Pollutant emission reductions deliver decreased PM_{2.5}-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 (O₃) 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_{2.5} trend of -3.4 $\mu g m^{-3} y ear^{-1}$, was well simulated by the model (-3.5 $\mu g m^{-3} y ear^{-1}$). With anthropogenic emissions fixed at 2015levels, the simulated trend was much weaker (-0.6 $\mu g m^{-3} y ear^{-1}$), demonstrating interannual variability in meteorology played
- 15 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⁻¹.

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 21st 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_{2.5}) 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 (NO₂) (Van der A et al., 2006), Sulfur Dioxide (SO₂) (Zhang et al., 2017) and PM_{2.5} (Ma et al., 2016) during the 1990s and early 2000s. Trends in aerosol optical depth have been used to estimate changes in PM_{2.5}, which peaked around 2011 (Ma et al., 2016). NO₂ 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 SO₂ concentrations in China peaked around 2006 (Van Der A et al., 2017;
- 35 Krotkov et al., 2016; Zhang et al., 2017), matching the period of maximum emissions (Duan et al., 2016; Li et al., 2017a; 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 SO₂ emissions and concentrations (Duan et al., 2016). Measurements of O₃ 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 O₃ has been increasing across China during the past two decades. More recently, measurements at urban sites, also show positive O₃ 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, PM_{2.5} concentrations
- 45 across China decreased by 28% (Silver et al., 2018). Zhai et al., (2019) reported a 30-40% decrease in PM_{2.5} concentrations during 2013-2017. In contrast O₃ concentrations have increased, with median concentration of O₃ across 74 key cities increasing from 141 μg m⁻³ in 2013 to 164 μg m⁻³ in 2017 (Huang et al., 2018). Silver et al. (2018) found that O₃ maximum 8 h mean concentrations (O₃MDA8) increased by 4.6 % year⁻¹ over 2015-2017. Lu et al., (2020) reported positive trends in April-September O₃MDA8 at 90% of sites during 2013 to 2019. Positive regional O₃ trends remain even after meteorological
- 50 variability has been removed (Li et al., 2019b). 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 NO₂ had negative 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

55 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 PM_{2.5}, SO₂ and NO_x (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.

60 Meteorological variability alters atmospheric mixing, deposition and transport, all of which can influence the concentration of 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 65 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 65 meteorological variables to be calculated (Tai et al., 2010). The second approach is to use an atmospheric chemistry transport 66 model to simulate pollutant concentrations (Ansari et al., 2019; Xing et al., 2011).

There are a limited number of modelling studies that attempt to separate the influence of meteorology and emissions changes on recent air quality trends in China. Chen et al. (2019) use WRF-Chem with 2010 emissions to examine the drivers of trends in wintertime PM. Ding et al. (2019) use WRF-CMAQ to evaluate importance of emissions, meteorology and demographic changes on PM_{2.5} related mortality during 2013-2017. Our paper adds to these previous studies by evaluating the ability of a online-coupled model (WRF-Chem) to capture trends in NO₂, O₃ and SO₂ as well as PM, using the most recent emissions and evaluated against a comprehensive measurement dataset.

75 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 concentrations and human health across China during 2015 to 2017.

2 Materials and Methods

80 2.1 Measurement dataset

We used hourly measurements from the CNEMC monitoring network (Wang et al., 2015) of PM_{2.5}, O₃, NO₂, and SO₂ for the period 2015-2017, which includes data from over 1600 monitoring stations across mainland China and is available to download from <u>http://beijingair.sinaapp.com/</u>. This was combined with data from the Hong Kong Environmental Protection Department (<u>https://cd.epic.epd.gov.hk/EPICDI/air/station/</u>) and Taiwan's Environmental Protection Administration

85 (<u>https://taqm.epa.gov.tw/taqm/en/YearlyDataDownload.aspx</u>). We conducted quality control on the measured data following the methods outlined in Silver et al. (2018), which include excluding data with a high proportion of repeated measurements and periods of low variability. The cleaned dataset included measurements from 1155 sites.

