

We thank the reviewers for their insightful comments. Our point-by-point responses to the reviewer comments (line numbers in this response refer to the clean manuscript) and the track-change manuscript are as follows.

Response to Review #1

Review of “2013-2019 increases of surface ozone pollution in China: anthropogenic and meteorological influences” by Ke Li, Daniel J. Jacob, Lu Shen, Xiao Lu, Isabelle De Smedt and Hong Liao, submitted to Atmospheric Chemistry and Physics.

Summary: Based on statistical analysis of recent surface measurements of ozone in China and meteorological conditions from a modern reanalysis product, the authors quantify the role of meteorology versus emissions on the positive ozone trends in Chinese megacities. Despite regulations targeting ozone pollution, ozone has continued to increase over the past decade.

While the results are interesting, they seem preliminary. Section 3 is a list of “Figure X shows” and I encourage the authors to add depth to their research and analysis and more fully develop their narrative. There are five figures, which are all multi-paneled, and only three pages of “Results and discussion”; this unequal balance of figure to text highlights the need for further exploration into the information contained in each figure. In addition, there is a supplemental figure and supplemental table which I found added to the analysis and I encourage the authors to include these in the main text.

This manuscript is within the scope for Atmospheric Chemistry and Dynamics; however, I suggest the authors expand the discussion of the results before I recommend it for publication.

We appreciate the reviewer’s constructive and thorough comments/suggestions. We have moved the supplementary figure and table to the main text. We also have detailed the description of MLR model and foehn wind effect. Figures have been also revised following your suggestions, thanks! Please find below our point-by-point response in **blue**.

Major comments:

Pg 2 Ln 15: The authors discuss VOCs as additional industrial sources but there can be natural sources of VOCs from plants. Have the authors considered the natural sources of VOCs in this analysis? There’s no discussion of natural VOC emissions on Page 6 or 7.

Thanks. We have added the discussion in P4L28-30: “The effect of biogenic VOCs on ozone trends depends on meteorological and land cover drivers. Meteorological drivers, in particular temperature, would be accounted for in the MLR model. The effect of land cover changes is expected to be small over the 7-year time horizon of our analysis (Fu and Tai, 2014).”

Pg2 Ln 27: The authors should include the MEE website here in the body of the text and in the Data Availability section at the end of the manuscript.
Added.

Pg 4 Ln 15, 20: These trend values are provided in Figure 2. What is novel about Table S1 is it highlights that a significance test was performed. In Line 20, the authors quote “significantly

enhanced” without a reference to a statistical test. The authors should describe this in the Data and Methods Section at the bottom of Page 3 and I would encourage the authors to include this table in the main text as it is referenced on Page 4 Line 21 to support a critical result.

Thanks! Now it (as Table 1) has been added in the main text. For statistical test, we have added in P4L24-25: “with the statistical significance of the anthropogenic trend determined by Student’s *t*-test”

Pg4 Ln 30: The authors reference a supplemental figure which I would argue should be in the main text as it highlights not only the change in maritime inflow which impacts the Shandong Peninsula, but it looks like the YRD and Northeast China as well. The authors should discuss how the meteorological conditions mitigate ozone pollution over western China (does that include SCB?) and northeast China.

Thanks! Now it (as Figure 3) has been added in the main text. We also have added the discussion on the decreasing ozone trends in P6L8-9: “Temperature decreased over northeastern China (Figure 3).”

Pg4 Lines 22,23 and Page 5 Ln15: The authors could go into more detail on not only the primary meteorological predictor variables, and also include the breakdown for all megacities and regions discussed. Only NCP and PRD’s principal predictors are given.

We have added the plots of leading meteorological variables in Supplementary Figures S1-3, which are also cited in the main text.

Figure comments:

Pg 4 Line 7: It would make it easier on the reader if the rectangles in 2019 Figure 1 top row were included in 2019 Figure 1 middle row since the text is discussing both mean MDA8 and the max MDA8 by region.

Added.

Pg 14 Figure 1:

While the figure caption includes the latitude and longitude for the four megacity clusters, the latitude and longitude ticks are not labelled. Can [some of] the ticks be labelled, or at least include in the caption what are the intervals of the major and minor ticks and some reference point?

Would the discussion of the ozone max and mean trends benefit from the max PM_{2.5} being included in Figure 1? Has the maximum PM_{2.5} decreased the same as the mean (Pg4 Lines 4-5). If that is the case, could state that and not show it.

We have revised Figure 1, and the maximum PM_{2.5} trends are also added to Figure 1. We have added in P5L9-10 about the decrease of maximum PM_{2.5}: “Maximum PM_{2.5} concentrations experienced a similar decrease trend.”

Page 15 Figure 2: Can the rectangles for the megacity clusters be added to Figure 2 or at least Figure 2a? I did my best to draw them on so I could follow the text referring to the trends in the four regions. Again, it would be helpful to have some of the latitude and longitude tick marks

labelled and/or the intervals of the major and minor ticks and some reference point defined in the figure caption.

Done.

Page 15 Line 10: Could add a reference to the Table S1 at the end of the caption.

Added.

Pg 5 Line 8: This sentence references the Table in Figure 3. The left figure and right table should be labelled as (a) and (b) to make referencing in the text clearer. Also, to save on white space, I would encourage the authors to include the table as an inset in Figure 3 or as a separate table.

The ozone trend has been already given in the main text and We have removed the right table now.

Pg 17 Line 7-9: The definition of the Foehn index and foehn-favorable conditions should be in Section 2 or in the text of Section 3; I suggest it is removed from the figure caption.

Thanks! We have included the definition of foehn index in P7L-10 in Section 3. We would like to still include this in the Figure caption to make the Figure self-explanatory.

Pg 17 Line 9-10: “The frequency of foehn wind under hot days increased by 85% over the period” is a result and should be in the main text and not in the figure caption. Can the authors go into more detail about this trend? Was it mainly driven by 2018 and 2019?

Yes. We have moved this into the main text in P7L11-13: “The frequency of foehn conditions under hot days in June increased by 85% over the 2013–2019 period (driven mainly by the increased frequency in 2018–2019), and ozone increase under foehn conditions is 1.2 ppb a⁻¹ larger than under no-foehn conditions.”

Minor and technical comments:

Pg1Ln19: The June-July temperatures over the NCP are higher than what? Other regions of China? Other months?

Higher than previous years. We have changed “higher” to “rising” for clarification.

Pg2Ln18: Can the authors describe how meteorological conditions may affect emissions? Are they referring to natural emissions from plants which do vary based on meteorological conditions, or do they mean anthropogenic emissions such as through energy consumption?

We intended to say “natural emissions”, which are much more meteorologically dependent than anthropogenic emissions. We have added “natural” in P2L19.

Pg 3 Line 1: No mention of NO₂ surface observations but these are referenced in Figure 5.

Added.

Pg 3 Line 12: There was a version change in the TROPOMI NO₂ data in March 2019 (<https://sentinel.esa.int/documents/247904/2474726/Sentinel-5P-Level-2-Product-UserManual-Nitrogen-Dioxide>). I am concerned this change could add a bias when comparing Summer 2018 (v1.2.0) vs Summer 2019 (v1.3.0).

Thank you for pointing this out. Now we have removed the results for TROPOMI NO₂ changes.

Pg 4 Line 2: A paragraph of preamble providing an overview of the results section would be good, to help the reader see that there are three subsections within Section 3.

Great suggestion.

We have added in this in the beginning of Section 3: “We first present the general 2013–2019 summer ozone trends in China and their statistically decomposed meteorological and anthropogenic contributions. Ozone trends over the major megacity clusters in China are highlighted. We go on to more specifically attribute the meteorological and anthropogenic drivers of recent ozone trends over the North China Plain, where the ozone increase is the highest.”.

Pg 4 Line 10: Do the authors have a hypothesis as to why the summer maximum MDA8 ozone values in YRD, PRD, and SCB were comparable to the NCP but not the means?

