

## Response to Review #2

This study quantifies the most recent trends in summertime O<sub>3</sub> concentrations in China and investigated the possible causes. This is a timely paper which has implications for the improvement of China's ongoing control policies. However, I have the following concerns which need to be addressed before the manuscript can be considered for publication in ACP.

We thank the reviewer's valuable comments which improve our manuscript greatly. We have detailed the MLR method and justified the application of this statistical approach. Please find below our point-by-point response in **blue**.

Major comments:

1. The multiple linear regression (MLR) is a key method used in this study to quantify the meteorological contribution. However, this paper lacks a lot of details regarding the data sources and results of the MLR method. In Section 2: "The regression model is first applied to select the key meteorological parameters driving the day-to-day variability of ozone for each grid cell." What meteorological parameters are considered in the selection? Which parameters show statistically significant contribution based on the regression? What criteria did you use to select the parameters used in the formal analysis? How much did the selected parameters explain the overall variability? In Section 3.1 and Section 3.2, you talked a lot about the dominant meteorological predictors in China and various metropolitan regions. However, no MLR results supporting these conclusions are shown. How much did these parameters contribute? Are the contributions from these parameters statistically significant?

Sorry for not making it clear. The statistical method follows our previous study (Li et al., 2019a). We now have detailed the MLR method in P4L5-21 in Section 2, and have included the meteorological candidates to be selected in the regression model, how the top three meteorological drivers are selected for each grid cell, and the explained variance by the MLR model.

"Firstly, the regression model is applied to select the key meteorological parameters driving the day-to-day variability of ozone for each grid cell. There are nine MERRA-2 meteorological variables considered as ozone covariates, including daily maximum 2-m air temperature (Tmax), 10-m zonal wind (U10) and meridional wind (V10), boundary layer height (PBLH), total cloud area fraction (TCC), rainfall (Rain), sea level pressure (SLP), relative humidity (RH), and 850-hPa meridional wind (V850), following (Li et al., 2019a). The meteorology fields are averaged over 24 h for use in the MLR model except for PBLH and TCC, which are averaged over daytime hours (8–20 local time), and for Tmax (daily maximum).

Secondly, to avoid overfitting, only the three locally dominant meteorological parameters are regressed onto the deseasonalized monthly MDA8 ozone to fit the role of 2013–2019 meteorological variability. The top three variables are selected based on their individual contribution to the regressed ozone, along with the requirement that they are statistically significant above the 95% confidence level in the MLR model. They will differ for each  $0.5^\circ \times 0.625^\circ$  grid cell. We show these top three meteorological drivers for ozone variability in Figure S1–S3 for different locations in China.

Thirdly, we fit the observed monthly ozone anomalies by applying these dominant meteorological drivers in the MLR model. The coefficients of determination ( $R^2$ ) for the MLR model are generally above 0.4–0.5 for polluted regions of China which are of most interest to us (Figure S4). Remote locations with background ozone levels have less ozone variability and are thus harder to fit.”

2. After reading the paper, my overall impression is that the author should tune down the statement that they have elucidated the relative contribution of meteorological and anthropogenic factors to the O<sub>3</sub> trend. The meteorologically driven trend is quantified by fitting O<sub>3</sub> to selected met parameters while the residual is regarded as the anthropogenically driven trend, so the anthropogenically driven trend is largely unconstrained. This attribution method is subject to a large uncertainty, especially for the anthropogenically driven part. I would not recommend the author to conduct a modeling simulation to test the anthropogenic contribution which requires a lot of additional work, but I am deeply concern that the quantitative attribution to the two parts may not be accurate without further constraint. Even for the meteorological part, you only considered a subset of met parameters in the MLR. Can these selected parameters represent the overall contribution of meteorology? This again points to my last comment that showing the results of the MLR analysis is important.

The application of MLR mode has been detailed in our response to the last comment. This method has been extensively applied to quantify the effect of meteorological variability on air pollutants, and statistical quantification of anthropogenic and meteorological contributions to air pollutants also has been well documented. We have clarified this in the main text.

In P4L21-22: “Similar MLR models have been extensively employed to quantify the effect of meteorological variability on air pollutants (e.g., Tai et al., 2010; Otero et al., 2018; Zhai et al., 2019; Han et al., 2020).”