2.2 WRF-Chem model setup

We used the Weather Research and Forecasting model with Chemistry (WRF-Chem) version 3.7.1 (Grell et al., 2005) to 90 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

estimates emissions using a database of activity rates across residential, industrial, electricity generation, transportation and

- 95 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
- 100 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 SO₂, 16% for PM_{2.5} and 8% for NOx. For regions outside the MEIC dataset, we used anthropogenic emissions from the EDGAR-HTAPv2.2 emission inventory for 2010.
- 105 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 km² emissions. Dust emissions were generated online the Georgia Institute of Technology-Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) model with Air Force Weather Agency (AFWA) modifications (LeGrand et al., 2019).
- 110 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; Hodzic and Knote, 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.

2.3 Model and measurement trend estimation

- 115 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), O₃ (NMB=-0.13) and SO₂ (NMB=0.07), while overestimating NO₂ concentrations by a factor of around 2 (NMB=1.17). Model biases were similar to previous model studies in China (Supplementary Table 1). We also evaluated the model against speciated aerosol measurements from the Surface PARTiculate mAtter Network (SPARTAN) (Snider et al., 2015, 2016) site in Beijing (https://www.spartan-
- 120 network.org/beijing-china, last accessed: 2nd July 2020) (Fig S4), as well as Zhou et al. (2019) (Figure S5) and from across China (Li et al., 2017b) (Fig S6). Measurements reported by Li et al. (2017b) were made from various years spanning 2006 to 2013 and do not match the years simulated by the model. Comparison against these data show that the model underestimates the sulfate fraction in PM_{2.5}, while overestimating the nitrate fraction. Underestimation of sulfate in comparison to Li et al.,

(2017b) will partly be caused by the large decline in SO_2 emissions that has occurred in the last decade (Zheng et al., 2018).

125 Underestimate of sulfate, particularly in winter, and overestimation of nitrate are consistent with previous modelling studies (Shao et al., 2019) including those using WRF-chem (Zhou et al., 2019). Newly proposed mechanisms to explain the rapid sulfate formation in China's winter haze (Gen et al., 2019; Shao et al., 2019; Xue et al., 2014; Zhang et al., 2019) need to be included and evaluated in models.

To separate the influence of changing anthropogenic emissions from interannual variability in meteorology, we conducted two

- 130 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. Both simulations include interannual variability in biogenic and biomass burning emissions, allowing us to isolate the impacts of changing anthropogenic emissions.
- Trends in the 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 using locally weighted scatterplot smoothing. The magnitude and direction of linear trends 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).</p>

2.4 Health impact estimation

- 140 Health impacts are estimated for ambient PM_{2.5} using the Global Exposure Mortality Model (GEMM) (Burnett et al., 2018), which uses cohort studies to estimate health risks integrated over a range of PM_{2.5} concentrations. GEMM applies a supralinear association between exposure and risk at lower concentrations and then a near-linear association at higher concentrations. 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 O₃, we used the
- 145 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, which is based on exposure and risk information from five epidemiological cohorts. It estimates a near-linear relationship between exposure and risk at lower concentrations of O_3 , and a sub-linear association at higher concentrations. The United Nations adjusted population count dataset for 2015 at 0.05° $\times 0.05^{\circ}$ resolution was obtained from the Gridded Population of the World, Version 4, along with population age distribution
- 150 from GBD2017. Health impacts depend on population count, population age, and baseline mortality rates which have changed over the period studied (Butt et al., 2017). To isolate the impacts of changing air pollution, other variables were kept constant for 2015-2017.

