



# Anthropogenic drivers of 2013–2017 trends in summer surface ozone in China

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**Observations of surface ozone available from ~1,000 sites across China for the past 5 years (2013–2017) show severe summertime pollution and regionally variable trends. We resolve the effect of meteorological variability on the ozone trends by using a multiple linear regression model. The residual of this regression shows increasing ozone trends of 1–3 ppbv a<sup>-1</sup> in megacity clusters of eastern China that we attribute to changes in anthropogenic emissions. By contrast, ozone decreased in some areas of southern China. Anthropogenic NO<sub>x</sub> emissions in China are estimated to have decreased by 21% during 2013–2017, whereas volatile organic compounds (VOCs) emissions changed little. Decreasing NO<sub>x</sub> would increase ozone under the VOC-limited conditions thought to prevail in urban China while decreasing ozone under rural NO<sub>x</sub>-limited conditions. However, simulations with the Goddard Earth Observing System Chemical Transport Model (GEOS-Chem) indicate that a more important factor for ozone trends in the North China Plain is the ~40% decrease of fine particulate matter (PM<sub>2.5</sub>) over the 2013–2017 period, slowing down the aerosol sink of hydroperoxy (HO<sub>2</sub>) radicals and thus stimulating ozone production.**

surface ozone | China | aerosol chemistry | emission reductions | air quality

Ozone in surface air is a major air pollutant harmful to human health (1) and to terrestrial vegetation (2, 3). Ozone pollution is a serious issue in China (4–8). Summer mean values of the maximum daily 8-h average (MDA8) ozone concentration exceed 60 ppbv over much of eastern China (9, 10), and episodes exceeding 120 ppbv occur frequently in megacities such as Beijing, Shanghai, and Guangzhou (4). Better understanding of the causes of elevated ozone in China is important for developing effective emission control strategies.

Ozone is produced rapidly in polluted air by photochemical oxidation of volatile organic compounds (VOCs) in the presence of nitrogen oxides (NO<sub>x</sub> ≡ NO + NO<sub>2</sub>). VOCs originate from both anthropogenic and biogenic sources. NO<sub>x</sub> is mainly from fuel combustion. Ozone sensitivity to anthropogenic emissions depends on the photochemical regime for ozone formation, i.e., whether ozone production is NO<sub>x</sub>-limited or VOC-limited (11). Observational and modeling studies suggest that ozone production in urban centers is VOC-limited, whereas ozone production in rural regions is NO<sub>x</sub>-limited, with megacity cluster regions in a transitional regime (4, 12).

Several studies have reported increasing ozone trends of 1–2 ppbv a<sup>-1</sup> at urban and background sites in eastern China over the 2001–2015 period (7, 13–15). Surface ozone data were very sparse before 2013. Starting in 2013 the surface monitoring network greatly expanded, and detailed hourly data across all of China became available from the China Ministry of Ecology and Environment. In the same year, the Chinese government launched the Air Pollution Prevention and Control Action Plan to reduce anthropogenic emissions ([www.gov.cn/zwqk/2013-09/12/content\\_2486773.htm](http://www.gov.cn/zwqk/2013-09/12/content_2486773.htm)). Fine particles with an aerodynamic diameter of 2.5 μm or smaller (PM<sub>2.5</sub>) concentration has decreased significantly since then, but ozone pollution has not decreased and is seemingly getting worse (8, 16). NO<sub>x</sub> emissions are estimated to have decreased by more than 20% over 2013–2017 (17), in part to decrease nitrate PM<sub>2.5</sub> (18–20),

but this could have had a counterproductive effect on ozone under VOC-limited conditions. Decreases in PM<sub>2.5</sub> could further affect ozone through changes in aerosol chemistry and photolysis rates (21, 22). On the other hand, meteorological variability could also have a large effect on ozone trends over a 5-y period.

