



Contrasting chemical environments in summertime for atmospheric ozone across major Chinese industrial regions: the effectiveness of emission control strategies

Zhenze Liu¹, Ruth M. Doherty¹, Oliver Wild², Michael Hollaway^{2,a}, and Fiona M. O'Connor³

¹School of GeoSciences, The University of Edinburgh, UK

²Lancaster Environment Centre, Lancaster University, UK

³Met Office Hadley Centre, UK

^anow at: Centre for Ecology & Hydrology, Lancaster Environment Centre, UK

Correspondence: Zhenze Liu (zhenze.liu@ed.ac.uk)

Abstract. The UKCA chemistry-climate model is used to quantify the differences in chemical environment for surface O₃ for six major industrial regions across China in summer 2016. We first enhance the UKCA gas-phase chemistry scheme by incorporating reactive VOC tracers that are necessary to represent urban and regional-scale O₃ photochemistry. We demonstrate that the model with the improved chemistry scheme captures the observed magnitudes and diurnal patterns of surface O₃ concentrations across these regions well. Simulated O₃ concentrations are highest in Beijing and Shijiazhuang on the North China Plain and in Chongqing, lower in Shanghai and Nanjing in the Yangtze River Delta, and lowest in Guangzhou in the Pearl River Delta despite the highest daytime O₃ production rates in Guangzhou. NO_x/VOC and H₂O₂/HNO₃ ratios indicate that O₃ production across all regions except Chongqing is VOC limited. We confirm this by constructing O₃ response surfaces for each region changing NO_x and VOC emissions and further contrast the effectiveness of measures to reduce surface O₃ concentrations. In VOC limited regions, reducing NO_x emissions by 20 % leads to a substantial O₃ increase (11 %) in Shanghai. We find that reductions in NO_x emissions alone of more than 70 % are required to decrease O₃ concentrations across all regions. Reductions in VOC emissions alone of 20 % produce the largest decrease (- 11 %) in O₃ levels in Shanghai and Guangzhou and the smallest decrease (- 1 %) in Chongqing. These responses are substantially different from those currently found in highly populated regions in other parts of the world, likely due to higher NO_x emission levels in these Chinese regions. Our work provides an assessment of the effectiveness of emission control strategies to mitigate surface O₃ pollution in these major industrial regions, and emphasizes that combined NO_x and VOC emission controls play a pivotal role in effectively offsetting high O₃ levels. It also demonstrates new capabilities in capturing regional air pollution that will permit this model to be used for future studies of regional air quality-climate interactions.

1 Introduction

Surface ozone (O₃) has become the main cause of atmospheric pollution in the summertime in China since 2013 and is particularly severe in industrial areas of China such as the North China Plain (NCP), the Yangtze River Delta (YRD), the Pearl River Delta (PRD) and the Sichuan basin where precursor emissions are high (Li et al., 2019a). The 90th percentile of the



maximum daily 8-hour average (MDA8) O_3 concentration in 30 of 74 major cities of China exceeded the National Ambient Air Quality Standard (100 ppb) in summer, 2017 (Wang et al., 2017; Lu et al., 2018; Silver et al., 2018; Li et al., 2019b; Lu et al., 2019a). During 2013-2017, the national Air Pollution Prevention and Control Action Plan has successfully reduced emissions of fine particulate matter ($PM_{2.5}$) and nitrogen oxides ($NO_x = NO + NO_2$) in China by 33 % and 21 %, respectively (Zheng et al., 2018). However, the reduction in NO_x emissions has led to an increase in O_3 levels in polluted areas due to the non-linear chemistry of O_3 and reduced titration of O_3 by NO (Li et al., 2019a; Wang et al., 2019). Volatile organic compounds (VOCs) are also important O_3 precursors and emissions of these have increased across China over the same period, exacerbating O_3 pollution (Zheng et al., 2018). VOC emissions are believed to have decreased in megacity regions such as Beijing (Cheng et al., 2019), but the resulting O_3 decrease is likely to have been offset by the O_3 increase due to reduced NO_x emissions. This poses a challenge to mitigate surface O_3 pollution. Therefore, the balance of emission controls on NO_x and VOC is critical to decreasing O_3 levels in these regions.

O_3 is a secondary photochemical pollutant in the troposphere that can be produced rapidly through oxidation of its precursors NO_x , VOCs and carbon monoxide (CO) in the presence of sunlight. Power plants, industry, residences, and transport are the main anthropogenic sources of NO_x and VOC emissions (Monks et al., 2015; Li et al., 2018a). Isoprene is the principal biogenic VOC and is released from trees, plants and crops (Sindelarova et al., 2014). O_3 formation is mainly initiated through oxidation of VOC species by hydroxyl radicals (OH). The resulting organic peroxy radicals (RO_2) and hydroperoxyl radicals (HO_2) can convert NO to NO_2 without destroying O_3 . O_3 is then created from the combination of $O(^3P)$ atoms, formed from photolysis of the resulting NO_2 , and oxygen (O_2) (Sillman, 1999; von Schneidmesser et al., 2015; Wang et al., 2017). Under low NO_x conditions, HO_2 radicals may combine to produce hydrogen peroxide (H_2O_2). However, at high NO_x concentrations, nitric acid (HNO_3) and organic nitrates (RO_2NO_2) are easily formed as NO_2 reacts with OH and RO_2 . HNO_3 and RO_2NO_2 are the main sinks of radicals and are readily removed from the atmosphere by deposition. Therefore, increasing NO_x concentrations increase O_3 production, but also accelerate the formation of HNO_3 and RO_2NO_2 , leading to less efficient O_3 formation. In addition, direct titration of O_3 by NO becomes increasingly important at higher levels of NO_x . There is hence a transition in the magnitude of O_3 production from low to high NO_x conditions. This turnover is dependent on the local chemical environment, and in particular on the relative abundance of NO_x and VOCs (Sillman, 1995; Kleinman et al., 1997; Thornton et al., 2002; Kleinman et al., 2005; Sillman and West, 2009).

A variety of O_3 sensitivity indicators have been proposed to characterise the O_3 response to changing precursor emissions. The simplest of these are based on the concentration ratios of the precursors, $NO_x/VOCs$, or of their oxidation products, H_2O_2/HNO_3 (Sillman, 1995). O_3 concentrations increase with NO_x emissions and are not sensitive to VOC emissions in a NO_x limited regime when NO_x concentrations are relatively low (Sillman et al., 1990). However, in a VOC limited regime, O_3 levels may increase with decreasing NO_x emissions, which is common in urban areas with high NO_x emissions, and this is reflected in high NO_x/VOC or low H_2O_2/HNO_3 ratios. Critical values of these indicators of O_3 sensitivity vary by region and by season (Sillman, 1995; Liu et al., 2010; Xing et al., 2019). Most major industrial regions in China are believed to be VOC limited and rural areas are NO_x limited or in a transition regime (Jin and Holloway, 2015; Wang et al., 2017). O_3 production efficiency (OPE) is another important metric to evaluate the impacts of NO_x emissions on O_3 concentrations (Liu



et al., 1987; Kleinman et al., 2002). This is defined as the number of O₃ molecules produced per molecule of NO_x oxidised. Low OPE values are typically associated with high NO_x conditions and indicate that there is less O₃ produced from a given amount of NO_x. OPE values generally increase as NO_x emissions decrease, reflecting greater O₃ production per molecule of NO_x oxidised at lower NO_x levels.

In this study, we develop new capabilities in a global scale model by incorporating higher VOC chemistry, allowing the model to represent the oxidation environment in major industrialised regions in China. We focus on the spatial and temporal variation of daytime O₃ in this study. We first evaluate the performance of this global chemistry-climate model in simulating regional O₃ across large industrialised regions. We use O₃ sensitivity indicators to compare and contrast the chemical oxidative environment across these different regions in China to identify emission control measures that would be most beneficial to reduce O₃ pollution levels. Using a global model novelly allows us to compare the impact of emission control measures in China with those in other major industrialised regions across the world. The value of this approach is that the same model set-up can be used to assess the impact of future emission and climate scenarios, studies of tropospheric and stratospheric O₃ influences and comparisons of O₃ in different parts of world.