We have added the explanation in P5L16-19: “These three megacity clusters are subject to similar ozone pollution episodes under stagnant conditions as the NCP (Wang et al., 2017), but they are more frequently ventilated by the summer monsoon bringing cleaner tropical air and precipitation hence the lower mean ozone.”.

Pg 4 Ln 14: Can the authors provide latitude and longitude regions for Shandong Peninsula and Northeast China. Line 28, the authors refer to ‘northeastern’ China. Is this different than Northeast China?

We now use “northeastern” instead of “Northeast” throughout the text for consistency. We intended to focus on the four major megacities clusters that are also the pollution control regions targeted by the Chinese government.

Shandong Peninsula typically refers to the east part of Shandong province; Northeast China typically includes Helongjiang, Jilin, and Liaoning provinces. We have added this information in the main text (P5L22).

Pg 5 Lines 2-3: It looks to me that the anthropogenic trend is more uniformly positive in Fig 2c than Figs 2a,b except for in the Shandong Peninsula and maybe the PRD and YRD regions. Can the authors confirm?

Yes, you are right. The meteorological role varies regionally. We have revised the text in P6L11-12: “This anthropogenic trend is more uniformly positive at a national scale...”.

Pg 5 Line 4: Why might the PRD experience a decrease? Is that because of the change in monsoon winds? Make connections between the figures and discussion where possible.

Yes, due to weakened monsoonal winds. We have added the explanation in P6L15-16: “The ozone increase in PRD is mainly meteorologically driven due to reduced monsoonal winds (**Figure 3**).”.

Pg 5 Line 16: While the June (August) temperatures clearly show increasing (decreasing) trends over the 2013-2019 period, the temperature pattern in July looks almost neutral if averaged over this period and not “a lesser increase”. This phrase is awkward.

In fact, the temperature trend in July ($0.22\text{ }^{\circ}\text{C a}^{-1}$) is comparable with trend in August ($-0.18\text{ }^{\circ}\text{C a}^{-1}$), while both of them are much lower than trend in June ($0.42\text{ }^{\circ}\text{C a}^{-1}$). The high temperature in 2018–2019 is the reason for an increasing temperature trend in July. We have added the temperature trends in P6L29 and P7L1.

Pg 5 Line 21: The reference following foehn winds gives me the impression that this paper is the first to define foehn winds. However, a foehn wind is the warming of air through adiabatic descent on the lee side of a mountain, much like a Chinook Wind on the lee side of the Rocky Mountains, so the reference is likely more appropriate at the end of the sentence. The authors should describe the foehn wind in meteorological terms, whereby air which is forced to rise over the mountains, loses much of its water vapor to condensation on the windward side, and subsequently warms dry adiabatically as it descends on the lee side.

Great suggestion.

We have added the introduction in P7L5-7: “Foehn wind conditions featuring warm and dry air subsiding from the mountains that are to the north and west of the NCP (Chen and Lu, 2016) also lead to high ozone pollution in the NCP.”

Pg 5 Line 22,23: The phrase “winds blow from the mountains to the north and west” is confusing. Either the mountains are to the north and west of NCP and the wind blows from the mountains to the NCP, or the wind blows to the north and west, from the mountains to NCP. Possibly this level of detail could be included in the meteorological definition for the foehn wind which would simplify this sentence, or the use of latitude and longitude for the mountain range versus the region defined for the NCP.

Revised. Please see our response to last comment.

Pg 6 Line 3: Are the MEE sites in the NCP average all full time series or are any partial records during the period? It would be good to state that like you did for Figure 2.

We have revised in P7L17: “...all MEE sites in the NCP and including sites with partial records.”

Pg 6 Line 6: Can the authors quantify the “much less month-to-month variability” (e.g., possibly through the standard deviation)?

Thanks! We have added this information in P7L21-22: “The standard deviation decreases from 8.8 ppb to 5.3 ppb after removal of meteorological influence.”.

Pg 6 Line 9,15: Provide a reference that NO₂ is a proxy for NO_x emissions. Could instead include this idea and a reference in the introduction (Pg 2).

Added. We also revised the text in P2L26-28: “bring in satellite and ground-based observations to relate the most recent ozone trends to those of VOC (Shen et al., 2019b) and NO_x emissions (Zheng et al., 2018; Shah et al., 2020).”

Pg 6 Line 14-15: The authors quote decreases in PM_{2.5} and NO_x emissions for 2017-2019 but the base year or period is not provided.

Base year is 2017. We have revised in P8L2: “Relative to 2017, we find for 2019 a 15% ...”

Pg 6 Line 28: add “and” between “Province and Northeast”
Done.

Pg 7 Line 5-6: How do these values compare to the Chinese and US National Ambient Air Quality Standards? Good to put this in perspective of the health standards.

We have added in P8L23-24: “In comparison, the Chinese air quality standard for annual maximum MDA8 ozone is 82 ppb.”

Pg 7 Line 9: Change “warning” to “warming”
Changed.

Response to Review #2

This study quantifies the most recent trends in summertime O₃ 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 O₃ trend. The meteorologically driven trend is quantified by fitting O₃ 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_{2.5} (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 O₃ trend is caused by decrease in PM_{2.5} or reduction in NO_x 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_{2.5}) 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_{2.5} (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_{2.5}...”.

5. In your regression analysis to determine the O₃ 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).”

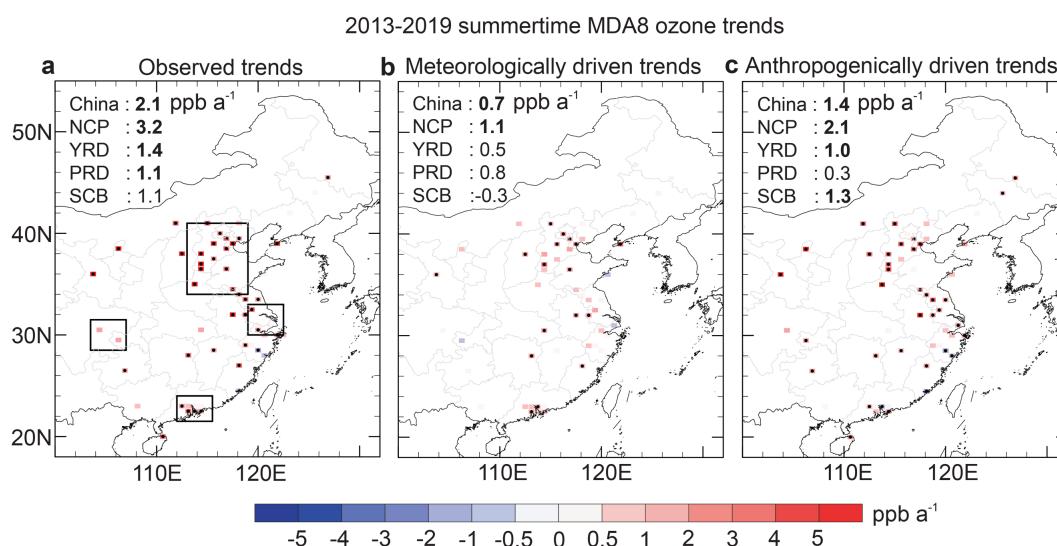


Figure S5. Same with Figure 2 but for the sites with continuous records from 2013.

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.

2013–2019 increases of surface ozone pollution in China: anthropogenic and meteorological influences

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Abstract. Surface ozone data from the Chinese Ministry of Ecology and Environment (MEE) network show sustained increases across the country over the 2013–2019 period. Despite Phase 2 of Clean Air Action targeting ozone pollution, ozone was higher in 2018–2019 than in previous years. The mean summer 2013–2019 trend of maximum 8-h average (MDA8) ozone was 1.9 ppb a⁻¹ ($p<0.01$) across China and 3.3 ppb a⁻¹ ($p<0.01$) in the North China Plain (NCP). Fitting ozone to meteorological variables with a multiple linear regression model shows that meteorology played a significant but not dominant role in the 2013–2019 ozone trend, contributing 0.70 ppb a⁻¹ ($p<0.01$) across China and 1.4 ppb a⁻¹ ($p=0.02$) in the NCP. HigherRising June-July temperatures over the NCP were the main meteorological driver, particularly in recent years (2017–2019), and were associated with increased foehn winds. NCP data for 2017–2019 show a 15% ~~continuing~~ decrease in fine particulate matter (PM_{2.5}) that may be driving the continued anthropogenic increase in ozone, and flat emissions of volatile organic compounds (VOCs), ~~which would explain the continued anthropogenic increase in ozone~~. VOC emission ~~controls~~reductions, as targeted by Phase 2 of the Chinese Clean Air Action, are needed to reverse the increase of ozone.

