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Unprecedented decline in summertime surface ozone over eastern China in 2020 comparably attributable to anthropogenic emission reductions and meteorology

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Abstract

China's nationwide monitoring network initiated in 2013 has witnessed continuous increases of urban summertime surface ozone to 2019 by about 5% year^{-1}, among the fastest ozone trends in the recent decade reported in the Tropospheric ozone assessment report. Here we report that surface ozone levels averaged over cities in eastern China cities decrease by 5.5 ppbv in May–August 2020 compared to the 2019 levels, representing an unprecedented ozone reduction since 2013. We combine the high-resolution GEOS-Chem chemical model and the eXtreme Gradient Boosting (XGBoost) machine learning model to quantify the drivers of this reduction. We estimate that changes in anthropogenic emissions alone decrease ozone by 3.2 (2.9–3.6) ppbv (57% of the total 5.5 ppbv reduction) averaged over cities in eastern China and by 2.5 $\sim$ 3.2 ppbv in the three key city clusters for ozone mitigation. These reductions appear to be driven by decreases in anthropogenic emissions of both nitrogen oxides (NO_x) and volatile organic compounds, likely reflecting the stringent emission control measures implemented by The Chinese Ministry of Environmental and Ecology in summer 2020, as supported by observed decline in tropospheric formaldehyde (HCHO) and nitrogen dioxides (NO_2) from satellite and by bottom-up emission estimates. Comparable to the emission-driven ozone reduction, the wetter and cooler weather conditions in 2020 decrease ozone by 2.3 (1.9–2.6) ppbv (43%). Our analyses indicate that the current emission control strategies can be effective for ozone mitigation in China yet tracking future ozone changes is essential for further evaluation. Our study also reveals important potential to combine the mechanism-based, state-of-art atmospheric chemical models with machine learning model to improve the attribution of ozone drivers.

1. Introduction

Surface ozone significantly damages human health and ecosystem (Jerrett *et al* 2009, Anenberg Susan *et al* 2012, Tai *et al* 2014). It is mainly generated from nitrogen oxides (NO_x) and volatile organic compounds (VOCs) through sunlight driven photochemical reactions (Monks *et al* 2015, Sun *et al* 2020, 2021). China is the present-day global hot spot of urban ozone pollution, with significantly higher daily maximum 8 h average (MDA8) ozone levels and faster increasing trends over 2013–2019 compared to other industrialized regions worldwide reported in the Tropospheric ozone assessment report (Fleming *et al* 2018, Lu *et al* 2018, 2020), even though the domestic anthropogenic NO_x emissions has been reduced by 20% since 2012 (Zheng *et al* 2018). Recent studies have reported that the observed ozone increases over China extended to spring 2020 in the coronavirus disease 2019 (COVID-19) lockdown period, highlighting the complexity of ozone mitigation (Le *et al* 2020, Huang *et al* 2021, Li *et al* 2021, Yin *et al* 2021). China's energy consumptions and travel demands resumed in May and returned to comparable levels of 2019 (Zheng *et al* 2020). In June 2020, the Chinese Ministry of Environmental and Ecology (MEE) implemented The 2020 action plan on VOCs mitigations (the Chinese MEE) (table S1 (available online at stacks.iop.org/ERL/16/124069/mmedia)), an action following the 2018–2020 action plan on defending the blue sky (Chinese State Council 2018) aiming at a 10% and 15% reduction of anthropogenic VOCs and NO_x emissions relative to the 2015 level. Here, we examine whether these measures can be effective to mitigate summertime (May–August) ozone pollution in 2020.

We first present an unprecedented widespread decrease in observed summertime ozone levels in Chinese cities in 2020 after a continuous increase from 2013 to 2019 since the nationwide monitoring network was launched. A critical underlying question is that to what extent the reduction of summertime ozone is driven by anthropogenic emission control measures rather than meteorology. A standard approach is to apply state-of-art chemical transport models (CTMs) with sensitivity simulations (e.g. fixed anthropogenic levels but with varied meteorology) to quantify ozone change drivers (Lu *et al* 2019a). However, uncertainties in emission estimates and model deficiencies can lead to systematic bias in CTMs that affect the accuracy of their sensitivity to changes in either emissions or meteorology (Fu *et al* 2015, Young *et al* 2018, Lu *et al* 2019b).

In this study, we introduce the eXtreme Gradient Boosting (XGBoost) machine learning model to correct the bias of ozone predictions from the GEOS-Chem CTM. We use the novel GEOS-Chem-XGBoost modeling approach to separate the role of emission and meteorology in ozone variability and estimate the uncertainties with the ensemble of sensitivity tests. Our analyses reveal for the first time that changes in domestic anthropogenic emissions contribute substantially to the summertime ozone reduction in 2020 relative to 2019 levels, thus delivers important policy implications for ozone mitigation over China.

2. Materials and methods

2.1. Ozone observation

We obtain hourly surface ozone observations in 2013–2020 from the China National Environmental Monitoring Center network (http://106.37.208.233:20035/). This nationwide network began operating in 74 major cities in 2013, and extended to 367 cities by 2020. We apply data quality control method to remove unreliable outliers following our previous studies (Lu *et al* 2018). Observations at monitoring sites within a city are averaged hourly to represent ozone air quality at city level. We report the MDA8 ozone values as the ozone metric.

2.2. Tropospheric nitrogen dioxide (NO_2) and formaldehyde (HCHO) observation

We use the daily tropospheric NO_2 and HCHO columns retrieved from TROPOspheric Monitoring Instrument (TROPOMI) on board the Copernicus Sentinel-5 Precursor satellite (Veefkind *et al* 2012) to examine the changes of ozone precursors in 2020 vs 2019. TROPOMI is a nadir-viewing spectrometer instrument with a ground pixel size of 7 km × 7 km and local passing time at approximately 13:30. TROPOMI retrievals of NO_2 and HCHO columns (de Smedt *et al* 2018, van Geffen *et al* 2020) have been applied in recent ozone studies over China (Li *et al* 2020, 2021). We use the Level 3 data with a spatial resolution of 25 km × 25 km. Pixels with quality assurance values less than 50% for HCHO and 75% for NO_2 have been removed.

2.3. GEOS-Chem CTM simulation

We use the nested-grid version of GEOS-Chem CTM 12.2.1 to simulate ozone and other atmospheric chemical components over China (http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem_12#New_data_directories_in_12.2.1). The model is conducted at a horizontal resolution of 0.25° × 0.3125° with the boundary conditions from a global simulation at 2° × 2.5° resolution, driven by GEOS-FP assimilated meteorological data. GEOS-Chem includes a detailed HO_x–NO_x–VOC–ozone–halogen–aerosol tropospheric chemistry mechanism (Wang *et al* 1998, Bey *et al* 2001, Park *et al* 2004, Mao *et al* 2013, Yin *et al* 2019, 2020) and has been widely used in ozone modeling studies over China (Ni *et al*

2018, Li *et al* 2019a, Lu *et al* 2019a, Dang *et al* 2021, Wang *et al* 2021b).

