



Pollutant emission reductions deliver decreased PM_{2.5}-caused mortality across China during 2015-2017

Ben Silver¹, Luke Conibear¹, Carly L. Reddington¹, Christoph Knote², Steve R. Arnold¹ and Dominick V. Spracklen¹

- ⁵ School of Earth and Environment, University of Leeds, Leeds, LS2 9JT, UK
 - ² Meteorological Institute, Ludwig-Maximilians-University Munich, Theresienstr. 37, Munich, 80333, Germany *Correspondence to*: Ben Silver (eebjs@leeds.ac.uk)

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}-attribrutable premature morality 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).

© Author(s) 2020. CC BY 4.0 License.



30

45

55



1.1 Previous studies of trends in China's air quality

positive trends in Nitrogen Dioxide (NO₂) (Van der A et al., (2006), Sulphur 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., 2017; Zheng et al., 2018). Analysis of measurements from the Acid Deposition Monitoring Network in East Asia (EANET) shows a negative pH trend (i.e., becoming more acidic) from 1999 until a reversal occurs in 2006, matching peak SO₂ emissions and concentrations (Duan et al., 2016). Measurements of O₃ concentrations at background monitoring sites indicate positive trends in western China during 1994-2013 (Xu et al., 2016), and Taiwan during 1994-2003 (Chang and Lee, 2007), suggesting that 40 O₃ has been increasing across China during the past two decades. More recently, measurements at urban sites, also show positive O₃ trends during 2005-2011 (Zhang et al., 2014). The establishment of China's air pollution monitoring network, operated by the China National Environmental Monitoring Centre (CNEMC) (Wang et al., 2015), which includes measurements from over 1600 locations, has enabled more detailed analysis of recent air pollution changes (Silver et al., 2018; Zhai et al., 2019). Between 2015 and 2017, PM_{2.5} concentrations 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 m3 in 2013 to 164 µg m3 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. Positive regional O₃ trends remain even after

Satellite remote sensing studies have been used to show large changes in air pollution across China in recent decades, with

1.2 Identifying drivers of recent trends

(SCB) and PRD, and no overall trend at the national scale.

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

meteorological variability has been removed (Li et al., 2019a). Trends in NO2 are more variable, with a negative trend reported in eastern China and positive trends in western areas (Li and Bai, 2019). Silver et al., (2018) found that NO₂ had negative trends in Hong Kong and North China Plain regions, but positive trends in the Yangtze River Delta (YRD), Sichuan Basin

© Author(s) 2020. CC BY 4.0 License.





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

There are two commonly used approaches to separate the influences of meteorology and emissions on variability in atmospheric pollutant abundances. The first approach uses statistical models, such as multi-linear regression, to control for the influence of meteorology and allowing the proportion of air pollutant concentration variability that can be explained by meteorological variables to be calculated (Tai et al., 2010). The second approach is to use an atmospheric chemistry transport model to simulate pollutant concentrations. Through a comparison of multiple simulations, where either annual variability in emissions or meteorology are held constant, the relative influence of the two factors can be estimated. Here we analyse measurements and a regional air quality model to explore the role of changing anthropogenic emissions on air pollutant 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). 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

© Author(s) 2020. CC BY 4.0 License.



100

105

110

115



agricultural emission sectors combined with China-specific emission factors (Hong et al., 2017). We used the 2015 MEIC dataset, then used sector-specific and species-specific scaling for 2016 and 2017 based on the emission totals estimated in B. Zheng et al. (2018). Table 1 shows emission totals for 2015, 2016 and 2017. Over the 2015 to 2017 period, Chinese emissions decreased by 38% for SO₂, 16% for PM_{2.5} and 8% for NOx. For regions outside the MEIC dataset, we used anthropogenic emissions from the EDGAR-HTAPv2.2 emission inventory for 2010.

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). 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), 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).

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

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

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

© Author(s) 2020. CC BY 4.0 License.



125



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

3 Measured and modelled trends comparison

3.1 Varying emissions scenario

Measured and simulated air quality trends over China during 2015 to 2017 largely compare well, and are shown in Figure 1 and 2. The measurements show widespread decline in PM_{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 O_3MDA8 , but underestimates the magnitude of the trend by a factor 2 (2.7 $\mu g \ m^{-3} \ year^{-1}$ in the measurements, versus 1.3 $\mu g \ m^{-3} \ year^{-1}$ simulated by WRF-Chem). WRF-Chem simulates negative O_3MDA8 trends in the Sichuan Basin and Taiwan, whereas in the measured data, all regions have positive median trends.

