

Response to comments on the revised submission of ‘Pollutant emission reductions deliver decreased PM2.5-caused mortality across China during 2015–2017’

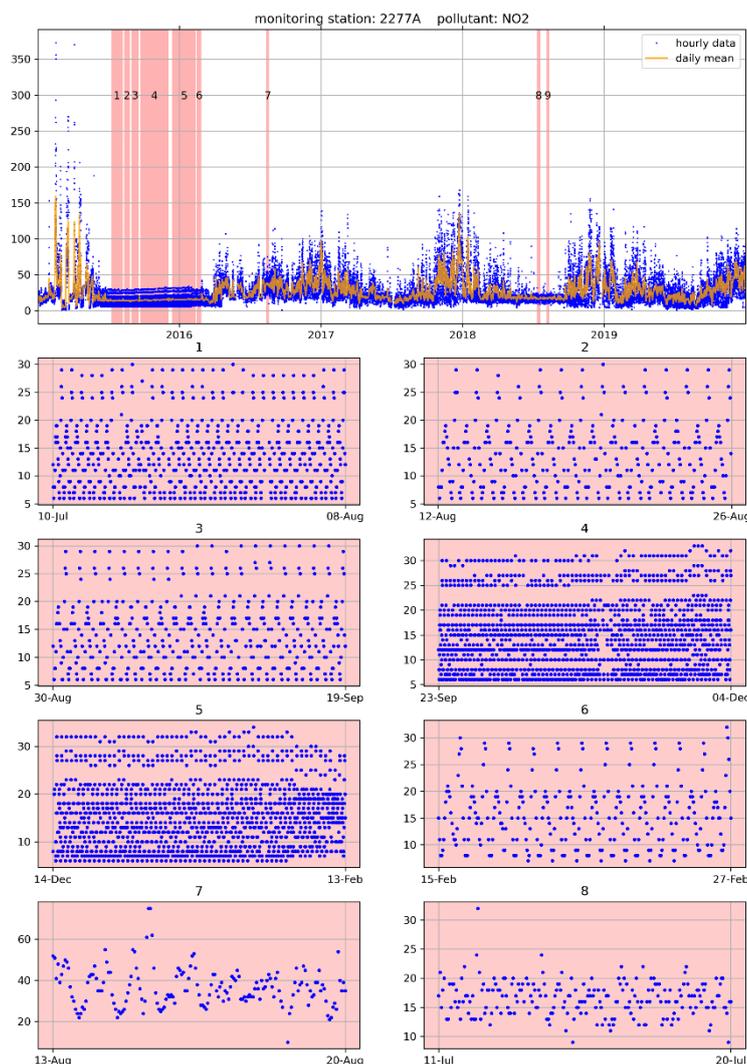
Response to Editor comments:

Concerning the data, I would recommend that you store the data behind your study on an open-accessible repository (with a DOI). The current way of referencing to data via web addresses is not optimal. Please have a look at the data guidelines of ACP: https://www.atmospheric-chemistry-and-physics.net/about/data_policy.html

We thank the editor for this suggestion and hope that it will be useful to share our data. We have deposited the data in ‘Research Data Leeds Repository,’ which is registered with <https://www.re3data.org/>. The repository has the DOI: <https://doi.org/10.5518/878>. Here we have included the calculated trends from the measurement data and both model runs, at each of the measurement station locations. We also include a copy of the measurement dataset containing data from 2014-05-13 to 2020-06-06, along with the results of our data cleaning process. We think having the measurements dataset in this convenient and accessible form will be very useful to other researchers. We have amended the data availability statement accordingly.

One further question from my side: In Sect. 2.1, you write "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." Why do you exclude data with low variability? Shouldn't it be the opposite (e.g. excluding local plumes/spikes in the observational data)?

The periods of low variability we refer to are periods where almost exactly the same measurements are repeated each day for long periods. This example shows the anomalous data found at some stations which we wish to exclude. The algorithm highlights the area of almost repeated measurements. If the station has more than 60 days of flagged data, we exclude it from the analysis. The algorithm is



described in our previous paper (<https://doi.org/10.1088/1748-9326/aae718>). We have amended lines 85-87 to better describe this as “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 identified as periods of low variability, which represent periods of missing or invalid data.”

Response to referee #2

The authors really worked hard on the new version of the manuscript considering comments from all reviewers. I believe the manuscript is in much better shape now and I only have minor comments.

We thank the reviewer for the positive comments.

Comments by line:

115-128. This paragraph corresponds to model evaluation, not to trend estimation, so consider having it in its own section (maybe even move it to results)

We thank the referee for pointing this out. We have moved that paragraph as suggested to the next section, and given it the sub-heading ‘3.1 Model Evaluation.’

Section 3.1. It would be informative to add the trends in aerosol composition for the Spartan Beijing site and compare it to a nearby PM_{2.5} trends, both in observations and model to add to the discussion.

The SPARTAN data is very useful in evaluating the ability of the model to capture aerosol speciation. However, the SPARTAN data is of limited temporal resolution and consistency. The SPARTAN data is collected every two weeks, but there are large areas of missing data throughout the time series. As a result, trends calculated from the SPARTAN data cannot be confidently compared with those calculated from the CNEMC data or model data. For this reason we would prefer not to add trends from the SPARTAN data to our paper.

159-161. Another reason could be linked to the representation of aerosol speciation, the fact that NO₂ trends are all negative and the model tends to overpredict the nitrate fraction could drive the PM_{2.5} trends. Same for overprediction of the magnitude of the SO₂ trend.