We use the community emissions data system inventory for global anthropogenic emissions at the latest 2017 level (McDuffie *et al* 2020), with the Chinese anthropogenic emissions from the multi-resolution emission inventory (Zheng *et al* 2018). We scale the Chinese anthropogenic emissions to 2019 levels based on observations of primary air pollutants and emission trends reported by the MEE following (Li *et al* 2021), resulting in a scaling factor of 87.3% for NO_2, 83.7% for CO, 85.1% for particulate matter (PM), and no scaler for VOCs. These scalers may remain uncertain and are not expected to represent precisely the anthropogenic emission levels in 2019–2020, but this is inconsequential here as we prefer to use machine learning model to correct the model bias as will be described later. Anthropogenic and biomass burning emissions (van der Werf *et al* 2017) are fixed for 2019 and 2020. Natural emissions of biogenic VOCs (Guenther *et al* 2012), lightning NO_x (Murray *et al* 2012), and soil NO_x (Hudman *et al* 2012, Lu *et al* 2021) are calculated online in the model and thus respond to meteorological parameters.

We run global simulation at $2^\circ \times 2.5^\circ$ resolution from January 2019 to August 2020. The model results on 1 April 2019 and 2020 were then interpolated to high resolution ($0.25^\circ \times 0.3125^\circ$) over the nested domain (70 °E–140 °E, 15 °N–55 °N) to initialize the nested model simulation. Results from the nested model for May–August in both years are analyzed. Since the anthropogenic emissions are fixed, simulated ozone difference between 2020 (GC_{2020}) and 2019 (GC_{2019}) provides a means to quantify ozone change due to changes in meteorological conditions (GC_Met), estimated as:

$$\mathrm{GC\_Met} = \mathrm{GC}_{2020} - \mathrm{GC}_{2019}. \quad (1)$$

We can sequentially quantify the role of emission changes (GC_Emis) as:

$$\mathrm{GC\_Emis} = (\mathrm{Obs}_{2020} - \mathrm{Obs}_{2019}) - (\mathrm{GC}_{2020} - \mathrm{GC}_{2019}) \quad (2)$$

where Obs_{2019} and Obs_{2020} represents observations for 2019 and 2020, respectively.

This widely-used method assumes that CTMs can accurately simulate ozone sensitivity to interannual changes in meteorological parameters, but this is very likely problematic because of model bias due to errors in emissions estimates, chemical mechanism, and sub-grid-scale local processes (Young *et al* 2018, Lu *et al* 2019a). We use a machine learning model in addition to the GEOS-Chem results to address this issue.

2.4. XGBoost machine learning model to correct GEOS-Chem bias

We apply the XGBoost model to correct the bias of hourly GEOS-Chem ozone predictions at each observation site. XGBoost uses the Gradient Boosting Decision Tree framework to iteratively train the systematic observation-model bias in a stagewise manner (Jerome 2001, Chen and Guestrin 2016, Keller *et al* 2021). We use hourly observations in 2019 from each site as learning samples, and GEOS-Chem input of emissions (total of anthropogenic and natural emissions) and meteorological parameters, output of concentrations of atmospheric chemical components, and time information (e.g. hour, day, month) as training input data (table S2), following (Keller *et al* 2021). These parameters are central to ozone chemistry and transport. All data have been standardized ($Z_i = \frac{X_i - \mu}{\sigma}$, where X_i represent the ith item in the dataset, and μ and σ are the average and standard deviation of dataset X, respectively). We choose a learning rate of 0.35, maximum tree depth of 6, L_1 and L_2 regularization terms of 0 and 1, the loss function of mean square, and evaluation metrics of root-mean-square error.

Our base estimates apply a five-fold cross validation strategy with the default learning rate and maximum tree depth following (Keller *et al* 2021). The five-fold cross validation is designed to reduce overfitting and auto-correlation signal (Kleinert *et al* 2021), and to test the robustness of XGBoost model. For each site, we first group all sample data into five subsets, and then train the model for five iterations, with each time taking four subsets as training dataset and the remaining one as testing data. We take the ensemble mean of these five iterations as the base estimate. We also conduct several sensitivity tests by altering the learning rate from 0.35 to 0.3 and 0.4 with the same five-fold cross validation strategy, and by altering the maximum tree depth of 6–4 and 8, yielding another 20 ensembles. The range of the total 25 ensemble are used to represent the uncertainty from the GEOS-Chem-XGBoost model.

The combination of GEOS-Chem and XGBoost model (hereafter GEOS-Chem-XGBoost, or GCB) is expected to provide a close fit to 2019 ozone observations. As GEOS-Chem simulation uses the same anthropogenic emissions in 2020 as in 2019, and observations for 2020 are not included in bias training, applying the GEOS-Chem-XGBoost model in 2020 then predicts ozone evolution in 2020 (GCB_{2020}) with 2019 anthropogenic emission levels at each observation site. Examination of the probability density functions of key ozone precursors and meteorological parameters shows that the training data in 2019 cover the range of variations in these factors in 2020 at individual cities (figures S1 and S2), supporting the application of the GEOS-Chem-XGBoost model to predict ozone in summer 2020 if anthropogenic emissions were unchanged from 2019 levels.

Comparison of the GEOS-Chem-XGBoost predictions to the actual 2020 observations then quantifies ozone changes due to perturbation of emissions (GCB_Emis), and sequentially the contribution from meteorology (GCB_Met):

$$\mathrm{GCB\_Emis} = \mathrm{Obs}_{2020} - \mathrm{GCB}_{2020} \quad (3)$$

$$\begin{aligned}\mathrm{GCB\_Met} &= (\mathrm{Obs}_{2020} - \mathrm{Obs}_{2019}) \\ &\quad - (\mathrm{Obs}_{2020} - \mathrm{GCB}_{2020}) \\ &= \mathrm{GCB}_{2020} - \mathrm{Obs}_{2019}. \end{aligned} \quad (4)$$

A major advantage by applying this approach is that it corrects the systematic bias in GEOS-Chem ozone predictions, allowing a more accurate ozone sensitivity to changes in meteorology and emissions, as will be discussed later. We report results based on the GEOS-Chem-XGBoost approach but also compare with the GEOS-Chem results for consistency check.

3. Results and discussion

3.1. Widespread surface ozone decreases in Chinese cities in 2020

Figures 1(a) and (b) present the May–August mean MDA8 ozone concentrations at all available urban sites in 2019 and 2020. High ozone levels are concentrated in eastern China, especially in the Beijing–Tianjin–Hebei (BTH), the Fenwei Plain (FWP), and the Yangtze River Delta (YRD) city clusters (table S3), reflecting high anthropogenic emissions of ozone precursors in the regions. These city clusters have been identified by the MEE as key regions for ozone mitigation.