The configuration of the model used in this study is described in section 2, along with its development and application to surface O₃ in China. We evaluate the model performance in reproducing the diurnal cycles of surface O₃ and NO₂ in section 3, and we investigate the O₃ chemical environment in China, including O₃ precursor concentrations and sensitivity ratios in section 4. We calculate the local O₃ production rates, O₃ loss rates, NO_x loss rates and OPE in section 5. We then quantify the O₃ responses to changing NO_x and VOC emissions in these regions and investigate the requirements of emission controls to reduce O₃ levels in each region in sections 6 and 7. To provide a global context we compare and contrast the effectiveness of emission control strategies with that in other parts of the world in section 7 and present our conclusions in section 8.

2 Materials and methods

2.1 Model description, development and application

The United Kingdom Chemistry and Aerosols (UKCA) model is a state-of-the-art chemistry and aerosol model that simulates atmospheric composition from the troposphere to the upper stratosphere. It is coupled to the Met Office Hadley Centre's Global Environment Model (HadGEM) family of climate models, all of which are based on the UK Unified Model (MetUM) (O'Connor et al., 2014). It is also the atmospheric composition component of the UK Earth System Model (UKESM) (Sellar et al., 2019). Version 10.6.1 of UKCA is used in this study, coupled with the Global Atmosphere 7.1 (GA7.1) configuration (Walters et al., 2019) of HadGEM3 (Hewitt et al., 2011). The spatial resolution is N96L85 with 1.875° by longitude and 1.25° by latitude, and there are 85 terrain-following hybrid height layers with a model top at 85 km. The model time step is 20 minutes for meteorology, and chemistry is calculated every hour. Wind speed and temperature are nudged with ERA-interim reanalyses from the European Centre for Medium-Range Weather Forecasts (ECMWF) every 6 hours (Dee et al., 2011). Sea surface temperature and sea ice fields are prescribed with the climatology mean of 1995-2004 (Reynolds et al., 2007).



90 The Stratosphere-Troposphere (Strat-Trop) gas-phase chemical scheme is used to simulate the inorganic odd oxygen (O_x),
hydrogen ($HO_x = OH + HO_2$) and NO_x chemical cycles, oxidation of CO and VOCs, chlorine and bromine chemistry, and
heterogeneous processes on aerosols (Archibald et al., 2020). The Global Model of Aerosol Processes (GLOMAP) aerosol
scheme is used with a two-moment pseudo-modal aerosol dynamics approach to simulate sulfate, sea-salt, dust, black carbon
and both primary and secondary organic aerosol (Mann et al., 2010). Interactive photolysis is represented with Fast-JX which
95 derives photolysis rates between 177 and 750 nm (Neu et al., 2007).

Global chemistry-climate models typically include simplified gas-phase chemistry schemes representing a limited number
of species to mitigate high computational demands. Major long-lived VOC species are selected and more reactive VOC species
are typically omitted from the chemistry scheme (Young et al., 2018). Eight discrete emitted VOC species (formaldehyde
(HCHO), ethane (C_2H_6), propane (C_3H_8), acetaldehyde (CH_3CHO), acetone ($(CH_3)_2CO$), methanol (CH_3OH), isoprene
100 (C_5H_8) and monoterpene ($C_{10}H_{16}$) are simulated in the Strat-Trop chemistry scheme of UKCA. This selection is appropriate
for simulating the global burden of O_3 but is less suitable for simulating O_3 concentrations in high emission areas. In industrial
regions of China, large abundances of more reactive VOCs such as alkenes and aromatics make substantial contributions to
 O_3 production (Wu and Xie, 2017; Tan et al., 2019; Liu et al., 2020). To address this, we incorporate more reactive classes
of VOC including alkenes, higher alkanes and aromatics, represented by propene (C_3H_6), butane (C_4H_{10}) and toluene (C_7H_8)
105 respectively in the chemistry scheme (Atkinson et al., 2006; Folberth et al., 2006). This permits a more realistic simulation
of photochemically active environments, and allows rapid O_3 formation in high VOC emission regions to be captured. The
improved chemistry scheme includes 101 species, 244 bimolecular reactions, 26 uni- and termolecular reactions, 70 photolytic
reactions, 5 heterogeneous reactions and 3 aqueous phase reactions for the sulfur cycle.

We perform model simulations for 2016 and focus our results on summer (June-July-August, JJA). We spin up the model
110 for 4 months and then simulate the full year nudged with ERA-interim reanalysis data for 2016. The new capabilities of the
model allow us to investigate regional O_3 chemical environment in industrial regions of China in the model. The relatively
coarse resolution of the model may lead to biases in surface O_3 associated with numerical diffusion (Wild and Prather, 2006;
Stock et al., 2014; Fenech et al., 2018; Mertens et al., 2020), but we note that the lifetime of O_3 makes it a regional-scale
pollutant except very close to high emission sources (Valari and Menut, 2008; Hodnebrog et al., 2011; Biggart et al., 2020).
115 This study demonstrates the first application of this improved chemistry scheme to high emission regions worldwide, and lays
the foundation for more detailed studies of the interactions between air quality and climate in a global chemistry-climate model
under future scenarios.

2.2 Emissions

The anthropogenic emission inventory of Hemispheric Transport of Air Pollution (HTAP) for 2010 is used for the globe
120 outside China (Janssens-Maenhout et al., 2015). The Multi-resolution Emission Inventory for China (MEIC) is used to provide
emissions over China for 2013 (Li et al., 2017). We apply independent diurnal and vertical profiles to each emission sector
(industry, power plants, transport and residences) according to European Monitoring and Evaluation Programme (EMEP)
emissions (Bieser et al., 2011; Mailler et al., 2013). Biogenic VOC (BVOC) emissions are calculated interactively through



the Joint UK Land Environmental Simulator (JULES) land-surface scheme in UKCA (Pacifico et al., 2011). The Global
125 Fire Emissions Database (GFED4) are used for biomass burning emissions (van der Werf et al., 2010). Other aspects of the
emissions used are as described in Archibald et al. (2020).

Given the rapid changes in anthropogenic emissions across China, we adjust NO_x , VOCs, CO, sulphur dioxide (SO_2), black
carbon (BC) and organic carbon (OC) emissions in MEIC from 2013 to 2016 by applying national and urban emission scaling
factors. NO_x emissions decreased by 18.8 % and VOC emissions increased slightly by 1.1 % between 2013 and 2016 across
130 China (Zheng et al., 2018). NO_x and VOC emissions are estimated to have decreased by 24.2 % and 12.8 % respectively in
Beijing and surrounding areas between 2013 and 2016 (Cheng et al., 2019). We apply the Beijing scaling factors to major
industrialised regions, reflecting tighter emission controls in these developed urban regions, and use national scaling factors
across the rest of the country.

2.3 Selected regions and observations

135 We focus on six heavily populated regions with high emissions within the major industrialised regions in China. These include
Beijing and Shijiazhuang on the North China Plain (32 – 40 °N, 114 – 121°E), Shanghai and Nanjing in the Yangtze River
Delta (28 – 33 °N, 118 – 123 °E), Guangzhou in the Pearl River Delta (21 – 25 °N, 111 – 115 °E) and Chongqing in the Sichuan
Basin (28 – 32 °N, 103 – 108 °E), see Fig. 1. Anthropogenic NO_x and VOC emissions are high in these regions (Fig. 2) due
to rapid industrialisation, urbanization and socio-economic development. Model grid cells that include observation stations
140 located in each of the urban and rural regions are selected to be representative of these regions, see Table 1. For comparison
with observations, we calculate a grid-weighted mean according to the number of measurement sites in each model grid cell
for the region.

We use observed hourly concentrations of air pollutants including O_3 and NO_2 from the surface monitoring networks of
China, obtained from the public website <https://quotsoft.net/air/> which mirrors data from the Chinese National Environmental
145 Monitoring Centre (CNEMC) <http://www.cnemc.cn/>. There are 1,670 national measurement stations that started operating in
2013.