1 Introduction

Surface ozone is a serious air pollution issue over much of eastern China (Ma et al., 2012; Fu et al., 2019). Measurements from the Chinese Ministry of Environment and Ecology (MEE) network of sites frequently exceed the national air quality standard of $160 \mu\text{g m}^{-3}$, corresponding to 82 ppb at 298 K and 1013 hPa (Li et al., 2017; Shen et al., 2019a; Fan et al., 2020). The Clean Air Action initiated in 2013 imposed rapid decreases in pollutant emissions (Chinese State Council, 2013) and resulted in large decreases in fine particulate matter ($\text{PM}_{2.5}$) concentrations (Zhai et al., 2019; Q. Zhang et al., 2019). However, ozone increased by $1\text{--}3 \text{ ppb a}^{-1}$ over the 2013–2017 period in megacity clusters of eastern China (Lu et al., 2018; Li et al., 2019a; Lu et al. 2020), partly offsetting the health benefits from improved $\text{PM}_{2.5}$ (Dang and Liao, 2019; Q. Zhang et al., 2019). Phase 2 of Clean Air Action starting in 2018 (Chinese State Council, 2018) imposed new emission controls targeted at ozone. Here we show that the increasing ozone trend in eastern China has continued through 2019, driven by both anthropogenic emission and meteorological trends, and stressing the urgent need for more vigorous emission controls.

Ozone in polluted regions is produced by photochemical reactions of volatile organic compounds (VOCs) and nitrogen oxides ($\text{NO}_x \equiv \text{NO} + \text{NO}_2$), enabled by hydrogen oxide radicals ($\text{HO}_x \equiv \text{OH} + \text{peroxy radicals}$) as oxidants. VOCs and NO_x are emitted by fuel combustion, and VOCs have additional industrial sources (Zheng et al., 2018) and biogenic sources (Guenther et al., 2012). HO_x is produced photochemically from ozone and water, formaldehyde (HCHO), nitrous acid, and other precursors (Tan et al., 2019). Ozone is highest in summer when photochemistry is most active (Wang et al., 2017). Meteorological conditions play an important role in modulating ozone concentrations, not only through transport but also by affecting natural emissions and chemical rates (Jacob and Winner, 2009; Shen et al., 2016; Fu et al., 2019; Lu et al., 2019).

A number of studies have investigated the roles of anthropogenic and meteorological factors in driving the 2013–2017 ozone trend, and concluded that meteorological factors were not negligible but anthropogenic factors were dominant (Ding et al., 2019; Li et al., 2019a; Liu et al., 2019; Yu et al., 2019; Liu et al., 2020). Our previous work (Li et al., 2019a, 2019b) found that the decrease of $\text{PM}_{2.5}$ was a major factor driving the increase of ozone due to the role of $\text{PM}_{2.5}$ as scavenger of hydroperoxy (HO_2) radicals and NO_x that would otherwise produce ozone. Here we extend the analysis of ozone trends to 2019, into the implementation of Clean Air Action Phase 2, and bring in satellite and ground-based observations to relate the most recent ozone trends to those of VOC (Shen et al., 2019b) and NO_x emissions (Zheng et al., 2018; Shah et al., 2020).

2 Data and methods

2.1 Surface measurements

Hourly concentrations of ozone, PM_{2.5}, and NO₂ are taken from the MEE website (<http://106.37.208.233:20035>) and archived at <http://beijingair.sinaapp.com>. The network was launched in 2013 as part of the Clean Air Action. It included 450 monitoring stations in 2013, growing to ~1500 stations by 2019. We compute maximum daily 8-h average (MDA8) ozone and 24-h average PM_{2.5} and NO₂ concentrations from the hourly data for June–July–August (JJA). Concentrations were reported by the MEE in units of $\mu\text{g m}^{-3}$ under standard conditions (273 K, 1013 hPa) until 31 August 2018. This reference state was changed on 1 September 2018 to (298 K, 1013 hPa) for gases and local ambient state for PM_{2.5} (MEE, 2018). We converted ozone and NO₂ concentrations to ppb, and rescaled post-August 2018 PM_{2.5} concentrations to standard conditions by assuming (298 K, 1013 hPa) as the local ambient state.

2.2 Satellite observations

We use observations of NO₂ and formaldehyde (HCHO) columns from the OMI and TROPOMI satellite instruments to track recent changes in anthropogenic emissions of NO_x and VOCs, respectively. Shen et al. (2019b) and Shah et al. (2020) previously found that OMI-derived trends of VOC and NO_x emissions were consistent with 2013–2017 bottom-up estimates from the Multi-resolution Emission Inventory for China (MEIC; Zheng et al., 2018). Here we extend the analysis using 2013–2019 OMI data from the European Quality Assurance for Essential Climate Variables project for NO₂ (Boersma et al., 2018) and HCHO (De Smedt et al., 2015). We further use TROPOMI HCHO data available for the summers of 2018–2019 for NO₂ (van Geffen et al., 2018) and HCHO (De Smedt et al., 2018). We do not use TROPOMI NO₂ data because of a version change in March 2019 from v1.2.0 to v1.3.0 that could bias the trend between the summers of 2018 and 2019 (<https://sentinel.esa.int/documents/247904/2474726/Sentinel-5P-Level-2-Product-UserManual-Nitrogen-Dioxide>, last access: 20 July 2020). The TROPOMI HCHO data are freely accessed from <https://s5phub.copernicus.eu/dhus/> (last access: 28 February 2020) and we only use observations with quality assurance value larger than 0.75 for NO₂ and larger than 0.5 for HCHO. These filters. This filter effectively removes data with cloud fraction larger than 0.5. Interannual trends in HCHO columns could be affected by temperature-dependent emissions of biogenic VOCs (Palmer et al., 2006). Following Zhu et al. (2017), we remove this contribution by regressing June–July–August (JJA) monthly mean HCHO columns onto noon (13:00 local time) surface air temperatures, and then subtracting this fitted temperature dependency.

2.3 Stepwise multiple linear regression (MLR) model

To quantify the role of meteorology in driving 2013–2019 ozone trends, we use the same stepwise multiple linear regression (MLR) modeling approach as Li et al. (2019a). This modeling approach relates the month-to-month variability of MDA8 ozone to that of meteorological variables. Consistent meteorological fields for 2013–2019 were obtained from the NASA Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA-2) product (https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2, last access: 28 February 2020) (Gelaro et al., 2017). The MERRA-2 data have a spatial resolution of 0.5° latitude \times 0.625° longitude. We average the daily MDA8 ozone from the MEE network onto the MERRA-2 grid. ~~The~~Firstly, the regression model is ~~first~~ applied to select the key meteorological parameters driving the day-to-day variability of ozone for each grid cell. ~~To~~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~~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. 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).

Finally, the trend in regressed ozone is taken to reflect the meteorological contribution, and the residual is then taken to reflect the ~~anthropogenic contribution (Li et al., 2019a; Zhai et al., 2019)~~presumed anthropogenic contribution, with the statistical significance of the anthropogenic trend determined by Student's t -test. We have followed this approach before to isolate the anthropogenic trends of ozone and $PM_{2.5}$ (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). The effect of biogenic VOCs on ozone

trends depends on meteorological and land cover drivers. Meteorological drivers, in particular temperature, would be accounted for in the MLR model. The effect of land cover changes is expected to be small over the 7-year time horizon of our analysis (Fu and Tai, 2014)

3 Results and discussion

5 We first present the general 2013–2019 summer ozone trends in China and their statistically decomposed meteorological and anthropogenic contributions. Ozone trends over the major megacity clusters in China are highlighted. We go on to more specifically attribute the meteorological and anthropogenic drivers of recent ozone trends over the North China Plain, where the ozone increase is the highest.