3 Measured and modelled trends comparison

3.1 Varying emissions scenario

- Figure 1 and 2 compare measured and simulated air quality trends over China during 2015 to 2017. The measurements show widespread decline in PM_{2.5} and SO₂ concentrations, widespread increase in O₃MDA8, and spatially variable trends in NO₂ concentrations, as reported previously (Silver et al., 2018). The model (Control simulation) simulates the widespread decline in PM_{2.5} concentrations, with the median measured trend across China (-3.4 μ g m⁻³ year⁻¹) well simulated by the model (-3.5 μ g m⁻³ year⁻¹). In the measurements, 90% of significant trends are negative and 10% of significant trends are positive, with
- 160 positive trends mostly being in the Fenwei Plain region, Jiangxi and Anhui. No significant 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 O₃MDA8, but underestimates the magnitude of the trend by a factor 2 (2.7 µg m⁻³ year⁻¹ in the measurements, versus 1.3 µg m⁻³ year⁻¹ simulated by WRF-Chem). WRF-Chem simulates negative O₃MDA8 trends in the Sichuan Basin and Taiwan, whereas in the measured data, all regions have positive median trends.
- 165 The measurements show zero overall median trend in NO₂ concentrations, with 46% of sites with significant trends being negative and 54% positive. In contrast, WRF-Chem simulates widespread reductions in NO₂ concentrations, with 100% of significant sites exhibiting negative trends and a negative nationwide median trend of -2.2 μg m⁻³ year⁻¹. The 7.0 % nationwide median decline in simulated NO₂ concentrations over 2015-2017, matches the 7.6 % decline in Chinese NOx emissions in the MEIC.
- 170 The measurements show a widespread decline in SO₂ concentrations, with a median nationwide trend of -1.9 μ g m⁻³ year⁻¹. 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 SO₂ concentrations over 2015-2017, matches the 37.8 % decline in SO₂ emissions in the MEIC.

3.2 Fixed emissions scenario

- 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⁻³ year⁻¹), 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_{2.5} across China during 2015-2017. Cheng et al., (2019) found that local and regional reductions in anthropogenic emissions were the dominant cause of reduced PM_{2.5} concentrations in Beijing between 2013 and 2017.
- The median O_3MDA8 trend in the fixed emission simulation is 0.0 µg m⁻³ year⁻¹. This suggests that interannual meteorological variation had little influence on O_3 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_3 . For example, in Guizhou province, a trend of -2.5 µg m⁻³ year⁻¹ was calculated in the fixed emissions simulation. Li et al. (2019a) also report that the positive ozone trend

- 185 over 2013 to 2017 is due to changes in anthropogenic emissions, and the magnitude of their estimated trend of 1-3 ppbv year⁻¹ (approximately 2-6 μg m⁻³ year⁻¹) is comparable to the 2.6 μg m⁻³ year⁻¹ trend found in this study. Lu et al. (2019) analysed changes in O₃ between 2016 and 2017 and concluded that hotter and drier conditions in 2017 contributed to higher O₃ concentrations in that year. Liu and Wang (2020) reported a complex O₃ response during 2013 to 2017, with changing anthropogenic emission increasing O₃MDA8 in urban areas and decreasing it in rural areas, whereas meteorological changes
- 190 drove regionally contrasting changes in O₃MDA8 through changes in cloud cover, wind, and temperature and through driving changes in biogenic emissions.

The fixed emission simulation also has a smaller NO₂ trend (-0.5 μ g m⁻³) compared to the control simulation (-2.2 μ g m⁻³ year⁻¹), demonstrating emission reductions that are estimated in the MEIC are also the main reason for the negative simulated NO₂ trend. However, unlike PM_{2.5} and O₃, the NO₂ trend calculated from the fixed emission simulation more closely matches

- 195 measured trend. This may suggest that MEIC has overestimated the NO₂ emission reductions during 2015-2017. This suggestion is supported by recent satellite studies which found a slowing down or even reversal of NO₂ reductions during 2016-2019 (Li et al., 2019c), no significant trend in NO₂ during 2013-2017 (Huang et al., 2018), and increases in NO₂ concentration in the YRD, PRD and FWP regions during 2015-2017 (Feng et al., 2019). If NO_x emissions decline too strongly in MEIC, this may contribute to the simulated underestimate of the positive observed O₃MDA8 trend in areas of China with a
- 200 NO_x limited or mixed Ozone regimes that cover the majority of China (Jin and Holloway, 2015). Other work has suggested that increased O₃ concentrations are possibly linked to the rapid decline in aerosol (Li et al., 2019b). Liu and Wang (2020b) found that the reasons for increased O₃ concentrations during 2013-2017 were regionally dependent and that anthropogenic VOC emission reductions of 16-24% would have been needed to avoid increased concentrations.
- Table 2 compares the control and fixed emission simulations against PM_{2.5}, O₃ and SO₂ and NO₂ measurements in 2015, 2016 and 2017. In the control simulation model biases remain similar during 2015-2017. In the fixed emission simulation, model biases for PM_{2.5}, O₃ and SO₂ increase between 2015 and 2017. This further suggests that changing anthropogenic emissions during 2015-2017 have been the dominant cause of changing concentrations.

