



# Pollutant emission reductions deliver decreased PM<sub>2.5</sub>-caused mortality across China during 2015-2017

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Abstract. Air pollution is a serious environmental issue and leading contributor to the disease burden in China. Rapid reductions in fine particulate matter ( $PM_{2.5}$ ) concentrations and increased ozone concentrations have occurred across China,

- 10 during 2015 to 2017. We used measurements of particulate matter with a diameter  $< 2.5 \ \mu m \ (PM_{2.5})$  and Ozone (O<sub>3</sub>) from >1000 stations across China along with Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) regional air quality simulations, to explore the drivers and impacts of observed trends. The measured nationwide median PM<sub>2.5</sub> trend of -3.4  $\mu g m^{-3}$  year<sup>-1</sup>, was well simulated by the model (-3.5  $\mu g m^{-3}$  year<sup>-1</sup>). With anthropogenic emissions fixed at 2015levels, the simulated trend was much weaker (-0.6  $\mu g m^{-3}$  year<sup>-1</sup>), demonstrating interannual variability in meteorology played
- a minor role in the observed  $PM_{2.5}$  trend. The model simulated increased ozone concentrations in line with the measurements, but underestimated the magnitude of the observed absolute trend by a factor of 2. We combined simulated trends in  $PM_{2.5}$ concentrations with an exposure-response function to estimate that reductions in  $PM_{2.5}$  concentrations over this period have reduced  $PM_{2.5}$ -attributable premature morality across China by 150 000 deaths year<sup>-1</sup>.

# **1** Introduction

- 20 Concentrations of particulate matter and ozone across China largely exceed international air quality standards (Reddington et al., 2019; Silver et al., 2018). This poor air quality is estimated to hasten the deaths of 870 000 2 470 000 people across China each year (Apte et al., 2015; Burnett et al., 2018; Cohen et al., 2017; Gu and Yim, 2016; Lelieveld et al., 2015). The Chinese government's efforts to improve air quality began in the 1990s, but emissions of pollutants continued to increase into the 21<sup>st</sup> century and air pollution worsened (Krotkov et al., 2016; Streets et al., 2008; Zhang et al., 2012). In 2013, China experienced
- 25 episodes of severe particulate matter pollution (Zhang et al., 2016). In response, the Chinese government announced the Action Plan on the Prevention and Control of Air Pollution which focused on the reduction of fine particulate matter (PM<sub>2.5</sub>) through stringent emission controls during 2012-2017 (Zheng et al., 2017).





# 1.1 Previous studies of trends in China's air quality

Satellite remote sensing studies have been used to show large changes in air pollution across China in recent decades, with 30 positive trends in Nitrogen Dioxide (NO<sub>2</sub>) (Van der A et al., (2006), Sulphur Dioxide (SO<sub>2</sub>) (Zhang et al., 2017) and PM<sub>2.5</sub> (Ma et al., 2016) during the 1990s and early 2000s. Trends in aerosol optical depth have been used to estimate changes in PM<sub>2.5</sub>, which peaked around 2011 (Ma et al., 2016). NO<sub>2</sub> across China peaked around 2011 (De Foy et al., 2016; Irie et al., 2016), although concentrations in the Pearl River Delta (PRD) peaked earlier and western regions may have peaked later (Cui et al., 2016). Several remote sensing studies show that  $SO_2$  concentrations in China peaked around 2006 (Van Der A et al.,

- 2017; Krotkov et al., 2016; Zhang et al., 2017), matching the period of maximum emissions (Duan et al., 2016; Li et al., 2017; 35 Zheng et al., 2018). Analysis of measurements from the Acid Deposition Monitoring Network in East Asia (EANET) shows a negative pH trend (i.e., becoming more acidic) from 1999 until a reversal occurs in 2006, matching peak SO<sub>2</sub> emissions and concentrations (Duan et al., 2016). Measurements of  $O_3$  concentrations at background monitoring sites indicate positive trends in western China during 1994-2013 (Xu et al., 2016), and Taiwan during 1994-2003 (Chang and Lee, 2007), suggesting that
- 40 O<sub>3</sub> has been increasing across China during the past two decades. More recently, measurements at urban sites, also show positive O<sub>3</sub> trends during 2005-2011 (Zhang et al., 2014).