We find significant reduction of MDA8 ozone by 3.8 ± 0.5 ppbv (6%) (mean ± standard deviation across cities) averaged over all 366 cities and 5.5 ± 0.5 ppbv (8%) for 149 cities in eastern China in May–August 2020 compared to 2019 levels (figure 1(c)). This ozone reduction is unprecedented in the 2013–2020 period, following a continuous increase of surface MDA8 ozone levels from 2013 to 2019 by approximately 5% year^{-1} (figure 1(d)) (Lu *et al* 2020, Li *et al* 2021). The ozone reductions are widespread across the cities as indicated by the significant decrease in number of sites with MDA8 ozone higher than 75 ppbv (approximately the Chinese grade II air quality standard) from 150 in 2019 to only 9 in 2020. In particular, ozone decreases by 4.7 ± 0.5 ppbv (7%), 5.4 ± 0.6 ppbv (8%), and 5.9 ± 0.6 ppbv (10%) in the BTH, FWP, and YRD regions, respectively. The reductions are most prominent in June–August (figure 3).

3.2. Model performance on capturing observed ozone levels

We first compare the ability of GEOS-Chem and GEOS-Chem-XGBoost in capturing the observed ozone levels in 2019. Figure 2 shows the time series of observed and model predicted ozone averaged for all Chinese cities over eastern China and for the BTH, FWP, and YRD regions. We focus on eastern China and the three key city cluster hereafter since summertime ozone levels are higher, ozone decreases are more prominent, and influences from foreign ozone transport are smaller in these cities than others. The GEOS-Chem predictions generally capture the synoptic-scale variability of MDA8 ozone in these regions, but they show high mean bias (MB) of 7.7 ppbv (17.5%) across all cities in eastern China, reflecting the model bias as discussed above.

The GEOS-Chem-XGBoost model significantly improves the prediction of ozone levels compared to the GEOS-Chem CTM (figure 2). It shows no bias to the training data (MB = 0.1 ppbv, 0.2%) and very small bias to the testing data (MB = 0.6 ppbv, 1.2%) (figure 4, table S4), resulting in a mean bias of 0.3 ppbv (0.7%) to all 2019 ozone observations (figure 2). We also estimate the importance of input parameters in the XGBoost model for bias correction, using the SHapely Additive exPlanations (SHAP) approach (Lundberg *et al* 2017). As shown in figure 5, meteorological parameters such as near-surface temperature and wind fields, the uncorrected GEOS-Chem predicted ozone, reactive nitrogen (e.g. NO_2, PAN), and non-methane VOCs are the main factors for explaining the GEOS-Chem ozone bias, consistent with (Keller *et al* 2021).

3.3. Attribution of summertime ozone reduction in 2020

We then attribute quantitatively the 2020 ozone reduction to changes in anthropogenic emissions and meteorological conditions following equations (3) and (4). Differences between the observed and GEOS-Chem-XGBoost predicted ozone in 2020 represent the emission-driven ozone changes in 2020 relative to 2019, as indicated by the shadings in figure 2. Figure 3 shows the spatial patterns of emission-driven versus meteorology-driven ozone changes, and figure 4 summarizes the mean contributions across eastern China and the key city clusters.

We find that the May–August ozone decreases in 2020 over eastern China can be largely attributed to anthropogenic emission changes, with 101 cities (69%) showing emission-driven ozone reduction of 0.2–15.3 ppbv though there are large variabilities in terms of magnitudes (figure 3). For the average of all eastern China cities, changes in emissions alone contribute to ozone decrease of 3.2 (2.9–3.6 from the ensemble range) ppbv, accounting for 57% (53%–66%) of the total ozone reduction (5.5 ± 0.5 ppbv) (figure 4). The estimated emission-driven ozone reductions are 2.5 (2.2–2.8) ppbv, 3.2 (2.9–3.6) ppbv, and 3.2 (2.8–3.6) ppbv in the BTH, FWP, and YRD city clusters, respectively.

