

Authors Response to Reviewer Comments on 'Pollutant emission reductions deliver decreased PM_{2.5}-caused mortality across China during 2015–2017'

Response to Anonymous Referee #1

Summary:

In this work, the authors use a chemical transport model (WRF-Chem) to demonstrate that emission controls rather than meteorology have been driving the air-quality improvement in China in recent years. Additionally, the authors calculate the number of lives saved from China's 'Air Pollution Prevention and Control Action Plan' between 2015 and 2017. This manuscript is of good scientific and presentation quality and in a highly-relevant area of research. However, there have already been several articles published on (1) whether meteorology or emissions are driving air quality changes in China and (2) the health impacts of the stringent emissions controls in China. Overall, a better case for the novelty of this work needs to be made in the motivation/introduction of the paper (see overall comment below). Additionally, there are several places where more detail and/or discussion is needed (see Line-by-line comments below).

General Comments:

There have already been several articles evaluating the impacts of meteorology vs emissions on changes of PM_{2.5} in China (e.g., <https://doi.org/10.5194/acp-19-7409-2019>, <https://doi.org/10.1289/EHP4157>) and several papers that calculated the health effects of the stringent emissions controls in China in recent years (e.g., <https://doi.org/10.1289/EHP4157>, <https://doi.org/10.1088/1748-9326/aa8a32>). Many of these papers were mentioned in the results/discussion section of this work. In the introduction, the authors should mention some of this closely-related previous work and discuss what distinguishes this work from previous studies.

We thank the reviewer for their comment. We have added a paragraph (on lines 69-74) which mentions similar papers, and discusses the distinguishing features of our study:

"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 WRF-Chem to simulate trends in NO₂, O₃ and SO₂ as well as PM, using the most recent emissions and evaluated against a comprehensive measurement dataset."

Specific Comments:

Line 75: What was done to clean the dataset? Please provide more information.

We already included some brief details in our manuscript and a full description is given in Silver et al. (2018). As suggested by the referee we provide more information (now on line 85-87) to further describe the data cleaning procedure, through the text:

'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'.

Line 106-108: Suggest providing context for these NMB. For example, how do they compare to previous work? How will they impact air pollution-mortality estimates?

We have added a table of NMB statistics from previous studies to the supplement (Table S1). We included studies that also use WRF-Chem in China, and have a similar model setup.

	PM _{2.5}	PM ₁₀	O ₃	NO ₂	SO ₂	CO
Zhang et al. (2016) (Hong Kong)		-0.47 to -0.07	0.88 to 1.6	-0.88 to -0.83	-0.84 to -0.59	-0.72 to -0.55
Zhang et al. (2016) (China)		-0.38 to -0.03			-0.8 to -0.72	
(Wang et al., 2016) (N China, January)	0.28 to 0.47	0.00 to 0.08		0.09 to 0.27	0.33 to 0.91	0.01 to 0.12
Zhou et al. (2017) (forecast)		-0.36		-0.05	-0.18	-0.4
This paper	0.49	-0.09	-0.15	1.2	0.09	-0.35

We add the following text to the main paper (Line 117-118):

“Model biases were similar to previous model studies in China (Supplementary Table 1).”

Are these NMB calculated using the “control” simulation? Is the NMB greater if the measurements are compared with the “fixed emissions” simulation?

This is a good suggestion. Yes, the NMB is further from zero in the fixed emissions scenario. We include the table below in the main paper (Table 2):

	PM2.5	O3	NO2	SO2
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.

Line 109-112: *What was the measurement/simulation bias for each year? If it does not change substantially, this would help validate the methodology used here for decoupling the impacts of emissions and meteorology on PM2.5 and O3 levels.*

This was a useful suggestion, and the information has been included in Table 2 (see above), which has been added to the manuscript. We also added these sentences (lines 204-207):

“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.”

Line 113-117: Please provide more information here. What is meant by “interpolated model data”? Did the authors look only at the model estimates that coincided with the measurements? Please provide some information about the method that was used for the measurements data, so the reader doesn’t need to look at the Silver et al, 2018, unless they are interested in a high level of detail of the methods. What method was used to deseasonalize the data?

We explain what we mean by interpolated model data on line 115:

“For comparison with the measurements, we sampled the model at the station locations using linear interpolation.”

We have amended the text (lines 134-135) to explain how we deseasonalise the data. For clarity, we remove mention of interpolated data here

“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.”