The establishment of China's air pollution monitoring network, operated by the China National Environmental Monitoring Centre (CNEMC) (Wang et al., 2015), which includes measurements from over 1600 locations, has enabled more detailed analysis of recent air pollution changes (Silver et al., 2018; Zhai et al., 2019). Between 2015 and 2017, PM25 concentrations

- across China decreased by 28% (Silver et al., 2018). Zhai et al., (2019) reported a 30-40% decrease in PM<sub>2.5</sub> concentrations 45 during 2013-2017. In contrast O<sub>3</sub> concentrations have increased, with median concentration of O<sub>3</sub> across 74 key cities increasing from 141  $\mu$ g m3 in 2013 to 164  $\mu$ g m3 in 2017 (Huang et al., 2018). Silver et al. (2018) found that O<sub>3</sub> maximum 8 h mean concentrations (O<sub>3</sub>MDA8) increased by 4.6 % year-1 over 2015-2017. Positive regional O<sub>3</sub> trends remain even after meteorological variability has been removed (Li et al., 2019a). Trends in NO2 are more variable, with a negative trend reported
- in eastern China and positive trends in western areas (Li and Bai, 2019). Silver et al., (2018) found that NO<sub>2</sub> had negative 50 trends in Hong Kong and North China Plain regions, but positive trends in the Yangtze River Delta (YRD), Sichuan Basin (SCB) and PRD, and no overall trend at the national scale.

#### 1.2 Identifying drivers of recent trends

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Changes in the concentrations of air pollutants may be caused by changing emissions or by interannual variability of meteorology. Stringent emission controls have started to reduce emissions of various pollutants across China. Between 2013 and 2017, emissions of PM2.5, SO2 and NOx (NO2 + Nitrogen Oxide) declined whereas emissions of Ammonia (NH3) and Non-Methane Volatile Organic Compounds (NMVOCs) remained fairly constant (Zheng et al., 2018). B. Zheng et al. (2018) also demonstrate that emission reductions were primarily driven by pollution controls, rather than decreasing activity rates. Meteorological variability alters atmospheric mixing, deposition and transport, all of which can influence the concentration of





60 pollutants. Separating the influence of meteorology and emissions on air pollutant concentrations is difficult, due to the interlinked nature of the chemistry-climate system (Jacob and Winner, 2009). However, to assess the efficacy of China's emissions reductions, it is necessary to separate these two factors.

There are two commonly used approaches to separate the influences of meteorology and emissions on variability in atmospheric pollutant abundances. The first approach uses statistical models, such as multi-linear regression, to control for the

- 65 influence of meteorology and allowing the proportion of air pollutant concentration variability that can be explained by meteorological variables to be calculated (Tai et al., 2010). The second approach is to use an atmospheric chemistry transport model to simulate pollutant concentrations. Through a comparison of multiple simulations, where either annual variability in emissions or meteorology are held constant, the relative influence of the two factors can be estimated. Here we analyse measurements and a regional air quality model to explore the role of changing anthropogenic emissions on air pollutant
- 70 concentrations and human health across China during 2015 to 2017.

#### 2 Materials and Methods

#### 2.1 Measurement dataset

We used hourly measurements from the CNEMC monitoring network (Wang et al., 2015) of PM<sub>2.5</sub>, O<sub>3</sub>, NO<sub>2</sub>, and SO<sub>2</sub> for the period 2015-2017, which includes data from over 1600 monitoring stations across mainland China and is available to download

75 from http://beijingair.sinaapp.com/. This was combined with data from the Hong Kong Environmental Protection Department (https://cd.epic.epd.gov.hk/EPICDI/air/station/) and Taiwan's Environmental Protection Administration (https://taqm.epa.gov.tw/taqm/en/YearlyDataDownload.aspx). We conducted quality control on the measured data following the methods outlined in Silver et al. (2018). The cleaned dataset included measurements from 1155 sites.