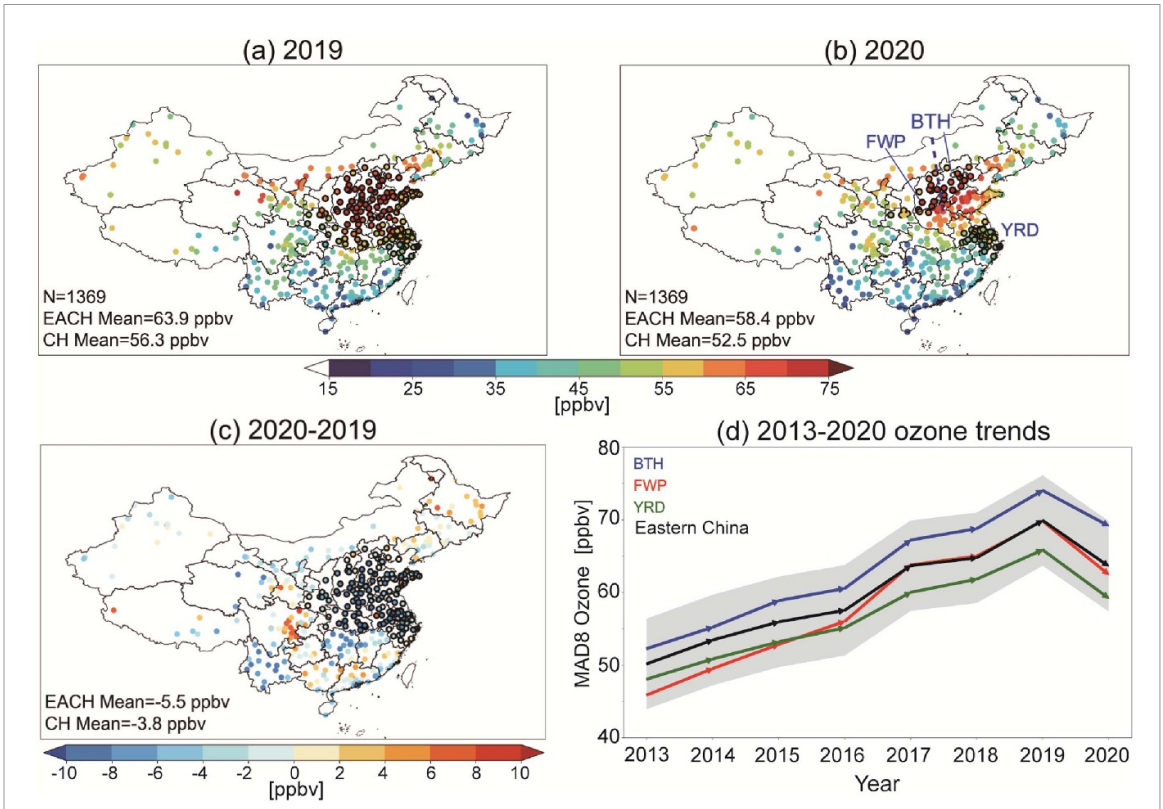

Figure 1. Decline in summertime surface MDA8 ozone over China (CH) in 2020. (a) Spatial distributions of May–August mean MDA8 ozone concentrations in 2019. Number (N) denotes available sites for this year. Observations at monitoring sites within a city are averaged hourly to represent ozone air quality at the city level, and the city-mean ozone levels are shown inset. Dots marked with black circles are sites in eastern China (EACH). (b) Same as (a), but for 2020. Dots marked with black circles are sites in the BTH, the FWP, and the YRD city clusters, the three key regions of ozone mitigation. (c) Ozone difference between 2020 and 2019. (d) Trends in summertime (May–August) mean MDA8 ozone concentrations in 2013–2020 averaged for eastern China cities and for the three key regions. Only 145 sites in 39 cities with continuous 8 year observations are included in trend analyses. Gray shadings represent the range of mean value ± standard deviation across all cities.

These estimates are sufficiently large compared to the GEOS-Chem-XGBoost model bias (0.3 ppbv). The reductions may include some contributions from global emission reductions during the COVID-19 restrictions, which have been found to reduce ozone in the free troposphere (Steinbrecht *et al* 2021). However, these foreign contributions to surface ozone are small due to the short ozone lifetime in polluted city boundary layers and in summer (Lu *et al* 2019a, 2021), and our focus of eastern China here further minimizes the effect of foreign ozone transport (Ni *et al* 2018, Lu *et al* 2019a). Thus, the emission-driven ozone reductions are more likely due to local anthropogenic emission changes in 2020.

We examine changes of HCHO and NO_2 from TROPOMI measurements to understand the anthropogenic driver of the ozone decreases from 2019 to 2020 in figure 5. We find notable decreases in observed tropospheric HCHO (4.1 ± 1.1% averaged for cities in eastern China) and NO_2 (6.0 ± 1.2%) columns in 2020 compared to 2019 levels in most regions of eastern China (figures 5(a) and (b)). These changes may partly come from meteorological influences on biogenic sources as will be discussed later, but also reflect the decline in anthropogenic emissions of NO_x and non-methane VOCs (NMVOCs) supported by the newly released bottom-up estimates (Zheng *et al* 2021) (figures 5(c) and (d)). The national total anthropogenic emissions of NO_x and NMVOCs in May-August decreased by 1%–3% and 2%–5% in 2020 compared to 2019, respectively, with most regions in eastern China showing decreasing emissions of about 5%. These reductions are linked to both the ongoing NO_x emission mitigation (Lu *et al* 2019a) and the new implementation of The 2020 action plan on VOCs mitigations by MEE in June 2020 (The Chinese Ministry of Environmental and Ecology (MEE) 2020), which issues a number of control measures including implementation of stringent VOCs emission standards, replacement of raw and auxiliary materials with low VOCs content, and mitigation of unorganized emissions, as summarized in table S1. The larger emission declines in areas surrounding Beijing may also be linked to the extension of COVID-19 lockdown measures (Zheng *et al* 2021), but at the country level these measures should play a much smaller role. Zheng *et al* (2021) shows that China's thermal electricity generation and travel demands in May–August 2020 are 1.6%–9.0% higher than those in the same months of 2019, with the loosening of COVID-19 lockdown measures from April in most regions.

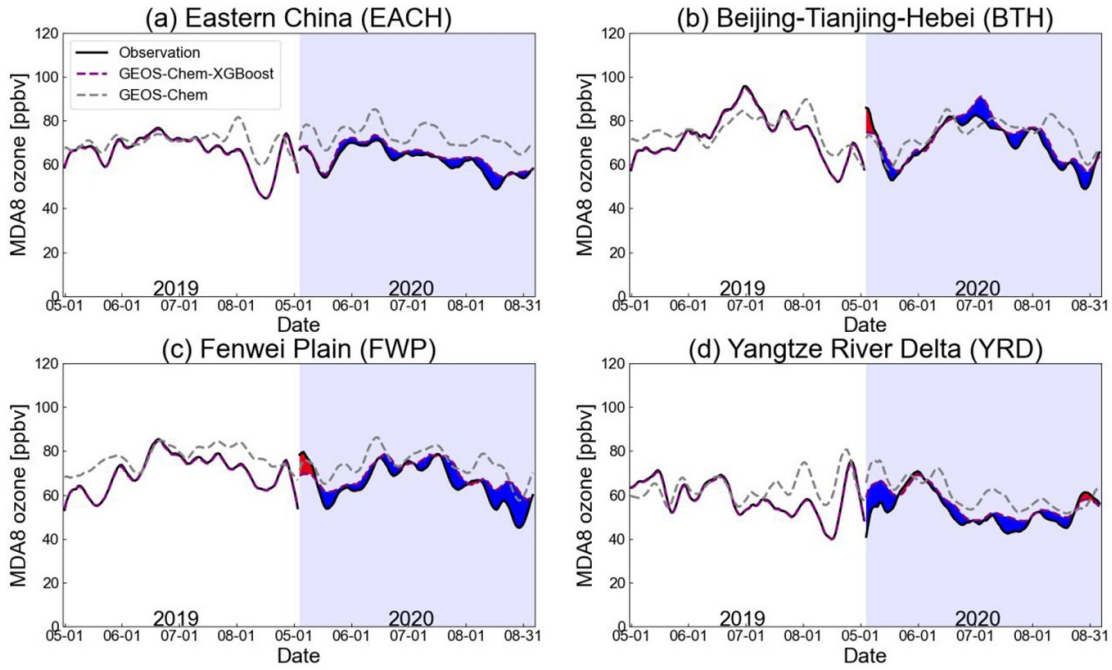

Figure 2. Observed and model predicted ozone evolution of summertime (May–August) ozone in 2019 and 2020. Figures are for (a) all eastern China cities, (b) the BTH region, (c) the FWP region, and (d) the YRD region. Observed ozone values are denoted by black solid lines. GEOS-Chem predictions are denoted by gray dashed lines. GEOS-Chem model predictions have notable bias to observations, and we corrected the bias in 2019 by applying the XGBoost machine learning model. The GEOS-Chem-XGBoost predictions as shown in the purple dashed lines thus present an improved fit to 2019 observations. GEOS-Chem-XGBoost predictions for 2020 then predicts ozone evolution in 2020 with 2019 emission levels. Comparison of the GEOS-Chem-XGBoost predictions to the actual 2020 observations then quantifies ozone changes due to perturbation of emissions. Blue (red) shadings represent where GEOS-Chem-XGBoost predictions are higher (lower) than the actual observations in 2020, indicating that emission changes lead to ozone decrease (increase) in 2020. All values shown are 7 d average for presentation purpose. See the text for more information.