140 The measurements show zero overall median trend in NO₂ concentrations, with 46% of sites with significant trends being negative and 54% positive. In contrast, WRF-Chem simulates widespread reductions in NO₂ concentrations, with 100% of significant sites exhibiting negative trends and a negative nationwide median trend of -2.2 μg m⁻³ year⁻¹. The 7.0 % nationwide median decline in simulated NO₂ concentrations over 2015-2017, matches the 7.6 % decline in Chinese NOx emissions in the MEIC.

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

150 The model simulation where anthropogenic emissions in China were fixed at 2015 levels has a weak negative PM_{2.5} trend (-0.6 μg m⁻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

155 The median O₃MDA8 trend in the fixed emission simulation is 0.0 μg m⁻³ year⁻¹. This suggests that interannual meteorological variation had no 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, a trend of -2.5 μg m⁻³ year⁻¹ was calculated in the fixed emissions simulation. Li et al. (2018) also report that the positive ozone trend over 2013 to 2017 is due to changes in anthropogenic emissions.

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 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., 2019b), 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. Other work has suggested that increased O₃ concentrations are possibly linked to the rapid decline in aerosol (Li et al., 2019a).

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

170 **4.1 PM_{2.5} health impacts**

175

180

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



190

195

200

205

210



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

4.2 O₃ health impacts

Increasing O_3 concentrations will result in an increase in health impacts that will act to offset some of the health benefits from declining PM_{2.5} concentrations. WRF-Chem underestimated the observed magnitude of the O_3 MDA8 trend during 2015-2017, so the simulated change in health impacts would also be underestimated. However, our model bias in O_3 across China during 2015-2017 was reasonable (NMB=-0.13). To provide an estimate of the change in health impacts due to increasing O_3 concentrations we used simulated concentrations to estimate average health impacts due to exposure to O_3 over the 2015-2017 period, and then multiplied by the measured relative change in O_3 MDA8. We estimate that exposure to O_3 caused an average of 143 000 (CI: 106 000 – 193 000) premature mortalities each year over 2015-2017. Assuming linear behaviour, the 15% measured increase in O_3 MDA8 would result in an increase of 21 000 premature mortalities per year. The exposure-outcome function is in reality sub-linear, so this is likely to be an overestimate. Regardless, this is substantially smaller than the 150 000 reduction in annual premature mortality due to reduced PM_{2.5}. We therefore suggest that changes in Chinese air pollution over 2015-2017 have likely had an overall beneficial impact on human health.

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

© Author(s) 2020. CC BY 4.0 License.



Atmospheric Chemistry And Physics

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 O_3 concentrations.

Data availability

220

225

235

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.

Acknowledgements

230 We gratefully acknowledge the AIA Group Limited and Natural Environment Research Council for funding for this research.

Model simulations were undertaken on ARC3, part of the High Performance Computing facilities at the University of Leeds.

We thank Qiang Zhang and Meng Li for providing MEIC data. We acknowledge use of the WRF-Chem preprocessor tools

bio emiss, anthro emiss, fire emiss, and mozbc provided by the Atmospheric Chemistry Observations and Modeling Lab

(ACOM) of the NCAR. We acknowledge use of NCAR/ACOM .MOZART-4 global model output available

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

References

van der A, R. J., Peters, D. H. M. U., Eskes, H., Boersma, K. F., Van Roozendael, M., De Smedt, I. and Kelder, H. M.:

Detection of the trend and seasonal variation in tropospheric NO<inf>2</inf> over China, J. Geophys. Res. Atmos.,

doi:10.1029/2005JD006594, 2006.