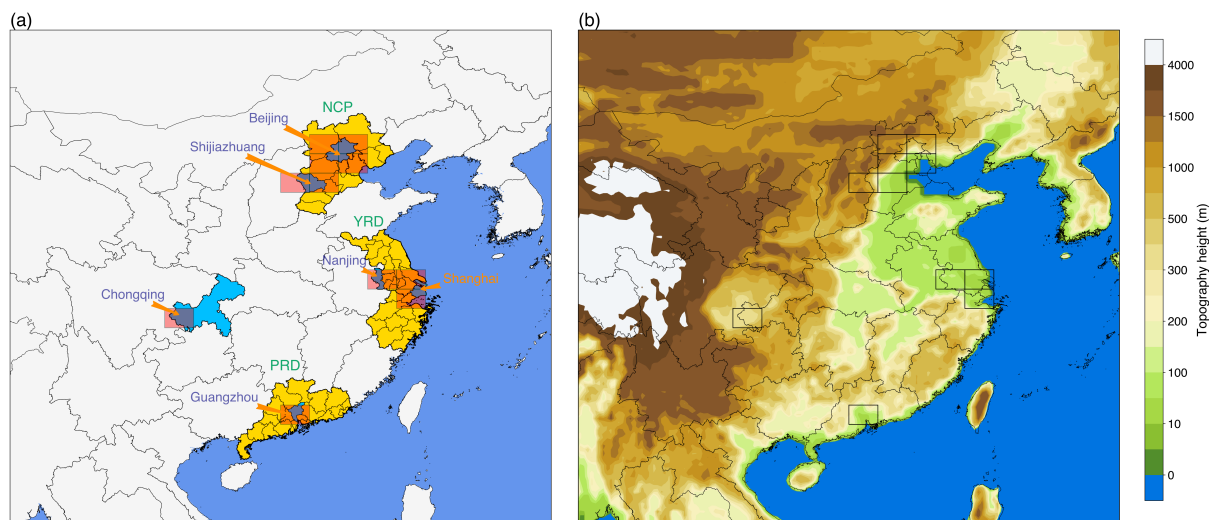


Figure 1. Map of China showing (a) the key provinces (yellow) representing the NCP, the YRD and the PRD and locations of the six regions (blue) – Beijing, Shijiazhuang, Shanghai, Nanjing, Guangzhou and Chongqing, and UKCA model grid cells co-located with these regions (red) (b) elevations across the whole of China.

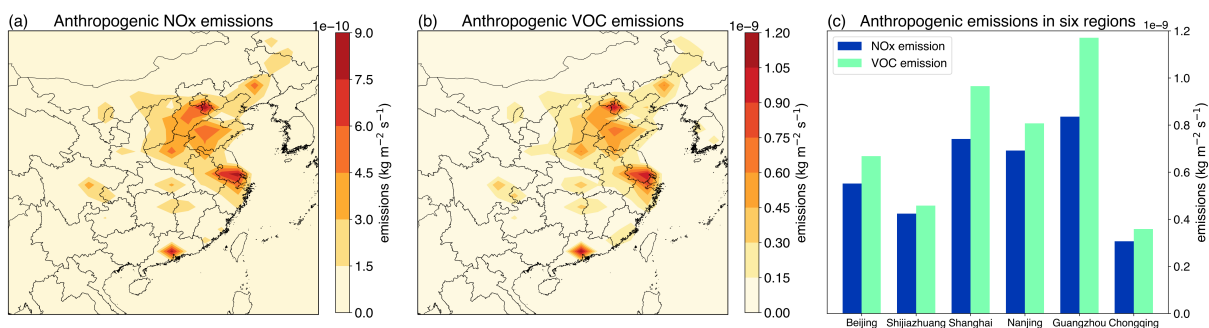


Figure 2. Spatial distributions of anthropogenic NO_x and VOC emissions (kg m⁻² s⁻¹) across China (a, b) and grid-weighted averaged emissions for the six regions within the four major industrialised regions (c) in JJA, 2016.



Table 1. Number of measurement sites and grid cells in the six industrial regions

Region	No. of measurement sites	No. of grid cells
Beijing	46	4
Shijiazhuang	28	2
Shanghai	58	2
Nanjing	45	1
Guangzhou	45	1
Chongqing	25	1

3 Model evaluation of surface O₃ and NO₂

We evaluate the diurnal variation in simulated surface O₃ and NO₂ concentrations against summertime observations for JJA, 2016 for the six industrialised regions (Fig. 3, 4). In general, the diurnal variation of observed O₃ is matched relatively well and the correlation coefficients are relatively high, see Table 2. Mean concentrations for O₃ and NO₂ over the lowest three model layers (from the surface up to 130 m) are also compared with observations. In the daytime, differences between the surface and three lowest layers are small due to efficient mixing in the planetary boundary layer (PBL). The height of the nocturnal PBL is typically underestimated in the model leading to overestimated NO_x concentrations and hence underestimated O₃ concentrations at nighttime due to excessive O₃ titration by NO (André et al., 1978; Petersen et al., 2019; Zhao et al., 2019). Fig. 3a shows a large difference in nighttime O₃ concentrations across the three layers, reflecting stable conditions that allow NO_x to accumulate at the surface. Simulated surface O₃ concentrations therefore tend to be underestimated at nighttime. In contrast, the peaks in daytime O₃ concentrations are captured relatively well, reflecting efficient O₃ production in the high VOC environment.

Daily mean O₃ concentrations for Beijing, Shijiazhuang, Shanghai and Guangzhou are reproduced well with small biases (~10 %; see Table 2). Simulated daily mean O₃ concentrations are highest (> 40 ppb) for Beijing, Shijiazhuang and Chongqing, lower in Shanghai and Nanjing (< 40 ppb), and lowest for Guangzhou (~30 ppb). Although daily mean O₃ concentrations are captured relatively well, as seen in Fig. 3a and 4a, daytime maximum O₃ concentrations are overestimated, associated with underestimated NO₂ concentrations. This overestimation is largest in Shijiazhuang where the underestimation of daytime NO₂ concentrations is larger than other regions. We find that there is a systematic bias in Chongqing where simulated O₃ levels are higher than observations. Chongqing is a mountainous inland region with complex topography that cannot be fully resolved, and surface O₃ here is thus representative of higher surface altitudes leading to a systematic bias high compared with observations (Su et al., 2018), and a corresponding bias low for NO₂ concentrations. In addition, simulated O₃ increases from biogenic emissions in the Sichuan basin are much larger in summertime than other regions (Lu et al., 2019b), and uncertainty in these emissions may contribute to the biases.

The diurnal patterns in NO₂ concentrations can also be captured as reflected by high levels at nighttime and low levels in the daytime for all regions. Daytime NO₂ concentrations can be reproduced relatively well, with a small underestimation. While



175 this may reflect underestimated NO_x emissions, it is more likely to arise from the effects of dilution on NO_x . High emissions in these regions are diluted over a large grid cell, resulting in lower NO_2 concentrations in the daytime. This is offset by high NO_2 concentrations in the PBL at nighttime as discussed above. The diurnal variation of NO_2 concentrations is hence stronger in the simulations than the observations (Fig. 4a).

180 Fig. 3 and 4 also show the frequency distribution of observed and modelled hourly O_3 and NO_2 concentrations. The simulated peaks in the distributions of O_3 and NO_2 are underestimated compared to observations for all six regions, reflecting the larger diurnal variation in the simulations. The diurnal variation is more closely simulated for O_3 concentrations (correlation coefficient $r > 0.7$) than for NO_2 concentrations. The Chongqing region has the closest correlation with observations ($r = 0.83$), which provides evidence that the overestimation of O_3 is systematic as suggested earlier. Overall, the magnitudes (see Table 2) and diurnal patterns (see Fig. 3 and 4) of both species can be simulated reasonably well, with differences between industrial regions clearly captured.

Table 2. Comparison of modelled and observed daily mean surface O_3 and NO_2 concentrations for the six industrial regions in JJA, 2016, China.