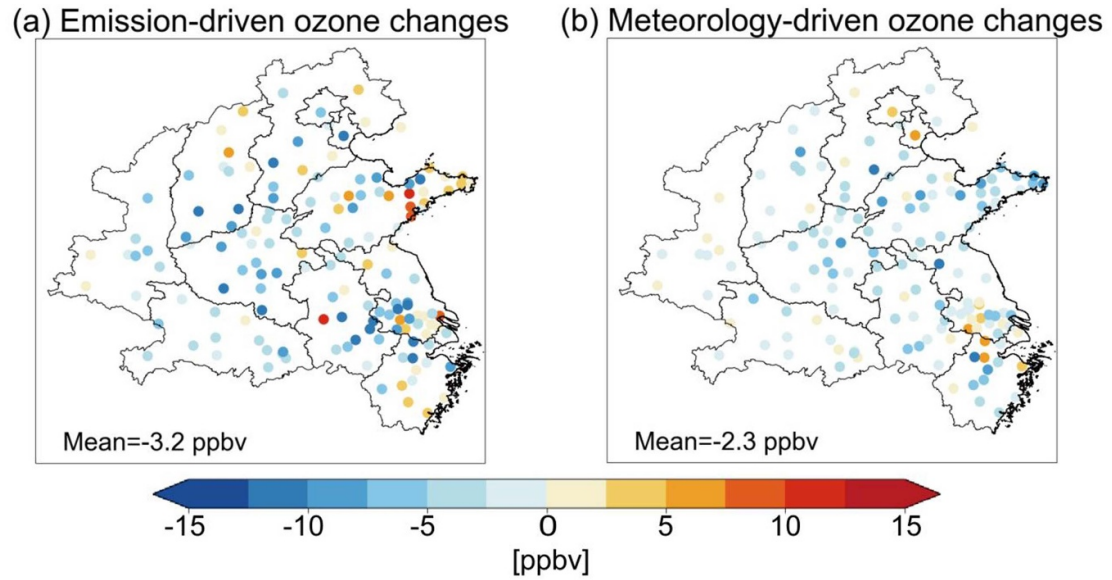

Figure 3. Attribution of summertime ozone changes for cities over eastern China in 2020 relative to the 2019 levels. (a) Ozone changes attributable to anthropogenic emissions, (b) ozone changes attributable to meteorological conditions.

Our analyses suggest that concurrent reductions of anthropogenic NO_x and VOCs emissions based on 2019 emission levels can be effective for ozone mitigation in eastern China. This result can be expected from several recent observational and modeling studies. Previous studies showed ozone increases in Chinese cities from 2013 were driven by the decline of PM through impacts on heterogeneous chemistry and radiation, and decline in anthropogenic NO_x emissions in regions where ozone chemical formation

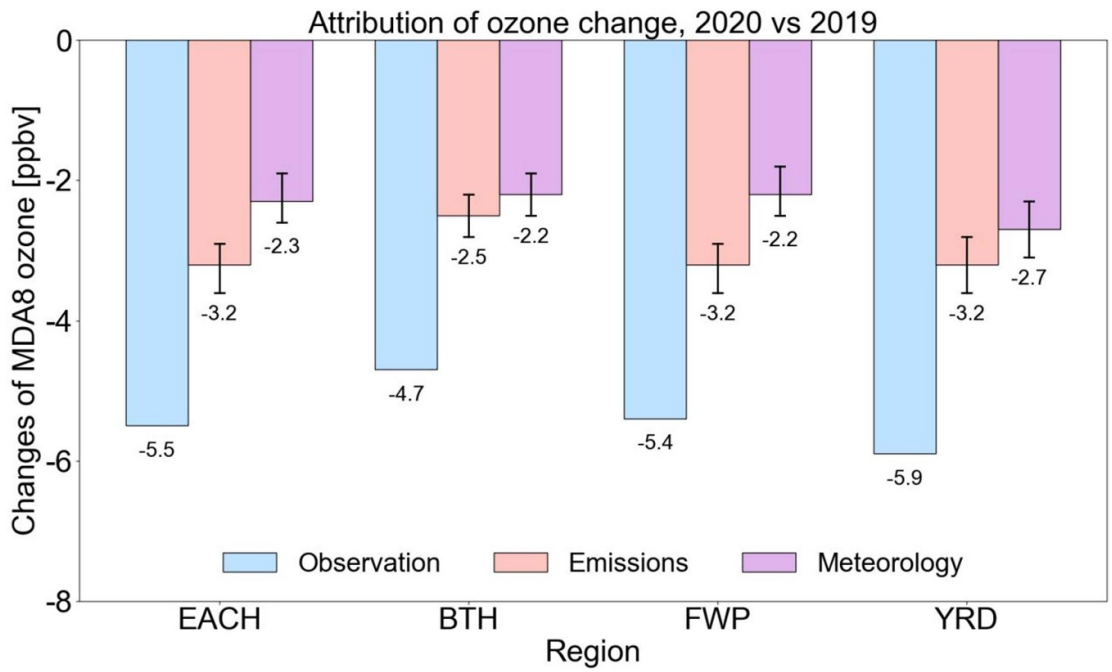

Figure 4. Attribution of summertime ozone changes for eastern China (EACH) cities and for the three key city clusters in 2020 relative to the 2019 levels. Filled colored bars denote ozone change as seen from observations, and due to changes in anthropogenic emissions and meteorological conditions estimated by the GEOS-Chem-XGBoost model. Black vertical bars represent the range estimated from the GEOS-Chem-XGBoost ensemble.

is in VOC-limited regime (Li *et al* 2019a, Liu and Wang 2020) while VOCs emissions have not been reduced. However, recent studies based on satellite observations of $HCHO/NO_2$ ratios have detected a shift of ozone chemical sensitivity from VOC-limited regime to transitional regime in eastern China from 2016 to 2019 with the continuous decline in NO_x levels (Wang *et al* 2021a), indicating that emission controls of both NO_x and VOCs based on 2019 levels would benefit ozone mitigation. Regional chemical models also predict decreases of summertime ozone levels with further reduction of VOCs emissions in years after 2017–2019 (Li *et al* 2019b, Liu and Wang 2020, Chen *et al* 2021, Kang *et al* 2021), when the impacts of PM on ozone formation also decline because of the continuous reductions of PM concentrations since 2013. However, our analyses could not distinguish which specific measures on controlling what chemical species are the most responsive for ozone reduction. We also note that there are still large variabilities of the ozone response to emission changes among the cities (figure 3), reflecting the different efficiency of city-level emission control measures and different ozone chemical regimes.