#### 2.2 WRF-Chem model setup

80 We used the Weather Research and Forecasting model with Chemistry (WRF-Chem) version 3.7.1 (Grell et al., 2005) to simulate trace gas and particulate pollution over China for 2015 to 2017. The model domain uses a Lambert Conformal grid (11-48 °N, 93-128 °E) centred on eastern China with a horizontal resolution of 30 km. The model has 33 vertical layers, with the lowest layer ~29 m above the surface, and the highest at 50 hPa (~19.6 km).

European Centre for Medium Range Weather Forecasts (ECMWF) ERA-Interim fields were used to provide meteorological
boundary and initial conditions, as well as to nudge the model temperature, winds and humidity above the boundary layer
every 6 hours. Restricting nudging to above the boundary layer, allowed a more realistic representation of vertical mixing
(Otte et al., 2012). Chemical boundary and initial conditions were provided by global fields from the Model for Ozone and
Related Chemical Tracers version 4 (MOZART-4) chemical transport model (Emmons et al., 2010).

Anthropogenic emissions were from the Multi-resolution Emission Inventory for China (MEIC; <u>www.meicmodel.org</u>). MEIC 90 estimates emissions using a database of activity rates across residential, industrial, electricity generation, transportation and



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agricultural emission sectors combined with China-specific emission factors (Hong et al., 2017). We used the 2015 MEIC dataset, then used sector-specific and species-specific scaling for 2016 and 2017 based on the emission totals estimated in B. Zheng et al. (2018). Table 1 shows emission totals for 2015, 2016 and 2017. Over the 2015 to 2017 period, Chinese emissions decreased by 38% for SO<sub>2</sub>, 16% for PM<sub>2.5</sub> and 8% for NOx. For regions outside the MEIC dataset, we used anthropogenic emissions from the EDGAR-HTAPv2.2 emission inventory for 2010.

- Biogenic emissions were generated online by the Model of Emissions of Gases and Aerosol from Nature (MEGAN) (Guenther et al., 2000). Biomass burning emissions were provided by the Fire Inventory from NCAR (FINN) version 1.5 (Wiedinmyer et al., 2011), which uses satellite fire observations of fires and land cover to estimate daily 1 km<sup>2</sup> emissions. Dust emissions were generated online the Georgia Institute of Technology-Goddard Global Ozone Chemistry Aerosol Radiation and Transport
- 100 (GOCART) model with Air Force Weather Agency (AFWA) modifications (LeGrand et al., 2019). Gas-phase chemistry is simulated using the MOZART-4 scheme and aerosol is treated by the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC; Zaveri *et al.*, 2008) scheme, including grid-scale aqueous chemistry and an extended treatment of organic aerosol (Hodzic and Jimenez, 2011; Knote et al., 2014). Four discrete size bins were used within MOSAIC (0.039–0.156 µm, 0.156–0.625 µm, 0.625–2.5 µm, 2.5–10 µm) to represent the aerosol size distribution.

#### 105 2.3 Model and measurement trend estimation

For comparison with the measurements, we sampled the model at the station locations using linear interpolation. Over 2015-2017, the model well simulated  $PM_{2.5}$  (normalised mean bias (NMB) = 0.45), O<sub>3</sub> (NMB=-0.13) and SO<sub>2</sub> (NMB=0.07), while overestimating NO<sub>2</sub> concentrations by a factor of around 2 (NMB=1.17).

To separate the influence of changing anthropogenic emissions from interannual variability in meteorology, we conducted two 3-year simulations, both for 2015-2017. The first simulation (Control) included interannual variability in both anthropogenic emissions and meteorology. The second simulation (Fixed emissions) included interannual variability in meteorology, but with anthropogenic emissions fixed at 2015 levels.

Trends in the interpolated model data were calculated using the same method as the measurement data (Silver et al., 2018). The hourly data are averaged to monthly means, which are then deseasonalised. The magnitude and direction of linear trends

115 were calculated using the Theil-Sen estimator, a non-parametric method that is resistant to outliers (Carslaw, 2015). The Mann-Kendall test was used to assess the significance of trends, using a threshold of p < 0.05. This stage of the analysis was performed using the R package '*openair*' (Carslaw and Ropkins, 2012).

# 2.4 Health impact estimation

Health impacts are estimated for ambient PM<sub>2.5</sub> using the Global Exposure Mortality Model (GEMM) (Burnett et al., 2018).