3.1 2013–2019 ozone trends: anthropogenic and meteorological contributions

10 **Figure 1** shows 2013–2019 trends of summer maximum ~~MDA8 ozone, summer and~~ mean MDA8 ozone, and ~~summer mean~~ PM_{2.5} from the MEE network. The Clean Air Action has dramatically improved PM_{2.5} pollution since 2013, with ~50% decrease of mean summertime mean PM_{2.5} concentrations across eastern China over the 2013–2019 period. Maximum PM_{2.5} concentrations experienced a similar decrease trend. In contrast, ozone has been steadily increasing over 2013–2019 and concentrations in 2019 are the highest in the record. The Clean Air Action focused specific attention on

15 the four megacity clusters identified by rectangles: North China Plain (NCP; 34°–41°N, 113°–119°E), Yangtze River Delta (YRD, 30°–33°N, 119°–122°E), Pearl River Delta (PRD, 21.5°–24°N, 112°–115.5°E), and Sichuan Basin (SCB, 28.5°–31.5°N, 103.5°–107°E). Mean MDA8 ozone in summer 2019 averaged 83 ppb across the North China Plain (NCP) sites and maximum MDA8 ozone averaged 129 ppb. Summer mean MDA8 ozone in 2019 was lower for the other megacity clusters (67 ppb for Yangtze River Delta (YRD), 46 ppb for Pearl River Delta (PRD), and 57 ppb for Sichuan Basin (SCB)) but summer maximum MDA8 ozone values were comparable to the NCP. These three megacity clusters are subject to similar ozone pollution episodes under stagnant conditions as the NCP (Wang et al., 2017), but they are more frequently ventilated by the summer monsoon bringing cleaner tropical air and precipitation hence the lower mean ozone.

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Figure 2 (left panel) 2a shows the 2013–2019 trends in summer mean MDA8 ozone obtained by ordinary linear regression of the data averaged over the $0.5^\circ \times 0.625^\circ$ MERRA-2 grid. Ozone increases almost everywhere in China. Decreases are largely restricted to the Shandong Peninsula and Northeast/northeastern China- (including Heilongjiang, Jilin, and Liaoning provinces). The mean trend for China is 1.9 ppb a^{-1} ($p < 0.01$). Trends in the four megacity clusters are 3.3 ppb a^{-1} ($p < 0.01$) for NCP, 1.6 ppb a^{-1} ($p < 0.01$) for YRD, 1.1 ppb a^{-1} ($p = 0.03$) for PRD, and 0.7 ppb a^{-1} ($p = 0.23$) for

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SCB (Table S1). The increases are largest in the NCP, ~~where the effects which could be explained by greater influence~~ of radical scavenging by PM_{2.5}. ~~would be largest~~ (Li et al., 2019a, 2019b).

Figure 2 (middle panel) 2b shows the meteorologically driven ozone trends, as determined by fitting ozone to meteorological variables with the MLR model. We find an average meteorologically driven trend of 0.7 ppb a⁻¹ ($p < 0.01$) for China. Ozone trends over 2013–2019 in the NCP and PRD are significantly ~~enhanced~~ contributed by meteorology, and this is particularly driven by 2018–2019 (Table S1). Similar to our previous study for 2013–2017 (Li et al., 2019a), the most important meteorological predictor variables in the MLR model are daily maximum temperature for the NCP and meridional wind at 850 hPa for the PRD. ~~(Figure S1)~~. These dominant meteorological parameters are also consistent with the findings from other studies (Gong and Liao, 2019; Wang et al., 2019; Han et al., 2020). Hot weather is the main meteorological driver for high ozone in the NCP, and we will elaborate on this in the next section. The main meteorological driver for the ozone increase in the PRD is the ~~weakened~~ weakening of the summer monsoonal ~~southwesterlies (i.e., increased northeasterlies, flow~~ (Figure S1) ~~that ventilate~~ ventilates the PRD with marine air.

On the other hand, we find that meteorology mitigated ozone pollution increases over ~~western China,~~ northeastern China, and the Shandong Peninsula. ~~Summer~~ As shown in Figure S2, summer ozone in the Shandong Peninsula is strongly affected by maritime inflow (J. Zhang et al, 2019; Han et al., 2020) which increased over the 2013–2019 period (**Figure S1)–3**). Temperature decreased over northeastern China (Figure 3).

Removing the meteorological contribution in the ozone trend leaves a residual trend that we interpret as anthropogenic (**Figure 2, right panel) 2c**), following Li et al. (2019a) and Zhai et al. (2019). This anthropogenic trend is more uniformly positive at a national scale than the observed and meteorologically driven trends. It averages 1.2 ppb a⁻¹ ($p < 0.01$) for all of China, as compared to 0.7 ppb a⁻¹ ($p < 0.01$) for the meteorologically driven trend. The observed 2013–2019 ozone increase in all the megacity clusters except the PRD is dominated by the anthropogenic contribution, averaging 1.9 ppb a⁻¹ ($p < 0.01$) for the NCP. This result still stands if only continuous records since 2013 are used in the analysis (Figure S5). The ozone increase in PRD is mainly meteorologically driven due to reduced monsoonal winds (**Figure 3**). The following sections present further analysis of the 2013–2019 ozone ~~trend~~ trend in the NCP, where both meteorological and anthropogenic contributions are particularly large.

3.2 Meteorologically driven 2013–2019 ozone increase in the North China Plain

Separating the observed 2013–2019 ozone trends by month (**Figure 3**) shows that the seasonal JJA trend of 3.3 ppb a⁻¹ ($p < 0.01$) over the NCP is driven by June and July. Observed trends are 5.5 ppb a⁻¹ ($p < 0.01$) for June, 3.7 ppb a⁻¹ ($p < 0.01$) for July, and 0.9 ppb a⁻¹ (~~statistically insignificant~~ $p = 0.34$) for August. This month-to-month difference is mainly driven

by meteorology. As derived from the MLR model, the meteorologically driven ozone trend of 1.4 ppb a^{-1} ($p=0.02$) for JJA breaks down to 3.1 ppb a^{-1} ($p<0.01$) for June, 2.2 ppb a^{-1} ($p=0.08$) for July, and -1.0 ppb a^{-1} ($p=0.16$) for August. The residual anthropogenic trend is much more similar across months (2.4 ppb a^{-1} ($p=0.02$) in June, 1.5 ppb a^{-1} ($p=0.07$) in July, 1.9 ppb a^{-1} ($p<0.01$) in August), as would be expected after removing meteorological influence.

Figure 34 shows the monthly mean time series of daily maximum temperature averaged over the NCP for 1980–2019, with 2013–2019 highlighted in shading. Temperature is the principal driver of the meteorologically driven ozone trend as indicated by the MLR model. We find a large 2013–2019 increase in temperature in June, ($0.42 \text{ }^{\circ}\text{C a}^{-1}$), a lesser increase in July, ($0.22 \text{ }^{\circ}\text{C a}^{-1}$), and a decrease in August, ($-0.18 \text{ }^{\circ}\text{C a}^{-1}$), reflected in the meteorologically driven ozone trend for each month. When placed in the context of the 1980–2019 record, we see that the 2013–2019 temperature trends reflect interannual climate variability rather than a long-term warming trend.