When evaluating model trends (Section 3) we match the model to the measurements. When exploring changes in exposure and impacts on health (Section 4) we calculate population-weighted exposure.

Line 125: It’s useful to calculate the changes in mortality based on exposure alone, but I also suggest calculating the number of PM_{2.5} and O₃ mortalities with population/age/baseline mortality data from 2017 to provide more realistic mortality estimates for 2017. It would be useful to see if the air pollution reductions in 2017 have increased benefits due to the increased population from 2015, for example.

We thank the reviewer for their suggestion and agree that it can be interesting to compare the effects of exposure and demography air pollution health impact. However, in the context of this study, which focusses on distinguishing the emissions and meteorological effects on air quality, we believe that including mortality changes due to changing age distribution, population and baseline mortality is outside the scope of the current study. We isolated the health impacts of the change in exposure by keeping the time frame constant thereby removing the influence of the confounding variations in population ageing, population size, and baseline disease levels. As the reviewer points out, health impact assessments are sensitive to the underlying epidemiological data and have rapidly developing methodologies. We now explain our approach more clearly (Line 150-152):

“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.”

Line 153: How did the Chen et al, 2019 trends compare and how did their emissions scaling compare?

Chen et al. (2019) did not scale emissions. They use 2010 emissions, which led to a ‘severe overestimation’ in PM, due to the decline in emissions that has occurred since 2010. Chen et al.

(2019) focused on wintertime (January). They also suggested that reductions in emissions had contributed to reduced wintertime PM_{2.5} concentrations (see Line 178-179):

“Chen et al. (2019) also concluded that emission reductions were the primary cause of reduced wintertime PM_{2.5} across China during 2015-2017.”

Line 157: Please provide more information on Guizhou and Li et al, 2018. Why do they see different trends? Did they use different emissions scaling? Did they look at different regions of China?

The reference to “Guizhou” is to the trend in the province of Guizhou in the fixed emissions run. The text has been amended to “Guizhou province” make this clear. For Li et al (2019a) [corrected], their trend estimate has been added in lines 184-186:

“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 μg m⁻³ year⁻¹) is comparable to the 2.6 μg m⁻³ year⁻¹ trend found in this study.”

Line 190-191: Suggest that the authors provide more context for what is meant by a “reasonable” NMB.

This word reasonable has been removed to remove subjectivity around the NMB. Additionally, a table has been added to the supplement (Table S1) that contains NMB recorded in other papers that have a similar WRF-Chem model setup.

Figure 2: Which color represents which region mentioned in the figure caption.

This has been added to the figure caption.

Figure 2: Suggest making the dots that don't have significant trends (i.e., the gray dots) a lighter blue or red color. Even if the trends are statistically significant the direction of the trend will still provide information.

Thank you for your suggestion, the figure caption has been updated to include the colour for each region, which corresponds to the legend in Figure 1.

Figure 4: I find this color scale to interpret because the mid-range yellows and greens all look similar. I suggest binning the color scale to make it easier to see how many lives are saved in each province.

Thank you for your suggestion, this has been changed to a binned colour scale.

Technical Corrections:

Line 157: Missing year of Guizhou. I also couldn't find this reference in the reference list.

Guizhou refers to the province of Guizhou, the text has been amended to make this clearer

Line 185: Should be “per year”.

This has been corrected

Line 247: The first two papers in the reference section don't seem to be in alphabetical order

These papers are ordered by “A” of “van der A”. We will check with the journal editorial team to ensure we are following journal guidelines.

Response to Anonymous Referee #2

The manuscript presents a study on estimating the changes of mortality due to air pollutants in China in 2015-2017 and explaining the causes of it using WRF-Chem simulations. For this, modeled trends are compared with observed ones to provide reliability in the model estimates. This study represents good contributions to the field and it's within the scope of ACP. I think the paper needs a bit more work before it's ready for publication based on the comments below. My main comment is the following. Given the issues in the modelled trends of the PM_{2.5} precursors (SO₂, NO_x), getting the right trend for PM_{2.5} could be do to a cancellation of errors, so you might be getting the right trend for the wrong reasons. I would like to encourage the authors to look into more details on this topic. For instance, analyze the model results by aerosol composition and how are the trends of each specie to assess the role of each of them. I would also encourage the authors to collaborate with other researchers that maintain sites where this speciation is observed and so the speciated comparison can be done as well. An example is the Beijing site from the Spartan network (<https://www.spartan-network.org/beijing-china>), but I'm sure there are many more. Even if a few sites are included this could provide useful information

We thank the reviewer for their comment, and agree that it is useful to evaluate simulated aerosol speciation by comparison with measurements. Unfortunately there is insufficient data to carefully evaluate the trend in aerosol speciation across China during the period analysed.