The aim of this work is to better understand the factors controlling ozone trends across China during 2013–2017, separating anthropogenic and meteorological influences, to diagnose the effect of emission reductions even though a 5-y record is relatively short. We focus on the summer season [June–July–August (JJA)] when ozone pollution in eastern China is most severe (4). We use a statistical model to isolate the meteorological contribution to month-to-month variability of ozone and infer a residual trend attributable to anthropogenic emissions. We interpret this residual trend in terms of changing emissions using the Goddard Earth Observing System Chemical Transport Model (GEOS-Chem) driven by 2013–2017 emissions from Multiresolution Emission Inventory for China (MEIC) (17).

## Results and Discussion

**Observed Summer Ozone Air Quality, Meteorologically Driven Variability, and Residual Trend.** Fig. 1 shows the 5-y average (2013–2017) values of the summer mean and maximum MDA8 ozone at the ensemble of sites operated by the China Ministry of Ecology and Environment. The Chinese National Ambient Air Quality Standard for MDA8 ozone is 160 μg m<sup>-3</sup>, corresponding to 82 ppbv at 298 K and

### Significance

**Drastic air pollution control in China since 2013 has achieved sharp decreases in fine particulate matter (PM<sub>2.5</sub>), but ozone pollution has not improved. After removing the effect of meteorological variability, we find that surface ozone has increased in megacity clusters of China, notably Beijing and Shanghai. The increasing trend cannot be simply explained by changes in anthropogenic precursor [NO<sub>x</sub> and volatile organic compound (VOC)] emissions, particularly in North China Plain (NCP). The most important cause of the increasing ozone in NCP appears to be the decrease in PM<sub>2.5</sub>, slowing down the sink of hydroperoxy radicals and thus speeding up ozone production. Decreasing ozone in the future will require a combination of NO<sub>x</sub> and VOC emission controls to overcome the effect of decreasing PM<sub>2.5</sub>.**

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The authors declare no conflict of interest.

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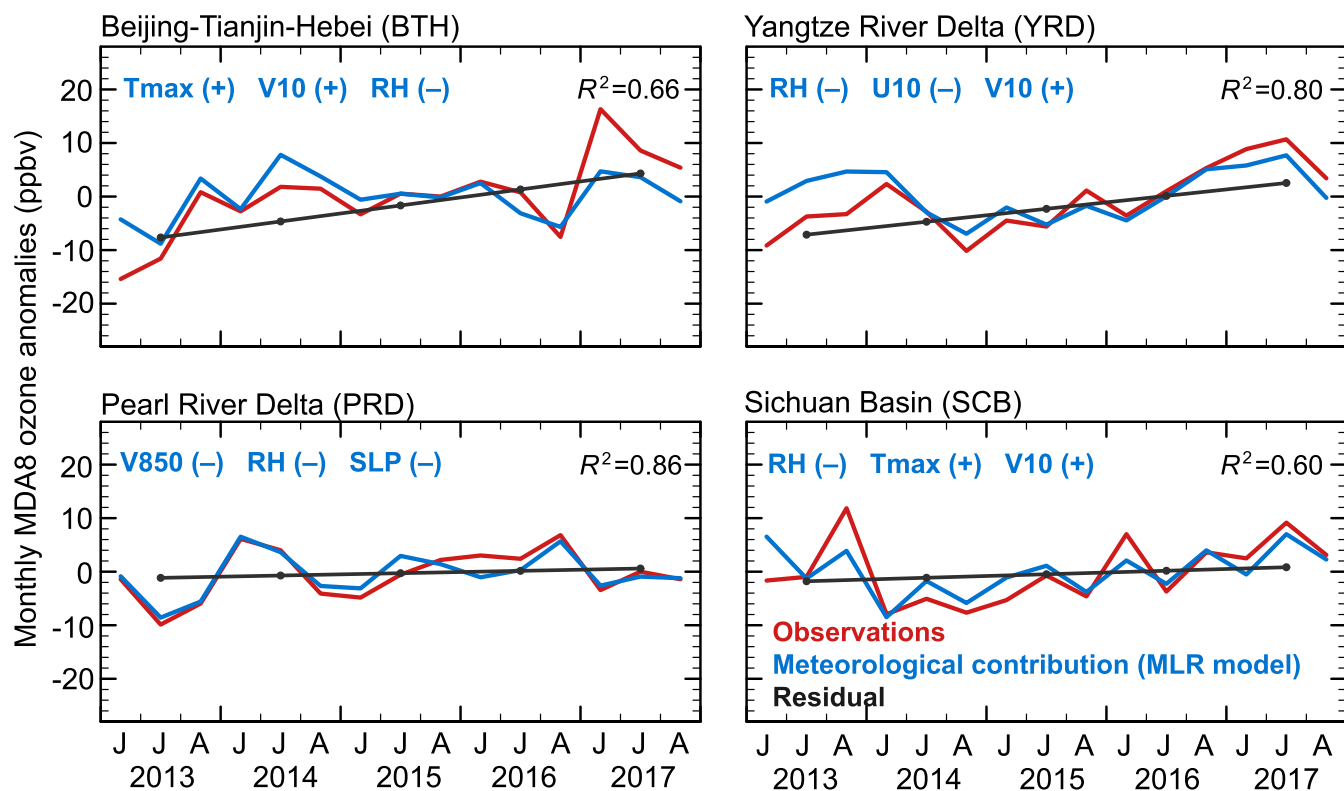
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## 2013–2017 summer ozone trends for the four megacity clusters