Changes in meteorological conditions contribute to 2.3 (1.9–2.6) ppbv (43%, 35%–47%) of the ozone reduction averaged over cities in eastern China, and reduce ozone by 2.2 (1.9–2.5) ppbv, 2.2 (1.8–2.5) ppbv, and 2.7 (2.3–3.1) ppbv in the BTH, FWP, and YRD city clusters, respectively (figures 3 and 4). Meteorological conditions influence surface ozone by modulating emissions from natural sources, chemical kinetics, dry deposition, and transport (Lu *et al* 2019b). In summer 2020, the eastern and southern China experienced anomaly strong or even record-breaking rainfalls (China Meterological Administration 2021), leading to significant decreases in daily maximum temperature and increases in specific humidity and cloud fractions compared to the 2019 levels (figure 5), especially in June–August (figures S9–S11). These weather conditions inhibit biogenic VOCs emissions (figure S12), slow down ozone chemical production, and facilitate the ventilation of ozone and its precursors (Gong and Liao 2019, Ma *et al* 2019, Lu *et al* 2019a, Jiang *et al* 2021), and therefore contribute to ozone decrease.

3.4. Comparison between GEOS-Chem-XGBoost and GEOS-Chem estimates

We compare the estimated ozone contribution of emissions and meteorology by the GEOS-Chem-XGBoost with those by the GEOS-Chem CTM only (figure 4 vs figure S14). Both methods agree that changes in anthropogenic emission play a significant role in explaining the ozone decrease over eastern China and for the three city clusters in 2020, supporting the main claim of this study, while the absolute magnitudes differ with each other (e.g. 3.2 vs 2.3 ppbv on the emission-driven ozone contribution). Ozone attribution estimated from the GEOS-Chem-only method using equations (1) and (2) assumes that the GEOS-Chem model can accurately simulate ozone sensitivity to changes in

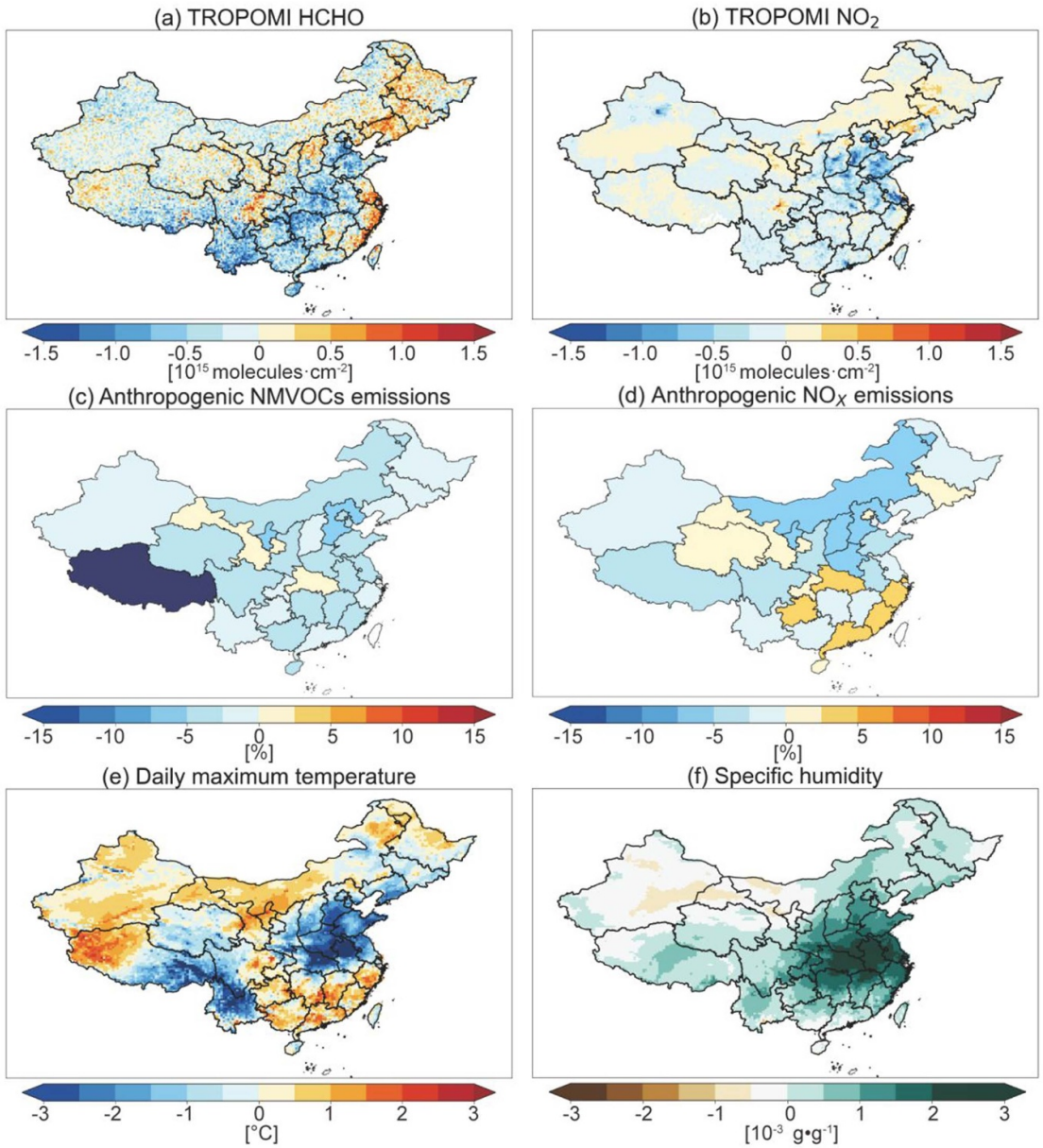

Figure 5. Differences (2020 versus 2019) in summertime (May–August) mean (a) tropospheric HCHO columns from TROPOMI, (b) tropospheric NO_2 columns from TROPOMI, (c) anthropogenic emissions for NMVOCs from (Zheng *et al* 2021), (d) anthropogenic emissions for NO_x from (Zheng *et al* 2021), (e) daily maximum 2 m air temperature, and (f) specific humidity from the GEOS-FP assimilated meteorological data.

meteorological parameters. We test this assumption by simply comparing the correlation coefficients (r) between key meteorological parameters and ozone concentration at afternoon (2 p.m.) in 2019 from observations and GEOS-Chem predictions (figure S14). The GEOS-Chem predictions successfully capture the expected overall positive correlation coefficients between ozone and temperature, and the negative correlations between ozone and specific humidity, cloud fraction, and precipitations, but large discrepancies are found in terms of the absolute values for individual cities, indicating that the model may overestimate/underestimate the ozone sensitivity to meteorological changes. We find that the GEOS-Chem-XGBoost predictions obtained from the testing dataset significantly improve the predicted ozone-meteorology relationship, although this improvement is not necessarily from a better representation of physical process. Thus, we expect that the GEOS-Chem-XGBoost method is more likely to provide an accurate estimate on attribution of ozone change, representing a major advantage in methodology in this study.

