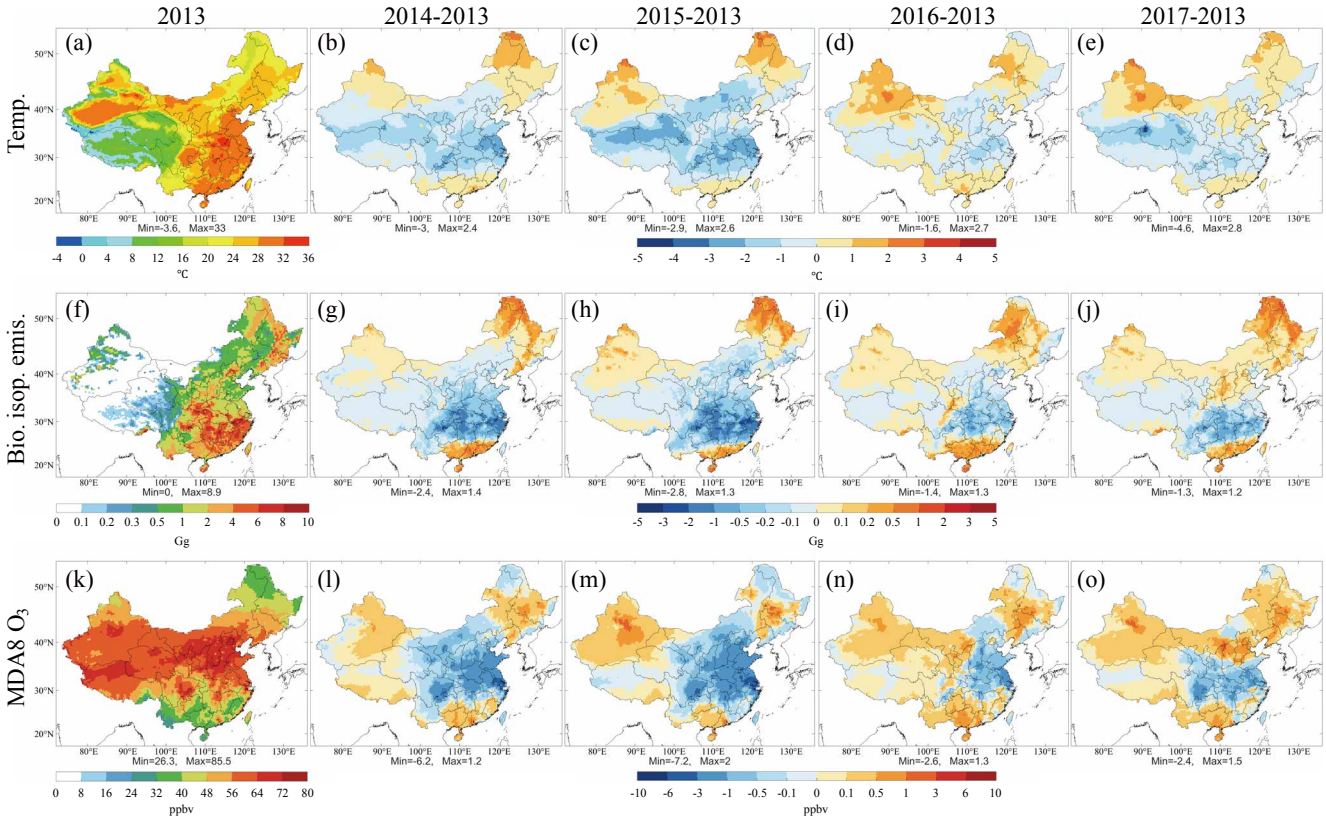

Figure 5: The simulated daytime averaged temperature at a height of 2 m (Temp., the top row) and total biogenic isoprene emissions (Bio. isop. emis., the central row) in summer over land areas of China in 2013 from the base simulations, and their changes in 2014, 2015, 2016 and 2017 relative to 2013. The bottom row (MDA8 O_3) shows the simulated summer surface MDA8 O_3 mixing ratios in 2013 from the base simulation, and their changes due to variations in biogenic emissions in 2014, 2015, 2016 and 2017 relative to 2013.