- Van Der A, R. J., Mijling, B., Ding, J., Elissavet Koukouli, M., Liu, F., Li, Q., Mao, H. and Theys, N.: Cleaning up the air: Effectiveness of air quality policy for SO2 and NO*x* emissions in China, Atmos. Chem. Phys., 17(3), 1775–1789, doi:10.5194/acp-17-1775-2017, 2017.
 - Apte, J. S., Marshall, J. D., Cohen, A. J. and Brauer, M.: Addressing Global Mortality from Ambient PM2.5, Environ. Sci. Technol., 49(13), 8057–8066, doi:10.1021/acs.est.5b01236, 2015.
- Burnett, R., Chen, H., Szyszkowicz, M., Fann, N., Hubbell, B., Pope, C. A., Apte, J. S., Brauer, M., Cohen, A., Weichenthal, S., Coggins, J., Di, Q., Brunekreef, B., Frostad, J., Lim, S. S., Kan, H., Walker, K. D., Thurston, G. D., Hayes, R. B., Lim, C. C., Turner, M. C., Jerrett, M., Krewski, D., Gapstur, S. M., Diver, W. R., Ostro, B., Goldberg, D., Crouse, D. L., Martin, R. V, Peters, P., Pinault, L., Tjepkema, M., van Donkelaar, A., Villeneuve, P. J., Miller, A. B., Yin, P., Zhou, M., Wang, L., Janssen, N. A. H., Marra, M., Atkinson, R. W., Tsang, H., Quoc Thach, T., Cannon, J. B., Allen, R. T., Hart, J. E., Laden, F.,
- 250 Cesaroni, G., Forastiere, F., Weinmayr, G., Jaensch, A., Nagel, G., Concin, H., Spadaro, J. V and Spadaro, J. V.: Global estimates of mortality associated with long-term exposure to outdoor fine particulate matter., Proc. Natl. Acad. Sci. U. S. A., 115(38), 9592–9597, doi:10.1073/pnas.1803222115, 2018.
 - Carslaw, D.: The openair manual open-source tools for analysing air pollution data, King's Coll. London, (January), 287 [online] Available from: http://www.openair-project.org/PDF/OpenAir_Manual.pdf, 2015.
- 255 Carslaw, D. C. and Ropkins, K.: openair Data Analysis Tools for the Air Quality Community, R J., 4(1), 20–29, 2012.
 Chang, S. C. and Lee, C. Te: Evaluation of the trend of air quality in Taipei, Taiwan from 1994 to 2003, Environ. Monit.
 Assess., doi:10.1007/s10661-006-9262-1, 2007.
 - Chen, D., Liu, Z., Ban, J., Zhao, P. and Chen, M.: Retrospective analysis of 2015-2017 wintertime PM2.5 in China: Response to emission regulations and the role of meteorology, Atmos. Chem. Phys., doi:10.5194/acp-19-7409-2019, 2019.
- Cohen, A. J., Brauer, M., Burnett, R., Anderson, H. R., Frostad, J., Estep, K., Balakrishnan, K., Brunekreef, B., Dandona, L., Dandona, R., Feigin, V., Freedman, G., Hubbell, B., Jobling, A., Kan, H., Knibbs, L., Liu, Y., Martin, R., Morawska, L., Pope, C. A., Shin, H., Straif, K., Shaddick, G., Thomas, M., van Dingenen, R., van Donkelaar, A., Vos, T., Murray, C. J. L. and Forouzanfar, M. H.: Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015, Lancet, 389(10082), 1907–1918, doi:10.1016/S0140-6736(17)30505-6, 2017.
 - Conibear, L., Butt, E. W., Knote, C., Arnold, S. R. and Spracklen, D. V.: Residential energy use emissions dominate health impacts from exposure to ambient particulate matter in India, Nat. Commun., doi:10.1038/s41467-018-02986-7, 2018.
 - Cui, Y., Lin, J., Song, C., Liu, M., Yan, Y., Xu, Y. and Huang, B.: Rapid growth in nitrogen dioxide pollution over Western China, Atmos. Chem. Phys, 16, 2005–2013, doi:10.5194/acp-16-6207-2016, 2016.
- Ding, D., Xing, J., Wang, S., Liu, K. and Hao, J.: Estimated Contributions of Emissions Controls, Meteorological Factors, Population Growth, and Changes in Baseline Mortality to Reductions in Ambient PM2.5 and PM2.5-Related Mortality in China, 2013-2017, Environ. Health Perspect., doi:10.1289/EHP4157, 2019.
 - Duan, L., Yu, Q., Zhang, Q., Wang, Z., Pan, Y., Larssen, T., Tang, J. and Mulder, J.: Acid deposition in Asia: Emissions,