Hot weather in the NCP in the summer is generally driven by large-scale anticyclonic conditions, and this has been viewed as the principal predictor of ozone pollution days (Gong and Liao, 2019). But foehn winds (Chen and Lu, 2016) are also important in June and to a lesser extent in July. Foehn winds blow wind conditions featuring warm and dry air subsiding from the mountains that are to the north and west, bringing warm of the NCP (Chen and Lu, 2016) also lead to high ozone pollution in the NCP. By categorizing the 2013–2019 June circulation patterns between foehn-favorable and no foehn conditions on the basis of the V850 foehn index (Foehn winds are most important in June. Following Chen and Lu, (2016), we find that diagnosed foehn-favorable conditions are to a large extent in the NCP with a foehn index defined by the 850 hPa northwesterly wind averaged along a section from (42°N , 108°E) to (38°N , 112°E) (Figure 5). The days with positive (negative) foehn index are taken as foehn (no-foehn) condition. We find that foehn conditions are largely responsible for the 2013–2019 increase in temperature in June (Figure 54). The frequency of foehn conditions under hot days in June increased by 85% over the 2013–2019 period (driven mainly by the increased frequency in 2018–2019), highlighting and ozone increase under foehn conditions is 1.2 ppb a^{-1} larger than under no-foehn conditions. Our result highlights the previously unrecognized effect of foehn winds on ozone pollution in the NCP.

3.3 Anthropogenically driven 2013–2019 ozone increase in the North China Plain

Figure 5a6a shows the observed time series of monthly mean JJA MDA8 ozone anomalies for 2013–2019 relative to the JJA 2013–2019 mean, averaged over all MEE sites in the NCP and including sites with partial records. We see large month-to-month variability superimposed on the long-term trend. Much of this month-to-month variability can be attributed to meteorological factors using the MLR model (blue line), as discussed in the previous section. The residual anthropogenic trend (red line) shows a 2013–2019 increasing trend with much less month-to-month variability than the

original observed time series. [The standard deviation decreases from 8.8 ppb to 5.3 ppb after removal of meteorological influence.](#)

Figure 5b6b shows the 2013–2019 observed trends of different quantities relevant to the anthropogenic ozone trend over the NCP: PM_{2.5} and NO₂ from the MEE network, and NO₂ and HCHO tropospheric columns from satellites. PM_{2.5} shows a steady decrease, 49% over the 2013–2019 period. NO₂ (a proxy for NO_x emissions; [Zheng et al., 2018](#)) shows a 25–30% decrease with some interannual variability that is consistent between the OMI satellite data and the surface MEE network. HCHO (a proxy ~~offor~~ for VOC emissions) shows no significant trend for the 2013–2019 period, with some interannual variability that could reflect noise in the measurement (Shen et al., 2019b).

Of particular interest are the trends for 2017–2019, extending beyond the currently available MEIC emission inventory (Zheng et al., 2018) and during which we find continued increase of ozone. [WeRelative to 2017, we](#) find for 2017–2019 a 15% decrease in PM_{2.5}, a 6–10% decrease in NO_x emissions (depending on which proxy record we use), and flat VOC emissions. Phase 2 of the Chinese government’s Clean Air Action (China State Council, 2018) called for a 18% decrease in PM_{2.5} over 2015–2020, a 15% decrease in NO_x emissions, and a 10% decrease in VOC emissions. Taking into account the already-achieved 2015–2017 gains in PM_{2.5} and NO_x emissions, Li et al. (2019b) inferred that those targets would require 2017–2020 decreases of 8% for PM_{2.5}, 9% for NO_x emissions, and 10% for VOCs emissions. They found from model simulations that the decrease in PM_{2.5} would cause further increase in ozone, but that decreasing VOC emissions would compensate and enable net improvement, with NO_x emission changes having relatively little effect. We find here that the observed 2017–2019 decrease in PM_{2.5} goes beyond the Clean Air Action target, while the satellite HCHO data show no evidence of a decrease in VOC emissions. Combination of these two effects is consistent with the observed anthropogenically driven increase in ozone over 2017–2019. Decrease of VOC emissions is the key to reverse the ozone increase (Li et al., 2019b).

4 Conclusions

Surface ozone data from the Chinese Ministry of Environment and Ecology (MEE) network show a sustained nationwide increase over the 2013–2019 period, with a few exceptions (Shandong Province, [Northeast and northeastern](#) China), and with particularly high concentrations in 2018–2019. Correction for ~~meteorological~~[meteorologically driven](#) trends with a multiple linear regression (MLR) model shows a general pattern of anthropogenically driven ozone increase across China, though meteorological influences are also significant. The mean summer (JJA) 2013–2019 increase in maximum daily 8-hour average (MDA8) ozone over China is 1.9 ppb a⁻¹, [\(p<0.01\)](#), including 0.7 ppb a⁻¹ [\(p<0.01\)](#) from ~~meteorological~~[meteorologically driven](#) trends (mostly temperature and circulation) and 1.2 ppb a⁻¹ [\(p<0.01\)](#) from

anthropogenic influence.- Ozone concentrations are highest in the North China Plain (NCP), where the summer mean MDA8 ozone averaged across sites was 83 ppb in 2019 and the summer maximum MDA8 ozone averaged across sites was 129 ppb. In comparison, the Chinese air quality standard for annual maximum MDA8 ozone is 82 ppb. Mean summer MDA8 ozone increased by 3.3 ppb a⁻¹ (p<0.01) in the NCP over the 2013–2019 period, which we attribute as 1.4 ppb a⁻¹ meteorological(p=0.02) meteorologically driven and 1.9 ppb a⁻¹ anthropogenic(p<0.01) anthropogenically driven.

Further investigation of the NCP trends shows that hot weather in June–July 2018–2019 was a major driver for the high ozone concentrations in those summers. Such hot weather does not relate to long-term warmingwarming but to interannual variability driven principally by foehn northwesterly winds. Removing this meteorological variability shows a sustained anthropogenic ozone increase over the NCP over the 2013–2019 record and persisting into 2018–2019.

Examination of ozone-relevant anthropogenic variables from the MEE network and from satellites shows a 49% decrease in PM_{2.5} for 2013–2019 (15% for 2017–2019), a 25–30% decrease in NO_x emissions for 2013–2019 (6–10% for 2017–2019) and flat VOC emissions. The sustained anthropogenic increase in ozone over the 2017–2019 period canmay be explained by the continued decrease of PM_{2.5}, which scavenges the radical precursors of ozone, combined with flat emissions of VOCs. Reducing VOC emissions should be the top priority for reversing the increase of ozone in the NCP and in other urban areas of China.

Data availability. Hourly surface concentrations of air pollutants are archived at <http://beijingair.sinaapp.com> (last access: 30 June 2020). The MERRA-2 reanalysis data are from <https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2> (last access: 28 February 2020). The L3 OMI satellite data for NO₂ and HCHO are available at <http://www.qa4ecv.eu/ecvsv> (last access: 28 February 2020). The L2 TROPOMI data for NO₂ and HCHO are freely available at <https://s5phub.copernicus.eu/dhus> (last access: 28 February 2020). The data used in this study can be accessed via doi (<https://doi.org/10.7910/DVN/T6D7YY>).

Author contributions. KL and DJJ designed the study. KL performed the analysis. LS and IDS provided the TROPOMI data. XL and HL contributed to the interpretation of the results. KL and DJJ wrote the paper with contributions from all co-authors

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. This work is a contribution from the Harvard-NUIST Joint Laboratory for Air Quality and Climate. HL is supported by the National Natural Science Foundation of China (91744311). We appreciate the efforts from the China Ministry of Ecology and Environment for supporting the nationwide observation network and publishing hourly

air pollutant concentrations. We acknowledge the QA4ECV project for the NO₂ and HCHO data. We appreciate the efforts from NASA GMAO for providing the MERRA-2 reanalysis data.

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Table and Figure captions

Table 1. MDA8 ozone trends in China (ppb a⁻¹), 2013–2019 and 2013–2017.