120 We used the GEMM for non–accidental mortality (non–communicable disease, NCD, plus lower respiratory infections, LRI), using parameters including the China cohort (GBD 2017 Risk Factor Collaborators, 2018). For ambient O<sub>3</sub>, we used the methodology of the Global Burden of Disease (GBD) study for 2017 (GBD 2017 Risk Factor Collaborators et al., 2018) to





estimate the mortality caused by chronic obstructive pulmonary disease. The United Nations adjusted population count dataset for 2015 at 0.05° × 0.05□ resolution was obtained from the Gridded Population of the World, Version 4 was used, along with
population age composition from GBD2017. Population count, population age, and baseline mortality rates were kept constant for 2015-2017 to estimate the variation due to changes in exposure only.

## 3 Measured and modelled trends comparison

## 3.1 Varying emissions scenario

Measured and simulated air quality trends over China during 2015 to 2017 largely compare well, and are shown in Figure 1 and 2. The measurements show widespread decline in PM<sub>2.5</sub> and SO<sub>2</sub> concentrations, widespread increase in O<sub>3</sub>MDA8, and spatially variable trends in NO<sub>2</sub> concentrations, as reported previously (Silver et al., 2018). The model (Control simulation) simulates the widespread decline in PM<sub>2.5</sub> concentrations, with the median measured trend across China (-3.4 µg m<sup>-3</sup> year<sup>-1</sup>) well simulated by the model (-3.5 µg m<sup>-3</sup> year<sup>-1</sup>). In the measurements, 90% of significant trends are negative and 10% of significant trends are positive, with positive trends mostly being in the Fenwei Plain region, Jiangxi and Anhui. No significant

135 positive trends are simulated by the model, possibly due to coarse resolution of the model and the simplified scaling we apply to emissions for 2016 and 2017.

WRF-Chem captures the widespread increase in  $O_3MDA8$ , but underestimates the magnitude of the trend by a factor 2 (2.7  $\mu$ g m<sup>-3</sup> year<sup>-1</sup> in the measurements, versus 1.3  $\mu$ g m<sup>-3</sup> year<sup>-1</sup> simulated by WRF-Chem). WRF-Chem simulates negative  $O_3MDA8$  trends in the Sichuan Basin and Taiwan, whereas in the measured data, all regions have positive median trends.

- 140 The measurements show zero overall median trend in NO<sub>2</sub> concentrations, with 46% of sites with significant trends being negative and 54% positive. In contrast, WRF-Chem simulates widespread reductions in NO<sub>2</sub> concentrations, with 100% of significant sites exhibiting negative trends and a negative nationwide median trend of -2.2 μg m<sup>-3</sup> year<sup>-1</sup>. The 7.0 % nationwide median decline in simulated NO<sub>2</sub> concentrations over 2015-2017, matches the 7.6 % decline in Chinese NOx emissions in the MEIC.
- 145 The measurements show a widespread decline in SO<sub>2</sub> concentrations, with a median nationwide trend of -1.9  $\mu$ g m<sup>-3</sup> year<sup>-1</sup>. WRF-Chem captures the direction of the trend, but the magnitude of the trend is overestimated by a factor 2. The 32.5 % decline in simulated nationwide median SO<sub>2</sub> concentrations over 2015-2017, matches the 37.8 % decline in SO<sub>2</sub> emissions in the MEIC.

## 3.2 Fixed emissions scenario

150 The model simulation where anthropogenic emissions in China were fixed at 2015 levels has a weak negative  $PM_{2.5}$  trend (-0.6 µg m<sup>-3</sup> year<sup>-1</sup>), a factor of six smaller than either the control simulation or the measurements (Figure 3). This suggests that the measured negative  $PM_{2.5}$  trend has largely been driven by decreased anthropogenic emissions, with limited impact from





interannual variability in meteorology. Chen et al. (2019) also concluded that emission reductions were the primary cause of reduced wintertime PM<sub>2.5</sub> across China during 2015-2017