4. Conclusion

In summary, we find that the unprecedented summertime ozone decrease (5.5 ppbv) over eastern China in 2020 relative to the 2019 levels are half attributed to domestic anthropogenic emissions

reductions (3.2, 2.9–3.6 ppbv from GEOS-Chem-XGBoost ensemble), while the wetter and cooler weather conditions contribute comparably (2.3, 1.9–2.6 ppbv). It indicates that the current emissions control measures on both NO_x and NMVOCs may have started to achieve initial success, but ozone changes in future years should be continuously tracked to evaluate emission control strategies rigorously. We also reveal important potential to combine the mechanism-based, state-of-art atmospheric chemical models with machine learning model to improve the attribution of ozone drivers. However, our correction strategy used here is only tested for 1 year, and does not directly improve the chemical model description of physical and chemical processes, which may consequently bring uncertainties to the estimated attribution of ozone drivers. Our analyses are not able to determine which specific measures on controlling what species are the most effective for ozone mitigation. More studies are required to address this important issue.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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References

Anenberg Susan C *et al* 2012 Global air quality and health co-benefits of mitigating near-term climate change through methane and black carbon emission controls *Environ. Health Perspect.* **120** 831–9

Bey I, Jacob D J, Yantosca R M, Logan J A, Field B D, Fiore A M, Li Q B, Liu H G Y, Mickley L J and Schultz M G 2001 Global modeling of tropospheric chemistry with assimilated meteorology: model description and evaluation *J. Geophys. Res. Atmos.* **106** 23073–95

Chen T and Guestrin C 2016 XGBoost: a scalable tree boosting system *Proc. 22nd ACM SIGKDD Int. Conf. on Knowledge Discovery and Data Mining* (*San Francisco, California, USA*)

Chen X *et al* 2021 Chinese regulations are working—why is surface ozone over industrialized areas still high? Applying lessons from northeast US air quality evolution *Geophys. Res. Lett.* **48** e2021GL092816

China Meterological Administration 2021 China climate bulletin 2020 (in Chinese) (available at: http://zwgk.cma.gov.cn/zfxxgk/gknr/qxbg/,http://zwgk.cma.gov.cn/zfxxgk/gknr/qxbg/202104/t20210406_3051288.html) (Accessed 1 May 2021)

Chinese State Council 2018 Three-year action plan on defending the blue sky (in Chinese) (available at: www.gov.cn/zhengce/content/2018-07/03/content_5303158.htm)

Dang R, Liao H and Fu Y 2021 Quantifying the anthropogenic and meteorological influences on summertime surface ozone in China over 2012–2017 *Sci. Total Environ.* **754** 142394

de Smedt I *et al* 2018 Algorithm theoretical baseline for formaldehyde retrievals from S5P TROPOMI and from the QA4ECV project *Atmos. Meas. Tech.* **11** 2395–426

Fleming Z L *et al* 2018 Tropospheric ozone assessment report: present-day ozone distribution and trends relevant to human health *Elem. Sci. Anth.* **6** 12

Fu T-M, Zheng Y, Paulot F, Mao J and Yantosca R M 2015 Positive but variable sensitivity of August surface ozone to large-scale warming in the southeast United States *Nat. Clim. Change* **5** 454–8

Gong C and Liao H 2019 A typical weather pattern for ozone pollution events in North China *Atmos. Chem. Phys.* **19** 13725–40

Guenther A B, Jiang X, Heald C L, Sakulyanontvittaya T, Duhl T, Emmons L K and Wang X 2012 The model of emissions of gases and aerosols from nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions *Geosci. Model Dev.* **5** 1471–92

Huang X *et al* 2021 Enhanced secondary pollution offset reduction of primary emissions during COVID-19 lockdown in China *Natl Sci. Rev.* **8**

Hudman R C, Moore N E, Mebust A K, Martin R V, Russell A R, Valin L C and Cohen R C 2012 Steps towards a mechanistic model of global soil nitric oxide emissions: implementation and space based-constraints *Atmos. Chem. Phys.* **12** 7779–95

Jerome H F 2001 Greedy function approximation: a gradient boosting machine *Ann. Stat.* **29** 1189–232

Jerrett M, Burnett R T, Pope C A, Ito K, Thurston G, Krewski D, Shi Y, Calle E and Thun M 2009 Long-term ozone exposure and mortality *New Engl. J. Med.* **360** 1085–95

Jiang Z, Li J, Lu X, Gong C, Zhang L and Liao H 2021 Impact of western Pacific subtropical high on ozone pollution over eastern China *Atmos. Chem. Phys.* **21** 2601–13

Kang M, Zhang J, Zhang H and Ying Q 2021 On the relevancy of observed ozone increase during COVID-19 lockdown to summertime ozone and $PM_{2.5}$ control policies in China *Environ. Sci. Tech. Lett.* **8** 289–94

Keller C A *et al* 2021 Global impact of COVID-19 restrictions on the surface concentrations of nitrogen dioxide and ozone *Atmos. Chem. Phys.* **21** 3555–92

Kleinert F, Leufen L H and Schultz M G 2021 IntelliO3-ts v1.0: a neural network approach to predict near-surface ozone concentrations in Germany *Geosci. Model Dev.* **14** 1–25

Le T, Wang Y, Liu L, Yang J, Yung Y, Li G and Seinfeld J 2020 Unexpected air pollution with marked emission reductions during the COVID-19 outbreak in China *Science* **369** eabb7431

Li K *et al* 2021 Ozone pollution in the North China Plain spreading into the late-winter haze season *Proc. Natl Acad. Sci.* **118** e2015797118

Li K, Jacob D J, Liao H, Shen L, Zhang Q and Bates K H 2019a Anthropogenic drivers of 2013–2017 trends in summer surface ozone in China *Proc. Natl Acad. Sci.* **116** 422–7

Li K, Jacob D J, Liao H, Zhu J, Shah V, Shen L, Bates K H, Zhang Q and Zhai S 2019b A two-pollutant strategy for improving ozone and particulate air quality in China *Nat. Geosci.* **12** 906–10

Li K, Jacob D J, Shen L, Lu X, de Smedt I and Liao H 2020 Increases in surface ozone pollution in China from 2013 to