- deposition, and ecosystem effects, Atmos. Environ., doi:10.1016/j.atmosenv.2016.07.018, 2016.
- Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L. and Kloster, S.: Geoscientific Model Development Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4). [online] Available from: www.geosci-model-dev.net/3/43/2010/, 2010.
- Feng, Y., Ning, M., Lei, Y., Sun, Y., Liu, W. and Wang, J.: Defending blue sky in China: Effectiveness of the "Air Pollution Prevention and Control Action Plan" on air quality improvements from 2013 to 2017, J. Environ. Manage., 252, 109603, doi:10.1016/J.JENVMAN.2019.109603, 2019.
 - De Foy, B., Lu, Z. and Streets, D. G.: Satellite NO2 retrievals suggest China has exceeded its NOx reduction goals from the twelfth Five-Year Plan, Sci. Rep., 6, doi:10.1038/srep35912, 2016.
 - GBD 2017 Risk Factor Collaborators, Stanaway, J. D., Afshin, A., Gakidou, E., Lim, S. S., Abate, D., Abate, K. H., Abbafati,
- C., Abbasi, N., Abbastabar, H., Abd-Allah, F., Abdela, J., Abdelalim, A., Abdollahpour, I., Abdulkader, R. S., Abebe, M., Abebe, Z., Abera, S. F., Abil, O. Z., Abraha, H. N., Abrham, A. R., Abu-Raddad, L. J., Abu-Rmeileh, N. M. E., Accrombessi, M. M. K., Acharya, D., Acharya, P., Adamu, A. A., Adane, A. A., Adebayo, O. M., Adedoyin, R. A., Adekanmbi, V., Ademi, Z., Adetokunboh, O. O., Adib, M. G., Admasie, A., Adsuar, J. C., Afanvi, K. A., Afarideh, M., Agarwal, G., Aggarwal, A., Aghayan, S. A., Agrawal, A., Agrawal, S., Ahmadi, A., Ahmadi, M., Ahmadieh, H., Ahmed, M. B., Aichour, A. N., Aichour,
- I., Aichour, M. T. E., Akbari, M. E., Akinyemiju, T., Akseer, N., Al-Aly, Z., Al-Eyadhy, A., Al-Mekhlafi, H. M., Alahdab, F., Alam, K., Alam, S., Alam, T., Alashi, A., Alavian, S. M., Alene, K. A., Ali, K., Ali, S. M., Alijanzadeh, M., Alizadeh-Navaei, R., Aljunid, S. M., Alkerwi, A., Alla, F., Alsharif, U., Altirkawi, K., Alvis-Guzman, N., Amare, A. T., Ammar, W., Anber, N. H., Anderson, J. A., Andrei, C. L., Androudi, S., Animut, M. D., Anjomshoa, M., Ansha, M. G., Antó, J. M., Antonio, C. A. T., Anwari, P., Appiah, L. T., Appiah, S. C. Y., Arabloo, J., Aremu, O., Ärnlöv, J., Artaman, A., Aryal, K. K., Asayesh, H.,
- Ataro, Z., Ausloos, M., Avokpaho, E. F. G. A., Awasthi, A., Quintanilla, B. P. A., Ayer, R., et al.: Global, regional, and national comparative risk assessment of 84 behavioural, environmental and occupational, and metabolic risks or clusters of risks for 195 countries and territories, 1990-2017: A systematic analysis for the Global Burden of Disease Stu, Lancet, doi:10.1016/S0140-6736(18)32225-6, 2018.
- Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C. and Eder, B.: Fully coupled "online" chemistry within the WRF model, Atmos. Environ., doi:10.1016/j.atmosenv.2005.04.027, 2005.
 - Gu, Y. and Yim, S. H. L.: The air quality and health impacts of domestic trans-boundary pollution in various regions of China, Environ. Int., 97, 117–124, doi:10.1016/j.envint.2016.08.004, 2016.
 - Guenther, A., Geron, C., Pierce, T., Lamb, B., Harley, P. and Fall, R.: Natural emissions of non-methane volatile organic compounds, carbon monoxide, and oxides of nitrogen from North America, Atmos. Environ., 34(12), 2205–2230, doi:10.1016/S1352-2310(99)00465-3, 2000.
 - Hodzic, A. and Jimenez, J. L.: Geoscientific Model Development Modeling anthropogenically controlled secondary organic aerosols in a megacity: a simplified framework for global and climate models, Geosci. Model Dev, 4, 901–917,