Region	Obs. (ppb)	Sim. (ppb)	Bias ppb/%	RMSE (ppb)	Correlation r
O_3					
Beijing	47.7±22.1	43.4±27.7	-4.4 (9.1%)	8.1	0.77
Shijiazhuang	42.9±18.4	47.6±28.7	4.7 (10.9%)	11.6	0.78
Shanghai	38.3±17.5	34.4±28.8	-3.9 (10.2%)	12.7	0.77
Nanjing	42.6±18.9	35.9±24.9	-6.8 (15.8%)	9.8	0.71
Guangzhou	29.8±18.3	28.0±25.9	-1.8 (-6.1%)	9.4	0.81
Chongqing	38.1±19.2	56.0±31.3	18.0 (47.2%)	22.3	0.83
NO_2					
Beijing	17.8±3.7	20.7±8.2	2.9 (16.2%)	5.8	0.69
Shijiazhuang	18.1±4.7	16.7±8.3	-1.4 (7.7%)	4.3	0.76
Shanghai	16.3±2.3	26.1±8.7	9.8 (60.0%)	12.1	0.50
Nanjing	17.2±3.6	21.3±9.0	4.1 (23.7%)	7.8	0.49
Guangzhou	16.1±3.0	19.9±9.3	3.8 (23.7%)	8.4	0.55
Chongqing	17.4±3.8	10.9±6.3	-6.4 (37.1%)	8.0	0.43

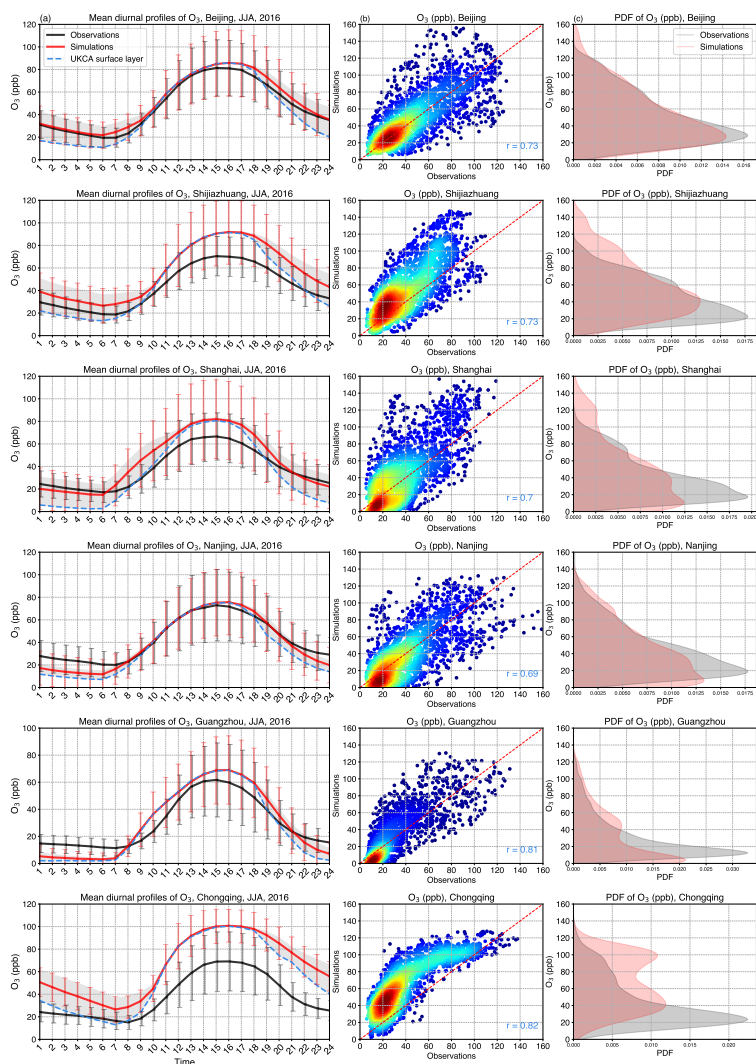


Figure 3. Comparison of observed and modelled O_3 concentrations for the six industrialised regions in JJA, 2016, China. **(a)** Mean diurnal cycles of observed and modelled O_3 concentrations (ppb). The shaded area represents the spread across the lowest three model layers. Error bars denote one standard deviation of hourly O_3 concentrations across all days **(b)** Observed and modelled hourly O_3 concentrations (ppb; three lowest model layers) and correlation coefficient values **(c)** PDF of O_3 concentrations (ppb) for modelled and observed hourly values.

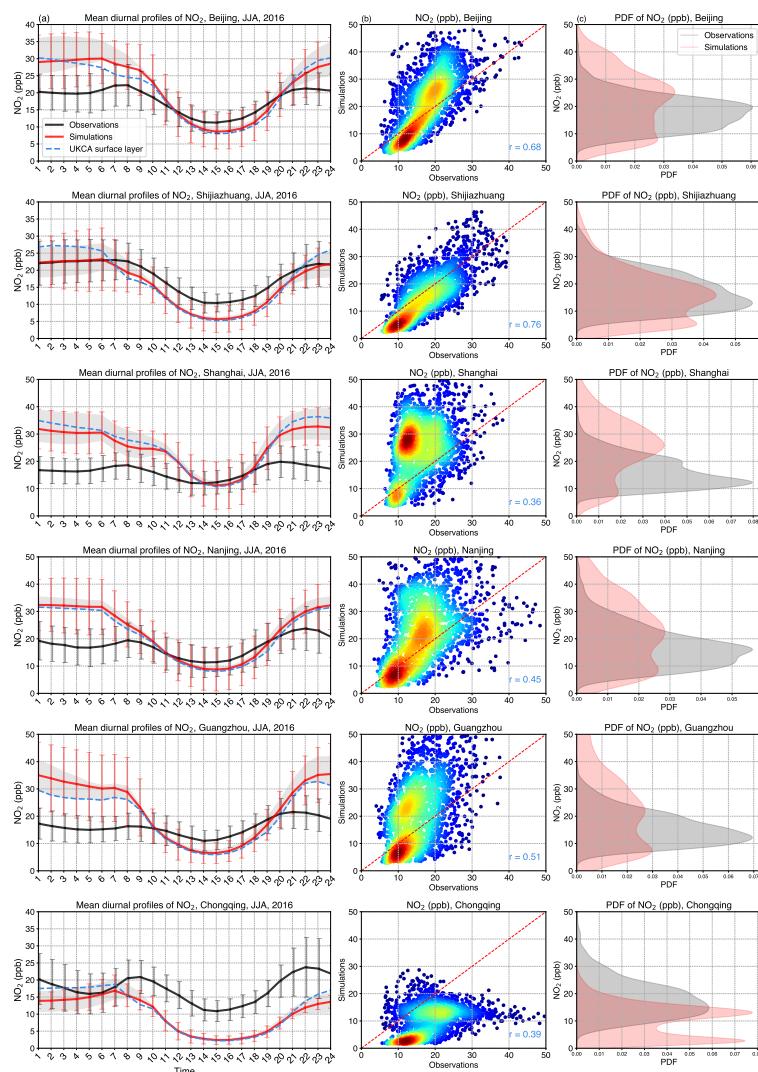


Figure 4. Comparison of observed and modelled NO_2 concentrations for the six industrialised regions in JJA, 2016, China. **(a)** Mean diurnal cycles of observed and modelled NO_2 concentrations (ppb). The shaded area represents the spread across the lowest three model layers. Error bars denote one standard deviation of hourly NO_2 concentrations across all days. **(b)** Observed and modelled hourly NO_2 concentrations (ppb; three lowest model layers) and correlation coefficient values **(c)** PDF of NO_2 concentrations (ppb) for modelled and observed hourly values.

4 Differences in chemical environment

Spatial distributions of modelled daytime concentrations of O_3 , NO_x , VOCs and O_3 sensitivity ratios (NO_x/VOCs and $\text{H}_2\text{O}_2/\text{HNO}_3$) are shown in Fig. 5 to illustrate the differences in chemical environment for the six regions. We focus on daytime hours with the highest O_3 concentrations using the MDA8 metric and consider this same time period for other species, which we refer to

hereafter as daytime concentrations. For the sensitivity ratio NO_x/VOCs we consider the sum of anthropogenic and biogenic daytime VOC concentrations.