**Fig. 2.** Time series of monthly mean MDA8 ozone anomalies in summer (JJA) 2013–2017 for the four megacity clusters of Fig. 1: BTH, YRD, PRD, SCB. MDA8 ozone values for individual  $0.5^\circ \times 0.625^\circ$  grid cells are averaged over each cluster and month, and anomalies are computed relative to the 2013–2017 means for that month of the year. In each panel, observations (red line) are compared with results from an MLR model driven by meteorological variability (blue line). The linear trend of the 3-mo average residuals for each year is shown in black. The MLR model uses the top three meteorological predictors (Table 1) for each  $0.5^\circ \times 0.625^\circ$  grid cell in the cluster, and the results are then averaged for each cluster. The dominant variables in each cluster are indicated in legend with the sign of their correlation to MDA8 ozone. The coefficients of determination ( $R^2$ ) for the MLR model are shown in the right corner of each plot for the detrended time series (removing the residual linear trend).

while decreasing ozone elsewhere, following expected patterns of VOC-limited and  $\text{NO}_x$ -limited conditions. Ozone production in urban areas is expected to be VOC-limited because  $\text{NO}_x$  concentrations are very high, but ozone production on a more regional scale in summer is expected to be  $\text{NO}_x$ -limited. The modeled ozone sensitivity is generally consistent with previous measurement-based, satellite-retrieved, and model inferences of  $\text{NO}_x$ - vs. VOC-limited conditions for ozone production in China (4, 12, 22).

However, we find that changes in  $\text{PM}_{2.5}$  are more important than changes in  $\text{NO}_x$  or VOC emissions in driving ozone trends, particularly in the North China Plain, and this is mainly due to aerosol chemistry rather than photolysis (Fig. 4). The relevant aerosol chemistry involves reactive uptake of the gaseous precursors to ozone formation, as described in GEOS-Chem by first-order reactive uptake coefficients  $\gamma$  (32). This includes reactive uptake of the hydroperoxy radical ( $\text{HO}_2$ ) with coefficient  $\gamma = 0.2$  and conversion to  $\text{H}_2\text{O}$  or  $\text{H}_2\text{O}_2$  (32–34) and reactive uptake of nitrogen oxides ( $\text{NO}_2$ ,  $\text{NO}_3$ , and  $\text{N}_2\text{O}_5$ ) with conversion to  $\text{HNO}_3$  (32, 35). Uptake of  $\text{HO}_2$  is by far the dominant effect (Fig. 4). It accounts in the model for most of the sink of hydrogen oxide radicals ( $\text{HO}_x \equiv \text{OH} + \text{peroxy}$ ) in eastern China (SI Appendix, Fig. S5). This suppresses the  $\text{HO}_2 + \text{NO}$  reaction by which ozone is produced. The effect is particularly important in the North China Plain where  $\text{PM}_{2.5}$  concentrations are highest.