We agree with the referee that inaccuracies in the model nitrate and sulphate estimations will affect the overall PM_{2.5} trend, among other model inaccuracies. However, due to the lack of speciated aerosol data across China, it is difficult to quantify the degree of inaccuracy in the PM_{2.5} across the model domain. To further highlight this issue, we add the sentence ‘However, as the above comparisons with speciated aerosol measurements show, the underlying trends in individual aerosol species may contain inaccuracies that affect the overall PM_{2.5} trend.’

Fig S4. Add to the caption for what period of time you are comparing

We have added ‘during 2015-2017’ to the caption.

Fig S5. Are statistics build with hourly or daily values?

According to the Zhou et al. paper, the aerosol components were measured at around 15 minute intervals. We have added the text 'The measurements had a time resolution of ~15 minutes and averaged by season' to the caption.

Minor Edits

69-72. *Since these are previous studies you could change the tense to past?*

We have amended these sentences to past tense, thank you for the correction.

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, during 2015 to 2017. We used measurements of particulate matter with a diameter < 2.5 μm (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 μg m⁻³ year⁻¹, was well simulated by the model (-3.5 μg m⁻³ year⁻¹). With anthropogenic emissions fixed at 2015-levels, the simulated trend was much weaker (-0.6 μg m⁻³ year⁻¹), 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 mortality across China by 150 000 deaths year⁻¹.

1 Introduction

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 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 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; 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 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 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 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

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. 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 atmospheric pollutant abundances. The first approach uses statistical models, such as multi-linear regression, to control for the 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 (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) used WRF-Chem with 2010 emissions to examine the drivers of trends in wintertime PM. Ding et al. (2019) used 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 an 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.

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

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 <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), which include excluding data with a high proportion of repeated measurements and periods of low variability, [which represent periods of missing or invalid data](#). 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 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; www.meicmodel.org). MEIC estimates emissions using a database of activity rates across residential, industrial, electricity generation, transportation and 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 NO_x. 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 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).

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

~~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 mAttor Network (SPARTAN) (Snider et al., 2015, 2016) site in Beijing (<https://www.spartan-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 SO₂ emissions that has occurred in the last decade (Zheng et al., 2018). 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 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).

2.4 Health impact estimation

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 O_3 , 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, 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 $0.05^\circ \times 0.05^\circ$ resolution was obtained from the Gridded Population of the World, Version 4, along with population age distribution 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 Model evaluation

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_3 (NMB=-0.13) and SO_2 (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-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 SO₂ emissions that has occurred in the last decade (Zheng et al., 2018). 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.

3.21 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 µg m⁻³ year⁻¹) well simulated by the model (-3.5 µg m⁻³ year⁻¹). However, as the above comparisons with speciated aerosol measurements show, the underlying trends in individual aerosol species may contain inaccuracies that affect the overall PM_{2.5} trend. 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 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.

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 NO_x emissions in the MEIC.

The measurements show a widespread decline in SO₂ concentrations, with a median nationwide trend of -1.9 µg 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.

3.3.2 Fixed emissions scenario

The model simulation where anthropogenic emissions in China were fixed at 2015 levels has a weak negative PM_{2.5} trend ($-0.6 \mu\text{g m}^{-3} \text{ year}^{-1}$), 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 O₃MDA8 trend in the fixed emission simulation is $0.0 \mu\text{g m}^{-3} \text{ year}^{-1}$. This suggests that interannual meteorological variation had little influence on O₃ trends at the China-wide scale during 2015-2017, which were largely driven by changing emissions. However, meteorological variability did drive regional changes in O₃. For example, in Guizhou province, a trend of $-2.5 \mu\text{g m}^{-3} \text{ year}^{-1}$ was calculated in the fixed emissions simulation. Li et al. (2019a) also report that the positive ozone trend 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 $2-6 \mu\text{g m}^{-3} \text{ year}^{-1}$) is comparable to the $2.6 \mu\text{g m}^{-3} \text{ year}^{-1}$ 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 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 ($-0.5 \mu\text{g m}^{-3}$) compared to the control simulation ($-2.2 \mu\text{g m}^{-3} \text{ year}^{-1}$), 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 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 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 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 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.

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

4.1 PM_{2.5} health impacts

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 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 per 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, largely due to strengthened industrial emission standards and cleaner residential fuels.

4.2 O₃ health impacts

Increasing O₃ 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 O₃ 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 O₃ trend. To provide an estimate of the health impacts due to exposure to O₃ we used simulated concentrations to estimate average exposure to O₃ over the 2015-2017 period. We estimate that exposure to O₃ caused an average of 143 000 (CI: 106 000 – 193 000) premature mortalities each year over 2015-2017. Applying the simulated change in O₃ concentrations would underestimate the change in exposure that has occurred. Instead, we estimated the impacts of increased O₃ by multiplying the average health impacts over 2015-2017 by the measured relative change in O₃MDA8. Assuming linear behaviour, the 15% measured increase 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 ~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

~~Data used to create all figures are available in the supplement.~~ [Data associated with this work has been made available at https://doi.org/10.5518/878, including the full trend results and the mainland China air quality stations measurement dataset.](https://doi.org/10.5518/878) ~~Air quality measurement data from mainland China's monitoring network is available from <http://beijingair.sinaapp.com/>.~~ Air quality measurement data from Hong Kong is available from <https://cd.epic.epd.gov.hk/EPICDI/air/station/>. Air quality monitoring data from Taiwan is available from <https://taqm.epa.gov.tw/taqm/en/YearlyDataDownload.aspx>. ~~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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