Acute and chronic health impacts of $PM_{2.5}$ in China and the influence of interannual meteorological variability

Yuanlin Wang[1,2,3]; Oliver Wild[2]; Huansheng Chen[1]; Meng Gao[4]; Qizhong Wu[5]; Yi Qi[6]; Xueshun Chen[1]; Zifa Wang[1,*]

1 The State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 10029, China

2 Lancaster Environment Centre, Lancaster University, LA1 4YQ, United Kingdom

3 University of Chinese Academy of Sciences, Beijing 100049, China

4 Department of Geography, Hong Kong Baptist University, Hong Kong SAR, China

5 College of Global Change and Earth System Science, Beijing Normal University, Beijing, 100875, China

6 School of Architecture and Urban Planning, Nanjing University, Nanjing, 210093, China

[*]*Corresponding author address:*

Prof. Zifa Wang

LAPC, Institute of Atmospheric Physics (IAP)

Chinese Academy of Sciences (CAS)

Beijing 100029, China

E-mail: zifawang@mail.iap.ac.cn

Abstract:

High concentrations of $PM_{2.5}$ in China have an adverse impact on human health and present a major problem for air quality control. Here we evaluate premature deaths attributable to chronic and acute exposure to ambient $PM_{2.5}$ at different scales in China over 2013-2017 with an air quality model at 5 km resolution and integrated exposure-response methods. We estimate that 1,210,000 (95% Confidence Interval: 720,000–1,750,000) premature deaths annually are attributable to chronic exposure to $PM_{2.5}$ pollution. Chongqing exhibits the largest chronic per capita mortality (1.4‰) among all provinces. A total of 116,000 (64,000–170,000) deaths annually are attributable to acute exposure during pollution episodes over the period, with Hubei province showing the highest acute per capita mortality (0.15‰). We also find that in urban areas premature deaths are 520,000 (320,000–760,000) due to chronic and 55,000 (3,000-81,000) due to acute exposure, respectively. At a provincial level, the annual mean $PM_{2.5}$ concentration varies by ±20% due to interannual variability in meteorology, and $PM_{2.5}$–attributable chronic mortality varies by ±8%, and by >±5% and ±1% at a national level. Meteorological variability shows larger impacts on interannual variations in acute risks than that in chronic exposure at both provincial (>±20%) and national (±4%) levels. These findings emphasize that tighter controls of $PM_{2.5}$ and precursor emissions are urgently needed, particularly under unfavorable meteorological conditions in China.

Keywords*:* High resolution; air quality model; exposure response functions; health impacts; acute and chronic exposure; urban and rural; meteorological variability

1. Introduction

With the rapid urbanization and industrialization of China over recent decades, air pollution has become a severe environmental and social issue, and regional-scale episodes of heavy air pollution have become more frequent (Chan and Yao, 2008; Zhang et al, 2013; Fu et al, 2017). $PM_{2.5}$ (particulate matter with aerodynamic diameter less than 2.5 μm) is one of the main constituents of air pollution. In most cities in China annual mean $PM_{2.5}$ concentrations are 4–10 times higher than the WHO Air Quality Guidelines of 10 μg/m³. High concentrations of $PM_{2.5}$ not only affect the environment, but also have a negative effect on human health. Both long-term and short-term exposure to particulate matter lead to health effects on human beings (Gordian et al., 1996; Dockery, 2001; Pope III et al., 2019). The 2015 Global Burden of Disease Study (GBD 2015) identified outdoor $PM_{2.5}$ pollution as the fifth greatest risk factor for health, with ambient $PM_{2.5}$ exposure responsible for 4.2 million deaths worldwide, and for nearly 1.1 million deaths annually in China (Cohen et al., 2017).

Several studies have investigated the health impacts induced by $PM_{2.5}$ exposure (Anenberg et al., 2010; Yang et al., 2013; Beelen et al., 2014; Apte et al., 2015; Lelieveld et al., 2015; Ghude et al., 2016). However, many of these studies rely on limited surface monitoring data (Song et al., 2017; Maji et al., 2018), and this may be insufficient to reproduce the long-term spatial and temporal variations of pollutants. Satellite observations provide valuable additional information with better spatial coverage, but there are substantial uncertainties in quantifying the surface concentrations relevant to health (Liu et al., 2017). Air quality models that are based on understanding of physical and chemical processes provide better temporal and spatial coverage than observations and are therefore useful for supplementing monitoring data. High spatial resolution allows sharp spatial gradients in PM concentrations to be captured, particularly in areas with diverse topography, demography and emissions (Chemel et al, 2010; Crippa et al, 2016). High resolution models are thus particularly valuable for evaluating the health impacts of ambient $PM_{2.5}$ in urban and rural areas (Hu et al., 2015).