The importance of aerosol chemistry as a sink for ozone precursors in China has been previously pointed out in model studies (21, 22), which found ozone decreases of 6–12 and 10–20 ppb, respectively, over eastern China as a result of this chemistry. Ref.

21 found the dominant effect to be the reactive uptake of nitrogen oxides, but we find that effect to be small in part because of VOC-limited conditions and in part because summertime conditions are not conducive to nighttime  $\text{NO}_3/\text{N}_2\text{O}_5$  chemistry.

The  $\text{HO}_2$  uptake coefficient  $\gamma = 0.2$  used in our simulation is consistent with a large body of experimental and modeling literature

**Table 1. Meteorological variables considered as ozone covariates**

| Variable name | Description                                    |
|---------------|--|
| Tmax          | Daily maximum 2-m air temperature (K)          |
| U10           | 10-m zonal wind ( $\text{m s}^{-1}$ )*         |
| V10           | 10-m meridional wind ( $\text{m s}^{-1}$ )†    |
| PBLH          | Mixing depth (m)                               |
| TCC           | Total cloud area fraction (%)                  |
| Rainfall      | Precipitation ( $\text{mm d}^{-1}$ )           |
| SLP           | Sea level pressure (Pa)                        |
| RH            | Surface air relative humidity (%)              |
| V850          | 850-hPa meridional wind ( $\text{m s}^{-1}$ )† |

Meteorological data from the NASA MERRA-2 reanalysis (23) with  $0.5^\circ \times 0.625^\circ$  grid resolution. The data are averaged over 24 h for use in the MLR model for ozone except for PBLH and TCC, which are averaged over daytime hours (8–20 local time), and for Tmax (daily maximum).

\*Positive westerly.

†Positive southerly.



and provide us with a full gridded ensemble of meteorological variables. We average them over either 24 h or daytime hours (8–20 local time), depending on the variable (Table 1). All data are normalized for use in the MLR model (see below) by subtracting their 2013–2017 mean for that day of the year and dividing by the standard deviation.

**Multiple Linear Regression Model.** A number of previous studies have examined meteorological influences on ozone variability in China (4, 9, 39, 40). On the basis of these studies we considered the correlation of MDA8 ozone across China with a large number of candidate meteorological variables from the MERRA-2 archive (*SI Appendix, Table S3 and Fig. S9*). This led us to adopt nine variables as featuring the strongest correlations (Table 1). We applied a stepwise MLR model for each  $0.5^\circ \times 0.625^\circ$  grid cell:

$$y = \beta_0 + \sum_{k=1}^9 \beta_k x_k + \text{interaction terms}, \quad [5]$$

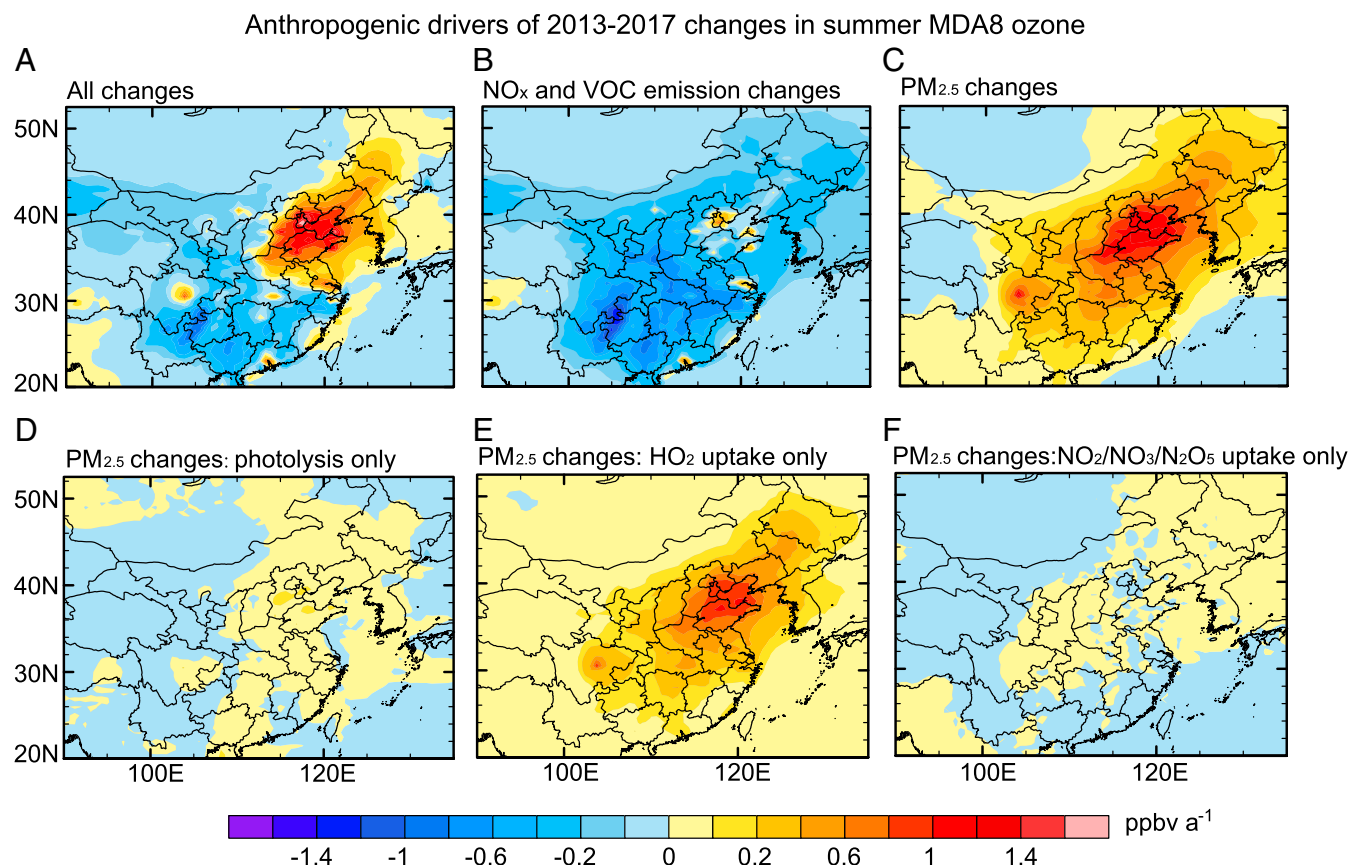
where  $y$  is the normalized daily MDA8 ozone concentration and  $(x_1, \dots, x_9)$  are the nine meteorological variables. The interaction terms are up to second order. The regression coefficients  $\beta_k$  are determined by a stepwise method adding and deleting terms based on Akaike information criterion statistics to obtain the best model fit (41). Similar MLR models have been successfully applied to quantify the effect of meteorological variability on air pollutants in North America, Europe, and China (42–44).

We first apply the MLR model to identify the key meteorological variables driving the variability of daily surface ozone for each grid cell. Only the three locally dominant meteorological variables are regressed onto deseasonalized monthly MDA8 ozone to fit the effect of 2013–2017 meteorological variability on ozone within a  $0.5^\circ \times 0.625^\circ$  grid cell. This is done to avoid overfitting. We find that the dominant meteorological variables driving ozone variability are consistent across grid cells on a regional scale.

**GEOS-Chem Simulations.** The ozone simulations use the nested-grid version of the GEOS-Chem chemical transport model with detailed oxidant–aerosol chemistry, driven by MERRA-2 assimilated meteorological data and with a horizontal resolution of  $0.5^\circ \times 0.625^\circ$  over East Asia (version 11-02; [acmg.seas.harvard.edu/geos/](http://acmg.seas.harvard.edu/geos/)). Anthropogenic emissions in China are from the MEIC inventory (see below). The base simulation is for the summer of 2013, and sensitivity simulations examine the effects of 2013–2017 changes in Chinese anthropogenic emissions,  $\text{PM}_{2.5}$ , and AOD, as described below. Additional sensitivity simulations isolate the effects of  $\text{PM}_{2.5}$  and AOD changes on photolysis rates,  $\text{NO}_x$  aerosol chemistry, and  $\text{HO}_2$  aerosol chemistry. Results presented in Fig. 4 are differences between the sensitivity simulations and the base simulation. Further details on the GEOS-Chem simulations are in *SI Appendix*.