An important future step is to understand how changing anthropogenic emissions, in terms of emission species or emission sectors, have contributed to observed trends in pollutant concentrations. Residential and industrial emissions are dominant

- 210 causes of PM_{2.5} concentrations across much of China (Reddington et al., 2019), but it is not clear which emission sectors have contributed most to observed PM_{2.5} trends. Cheng et al. (2019) suggests that emission controls in the residential and industrial sectors were the dominant causes for reduced PM_{2.5} in Beijing between 2014 and 2017. Measurements of aerosol composition (Li et al., 2017b; Weagle et al., 2018) add confidence to model simulations and can inform our understanding of how aerosol chemistry responds to emission changes. However, except for Beijing, there is insufficient measurement data of how aerosol
- 215 composition has changed across China in recent years. Li et al. (2019a) found large declines in wintertime organics and sulfate and smaller declines in nitrate and ammonium in Beijing between 2014 and 2017. Zhou et al. (2019) also analysed aerosol composition data from Beijing and found large declines in all aerosol components except nitrate between 2011-12 and 2017-

18. Continuous measurements of aerosol composition across China are required to determine how different aerosol components are contributing to the observed PM_{2.5} trend and to evaluate simulated responses to emission changes.

220 4 Health impacts of changes to PM_{2.5} and O₃ concentrations

4.1 PM_{2.5} health impacts

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The control run simulated nation-wide population-weighted mean $PM_{2.5}$ concentration decreased by 12.8 % (10.1 µg m⁻³), from 79.2 µg m⁻³ in 2015 to 69.1 µg m⁻³ in 2017. Greater decreases were simulated in more polluted and highly populated regions such as Beijing (-15.3 µg m⁻³), Tianjin (-19.4 µg m⁻³), Chongqing (province) (-14.2 µg m⁻³) and Henan (-22.3 µg m⁻³). Using the methodology of Burnett *et al.*, (2018), we estimate that mortality due to exposure to $PM_{2.5}$ decreased from 2 800 000 (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

- 230 higher exposure ranges (Conibear et al., 2018). The largest absolute reductions in premature mortality occur in Henan (15 000 deaths year⁻¹), Sichuan, Hebei and Tianjin (11 000 deaths year⁻¹) (Figure 4). The decline in PM_{2.5} 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_{2.5} concentrations decreased 21.5% during 2013 2015, resulting
- in a premature mortality decrease of 120 000 deaths year⁻¹. Ding et al., (2019) estimated that during 2013-2017, a nationwide PM_{2.5} decrease of 9 µg m⁻³ year⁻¹ 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. Yue et al. (2020) estimated that the annual number of mortalities in China attributable to PM_{2.5} decreased by 64 000 (7%) from 2013 to 2017. Zhang et al. (2019) reported a 32% decline in population-weighted PM_{2.5} concentration during 2013 to 2017, 240 largely due to strengthened industrial emission standards and cleaner residential fuels.

4.2 O₃ health impacts

Increasing O_3 concentrations will result in an increase in health impacts that will act to offset some of the health benefits from declining PM_{2.5} concentrations. WRF-Chem simulated O_3 concentrations across China during 2015-2017 to within 15% (NMB=-0.13), which is consistent with previous studies, but underestimated the magnitude of the observed O_3 trend. To provide an estimate of the health impacts due to exposure to O_3 we used simulated concentrations to estimate average exposure

to O_3 over the 2015-2017 period. We estimate that exposure to O_3 caused an average of 143 000 (CI: 106 000 – 193 000) premature mortalities each year over 2015-2017. Applying the simulated change in O_3 concentrations would underestimate the change in exposure that has occurred, Instead, we estimated the impacts of increased O_3 by multiplying the average health

impacts over 2015-2017 by the measured relative change in O₃MDA8. Assuming linear behaviour, the 15% measured increase

250 in O₃MDA8 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_{2.5}. We therefore suggest that changes in Chinese air pollution over 2015-2017 have likely had an overall beneficial impact on human health. The dominance of the PM_{2.5} reduction over the O₃ increase on health impacts is also found in Dang and Liao (2019) who reported a 21% reduction in PM_{2.5} and 12% increase in O₃ concentrations between 2012 and 2017 resulted in 268 000 fewer annual mortalities overall.