- doi:10.5194/gmd-4-901-2011, 2011.
- Hong, C., Zhang, Q., He, K., Guan, D., Li, M., Liu, F. and Zheng, B.: Variations of China's emission estimates: Response to uncertainties in energy statistics, Atmos. Chem. Phys., 17(2), 1227–1239, doi:10.5194/acp-17-1227-2017, 2017.
 - Huang, J., Pan, X., Guo, X. and Li, G.: Health impact of China's Air Pollution Prevention and Control Action Plan: an analysis of national air quality monitoring and mortality data, Lancet Planet. Heal., doi:10.1016/S2542-5196(18)30141-4, 2018.
 - Irie, H., Muto, T., Itahashi, S., Kurokawa, J. and Uno, I.: Turnaround of Tropospheric Nitrogen Dioxide Pollution Trends in China, Japan, and South Korea, Sola, 12(0), 170–174, doi:10.2151/sola.2016-035, 2016.
- 315 Jacob, D. J. and Winner, D. A.: Effect of climate change on air quality, Atmos. Environ., 43(1), 51–63, doi:10.1016/j.atmosenv.2008.09.051, 2009.
 - Knote, C., Hodzic, A., Jimenez, J. L., Volkamer, R., Orlando, J. J., Baidar, S., Brioude, J., Fast, J., Gentner, D. R., Goldstein, A. H., Hayes, P. L., Knighton, W. B., Oetjen, H., Setyan, A., Stark, H., Thalman, R., Tyndall, G., Washenfelder, R., Waxman, E. and Zhang, Q.: Simulation of semi-explicit mechanisms of SOA formation from glyoxal in aerosol in a 3-D model, Atmos.
- 320 Chem. Phys., 14(12), 6213–6239, doi:10.5194/acp-14-6213-2014, 2014.
 - Krotkov, N. A., McLinden, C. A., Li, C., Lamsal, L. N., Celarier, E. A., Marchenko, S. V., Swartz, W. H., Bucsela, E. J., Joiner, J., Duncan, B. N., Folkert Boersma, K., Pepijn Veefkind, J., Levelt, P. F., Fioletov, V. E., Dickerson, R. R., He, H., Lu, Z. and Streets, D. G.: Aura OMI observations of regional SO2 and NO2 pollution changes from 2005 to 2015, Atmos. Chem. Phys., 16(7), 4605–4629, doi:10.5194/acp-16-4605-2016, 2016.
- LeGrand, S. L., Polashenski, C., Letcher, T. W., Creighton, G. A., Peckham, S. E. and Cetola, J. D.: The AFWA dust emission scheme for the GOCART aerosol model in WRF-Chem v3.8.1, Geosci. Model Dev., doi:10.5194/gmd-12-131-2019, 2019. Lelieveld, J., Evans, J. S., Fnais, M., Giannadaki, D. and Pozzer, A.: The contribution of outdoor air pollution sources to premature mortality on a global scale, Nature, 525(7569), 367–371, doi:10.1038/nature15371, 2015.
 - Li, K. and Bai, K.: Spatiotemporal Associations between PM2.5 and SO2 as well as NO2 in China from 2015 to 2018, Int. J.
- 330 Environ. Res. Public Health, 16(13), 2352, doi:10.3390/ijerph16132352, 2019.
 - Li, K., Jacob, D. J., Liao, H., Shen, L., Zhang, Q. and Bates, K. H.: Anthropogenic drivers of 2013–2017 trends in summer surface ozone in China, Proc. Natl. Acad. Sci. U. S. A., doi:10.1073/pnas.1812168116, 2019a.
 - Li, M., Liu, H., Geng, G., Hong, C., Liu, F., Song, Y., Tong, D., Zheng, B., Cui, H., Man, H., Zhang, Q. and He, K.: Anthropogenic emission inventories in China: A review, Natl. Sci. Rev., doi:10.1093/nsr/nwx150, 2017.
- Li, R., Bo, H. and Wang, Y.: Slowing-down reduction and Possible Reversal Trend of Tropospheric NO2 over China during 2016 to 2019, [online] Available from: http://arxiv.org/abs/1907.06525 (Accessed 23 September 2019b), 2019.
 - Ma, Z., Hu, X., Sayer, A. M., Levy, R., Zhang, Q., Xue, Y., Tong, S., Bi, J., Huang, L. and Liu, Y.: Satellite-based spatiotemporal trends in PM2.5concentrations: China, 2004-2013, Environ. Health Perspect., 124(2), 184–192, doi:10.1289/ehp.1409481, 2016.
- Otte, T. L., Nolte, C. G., Otte, M. J. and Bowden, J. H.: Does nudging squelch the extremes in regional climate modeling?, J. Clim., doi:10.1175/JCLI-D-12-00048.1, 2012.