- 155 The median O<sub>3</sub>MDA8 trend in the fixed emission simulation is 0.0 μg m<sup>-3</sup> year<sup>-1</sup>. This suggests that interannual meteorological variation had no influence on O<sub>3</sub> trends at the China-wide scale during 2015-2017, which were largely driven by changing emissions. However, meteorological variability did drive regional changes in O<sub>3</sub>. For example, in Guizhou, a trend of -2.5 μg m<sup>-3</sup> year<sup>-1</sup> was calculated in the fixed emissions simulation. Li et al. (2018) also report that the positive ozone trend over 2013 to 2017 is due to changes in anthropogenic emissions.
- 160 The fixed emission simulation also has a smaller NO<sub>2</sub> trend (-0.5 μg m<sup>-3</sup>) compared to the control simulation (-2.2 μg m<sup>-3</sup> year<sup>-1</sup>), demonstrating emission reductions that are estimated in the MEIC are also the main reason for the negative simulated NO<sub>2</sub> trend. However, unlike PM<sub>2.5</sub> and O<sub>3</sub>, the NO<sub>2</sub> trend calculated by the fixed emission simulation more closely matches measured trend. This may suggest that MEIC has overestimated the NO<sub>2</sub> emission reductions during 2015-2017. This suggestion is supported by recent satellite studies which found a slowing down or even reversal of NO<sub>2</sub> reductions during
- 165 2016-2019 (Li et al., 2019b), no significant trend in NO<sub>2</sub> during 2013-2017 (Huang et al., 2018), and increases in NO<sub>2</sub> concentration in the YRD, PRD and FWP regions during 2015-2017 (Feng et al., 2019). If NO<sub>X</sub> emissions decline too strongly in MEIC, this may contribute to the simulated underestimate of the positive observed O<sub>3</sub>MDA8 trend. Other work has suggested that increased O<sub>3</sub> concentrations are possibly linked to the rapid decline in aerosol (Li et al., 2019a).

#### 4 Health impacts of changes to PM<sub>2.5</sub> and O<sub>3</sub> concentrations

# 170 4.1 PM<sub>2.5</sub> health impacts

The control run simulated nation-wide population-weighted mean PM<sub>2.5</sub> concentration decreased by 12.8 % (10.1  $\mu$ g m<sup>-3</sup>), from 79.2  $\mu$ g m<sup>-3</sup> in 2015 to 69.1  $\mu$ g m<sup>-3</sup> in 2017. Greater decreases were simulated in more polluted and highly populated regions such as Beijing (-15.3  $\mu$ g m<sup>-3</sup>), Tianjin (-19.4  $\mu$ g m<sup>-3</sup>), Chongqing (province) (-14.2  $\mu$ g m<sup>-3</sup>) and Henan (-22.3  $\mu$ g m<sup>-3</sup>). Using the methodology of Burnett *et al.*, (2018), we estimate that mortality due to exposure to PM<sub>2.5</sub> decreased from 2 800

- 175 000 (CI: 2 299 000 3 302 000) premature mortalities in 2015, to 2 650 000 premature mortalities in 2017. The simulated reduction in  $PM_{2.5}$  concentrations therefore reduced the number of premature mortalities attributable to  $PM_{2.5}$  exposure by 150 000 (CI: 129 000 170 000) annual premature mortalities across China. The 12.8% reduction in  $PM_{2.5}$  exposure only led to a 5% reduction in attributable mortality due to the non-linearity of the exposure-response function, which is less sensitive at higher exposure ranges (Conibear et al., 2018). The largest absolute reductions in premature mortality occur in Henan (15 000
- 180 deaths year<sup>-1</sup>), Sichuan, Hebei and Tianjin (11 000 deaths year<sup>-1</sup>) (Figure 4). The decline in PM<sub>2.5</sub> exposure also led to reduced morbidity with the Disability Adjusted Life Years (DALYs) rate per 100,000 population reduced from 159 to 150, with the largest changes occurring in central provinces such as (Supplementary Figure S3). Our results are comparable to Zheng *et al.*, (2017), who found that population weighted annual mean PM<sub>2.5</sub> concentrations decreased 21.5 % during 2013 2015, resulting in a premature mortality decrease of 120 000 deaths year<sup>-1</sup>. Ding et al., (2019) estimated that during 2013-2017, a nationwide





185  $PM_{2.5}$  decrease of 9 µg m<sup>-3</sup> caused premature mortalities pear year to decrease by 287 000, using the methodology from the GBD 2015 study, which estimates health impacts as having a weaker and less linear relationship to  $PM_{2.5}$  concentrations.