In P4L25-28: “We have followed this approach before to isolate the anthropogenic trends of ozone and PM<sub>2.5</sub> (Li et al., 2019a; Zhai et al., 2019). Similar statistical decomposition of anthropogenic and meteorological contributions to air pollutant trends has been also employed by previous studies (e.g., Chen et al. 2019; Yu et al., 2019; X. Zhang et al., 2019).”

3. Section 3.1: When you talk about the observational trends, you need to point out whether these trends are statistically significant. Fig. 2 shows some significance testing results, but it’s also important to incorporate such information in your description.

We have moved Table S1 into the main text, as also suggested by Reviewer #1. A p-value showing significance is also given wherever applicable.

4. Abstract Line 20-22: Whether the anthropogenically driven O<sub>3</sub> trend is caused by decrease in PM<sub>2.5</sub> or reduction in NO<sub>x</sub> is a controversial issue. This study actually did not carefully investigate this issue but just referred to a previous study. Therefore, you may at most infer that this might be a cause rather than state with certainty that this is the actual explanation.

Thanks. We agree with the reviewer. We have revised the text accordingly.

In P1L21: “fine particulate matter (PM<sub>2.5</sub>) that may be driving the continued anthropogenic increase in ozone”

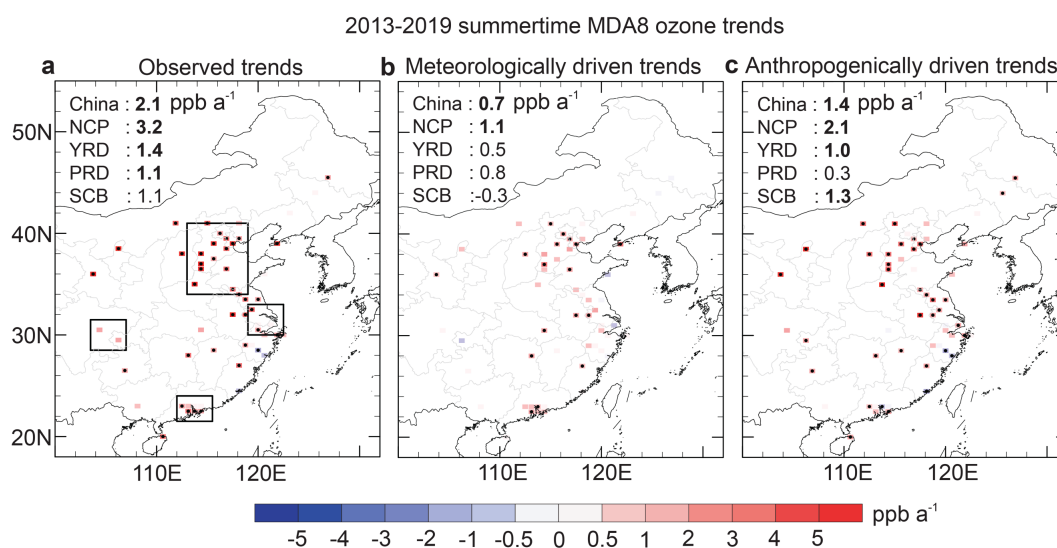
In P5L24-26: “The increases are largest in the NCP, which could be explained by greater influence of radical scavenging by PM<sub>2.5</sub> (Li et al., 2019a, 2019b).”

In P9L3-4: “The sustained anthropogenic increase in ozone over the 2017–2019 period may be explained by the continued decrease of PM<sub>2.5</sub>...”

5. In your regression analysis to determine the O<sub>3</sub> trend, you included sites with partial records. Since the number of observational sites grow dramatically from 2013 to 2019, the trends can be biased by the differences in observational sites. I suggest that you repeat the analysis using only continuous sites and examine whether this affects your results significantly.

The sites are basically stable after 2014. Our results still stand if only continuous records are used, as shown in the following plot.

We have added the plot in the Supplementary Information, and description in main text P6L15: “This result still stands if only continuous records since 2013 are used in the analysis (Figure S5).”



**Figure S5.** Same with Figure 2 but for the sites with continuous records from 2013.

Minor comments:

1. Sometimes you abbreviated “meteorologically driven trends” to “meteorological trends”, which I think is not accurate.

Corrected throughout the text. Thanks.

2. The spatial extents of NCP, YRD, PRD, and SCB are not defined in the paper

Added in P5L12-13.