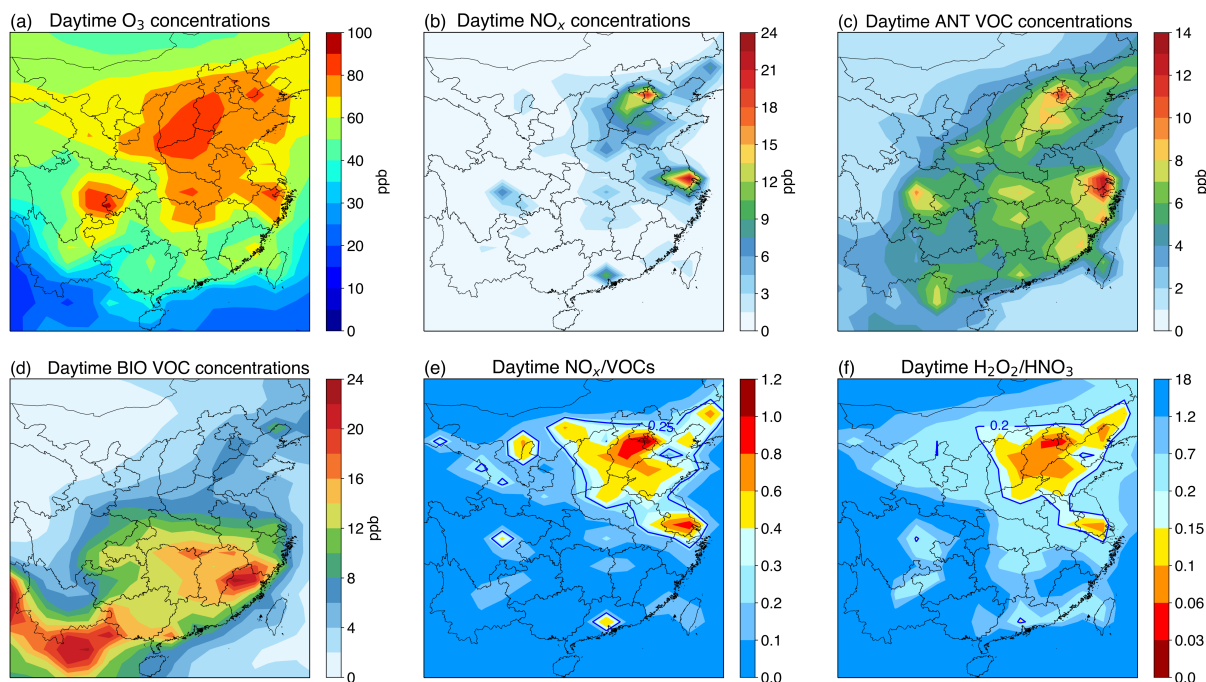


Figure 5. Spatial distributions of simulated surface daytime O_3 , NO_x , anthropogenic VOCs, biogenic VOCs (ppb) (a, b, c, d) and two O_3 sensitivity ratios – NO_x/VOCs and $\text{H}_2\text{O}_2/\text{HNO}_3$ (e, f) in JJA, 2016, China.

Figure 5a shows high daytime O_3 levels (> 80 ppb) across northern China, eastern China and the Sichuan basin in JJA, 2016. O_3 levels in the PRD (~ 40 ppb) are much lower despite high emissions likely due to transport of clean air from the South China Sea associated with the East Asian summer monsoon (Zhao et al., 2010; Li et al., 2018b). Areas with high anthropogenic NO_x and VOC concentrations generally coincide with high emission regions (Fig. 2, Fig. 5b, 5c). High daytime NO_x concentrations (> 12 ppb) are simulated in Beijing and Shijiazhuang, Shanghai and Nanjing. Chongqing has the lowest NO_x concentrations of 3–6 ppb due to relatively low NO_x emissions. High anthropogenic daytime VOCs concentrations are simulated across the main industrial regions, in particular in Shanghai with the highest levels (> 12 ppb; Fig 5c). 195

The distribution of biogenic VOC concentrations differs from that of anthropogenic VOCs (Fig. 5c, 5d). There is a strong latitudinal gradient, reflecting differences in climate and the spatial distribution of vegetation (Li et al., 2013). The highest biogenic VOC levels (> 16 ppb) are simulated in south-eastern China where deciduous and mixed broadleaf trees are the main source of biogenic VOCs. The YRD, the PRD and the Sichuan basin have higher biogenic VOC concentrations than the NCP. Chongqing has the highest biogenic VOC levels (> 12 ppb) of the regions considered here. However, highest biogenic VOC levels are found south of China in Laos, Vietnam and Cambodia. 200



High NO_x/VOC ratios and low $\text{H}_2\text{O}_2/\text{HNO}_3$ ratios typically indicate VOC limited O_3 production. The transition between VOC and NO_x limited regimes is typically about 0.25 for the NO_x/VOC ratio and about 0.2 for the $\text{H}_2\text{O}_2/\text{HNO}_3$ ratio (Liu et al., 2010; Xing et al., 2019). From these two thresholds for the O_3 sensitivity ratios, it can be seen that VOC limited regions cover most areas of the NCP, parts of the YRD including Shanghai and Nanjing, and Guangzhou in the PRD (Fig. 5e, 5f). All six regions except Chongqing have NO_x/VOCs ratios ≥ 0.6 and $\text{H}_2\text{O}_2/\text{HNO}_3$ ratios ≤ 0.18 (Table 3). This suggests that these regions have a chemical environment that is strongly VOC limited. In addition, VOC limited regimes shown by both indicators are quite similar, showing that these two O_3 sensitivity ratios may be useful to directly diagnose different O_3 sensitivity regimes in China. Regions with high NO_x/VOC ratios and low $\text{H}_2\text{O}_2/\text{HNO}_3$ ratios typically occur where NO_x concentrations are high. Overall, these transition values delineate the different O_3 sensitivity regions across China well, showing VOC limited regimes in the major industrial regions with high emissions. However, we note that these O_3 sensitivity ratios only provide an estimate of the chemical environment, and further, more detailed investigation of O_3 responses to emission changes are required.

Table 3. Simulated surface daytime concentrations of species, radicals, O_3 sensitivity ratios and the photolysis rate $j(\text{O}^1\text{D})$ for the six industrial regions in JJA, 2016, China.

Region	Beijing	Shijiazhuang	Shanghai	Nanjing	Guangzhou	Chongqing
Species (ppb)						
O_3	78.0	83.5	70.1	66.8	60.2	93.8
NO_x	12.8	8.7	19.2	12.9	10.7	3.8
VOC (ANT)	8.7	7.0	12.7	7.6	7.5	7.7
VOC (BIO)	5.5	4.3	10.6	9.2	10.2	13.5
VOC (Total)	14.3	11.3	23.3	16.9	17.7	21.3
Sensitivity ratios						
NO_x/VOCs	0.79	0.73	0.89	0.78	0.60	0.18
$\text{H}_2\text{O}_2/\text{HNO}_3$	0.18	0.08	0.10	0.11	0.09	0.29
Radicals						
$\text{OH} / 10^6 \text{ cm}^{-3}$	7.8	10.3	8.4	9.8	13.0	16.6
$\text{HO}_2 / 10^8 \text{ cm}^{-3}$	2.6	2.9	2.3	2.2	2.2	7.4
$\text{RO}_2 / 10^8 \text{ cm}^{-3}$	1.0	0.9	0.8	0.8	0.9	2.5
Photolysis rate						
$j(\text{O}^1\text{D}) / 10^{-5} \text{ s}^{-1}$	2.3	2.6	2.3	2.5	3.1	3.4

5 Differences in local O_3 production rates

In this section, we calculate the daytime production rates for surface O_3 to investigate how the local O_3 production compares across the six regions. We define the net O_3 production rate (ppb/h) as the gross rate of production $P(\text{O}_3)$ from the reactions



$\text{HO}_2 + \text{NO}$ and $\text{RO}_2 + \text{NO}$ minus the gross rate of loss $L(\text{O}_3)$ from the reactions $\text{O}(^1\text{D}) + \text{H}_2\text{O}$, $\text{O}_3 + \text{OH}$, $\text{O}_3 + \text{HO}_2$ and $\text{O}_3 + \text{VOCs}$. We assume that the pathways above represent the net O_3 production rate under O_3 photochemical steady state between NO and NO_2 , and are shown as follows:

$$\text{Net } P(\text{O}_3) = P(\text{O}_3) - L(\text{O}_3) =$$

$$k_1[\text{HO}_2][\text{NO}] + k_2[\text{RO}_2][\text{NO}] - (k_3[\text{O}(^1\text{D})][\text{H}_2\text{O}] + k_4[\text{O}_3][\text{OH}] + k_5[\text{O}_3][\text{HO}_2] + k_6[\text{O}_3][\text{VOCs}]) \quad (1)$$

where k_i represents the rate coefficient of reaction i .