Figure 1. Summer (JJA) concentrations of maximum MDA8 ozone (~~top~~–~~summera~~), mean MDA8 ozone (~~middleb~~), maximum PM_{2.5} (c), and ~~summer~~ mean PM_{2.5} (~~bottomd~~) for 2013–2019 at the network operated by the China Ministry of Ecology and Environment (MEE). Rectangles denote the four megacity clusters discussed in the text: North China Plain

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(NCP; 34°–41°N, 113°–119°E), Yangtze River Delta (YRD, 30°–33°N, 119°–122°E), Pearl River Delta (PRD, 21.5°–24°N, 112°–115.5°E), and Sichuan Basin (SCB, 28.5°–31.5°N, 103.5°–107°E).

Figure 2. Summertime ozone trends in China, 2013–2019. The left panel (a) shows observed trends of summer mean MDA8 ozone at MEE sites averaged on the $0.5^\circ \times 0.625^\circ$ ($\approx 50 \times 50$ km²) MERRA-2 grid. The trends are obtained by ordinary linear regression and include sites with partial records. The middle panel (b) shows meteorologically driven trends determined by fitting ozone to meteorological covariates in the multiple linear regression (MLR) model. The right panel (c) shows anthropogenic trends as inferred from the residual of the MLR model. Statistically significant trends above the 90% confidence level are marked with black dots. The mean trends for all of China and for the four megacity clusters (rectangles) are inset, where the regression is applied to the spatially averaged MDA8 ozone for the cluster. Numbers in bold are statistically significant above the 90% confidence level- (Table 1).

~~Figure 3. Time series of June–August~~**Figure 3.** Summer mean trends of 850 hPa wind vectors ($\text{m s}^{-1} \text{ a}^{-1}$) and surface daily maximum temperature ($^\circ\text{C a}^{-1}$, shaded) over the period 2013–2019. Data are from the MERRA-2 reanalysis. The trends are obtained by ordinary linear regression of mean JJA data for individual years.

~~Figure 4. Time series of JJA~~**Figure 4.** Time series of JJA daily maximum surface air temperatures over the North China Plain (NCP) for 1980–2019. Values are monthly means from the MERRA-2 reanalysis. The 2013–2019 period for the ozone trend analysis is shaded in grey. ~~The observed (OBS), meteorologically driven (MET), and anthropogenically driven (ANTH) monthly ozone trends (ppb a^{-1}) in the NCP for 2013–2019 are shown in the table to the right, where numbers in bold are statistically significant above the 90% confidence level.~~

~~Figure 45.~~**Figure 45.** June mean trends in meteorological variables over 2013–2019 under foehn-favorable (top) and non-foehn (bottom) conditions. (a) Trends in 850 hPa winds ($\text{m s}^{-1} \text{ a}^{-1}$) and surface daily maximum temperature ($^\circ\text{C a}^{-1}$, shaded) under foehn-favorable conditions; (b) Trends in 500 hPa winds ($\text{m s}^{-1} \text{ a}^{-1}$) and surface relative humidity ($\% \text{ a}^{-1}$, shaded) under foehn-favorable conditions; (c, d) are the same as (a, b) but for non-foehn conditions. Data are from the MERRA-2 re-analysis and trends are obtained by ordinary linear regression. Foehn conditions are diagnosed by a foehn index-~~V850~~ defined by the 850 hPa northwesterly wind averaged along a section from (42°N, 108°E) to (38°N, 112°E) (green line in a). The days with positive (negative) ~~V850~~foehn index are taken as foehn-favorable_ (no-foehn) condition. ~~The frequency of foehn wind under hot days increased by 85% over the period. Data~~conditions. Meteorological data are from the MERRA-2 reanalysis.

Figure 56. Trends in summertime ozone and related anthropogenic drivers in the North China Plain (NCP). The left panel (a) shows time series of monthly mean MDA8 ozone (ppb) anomalies averaged over the MEE sites relative to the 2013–2019 summer (JJA) mean. Values are shown as anomalies for individual JJA months (3 points per year). Observed trends are compared to the meteorologically driven trends diagnosed by the MLR model, and to the residuals determining

the anthropogenically driven trend. The right panel **(b)** shows time series of observed JJA mean quantities averaged over the NCP: PM_{2.5} and NO₂ concentrations from the MEE sites, ~~and~~ tropospheric NO₂ and HCHO column densities from the OMI satellite instrument, and HCHO column ~~densities~~density from the ~~OMI and~~ TROPOMI satellite ~~instruments~~instrument. Values are presented as ratios relative to 2013. The TROPOMI HCHO data for 2018 have been scaled to the OMI data for that year with the multiplicative factor indicated in legend. ~~The low bias for TROPOMI NO₂ is similar with the finding by Lorente et al. (2019).~~

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Table 1. MDA8 ozone trends in China (ppb a⁻¹), 2013–2019 and 2013–2017.

Regions	JJA 2013–2019 trends			JJA 2013–2017 trends		
	Observed ^a	Meteorological ^b	Anthropogenic ^c	Observed	Meteorological	Anthropogenic
China	1.9 (<0.01) ^d	0.7 (<0.01)	1.2 (<0.01)	1.7 (<0.01)	0.4 (0.22)	1.3 (<0.01)
NCP	3.3 (<0.01)	1.4 (0.02)	1.9 (<0.01)	2.7 (0.01)	0.7 (0.43)	2.0 (<0.01)
YRD	1.6 (<0.01)	0.7 (0.12)	0.9 (<0.01)	1.7 (0.03)	0.2 (0.82)	1.5 (<0.01)
PRD	1.1 (0.03)	0.8 (0.07)	0.3 (0.29)	0.6 (0.44)	0.4 (0.65)	0.3 (0.51)
SCB	0.7 (0.23)	-0.2 (0.59)	1.0 (<0.01)	0.9 (0.42)	0.1 (0.90)	0.8 (0.20)

^aObserved trends (OBS) are obtained by ordinary linear regression on summer (JJA) mean values of maximum daily 8-h average (MDA8) ozone measured at the sites of the Ministry of Ecology and Environment (MEE) network. The MDA8 ozone data are first averaged spatially over the 0.5° × 0.625° MERRA-2 grid (Figure 2), and then averaged nationally (China) and over four megacity clusters: North China Plain (NCP), Yangtze River Delta (YRD), Pearl River Delta (PRD), Sichuan Basin (SCB).

^bMeteorologically-driven trends are obtained by fitting the ozone data to a multiple linear regression (MLR) model with the three most important meteorological covariates (see text).

^cThe anthropogenically-driven trends are obtained by ordinary linear regression of the residual ozone after removing the MLR-fitted value.

^dp-values for the trends are in italics; trends in bold are those with p-value smaller than 0.1.