- Reddington, C. L., Conibear, L., Knote, C., Silver, B. J., Arnold, S. R. and Spracklen, D. V.: Exploring the impacts of anthropogenic emission sectors on PM<sub>2.5</sub> and human health in South and East Asia, Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2019-147, 2019.
- Silver, B., Reddington, C. L., Arnold, S. R. and Spracklen, D. V.: Substantial changes in air pollution across China during 2015-2017, Environ. Res. Lett., doi:10.1088/1748-9326/aae718, 2018.
 - Streets, D. G., Yu, C., Wu, Y., Chin, M., Zhao, Z., Hayasaka, T. and Shi, G.: Aerosol trends over China, 1980–2000, Atmos. Res., 88(2), 174–182, doi:10.1016/J.ATMOSRES.2007.10.016, 2008.
- Tai, A. P. K., Mickley, L. J. and Jacob, D. J.: Correlations between fine particulate matter (PM2.5) and meteorological variables in the United States: Implications for the sensitivity of PM2.5 to climate change, Atmos. Environ., 44(32), 3976–3984, doi:10.1016/j.atmosenv.2010.06.060, 2010.
 - Wang, S., Li, G., Gong, Z., Du, L., Zhou, Q., Meng, X., Xie, S. and Zhou, L.: Spatial distribution, seasonal variation and regionalization of PM2.5 concentrations in China, Sci. China Chem., doi:10.1007/s11426-015-5468-9, 2015.
 - Wiedinmyer, C., Akagi, S. K., Yokelson, R. J., Emmons, L. K., Al-Saadi, J. A., Orlando, J. J. and Soja, A. J.: The Fire
- INventory from NCAR (FINN): A high resolution global model to estimate the emissions from open burning, Geosci. Model Dev., 4(3), 625–641, doi:10.5194/gmd-4-625-2011, 2011.
 - Xu, W., Lin, W., Xu, X., Tang, J., Huang, J., Wu, H. and Zhang, X.: Long-term trends of surface ozone and its influencing factors at the Mt Waliguan GAW station, China-Part 1: Overall trends and characteristics, Atmos. Chem. Phys., 16(10), 6191–6205, doi:10.5194/acp-16-6191-2016, 2016.
- Zaveri, R. A., Easter, R. C., Fast, J. D. and Peters, L. K.: Model for Simulating Aerosol Interactions and Chemistry (MOSAIC), J. Geophys. Res. Atmos., doi:10.1029/2007JD008782, 2008.
 - Zhai, S., Jacob, D. J., Wang, X., Shen, L., Li, K., Zhang, Y., Gui, K., Zhao, T. and Liao, H.: Fine particulate matter (PM<sub>2.5</sub>) trends in China, 2013–2018: separating contributions from anthropogenic emissions and meteorology, Atmos. Chem. Phys., doi:10.5194/acp-19-11031-2019, 2019.
- Zhang, H., Wang, S., Hao, J., Wang, X., Wang, S., Chai, F. and Li, M.: Air pollution and control action in Beijing, J. Clean. Prod., 112, 1519–1527, doi:10.1016/j.jclepro.2015.04.092, 2016.
 - Zhang, L., Lee, C. S., Zhang, R. and Chen, L.: Spatial and temporal evaluation of long term trend (2005–2014) of OMI retrieved NO2and SO2concentrations in Henan Province, China, Atmos. Environ., doi:10.1016/j.atmosenv.2016.11.067, 2017.
- Zhang, Q., Geng, G. N., Wang, S. W., Richter, A. and He, K. Bin: Satellite remote sensing of changes in NO x emissions over
- China during 1996-2010, Chinese Sci. Bull., 57(22), 2857–2864, doi:10.1007/s11434-012-5015-4, 2012.
 Zhang, Q., Yuan, B., Shao, M., Wang, X., Lu, S., Lu, K., Wang, M., Chen, L., Chang, C. C. and Liu, S. C.: Variations of ground-level O3 and its precursors in Beijing in summertime between 2005 and 2011, Atmos. Chem. Phys., doi:10.5194/acp-14-6089-2014, 2014.
 - Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng,
- 375 Y., He, K. and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions,