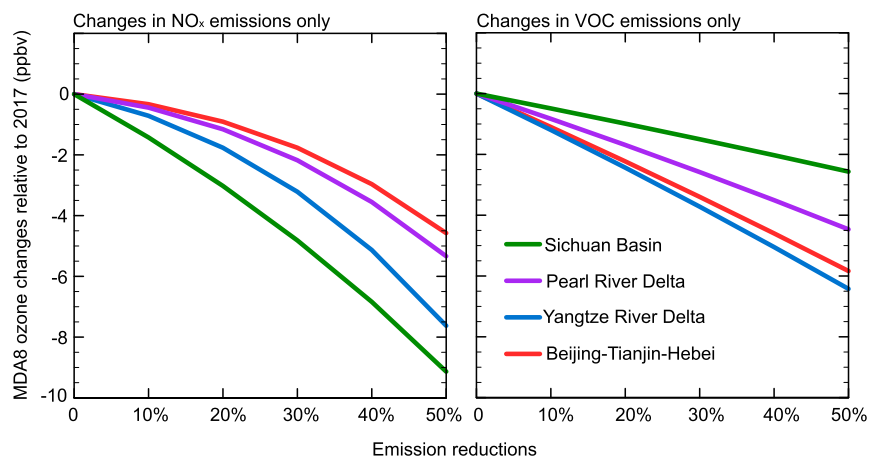
**Anthropogenic Emission Inventory.** The MEIC ([www.meicmodel.org](http://www.meicmodel.org)) is used to estimate China's anthropogenic emissions and their trends from 2013 to 2017 (17, 31). MEIC is a widely used bottom-up emission inventory framework that follows a technology-based methodology to calculate emissions from more than 700 anthropogenic source types in China.

**$\text{PM}_{2.5}$  and Aerosol Optical Depth Data.** Observed  $\text{PM}_{2.5}$  concentrations during 2013–2017 are from the same MEE observation network as ozone. Local changes in  $\text{PM}_{2.5}$  concentrations from 2013 to 2017 affecting aerosol chemistry are applied as scaling factors to GEOS-Chem aerosol surface areas in the boundary layer below 1.3 km. AOD trends for 2013–2017 are from the monthly level 3 product of the Moderate Resolution Imaging Spectroradiometer (MODIS) instrument aboard the Aqua satellite, reported at 550-nm wavelength with a resolution of  $1^\circ \times 1^\circ$  (<https://ladsweb.modaps.eosdis.nasa.gov/>). These trends in AOD are applied as scaling factors to simulated AOD in the GEOS-Chem calculation of photolysis rates (see details in *SI Appendix, sections 1 and 2*).



**Fig. 4.** Anthropogenic drivers of 2013–2017 changes in mean summertime MDA8 ozone in China. (A–C) GEOS-Chem model results for the changes in MDA8 ozone resulting from: (A) combined effects of 2013–2017 changes in  $\text{NO}_x$  and VOC emissions together with changes in  $\text{PM}_{2.5}$ , (B) effects of 2013–2017 changes in  $\text{NO}_x$  and VOC emissions alone, and (C) effects of 2013–2017  $\text{PM}_{2.5}$  changes alone including contributions from aerosol chemistry and photolysis rates. (D–F) The different effects of 2013–2017  $\text{PM}_{2.5}$  changes on ozone are separated: (D) radiative effect on photolysis rates, (E) effect of  $\text{HO}_2$  uptake, and (F) effect of nitrogen oxide ( $\text{NO}_2$ ,  $\text{NO}_3$ , and  $\text{N}_2\text{O}_5$ ) uptake.

Ozone changes under future emission reductions



**Fig. 5.** Response of ozone to decreases of anthropogenic  $\text{NO}_x$  and VOC emissions in China relative to 2017 values. Values are GEOS-Chem model results for summertime mean MDA8 ozone in the four mega-city clusters of Fig. 1. The simulations decrease either (Left)  $\text{NO}_x$  or (Right) VOC emissions by a uniform fraction across China.

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