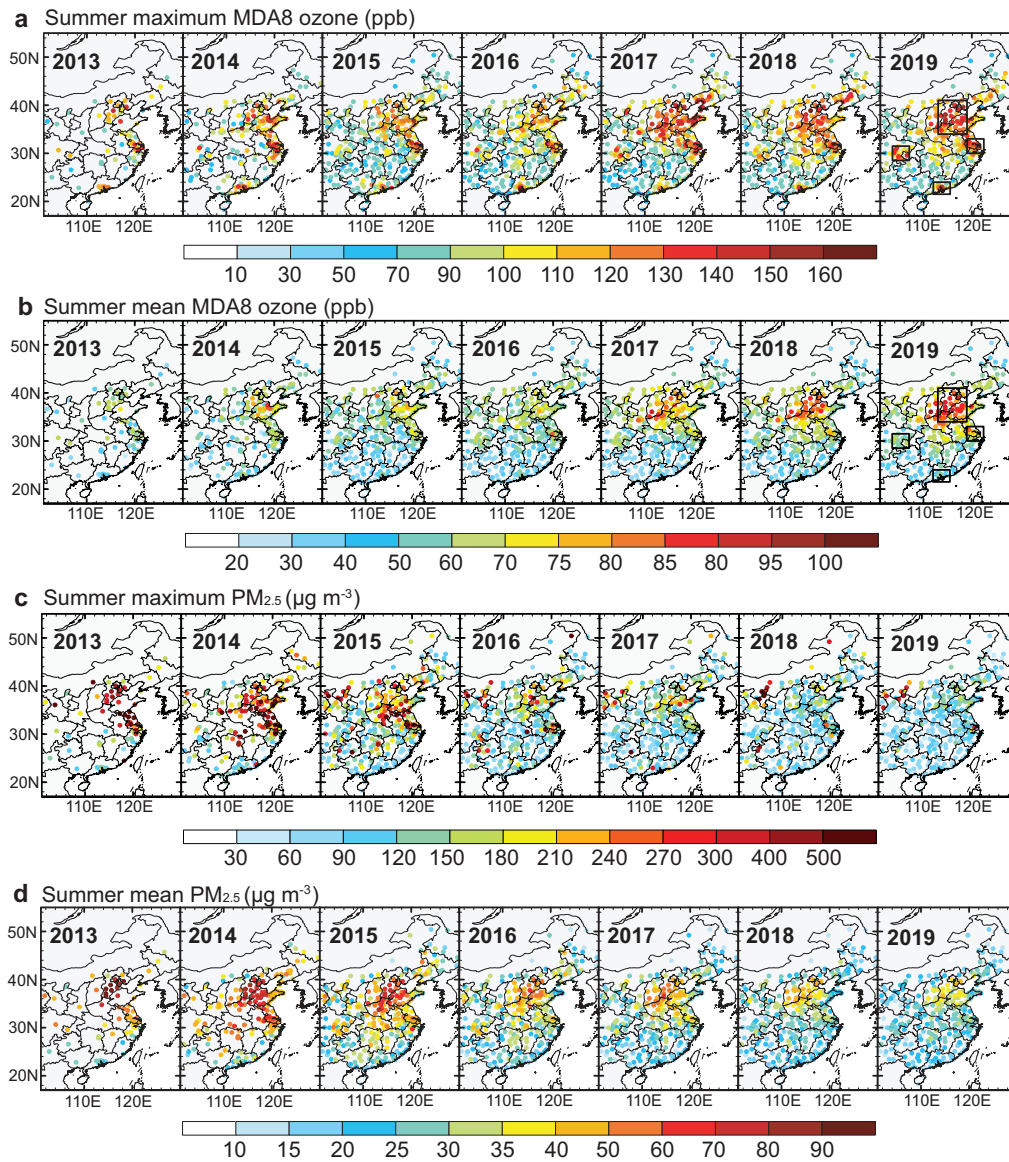


Figure 1. Summer (JJA) concentrations of maximum MDA8 ozone (a), mean MDA8 ozone (b), maximum PM_{2.5} (bottom c), and mean PM_{2.5} (d) for 2013–2019 at the network operated by the China Ministry of Ecology and Environment (MEE). Rectangles denote the four megacity clusters discussed in the text: North China Plain (NCP; 34°–41°N, 113°–119°E), Yangtze River Delta (YRD, 30°–33°N, 119°–122°E), Pearl River Delta (PRD, 21.5°–24°N, 112°–115.5°E), and Sichuan Basin (SCB, 28.5°–31.5°N, 103.5°–107°E).

2013-2019 summertime MDA8 ozone trends

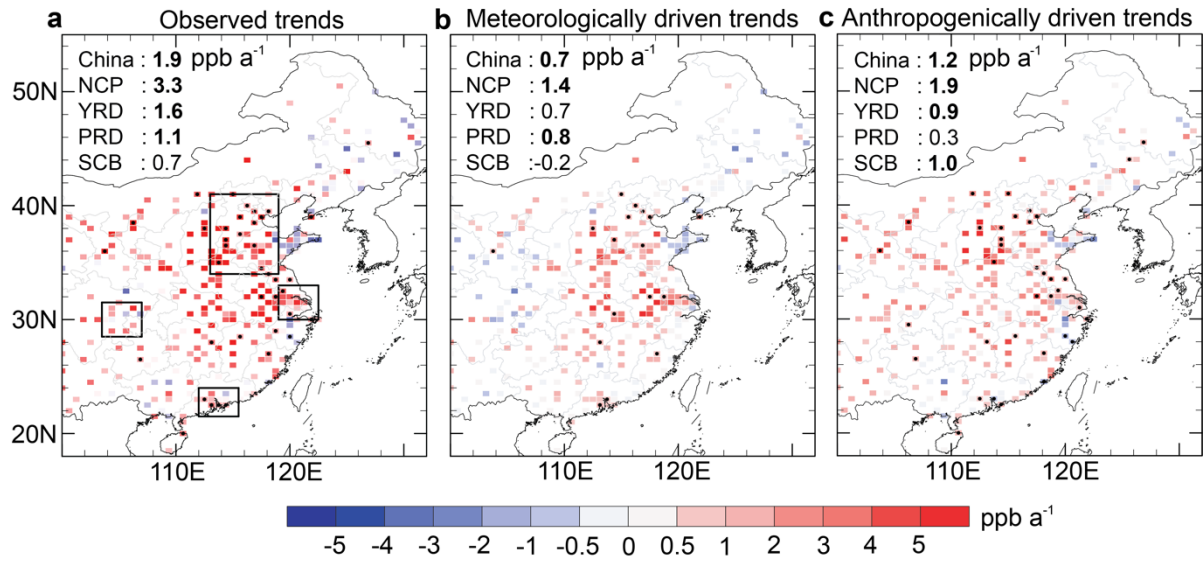


Figure 2. Summertime ozone trends in China, 2013–2019. The left panel (a) shows observed trends of summer mean MDA8 ozone at MEE sites averaged on the $0.5^\circ \times 0.625^\circ$ ($\approx 50 \times 50$ km²) MERRA-2 grid. The trends are obtained by ordinary linear regression and include sites with partial records. The middle panel (b) shows meteorologically driven trends determined by fitting ozone to meteorological covariates in the multiple linear regression (MLR) model. The right panel (c) shows anthropogenic trends as inferred from the residual of the MLR model. Statistically significant trends above the 90% confidence level are marked with black dots. The mean trends for all of China and for the four megacity clusters (rectangles) are inset, where the regression is applied to the spatially averaged MDA8 ozone for the cluster. Numbers in bold are statistically significant above the 90% confidence level- (Table 1).

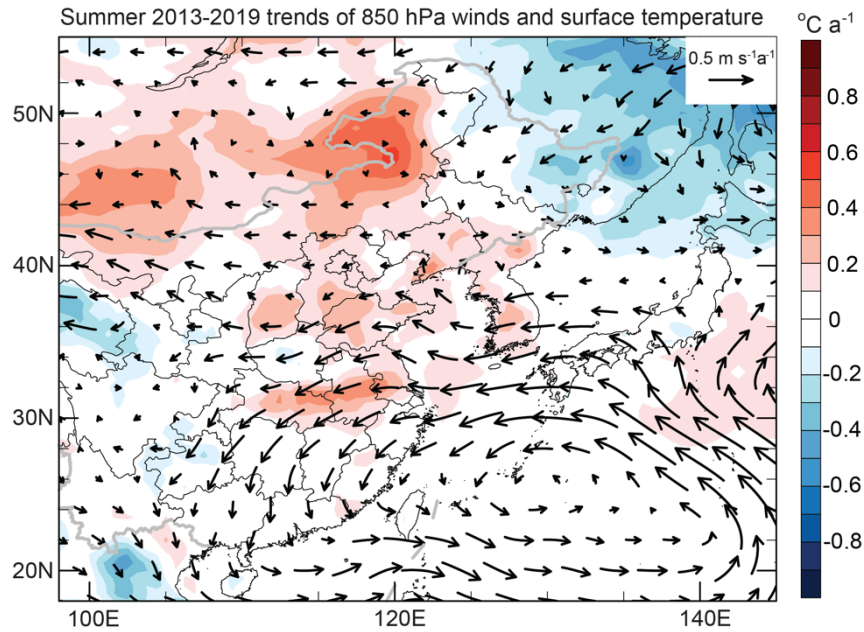


Figure 3. Summer mean trends of 850 hPa wind vectors ($\text{m s}^{-1} \text{a}^{-1}$) and surface daily maximum temperature ($^{\circ}\text{C a}^{-1}$, shaded) over the period 2013–2019. Data are from the MERRA-2 reanalysis. The trends are obtained by ordinary linear regression of mean JJA data for individual years.