2019: anthropogenic and meteorological influences *Atmos. Chem. Phys.* **20** 11423–33
Liu Y and Wang T 2020 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.* **20** 6323–37
Lu X *et al* 2021 The underappreciated role of agricultural soil nitrogen oxide emissions in ozone pollution regulation in North China *Nat. Commun.* **12** 5021
Lu X, Hong J, Zhang L, Cooper O R, Schultz M G, Xu X, Wang T, Gao M, Zhao Y and Zhang Y 2018 Severe surface ozone pollution in China: a global perspective *Environ. Sci. Tech. Lett.* **5** 487–94
Lu X, Zhang L, Chen Y, Zhou M, Zheng B, Li K, Liu Y, Lin J, Fu T M and Zhang Q 2019a Exploring 2016–2017 surface ozone pollution over China: source contributions and meteorological influences *Atmos. Chem. Phys.* **19** 8339–61
Lu X, Zhang L and Shen L 2019b Meteorology and climate influences on tropospheric ozone: a review of natural sources, chemistry, and transport patterns *Curr. Pollut. Rep.* **5** 238–60
Lu X, Zhang L, Wang X, Gao M, Li K, Zhang Y, Yue X and Zhang Y 2020 Rapid increases in warm-season surface ozone and resulting health impact in China since 2013 *Environ. Sci. Tech. Lett.* **7** 240–7
Lundberg S M, Lee S-I and Unified A 2017 Approach to interpreting model predictions (available at: https://arxiv.org/pdf/1705.07874.pdf)
Ma M *et al* 2019 Substantial ozone enhancement over the North China Plain from increased biogenic emissions due to heat waves and land cover in summer 2017 *Atmos. Chem. Phys.* **19** 12195–207
Mao J, Paulot F, Jacob D J, Cohen R C, Crounse J D, Wennberg P O, Keller C A, Hudman R C, Barkley M P and Horowitz L W 2013 Ozone and organic nitrates over the eastern United States: sensitivity to isoprene chemistry *J. Geophys. Res. Atmos.* **118** 11256–68
McDuffie E E, Smith S J, O'Rourke P, Tibrewal K, Venkataraman C, Marais E A, Zheng B, Crippa M, Brauer M and Martin R V 2020 A global anthropogenic emission inventory of atmospheric pollutants from sector- and fuel-specific sources (1970–2017): an application of the community emissions data system (CEDS) *Earth Syst. Sci. Data* **12** 3413–42
Monks P S *et al* 2015 Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer *Atmos. Chem. Phys.* **15** 8889–973
Murray L T, Jacob D J, Logan J A, Hudman R C and Koshak W J 2012 Optimized regional and interannual variability of lightning in a global chemical transport model constrained by LIS/OTD satellite data *J. Geophys. Res. Atmos.* **117**
Ni R, Lin J, Yan Y and Lin W 2018 Foreign and domestic contributions to springtime ozone over China *Atmos. Chem. Phys.* **18** 11447–69
Park R J, Jacob D J, Field B D, Yantosca R M and Chin M 2004 Natural and transboundary pollution influences on sulfate-nitrate-ammonium aerosols in the United States: implications for policy *J. Geophys. Res. Atmos.* **109** D15204
Steinbrecht W *et al* 2021 COVID-19 crisis reduces free tropospheric ozone across the Northern Hemisphere *Geophys. Res. Lett.* **48** e2020GL091987
Sun Y *et al* 2020 Mapping the drivers of formaldehyde (HCHO) variability from 2015-2019 over eastern China: insights from FTIR observation and GEOS-Chem model simulation *Atmos. Chem. Phys. Discuss.* **2020** 1–24
Sun Y *et al* 2021 The reduction in C_2H_6 from 2015 to 2020 over Hefei, eastern China, points to air quality improvement in China *Atmos. Chem. Phys.* **21** 11759–79
Tai A P K, Martin M V and Heald C L 2014 Threat to future global food security from climate change and ozone air pollution *Nat. Clim. Change* **4** 817–21
The Chinese Ministry of Environmental and Ecology (MEE) 2020 The volatile organic compound management attack program in 2020 (in Chinese) (available at: www.mee.gov.cn/xxgk2018/xxgk/xxgk03/202006/t20200624_785827.html)
van der Werf G R *et al* 2017 Global fire emissions estimates during 1997–2016 *Earth Syst. Sci. Data* **9** 697–720
van Geffen J, Boersma K F, Eskes H, Sneep M, Ter Linden M, Zara M and Veefkind J P 2020 S5P TROPOMI NO_2 slant column retrieval: method, stability, uncertainties and comparisons with OMI *Atmos. Meas. Tech.* **13** 1315–35
Veefkind J P *et al* 2012 TROPOMI on the ESA sentinel-5 precursor: a GMES mission for global observations of the atmospheric composition for climate, air quality and ozone layer applications *Remote Sens. Environ.* **120** 70–83
Wang W, van der A, R, Ding J, van Weele M and Cheng T 2021a Spatial and temporal changes of the ozone sensitivity in China based on satellite and ground-based observations *Atmos. Chem. Phys.* **21** 7253–69
Wang X *et al* 2021b Sensitivities of ozone air pollution in the Beijing–Tianjin–Hebei area to local and upwind precursor emissions using adjoint modeling *Environ. Sci. Technol.* **55** 5752–62
Wang Y H, Jacob D J and Logan J A 1998 Global simulation of tropospheric O_3-NO_x-hydrocarbon chemistry 3. Origin of tropospheric ozone and effects of nonmethane hydrocarbons *J. Geophys. Res. Atmos.* **103** 10757–67
Yin H *et al* 2019 FTIR time series of stratospheric NO_2 over Hefei, China, and comparisons with OMI and GEOS-Chem model data *Opt. Express* **27** A1225–40
Yin H *et al* 2020 Ground-based FTIR observation of hydrogen chloride (HCl) over Hefei, China, and comparisons with GEOS-Chem model data and other ground-based FTIR stations data *Opt. Express* **28** 8041–55
Yin H, Liu C, Hu Q, Liu T, Wang S, Gao M, Xu S, Zhang C and Su W 2021 Opposite impact of emission reduction during the COVID-19 lockdown period on the surface concentrations of $PM_{2.5}$ and O_3 in Wuhan, China *Environ. Pollut.* **289** 117899
Young P J *et al* 2018 Tropospheric ozone assessment report: assessment of global-scale model performance for global and regional ozone distributions, variability, and trends *Elem. Sci. Anth.* **6**
Zheng B *et al* 2018 Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions *Atmos. Chem. Phys.* **18** 14095–111
Zheng B, Zhang Q, Geng G, Chen C, Shi Q, Cui M, Lei Y and He K 2021 Changes in China's anthropogenic emissions and air quality during the COVID-19 pandemic in 2020 *Earth Syst. Sci. Data* **13** 2895–907
Zheng B, Zhang Q, Geng G, Shi Q, Lei Y and He K 2020 Changes in China's anthropogenic emissions during the COVID-19 pandemic *Earth Syst. Sci. Data Discuss.* **2020** 1–20