The loss of NO_x , $L(\text{NO}_x)$, is principally determined by the reactions $\text{OH} + \text{NO}_2$ and $\text{RO}_2 + \text{NO}_2$ which produce HNO_3 and RO_2NO_2 , respectively. OPE is then defined as the number of O_3 molecules produced per molecule of NO_x consumed (Liu et al., 1987).

$$\text{OPE} = \frac{P(\text{O}_3)}{L(\text{NO}_x)} \quad (2)$$

As shown in Fig. 6, local O_3 production varies across the six regions with O_3 net production rates ranging from 4-10 ppb/h. Simulated daytime net O_3 production rates are highest (> 8 ppb/h) in Shanghai and Guangzhou mainly due to high precursor emissions, and this is reflected by higher O_3 concentrations in Shanghai than in nearby Nanjing. While O_3 production is high in Guangzhou, the O_3 concentrations are much lower than in other regions, indicating that meteorological impacts in this coastal region are important to transport O_3 produced locally. O_3 net production in Beijing and Shijiazhuang is similar to that in Nanjing (~ 5 ppb/h). O_3 production in Chongqing is also high, reflecting high radical concentrations (see Table 3) that promote O_3 production despite lower precursor emissions. High photolysis rates $j(\text{O}(^1\text{D}))$ in Chongqing and Guangzhou contribute to high concentrations of OH radicals (Table 3). O_3 destruction rates are fairly similar (< 4 ppb/h) across these regions, but are higher in Chongqing, offsetting its high O_3 production rates.

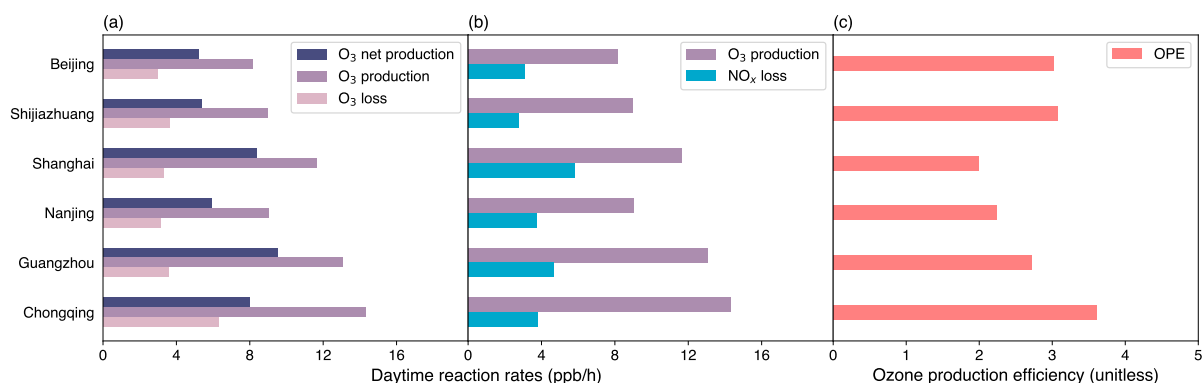


Figure 6. Simulated surface daytime (a) net O_3 production rates, gross O_3 production rates and gross O_3 loss rates (ppb/h) (b) gross O_3 production rates and NO_x loss rates (ppb/h) (c) OPE (unitless) for the six industrial regions in JJA, 2016, China.



235 The simulated NO_x loss rates (Fig. 6b) show the highest removal of NO_x in Shanghai, where NO_x concentrations are also
highest. This influences OPE, which is strongly dependent on NO_x loss, and leads to the lowest OPE in Shanghai and highest
in Chongqing (Fig. 6c). The low OPE in Shanghai and Nanjing shows the low efficiency in O_3 production per molecule of
 NO_x consumed. However, the OPE values in all six regions are generally lower than those in other remote and rural regions, in
agreement with Wang et al. (2018), indicating that high precursor emissions in these regions are the main cause of high surface
240 O_3 concentrations.

6 Response of O_3 to emission controls

We quantify the response of daytime O_3 to emission changes to investigate the relationship between the chemical environ-
ment and the magnitude of O_3 changes for the six industrial regions of China. We implement three scenarios applying 20 %
reductions in anthropogenic NO_x emissions, VOC emissions and combined NO_x and VOC emissions.

245 Spatial distributions of simulated daytime surface O_3 responses vary across China (Fig. 7). In the 20 % NO_x emission control
scenario, substantial O_3 increases (2-10 ppb) can be seen in the NCP, the YRD and the PRD, and O_3 concentrations decrease
(0-8 ppb) in the Sichuan basin. In the 20 % VOC emission control scenario, there are small O_3 changes in most non-industrial
regions of China (-1-2 ppb) but O_3 concentrations generally decrease by 1-9 ppb across the NCP, the YRD and the PRD. The
Sichuan basin shows relatively small O_3 decreases. Areas showing O_3 increases in the 20 % NO_x emission control experiment
250 match well with VOC limited areas indicated by the NO_x/VOCs and $\text{H}_2\text{O}_2/\text{HNO}_3$ ratios (cf. Fig. 5e, 5f vs Fig. 7a) suggesting
that all the industrial regions considered here are VOC limited except Chongqing in the Sichuan basin that is NO_x limited.

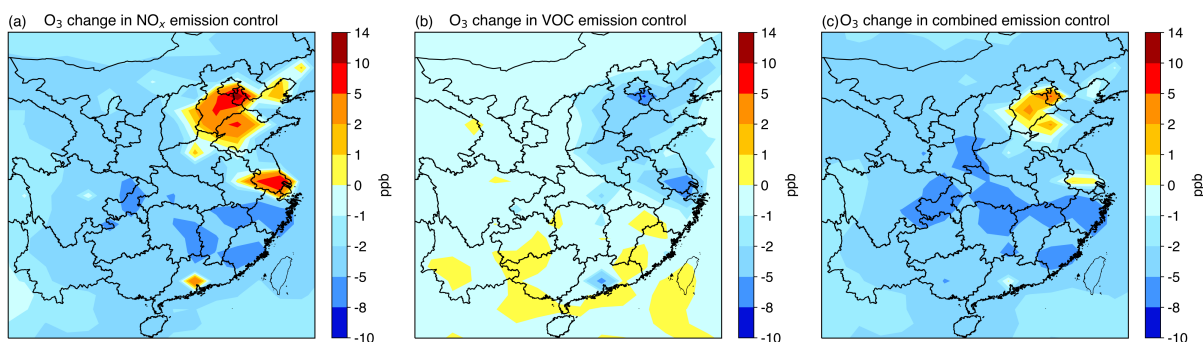


Figure 7. Spatial distributions of simulated surface daytime O_3 concentration changes (ppb) for (a) the 20 % NO_x emission control, (b) the
20 % VOC emission control and (c) the 20 % combined NO_x and VOC emission control compared to the present-day results in JJA, 2016,
China.

In general, the greatest O_3 increases in the 20 % NO_x control scenario occur in areas with high precursor concentrations.
Shanghai shows the largest O_3 increases (11 %) (Table 4) and has the highest underlying NO_x concentrations (Table 3). O_3
increases in Beijing and Guangzhou are similar (~8 %) possibly because of their similar NO_x concentrations. Shijiazhuang
255 in the NCP shows the smallest O_3 increase (4 %) because of its lower NO_x concentrations. In contrast, an O_3 decrease of



4 % is seen in Chongqing that is NO_x limited. In the 20 % VOC control scenario, the largest O_3 decreases are simulated in Shanghai and Guangzhou (-10 %) while minimal O_3 decreases (-1 %) are simulated in Chongqing. The chemical environment in Chongqing is NO_x limited and therefore the O_3 changes are not sensitive to VOC emissions.

Table 4. Simulated daytime mean O_3 concentrations and changes in NO_x , VOC and combined NO_x and VOC emission controls for the six industrial regions in JJA, 2016, China.

O_3 (ppb)/ Region	Base	NO_x control	Change (%)	VOC control	Change (%)	NO_x +VOC control	Change (%)
Beijing	78.0	84.7	8.6%	72.5	-7.0%	79.7	2.2%
Shijiazhuang	83.5	86.6	3.8%	80.2	-3.9%	84.6	1.4%
Shanghai	70.1	77.8	11.0%	63.1	-10.0%	69.4	-1.0%
Nanjing	66.8	72.4	8.5%	61.4	-8.0%	67.8	1.6%
Guangzhou	60.2	64.8	7.6%	53.8	-10.7%	60.4	0.3%
Chongqing	93.8	89.5	-4.6%	92.5	-1.4%	88.5	-5.6%

In addition to separate 20 % reductions in NO_x and VOC emissions, we demonstrate the importance of combined NO_x and VOC emission controls to mitigate O_3 pollution in VOC limited regions. This effectively offsets the higher levels of O_3 that arise with NO_x emission reductions alone. The O_3 increase in Shanghai is fully offset in the combined emission control (-1 %). While O_3 increases still occur in the other VOC limited regions, these increases are minimal (< 3 %). Reducing both NO_x and VOC emissions decreases O_3 levels in Chongqing by 6 %. Therefore, combined emission controls are necessary to efficiently mitigate O_3 pollution in all these industrial regions, and VOC emission controls should be at least as stringent as NO_x emission controls to address rising O_3 levels in these industrial regions.