Atmos. Chem. Phys. Discuss., (X), 1–27, doi:10.5194/acp-2018-374, 2018.

Zheng, Y., Xue, T., Zhang, Q., Geng, G., Tong, D., Li, X. and He, K.: Air quality improvements and health benefits from China's clean air action since 2013, Environ. Res. Lett., 12(11), doi:10.1088/1748-9326/aa8a32, 2017.

Table 1. Chinese pollutant emissions (Tg yr^{-1}) during 2015 to 2017 from MEIC.

	SO_2	NO _x	NMVOC	NH ₃	СО	TSP	PM_{10}	PM _{2.5}	BC	OC	CO_2
2015	16.9	23.7	28.6	10.5	153.6	21.9	12.3	9.1	1.4	2.5	10347.2
2016	13.4	22.5	28.4	10.2	142	17.9	10.8	8.1	1.3	2.3	10290.7
2017	10.5	21.9	28.6	10.2	136.2	16.7	10.2	7.6	1.2	2.1	10434.3





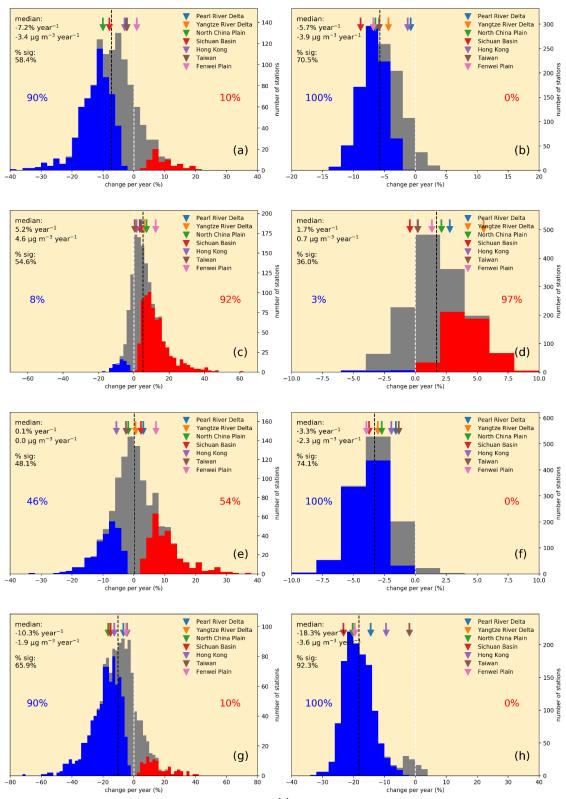


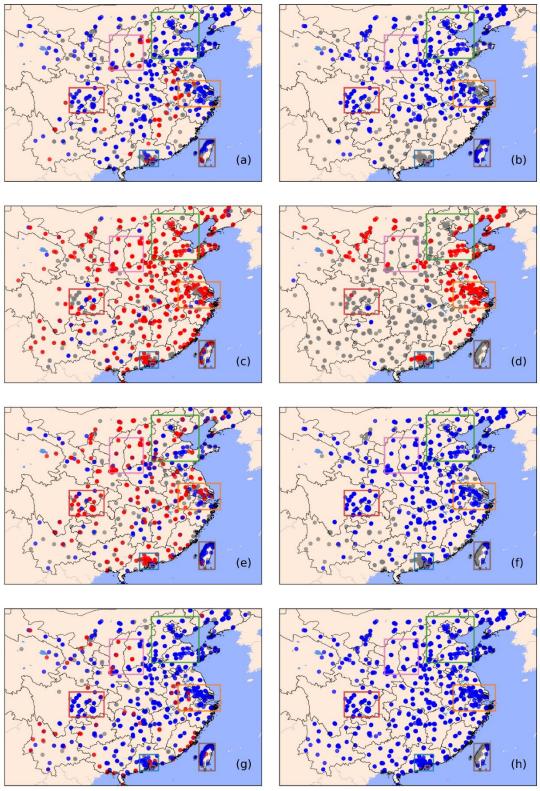




Figure 1: Histograms showing the frequency distribution of trends in concentrations of (a,b) PM_{2.5}, (c,d) O₃MDA8, (e,f) NO₂, (g,h) SO₂ across China and Taiwan during 2015–2017. Measured trends (left hand panels) are compared to simulated trends (right hand panels). The median relative and absolute trend as well as the percentage of stations with significant trends is shown on each panel. The percentage of significant trends that are negative (blue) or positive (red) are also shown. The black dotted line shows the median trend across all sites, while the white dotted line shows zero. Arrows show the median trend for the regional domain: Pearl River Delta (PRD), Yangtze River Delta (YRD), North China Plain (NCP), Sichuan Basin (SCB), Hong Kong (HK), Taiwan (TW) and the Fenwei Plain (FWP).