5 Conclusions

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 SO₂ and NO₂, and underestimating the positive trend in O₃. 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 pollutants that were observed across China during 2015 – 2017. Our work shows that reduced anthropogenic emissions are the main cause of reduced PM_{2.5} 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_{2.5} concentrations that occurred during 2015-2017 has reduced premature mortality due to exposure

- to PM_{2.5} by 5.3%, preventing 150 000 premature mortalities across China annually. Despite these substantial reductions, PM_{2.5} concentrations still exceed air quality guidelines and cause negative impacts on human health. We estimate that exposure to O₃ during 2015-2017 causes on average 143 000 premature mortalities across China each year. Increases in O₃ 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 \sim 5 % reduction of the ambient air pollution disease burden. However, to achieve larger reductions in the disease burden, further reductions in PM_{2.5} concentrations are required, and pollution controls need to be designed that simultaneously reduce PM_{2.5} and O₃ concentrations.

Data availability

275 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 <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.

280 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

285 The authors declare that they have no conflict of interest.

Acknowledgements

We gratefully acknowledge the AIA Group Limited and Natural Environment Research Council (NE/S006680/1; NE/N006895/1) for funding for this research. Model simulations were undertaken on ARC3, part of the High Performance Computing facilities at the University of Leeds. We thank Qiang Zhang and Meng Li for providing MEIC data. We

290 acknowledge use of the WRF-Chem preprocessor tools bio_emiss, anthro_emiss, fire_emiss, and mozbc provided by the Atmospheric Chemistry Observations and Modeling Lab (ACOM) of the NCAR. We acknowledge use of NCAR/ACOM .MOZART-4 global model output available at http://www.acom.ucar.edu/wrf-chem/mozart.shtml (last accessed 12th December 2018). We thank the SPARTAN project for its effort in establishing and maintaining the site in Beijing. The SPARTAN network was initiated with funding from the Natural Sciences and Engineering Research Council of Canada.

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Table 1. Chinese pollutant emissions (Tg yr⁻¹) during 2015 to 2017 from MEIC.

	SO ₂	NO _x	NMVOC	NH ₃	СО	TSP	PM ₁₀	PM _{2.5}	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

	PM _{2.5}	O ₃	NO ₂	SO ₂
Control				
2015-2017	0.49	-0.15	1.2	0.09
2015	0.5	-0.12	1.32	0.17
2016	0.47	-0.14	1.20	0.05
2017	0.5	-0.21	1.10	0.04
Fixed emissions				
2015-2017	0.57	-0.18	1.26	0.35
2015	0.50	-0.12	1.32	0.17
2016	0.56	-0.16	1.28	0.31
2017	0.66	-0.24	1.20	0.65

Table 2. Model evaluation shown as a normalised mean bias (NMB). Evaluation is shown separately for the control and fixed emission simulations. The NMB for 2015-2017 is compared to individual years.



- 515 Figure 1: Histograms showing the frequency distribution of trends in concentrations of (a,b) PM_{2.5}, (c,d) O₃MDA8, (e,f) NO₂, (g,h) SO₂ 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
- 520 Delta (PRD), Yangtze River Delta (YRD), North China Plain (NCP), Sichuan Basin (SCB), Hong Kong (HK), Taiwan (TW) and the Fenwei Plain (FWP).



525 Figure 2. Map showing the spatial distribution of trends in concentrations of (a,b) PM_{2.5}, (c,d) O₃MDA8, (e,f) NO₂, (g,h) SO₂ 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. Lighted coloured circles indicated a statistically insignificant trend. Coloured boxes show the regional domains: Pearl River Delta (blue), Yangtze River Delta (orange), North China Plain (green), Sichuan Basin (red), Hong Kong (purple), Taiwan (brown) and the Fenwei Plain (pink).



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_{2.5}, b) O₃MDA8, c) NO₂, d) SO₂. 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_{2.5}. Results are shown at the province scale.