7 Effectiveness of emission controls in reducing surface O_3 levels

To provide a more complete exploration of the effectiveness of emission controls, we construct a response surface of summertime daytime O_3 for each region to show the effect of changing NO_x and VOC emissions. We do this by performing a set of 64 model simulations with global anthropogenic NO_x and VOC emissions scaled independently over the range 0-140 % in increments of 20 %.

Figure 8 shows the magnitude and direction of O_3 changes in the six regions as NO_x and VOC emissions change. For context, Fig. 8a also shows the simulated daytime O_3 changes between 2013 and 2019 in the Beijing region following estimated emission changes (Cheng et al., 2019). We find that simulated O_3 concentrations in Beijing increase from 71.6 ppb in 2013 to 82.6 ppb in 2019, an increase of 1.8 ppb/year. This is consistent with observed changes of 1.9 ppb/year over this period due to anthropogenic emission changes (Li et al., 2020). The observed daytime O_3 concentrations are 83 ppb in the Beijing region in 2019. This demonstrates that the model captures not only the magnitude and diurnal pattern of O_3 in summer 2016 well but also the observed O_3 changes in recent years.

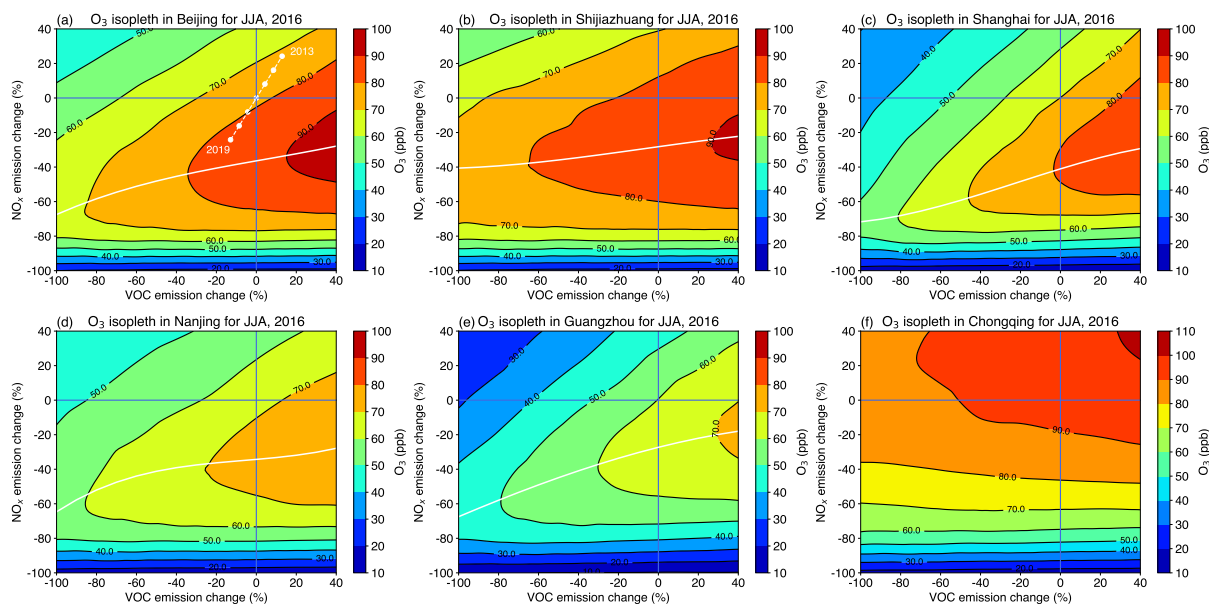


Figure 8. Simulated daytime surface O_3 responses (ppb) to anthropogenic NO_x and VOC emission changes for the six industrial regions across China (a – f) in JJA, 2016. The intersection of the vertical and horizontal lines marks current O_3 levels. White ridge lines mark the peak in O_3 concentrations for given VOC emissions, and show the approximate transition between VOC limited (above the ridge) and NO_x limited (below the ridge) regimes. White dots in (a) represent simulated daytime O_3 levels in the Beijing region in JJA between 2013 and 2019 following estimated NO_x and VOC emission changes.

The patterns of O_3 response seen in the VOC limited regions (Fig. 8a-e) are similar, such that decreases in NO_x emissions from their current levels increase O_3 concentrations. Large O_3 increases occur in Shanghai and Beijing, highlighting that it is not beneficial to reduce NO_x emissions unless VOC emissions are also reduced. Large reductions (~40 %) in NO_x emissions are required to shift the chemical environment from VOC limited to NO_x limited for these two regions. The large decrease in O_3 in Shanghai and Guangzhou when reducing VOC emissions indicates that the efficiency in lowering O_3 levels by decreasing VOC emissions is high in these regions. In contrast, the efficiency of VOC emissions alone in reducing O_3 levels is lower in Shijiazhuang and Chongqing.

Figure 9 shows the O_3 responses in each region to changes in NO_x emissions, VOC emissions and combined NO_x and VOC emissions, which represent cross-sections through the O_3 response surfaces shown in Fig 8. It is difficult to decrease O_3 concentrations in Shanghai by reducing NO_x emissions alone because there is a steep rise in surface O_3 (~15 %) when NO_x emissions are reduced by 40 % from the current state. Decreasing O_3 from current levels requires reductions in NO_x emissions of more than 50 % for Shijiazhuang and Guangzhou and more than 70 % for Beijing, Shanghai and Nanjing. This suggests that mitigating poor O_3 air quality in these VOC limited regions through NO_x emission controls alone would require much greater reductions than the 21 % reductions in NO_x emissions that are reported to have occurred in China from 2013 to 2017 (Zheng et al., 2018).

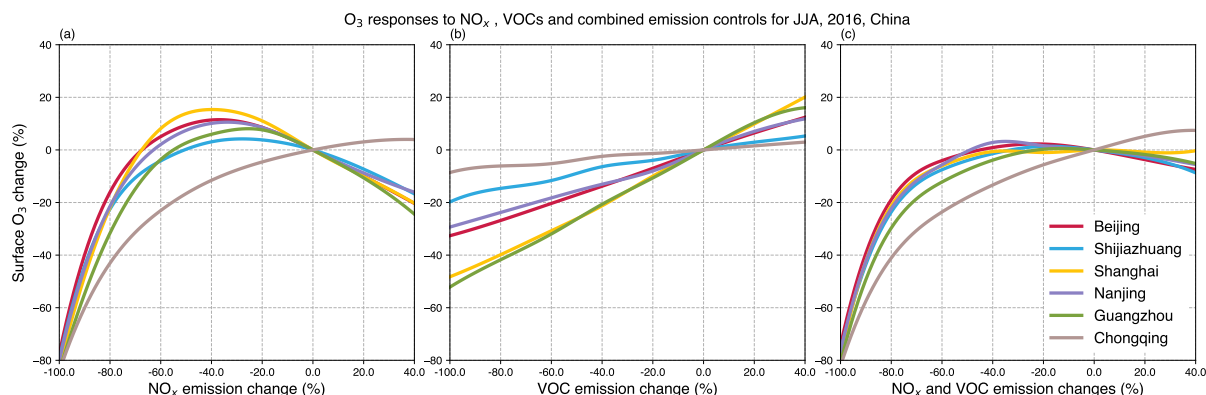


Figure 9. Simulated daytime surface O_3 responses to changes in anthropogenic emissions of (a) NO_x , (b) VOC and (c) combined NO_x and VOC emissions for the six industrial regions in JJA, 2016, China.

O_3 responses to VOC emission changes are smaller and more linear than the responses seen for NO_x emissions changes (Fig. 9a, 9b). Reducing VOC emissions by 40 % gives large decreases in O_3 concentrations (20 %) in Shanghai and Guangzhou and smaller decreases (< 10 %) in Shijiazhuang and Chongqing (Fig. 9b). Reductions in VOC emissions are key to reducing present-day O_3 concentrations as they effectively offset the rising O_3 levels due to decreasing NO_x emissions (Fig. 9c). Emission reductions of 50 % or more are required to reduce O_3 levels for all regions if controls on NO_x and VOC emissions are applied simultaneously.