As suggested, we compare the model with the Beijing SPARTAN data (Snider et al., 2015, 2016). The model underestimates sulfate and overestimates nitrate at this particular site. The results of the comparison have been added to the supplement (Figure S4). We also compare to another set of measurements from Beijing that were reported by Zhou et al. (2019) (Figure S5), which point to an underestimate of sulfate, nitrate and ammonium in winter, but reasonable estimations in other seasons.

To extend the evaluation beyond Beijing, we also use a dataset of speciated aerosol data from different field campaigns compiled by Li et al. (2017) (Figure S6). Data spans multiple years from 2006-2013, so the comparison is complicated by comparing the model for 2015 against years of different meteorology and emissions. For this reason, we only compare means values across the campaign which will reduce the impacts of different meteorological conditions between the measurement and model. Nevertheless, this comparison also suggests the model underestimates sulfate and overestimate nitrate. The large changes in emissions (in particular the large decline in SO₂ emissions) over this period is likely to cause at least some of the underestimate in sulfate. The results of this comparison are added to the Supplement (Fig. S6). We add the following text to the paper on lines (118-128):

“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), Aerosol Chemical Speciation Monitor measurements from Beijing (Zhou et al. (2019)) (Figure S5) and Aerosol Mass Spectrometer measurements from across China (Li et al., 2017) (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."

There is insufficient data to evaluate the trends in aerosol speciation over the 2015 to 2017 period. We add the following text to the paper (Line 208-19):

"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., 2017; 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."

Comments by line:

66-70. Please list some references on the second approach.

Two references, Ansari et al. (2019) and Xing et al. (2011) have been added as examples of this approach on line 68.

79. Please briefly summarize the quality control process

A brief summary of the quality control process has been added on lines 86-87: '...which include excluding data with a high proportion of repeated measurements and periods of low variability'

Section 2.2. Any previous work where you have used this or similar configuration with positive results in terms of meteorology, PM_{2.5} and O₃? In this work you are not much model evaluation other than the evaluation of the trends and brief statistics in sections

Further detail of the model evaluation has been added in Table 2 (see comments to Referee #1). Previous studies using a similar model set-up that demonstrate the model's ability to capture PM include Reddington et al. (2019). A table (Table S1) has been added to the supplementary showing the comparable NMB statistics of published research that use a similar WRF-Chem setup to simulate air pollution in China.

2.3. Adding evaluation on the ability of this model configuration to capture aerosol speciation would also be desirable.

Two plots that show the ability of the model to capture aerosol speciation, Figures S4 and S5, have been added to the supplement. We have also added text to the manuscript (see response to Referee #1).

103 Hodzic and Jimenez, and Knote et al. papers described two very different SOA schemes, please specify which one you are using.

We use the scheme described in Hodzic and Jimenez. The second reference on line 112 has been corrected to Hodzic and Knote (2014).

Section 2.4. Can you briefly describe the exposure response functions used for PM_{2.5} and O₃?

We have added more information to the paper on the shape of the exposure-response functions. For PM_{2.5} on lines 140-142:

“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.”

And for O₃ on lines 144-148:

For ambient O₃, 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 O₃, and a sub-linear association at higher concentrations.

129. After reading section 3.1, I don't think the trends compare largely well as stated in this sentence. You could make this point for PM_{2.5}, but for the others, although the sign is generally correct, the magnitudes tends to be off by at least factor of 2. For the case of NO₂ the sign of the trend is not even well captured. Please revise to better represent the actual results

We have changed this sentence to remove the statement that all the trends compare well. The sentence now reads (line 155):

“Figure 1 and 2 compare measured and simulated air quality trends over China during 2015 to 2017”

129-136. Can you add additional analysis in whether the model captures the regions with more negative (and more positive) trends? I see this info in the plots but it's not discussed.

The regional distribution of trends for PM are discussed in lines 160-161 and for O₃ in lines 163-164. Line 166-167 mentions the fact that the model does not capture the regional distribution of trends for NO₂.