## 4.2 O<sub>3</sub> health impacts

Increasing O<sub>3</sub> concentrations will result in an increase in health impacts that will act to offset some of the health benefits from declining PM<sub>2.5</sub> concentrations. WRF-Chem underestimated the observed magnitude of the O<sub>3</sub>MDA8 trend during 2015-2017, so the simulated change in health impacts would also be underestimated. However, our model bias in O<sub>3</sub> across China during 2015-2017 was reasonable (NMB=-0.13). To provide an estimate of the change in health impacts due to increasing O<sub>3</sub> concentrations we used simulated concentrations to estimate average health impacts due to exposure to O<sub>3</sub> over the 2015-2017 period, and then multiplied by the measured relative change in O<sub>3</sub>MDA8. We estimate that exposure to O<sub>3</sub> caused an average of 143 000 (CI: 106 000 – 193 000) premature mortalities each year over 2015-2017. Assuming linear behaviour, the 15% measured increase in O<sub>3</sub>MDA8 would result in an increase of 21 000 premature mortalities per year. The exposure-outcome function is in reality sub-linear, so this is likely to be an overestimate. Regardless, this is substantially smaller than the 150 000 reduction in annual premature mortality due to reduced PM<sub>2.5</sub>. We therefore suggest that changes in Chinese air pollution

#### **5** Conclusions

200 We used the WRF-Chem model to explore the drivers and impacts of changing air pollution across China during 2015-2017. A simulation with annually updated emissions was able to reproduce the measured negative trends in  $PM_{2.5}$  concentrations over China during 2015 – 2017, while overestimating the negative trend in SO<sub>2</sub> and NO<sub>2</sub>, and underestimating the positive trend in O<sub>3</sub>. By comparing this with a simulation where emissions are held constant at 2015 levels, but meteorological forcing was updated, we show that interannual meteorological variation was not the main driver of the substantial trends in air

over 2015-2017 have likely had an overall beneficial impact on human health.

- 205 pollutants that were observed across China during 2015 2017. Our work shows that reduced anthropogenic emissions are the main cause of reduced PM<sub>2.5</sub> concentrations across China, suggesting that the Chinese government's 'Air Pollution Prevention and Control Action Plan' has been effective at starting to control particulate pollution. We estimate that the 12.8% reduction in population-weighted PM<sub>2.5</sub> concentrations that occurred during 2015-2017 has reduced premature mortality due to exposure to PM<sub>2.5</sub> by 5.3%, preventing 150 000 premature mortalities across China annually. Despite these substantial reductions, PM<sub>2.5</sub>
- 210 concentrations still exceed air quality guidelines and cause negative impacts on human health. We estimate that exposure to O<sub>3</sub> during 2015-2017 causes on average 143 000 premature mortalities across China each year. Increases in O<sub>3</sub> concentration over 2015-2017, may have increased this annual mortality by about 20 000 premature mortalities per year, substantially less than the reduction in premature mortality due to declining particulate pollution. Changes in air pollution across China during 2015-2017 are therefore likely to have led to overall positive benefits to human health, amounting to a ~5 % reduction of the





ambient air pollution disease burden. However, to achieve larger reductions in the disease burden, further reductions in PM<sub>2.5</sub> concentrations are required, and pollution controls need to be designed that simultaneously reduce PM<sub>2.5</sub> and O<sub>3</sub> concentrations.

# Data availability

Data used to create all figures are available in the supplement. Air quality measurement data from mainland China's monitoring network is available from <u>http://beijingair.sinaapp.com/</u>. Air quality measurement data from Hong Kong is available from

220 <u>https://cd.epic.epd.gov.hk/EPICDI/air/station/</u>. Air quality monitoring data from Taiwan is available from <u>https://taqm.epa.gov.tw/taqm/en/YearlyDataDownload.aspx</u>. Data from all WRF-Chem model simulations and post-processing codes are available from the corresponding author on request.

#### Author contributions

BS, CLR, DVS and SRA designed the research. BS performed the WRF-Chem model simulations, analysed all the model data
 and wrote the manuscript. LC performed the health impact calculations. All authors contributed to scientific discussions and to the manuscript.