To place our results in a wider global context, Figure 10 shows summer-mean surface O_3 changes over different regions with high emissions in other parts of the world compared with those in China. We consider six major industrialised regions outside of China and select the model grid-cell that is most closely co-located with the region. We note that proportional increases in summer-mean O_3 are larger than that of daytime O_3 increases when NO_x emissions are reduced (see Fig. 9), principally because absolute O_3 concentrations are smaller with the inclusion of nighttime conditions. We find that all selected high emission regions across the globe outside of China are NO_x limited, such that NO_x emissions decreases yield regional O_3 decreases. Current levels of NO_x emissions in these regions are considerably lower than for the industrial regions of China, reflecting the different O_3 sensitivity regimes (Table 5).

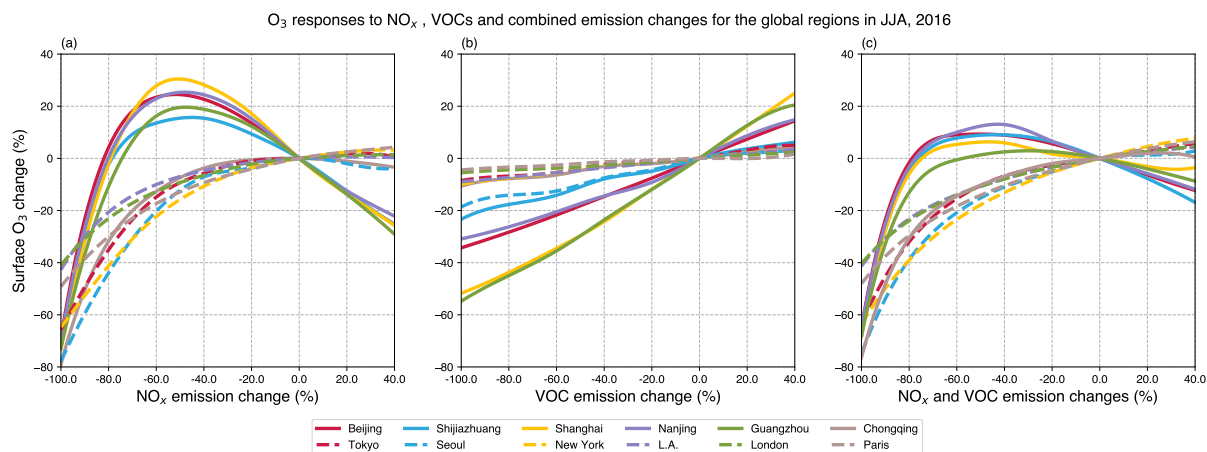


Figure 10. Simulated summertime mean surface O₃ responses to anthropogenic (a) NO_x, (b) VOC and (c) combined NO_x and VOC emission changes in regions across the globe: Tokyo, Seoul, New York, L.A., London, Paris (dashed lines) and those in major industrial regions of China (solid lines) in JJA, 2016.

Reductions of both NO_x and VOC emissions substantially decrease O₃ levels for these selected regions outside of China, and the magnitude of the O₃ decreases are similar to those found for Chongqing (Fig 10). Conversely, the magnitude of O₃ decreases when reducing VOC emissions are smaller than all five VOC limited regions in China. This indicates that O₃ concentrations are less sensitive to VOC emissions in these other world regions due to their lower VOC emissions (Table 5).

Despite lower NO_x and VOC emissions in the regions outside of China, surface O₃ concentrations, particularly in the Seoul and New York regions, are similar to those for China. This highlights that regional O₃ levels also depend on background O₃ concentrations, despite localised NO_x and VOC emissions that lead to different O₃ production regimes. The O₃ levels in European regions e.g. London and Paris are lowest, in accordance with the lowest NO_x and VOC emission levels. Overall, these results show that there are substantial differences in the efficiency of emission control scenarios to reduce surface O₃ levels in different parts of the world. For many industrial regions of China, the extended regions are VOC limited and hence, reductions of VOC emissions are the key to reducing poor O₃ air quality. For other regions selected in this study NO_x emission reductions are still pertinent to improving O₃ pollution.



Table 5. Anthropogenic NO_x and VOC emissions ($\times 10^{-10}$ kg m⁻² s⁻¹) and summertime mean surface O₃ concentrations (ppb) in regions across the industrial regions of China and the globe. MEIC emissions of 2013 adjusted for 2016 are used for Chinese industrial regions. HTAP emissions of 2010 are used for other regions of the globe.

Region	NO _x emissions	VOC emissions	O ₃ conc.
China			
Beijing	5.5	6.7	43.4
Shijiazhuang	4.2	4.6	47.6
Shanghai	7.4	9.6	34.4
Nanjing	6.9	8.1	35.9
Guangzhou	8.4	12.0	28.0
Chongqing	3.1	3.6	56.0
Global			
Tokyo	2.0	2.6	38.9
Seoul	1.5	2.1	45.5
New York	2.3	3.1	45.3
L.A.	1.1	1.3	40.1
London	1.1	1.5	30.6
Paris	0.8	1.0	32.6

8 Conclusions

320 This study presents the application of the global chemistry-climate UKCA model with an improved gas-phase chemistry
scheme including more reactive VOCs to simulate regional summertime O₃ pollution across major industrialised regions in
China for the first time. Differences in atmospheric chemical environments are investigated, and the effectiveness of different
emission control strategies in reducing O₃ concentrations is quantified. The model captures the magnitude, diurnal profiles and
diurnal variation of O₃ concentrations across most industrial regions well. We highlight that peak O₃ concentrations can be
325 captured well, indicating that O₃ production can be effectively simulated with more highly active VOC oxidation environments
for high emission regions of China.

Simulated daytime O₃ levels are highest on the North China Plain (Beijing and Shijiazhuang), and in the Sichuan Basin
(Chongqing), and are lowest in the Pearl River Delta (Guangzhou). We find that there is a systematic bias in O₃ throughout
the diurnal cycle in Chongqing reflecting the mountainous inland area that is inadequately captured by the topography in the
330 model. The O₃ production rates are highest in the Pearl River Delta compared to other regions. However, its much lower O₃
levels reflect the importance of meteorological impacts in this coastal region. OPE values in these industrial regions are low,
indicating that their high O₃ levels are mainly caused by high precursor emissions. Both O₃ sensitivity ratios we apply here
(NO_x/VOCs and H₂O₂/HNO₃) suggest that all the industrial regions except Chongqing are VOC limited.



335 A set of simulations are performed with a range of NO_x and VOC emissions to construct O_3 response surfaces to assess
the impacts of different emission control strategies in different regions. Reducing NO_x emissions alone by 20 % leads to a
substantial O_3 increase (11 %) in Shanghai. Reductions in VOC emissions alone of 20 % produce the largest decrease (-
11 %) in O_3 levels in Shanghai and Guangzhou and the smallest decrease (- 1 %) in Chongqing. We find that reducing O_3
concentrations across all industrial regions of China would require more than 70 % reductions if reducing NO_x emissions
alone, and therefore VOC emission controls are important to reduce O_3 levels. We also find that combined emission controls
340 effectively offset high O_3 levels that arise from reduced NO_x emissions alone. These responses are substantially different
from those currently found in major highly populated regions in other parts of the world. The results show NO_x limited O_3
production in these global areas, which also reflects the predominance of heavily populated VOC limited areas across the
industrial regions in China. Therefore, O_3 pollution in the industrial regions of China should be treated as a regional issue and
regional VOC emission control strategies should be considered.

345 The new capabilities for simulating regional surface O_3 pollution developed here will be helpful for future model studies
to investigate the regional O_3 impacts on climate. The magnitude of O_3 changes over recent years in the Beijing region can
be reproduced well. There remain model biases in regions with complex topography and high elevation – a common issue for
global and regional models. Another source of uncertainty is the rapid change in anthropogenic emissions in recent years in
China, which presents a particular challenge for inventory development. Recently, while NO_x emissions have been successfully
350 reduced across many regions in China, changes in VOC emissions have been relatively small, and this has led to an increase in
 O_3 concentrations in many regions. Regional VOC emission controls are hence urgently needed to maximise the effectiveness
in reducing surface O_3 pollution in China.

Data availability. The data generated in this study will be made available through the Edinburgh DataShare at <https://datashare.is.ed.ac.uk/handle/xxxxx>.

355 *Author contributions.* ZL, RD and OW designed the study. ZL, MH and FO'C set up the model. ZL ran model simulations and performed
the analysis. ZL, RD and OW prepared the paper with contributions from all co-authors.

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

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