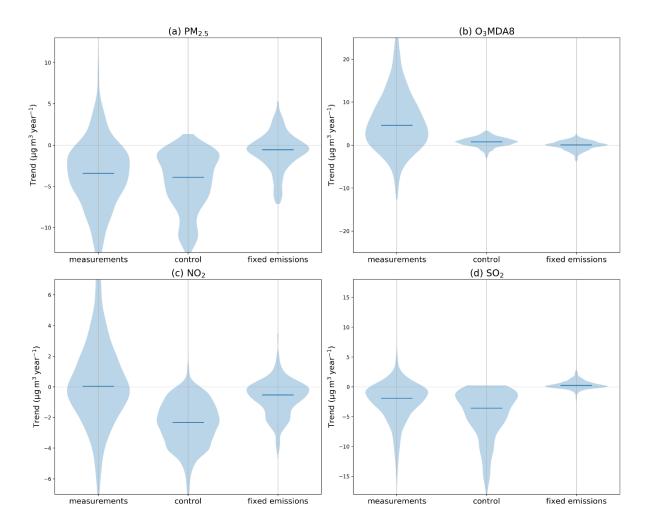




395



Figure 2. Map showing the spatial distribution of trends in concentrations of (a,b) PM_{2.5}, (c,d) O₃MDA8, (e,f) NO₂, (g,h) SO₂ across China and Taiwan during 2015–2017. Measured trends (left hand panels) are compared to simulated trends (right hand panels). Red indicates a significant positive trend, whereas blue indicates a significant negative trend, and grey an insignificant trend. Coloured boxes show the regional domains: Pearl River Delta (PRD), Yangtze River Delta (YRD), North China Plain (NCP), Sichuan Basin (SCB), Hong Kong (HK), Taiwan (TW) and the Fenwei Plain (FWP).



400 Figure 3. Comparison of measured and simulated concentration trends during 2015 to 2017. The left violin shows the measured trend, the centre shows the simulated trend with varying emissions and meteorology (control), and the right shows the simulated trends for the fixed emissions simulation. a) PM2.5, b) O3MDA8, c) NO2, d) SO2. The solid line shows the median absolute trend, and the shaded area shows a smoothed relative frequency distribution.





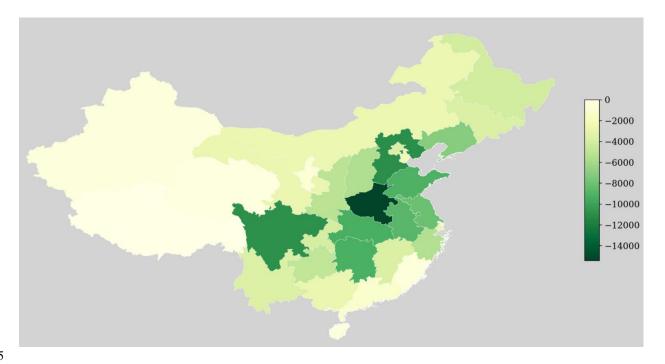


Figure 4. Simulated change during 2015-2017 in annual premature mortality per year due to changes in exposure to ambient $PM_{2.5}$. Results are shown at the province scale.