# **Competing interests**

The authors declare that they have no conflict of interest.

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- 235 at <u>http://www.acom.ucar.edu/wrf-chem/mozart.shtml</u> (last accessed 12<sup>th</sup> December 2018).

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# 380 Table 1. Chinese pollutant emissions (Tg yr<sup>-1</sup>) during 2015 to 2017 from MEIC.

	SO <sub>2</sub>	NO <sub>x</sub>	NMVOC	NH <sub>3</sub>	СО	TSP	PM <sub>10</sub>	PM <sub>2.5</sub>	BC	OC	$CO_2$
2015	16.9	23.7	28.6	10.5	153.6	21.9	12.3	9.1	1.4	2.5	10347.2
2016	13.4	22.5	28.4	10.2	142	17.9	10.8	8.1	1.3	2.3	10290.7
2017	10.5	21.9	28.6	10.2	136.2	16.7	10.2	7.6	1.2	2.1	10434.3





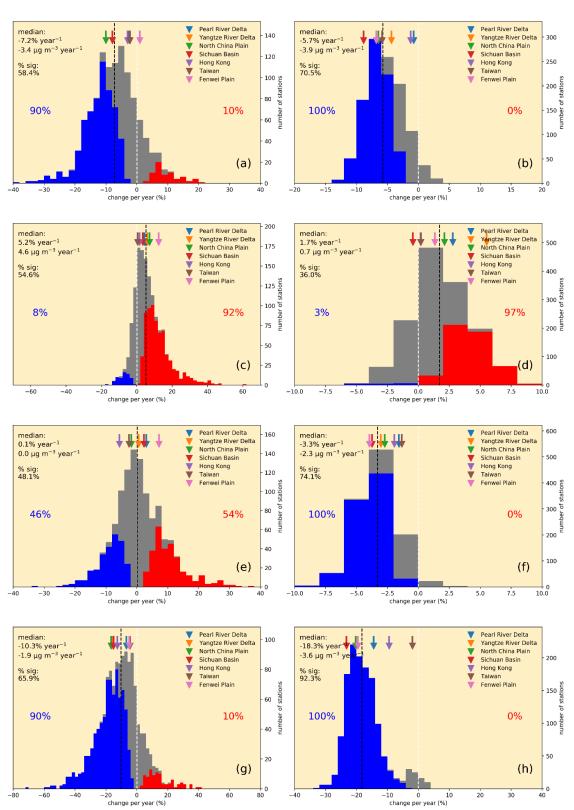






Figure 1: Histograms showing the frequency distribution of trends in concentrations of (a,b) PM<sub>2.5</sub>, (c,d) O<sub>3</sub>MDA8, (e,f) NO<sub>2</sub>, (g,h)
SO<sub>2</sub> across China and Taiwan during 2015–2017. Measured trends (left hand panels) are compared to simulated trends (right hand panels). The median relative and absolute trend as well as the percentage of stations with significant trends is shown on each panel. The percentage of significant trends that are negative (blue) or positive (red) are also shown. The black dotted line shows the median trend across all sites, while the white dotted line shows zero. Arrows show the median trend for the regional domain: Pearl River Delta (PRD), Yangtze River Delta (YRD), North China Plain (NCP), Sichuan Basin (SCB), Hong Kong (HK), Taiwan (TW) and the Senwei Plain (FWP).