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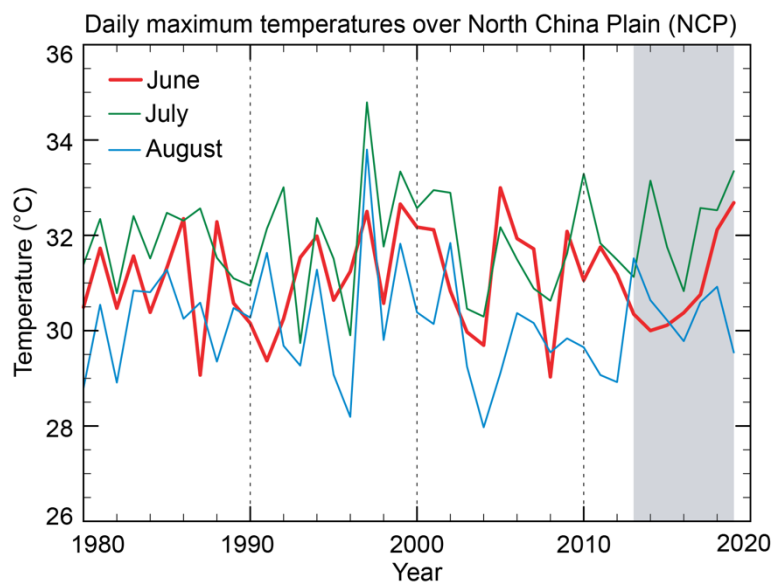


Figure 4. Time series of JJA daily maximum surface air temperatures over the North China Plain (NCP) for 1980–2019. Values are monthly means from the MERRA-2 reanalysis. The 2013–2019 period for the ozone trend analysis is shaded in grey. The observed (OBS), meteorologically driven (MET), and anthropogenically driven (ANTH) monthly ozone trends (ppb a^{-1}) in the NCP for 2013–2019 are shown in the table to the right, where numbers in bold are statistically significant above the 90% confidence level.

June meteorology trends over 2013–2019 under foehn-favorable (top) and non-foehn (bottom) conditions

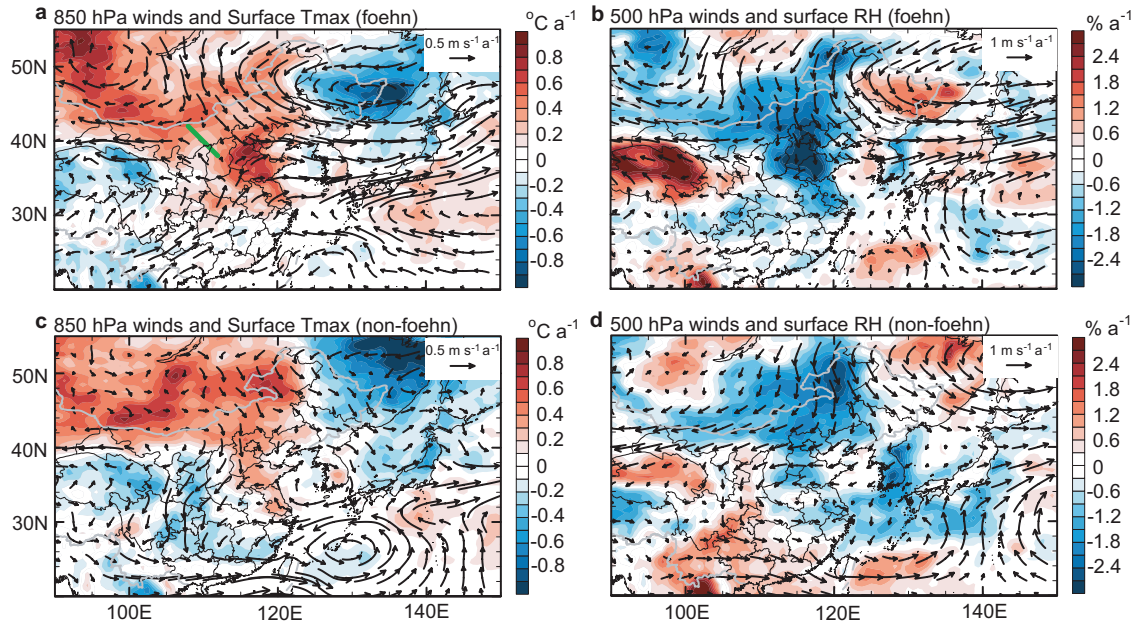


Figure 45. June mean trends in meteorological variables over 2013–2019 under foehn-favorable (top) and non-foehn (bottom) conditions. (a) Trends in 850 hPa winds ($\text{m s}^{-1} \text{a}^{-1}$) and surface daily maximum temperature ($^{\circ}\text{C a}^{-1}$, shaded) under foehn-favorable conditions; (b) Trends in 500 hPa winds ($\text{m s}^{-1} \text{a}^{-1}$) and surface relative humidity ($\% \text{a}^{-1}$, shaded) under foehn-favorable conditions; (c, d) are the same as (a, b) but for non-foehn conditions. Data are from the MERRA-2 re-analysis and trends are obtained by ordinary linear regression. Foehn conditions are diagnosed by a foehn index $\nabla 850$ defined by the 850 hPa northwesterly wind averaged along a section from (42°N, 108°E) to (38°N, 112°E) (green line in a). The days with positive (negative) $\nabla 850$ foehn index are taken as foehn-favorable_ (no-foehn) condition. The frequency of foehn wind under hot days increased by 85% over the period. Data conditions. Meteorological data are from the MERRA-2 reanalysis.

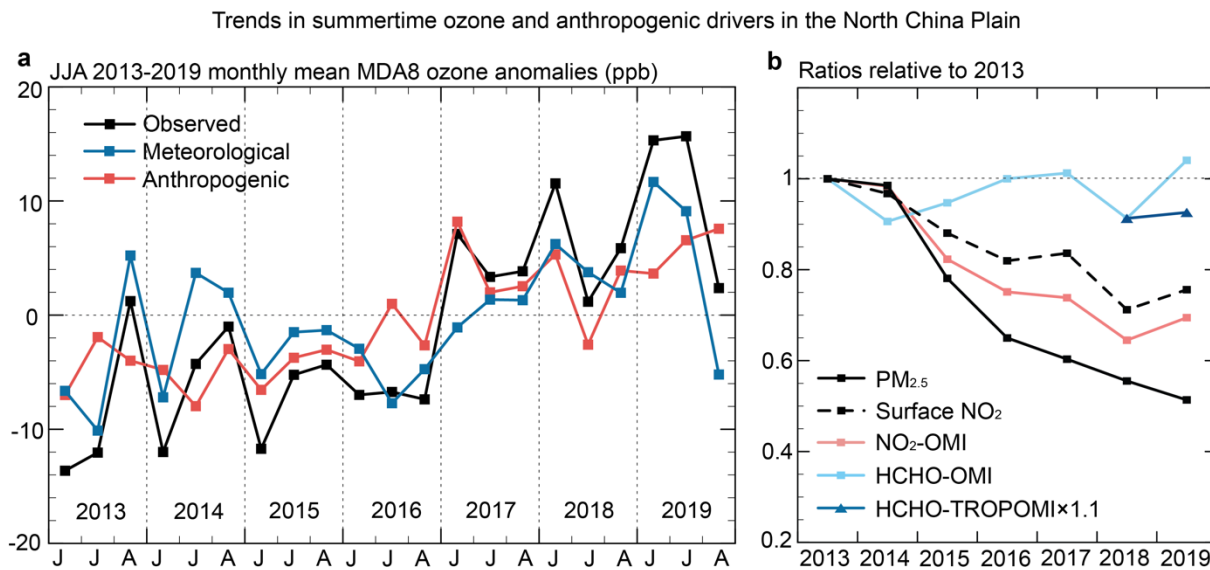


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