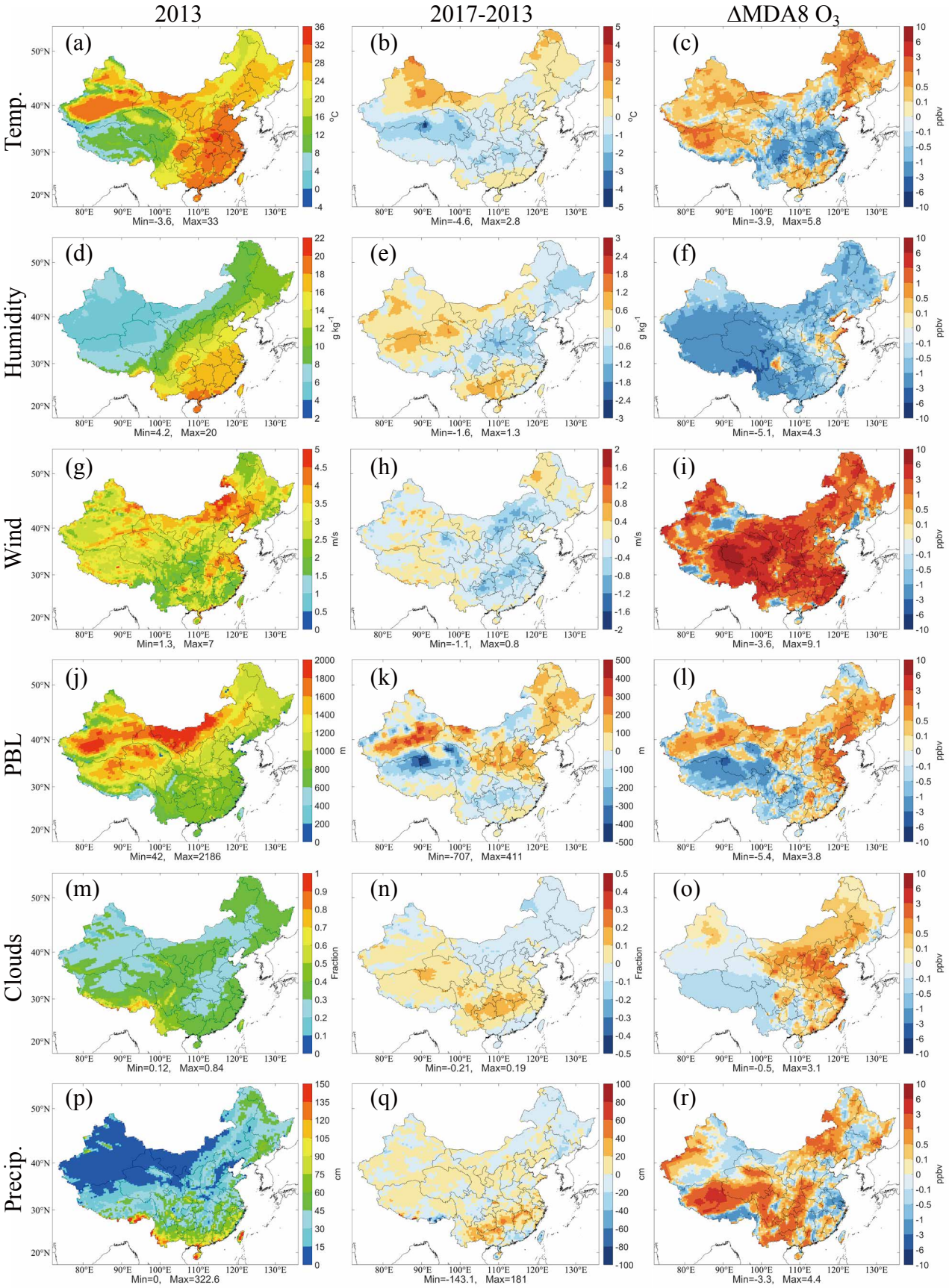

Figure 6: The simulated averaged temperature (Temp.) and specific humidity (Humidity) at a height of 2 m, wind speed at a height of 10 m (Wind), planetary boundary layer (PBL) height, total clouds fraction (Clouds), and accumulated precipitation (Precip.) in the daytime in summer of 2013 from the base simulation (the left column), and their changes in 2017 relative to 2013 (the central column). The right column shows the changes in simulated summer surface MDA8 O_3 mixing ratios due to variations in temperature, specific humidity, wind fields, PBL height, clouds and precipitation in 2017 relative to 2013.

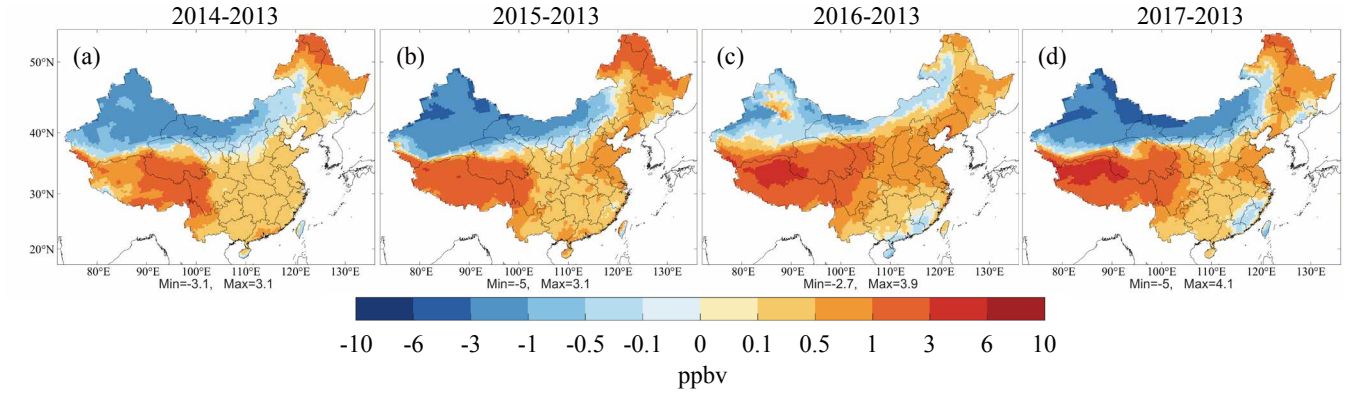

Figure 7: Changes in the simulated summer surface MDA8 O_3 mixing ratios due to variations in long-range transport over land areas of China in 2014, 2015, 2016, and 2017 relative to 2013.