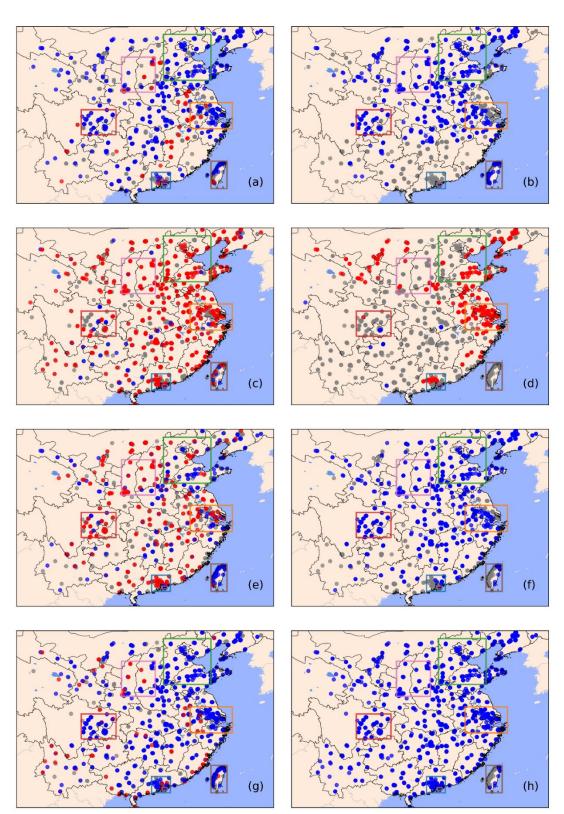
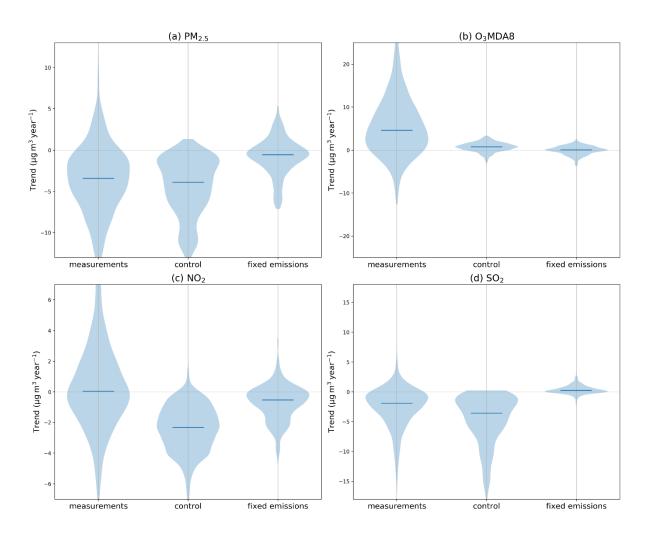






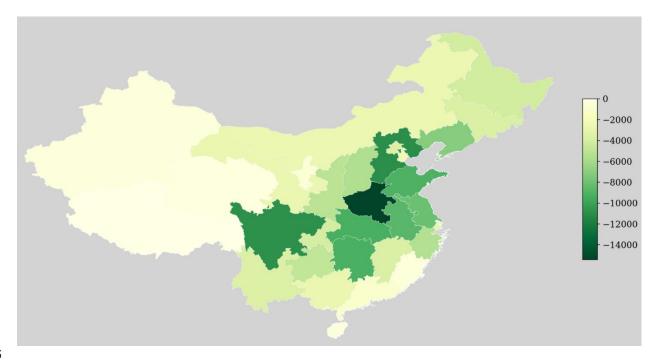
Figure 2. Map showing the spatial distribution of trends in concentrations of (a,b) PM<sub>2.5</sub>, (c,d) O<sub>3</sub>MDA8, (e,f) NO<sub>2</sub>, (g,h) SO<sub>2</sub> across China and Taiwan during 2015–2017. Measured trends (left hand panels) are compared to simulated trends (right hand panels).
Red indicates a significant positive trend, whereas blue indicates a significant negative trend, and grey an insignificant trend. Coloured boxes show the regional domains: Pearl River Delta (PRD), Yangtze River Delta (YRD), North China Plain (NCP), Sichuan Basin (SCB), Hong Kong (HK), Taiwan (TW) and the Fenwei Plain (FWP).



400 Figure 3. Comparison of measured and simulated concentration trends during 2015 to 2017. The left violin shows the measured trend, the centre shows the simulated trend with varying emissions and meteorology (control), and the right shows the simulated trends for the fixed emissions simulation. a) PM<sub>2.5</sub>, b) O<sub>3</sub>MDA8, c) NO<sub>2</sub>, d) SO<sub>2</sub>. The solid line shows the median absolute trend, and the shaded area shows a smoothed relative frequency distribution.







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Figure 4. Simulated change during 2015-2017 in annual premature mortality per year due to changes in exposure to ambient PM<sub>2.5</sub>. Results are shown at the province scale.