150. Is clear to me that natural emissions remained equal in the two emission scenarios, but what did the authors do for biomass burning emissions?

Biomass burning emissions are from the FINN inventory in both scenarios. We now clarify this in the manuscript (Line 132-133):

“Both simulations include interannual variability in biogenic and biomass burning emissions, allowing us to isolate the impacts of changing anthropogenic emissions.”

153-154. Chen et al. (2019) also found that there were periods where meteorology did play a role, can you compare your results to theirs?

We add the following sentence to our manuscript (line 178-179):

Chen et al. (2019) also concluded that emission reductions were the primary cause of reduced wintertime PM_{2.5} across China during 2015-2017

155-156. I would say "little influence" rather than "no influence" as you are basing your analysis in a model that contains uncertainties.

This has been corrected in the text.

166-167. This statement depends if the region is NO_x or VOC limited. Might be good to include these indicators from the model perspective to shows that this is what's actually happening in the model.

We agree with the reviewer that this statement depends on the ozone regime, and we have added a reference to a satellite study which finds that a NO_x limited or mixed regime dominates across China. We amended the sentence (lines 198-200):

"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)."

168. The Li et al. (2019a) study blames heterogeneous chemistry happening in declining particles for the negative trend. Is this process included in this WRF-Chem configuration and how this influences your results? An attempt to compute similar metrics as in the Li study might be good to intercompare results.

In our version of WRF-Chem, the heterogeneous HO₂ uptake process is not included. Li et al. (2019b) conclude that this process is the main driver for the positive O₃ trend across China, which has been disputed in Tan et al. (2020), who find that this process was not significant in the NCP region. The main aim of our study was to compare the importance of meteorology and emissions in driving trends of major air pollutants. Therefore, we believe that performing additional model runs to specifically examine the chemistry driving the O₃ trend is beyond the scope of this study.

191-196. Is not clear in this paragraph where you consider the model issues on O3 trends.

In this paragraph, we acknowledge the underestimation of the positive O₃ trend, which is why we do not use the modelled trends to estimate the change in O₃ caused mortality. We instead apply the measured trend to the 2015 model fields. We have clarified this in the paper.

Minor Edits

75. I believe ACP policy is to not use links but references, please check.

We have seen these data sources hyperlinked in other ACP articles, and will verify this is acceptable during the editing process.

124. Fix issue with the symbol after 0.05

This has been corrected

329. Should this be 2019b?

The references have been checked to ensure they correspond to the correct paper.

List of major changes

- Added a paragraph in Section 1.2 to better define the scope of the article with reference to recent similar articles
- Extended Section 2.3 by adding detail of comparison of our simulated aerosol with speciated aerosol measurement datasets. The comparisons are shown in the newly added figures to the supplement, as Figures S4-6.
- Added a new table (Table 2) that gives more detail of the model biases and is referenced in Section 3.2.
- Added a paragraph to the end of Section 3.2 that identifies areas for future research into how changing emissions are driving trends in particulate matter species.

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, 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. \(2019\) 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.

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. 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; Knote et al., 2014). (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 $\text{PM}_{2.5}$ (normalised mean bias (NMB) = 0.45), O_3 (NMB=-0.13) and SO_2 (NMB=0.07), while overestimating NO_2 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 $\text{PM}_{2.5}$, 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). 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 $\text{PM}_{2.5}$ using the Global Exposure Mortality Model (GEMM) (Burnett et al., 2018), which uses cohort studies to estimate health risks integrated over a range of $\text{PM}_{2.5}$ 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 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 O₃, and a sub-linear association at higher concentrations. 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, 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 Varying emissions scenario

~~Measured~~ Figure 1 and 2 compare 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_{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). 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.

3.2 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 O₃MDA8 trend in the fixed emission simulation is 0.0 µg m⁻³ 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 µg m⁻³ 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 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 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 µg m⁻³) compared to the control simulation (-2.2 µg 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 by 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 ~~underestimated the observed magnitude of the O₃MDA8 trend during 2015-2017, so the simulated change in health impacts would also be underestimated. However, our model bias in O₃-O₃ concentrations across China during 2015-2017 was reasonable 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 ~~change in~~ health impacts due to ~~increasing exposure to O₃ concentrations~~ we used simulated concentrations to estimate average ~~health impacts due to~~ exposure to O₃ over the 2015-2017 period, ~~and then multiplied by the measured relative change in O₃MDA8.~~ 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. 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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