An integrated exposure-response (IER) function for $PM_{2.5}$ was developed by Burnett et al (2014) for the GBD 2010 study and has been applied widely to estimate chronic health impacts in China. This approach combines information from cohort studies on ambient air pollution, second-hand smoke, household solid fuel and smoking to predict the relative risk over the entire $PM_{2.5}$ exposure range, covering low and high concentrations. Using these IER curves, Lelieveld et al (2015) estimated that $PM_{2.5}$ pollution was responsible for 1.36 million premature deaths in 2010 across China, based on coarse resolution (>100 km) model simulations of $PM_{2.5}$ concentration. GBD 2010 used a higher resolution (10 km) data set (Lim et al., 2012) to estimate that 1.23 million premature deaths occurred in 2010, while premature deaths attributable to ambient $PM_{2.5}$ in 2013 were estimated at 1.37 million based on assimilated $PM_{2.5}$ concentrations (Liu et al, 2016). Other studies have analyzed the long-term spatial and temporal variation of mortality induced by ambient $PM_{2.5}$ using satellite data (Liu et al, 2017). However, these studies used an earlier parameterization of IER, and the updated parameterizations of Cohen et al. (2017) are expected to be more accurate. Many of these previous studies have employed national-level baseline rates of disease incidence over China, neglecting the differences between different provinces which are known to be substantial (Xie et al., 2016).

To mitigate serious air pollution and the adverse health impacts of pollutant exposure, stringent air pollution control policies were implemented in China in 2013 that required a reduction in $PM_{2.5}$ concentrations of 10% by 2017 (Huang et al., 2018; Cheng et al., 2019). Previous studies have evaluated the health benefits of the reductions in ambient $PM_{2.5}$ over this period, which were driven mainly by the emission control measures (Zheng et al., 2017; Huang et al., 2018; Ding et al., 2019). However, meteorological processes play an important role in the transport and accumulation of $PM_{2.5}$. Interannual variations in meteorology may thus lead to substantial variations in surface $PM_{2.5}$ that affect the health burden and this has not been thoroughly explored.

While long-term cumulative exposure to elevated levels of $PM_{2.5}$ on annual timescales is known to be damaging to health, short-term acute exposure to high $PM_{2.5}$ on daily timescales can also be important (Pope III, 2000), and common health endpoints include mortality, hospitalization, outpatient visits and respiratory symptoms. In China, studies have estimated the health impacts of severe haze events, such as the extreme episode in January 2013 (Li et al., 2013; Xie et al., 2014; Gao et al., 2015). However, few studies have focused on short-term exposure over annual timescales across the country.

In this study, we estimate the premature human mortality over the 2013 – 2017 period attributable to ambient $PM_{2.5}$ across China using updated IER functions and population data, provincial level disease incidence data, and a high-resolution air quality model.

We use the terminology “chronic health impacts” to describe premature deaths attributable to long-term exposure to $PM_{2.5}$ and “acute health impacts” to refer to premature deaths attributable to short-term exposure to very high $PM_{2.5}$ concentrations. We explore how $PM_{2.5}$-induced premature mortality varies at different scales, including the contrasts between urban and rural districts. We then quantify the spatial and interannual variation of chronic and acute health impacts due to the variability in meteorological conditions, and identify the uncertainties. We focus on providing a contemporary assessment of health impacts and a full characterization of the spatial and temporal variations imposed by meteorology, and we therefore neglect year to year changes in emissions, population and land cover which also influence the health impacts. Section 2 introduces the methods used, including the environmental data and functions used for evaluating the chronic and acute health impacts. Section 3 describes and explains the health impacts attributable to ambient $PM_{2.5}$, and assesses the impact of interannual meteorological variability. Section 4 discusses uncertainties, and section 5 presents the conclusions.

2. Materials and methods

2.1 NAQPMS atmospheric model

The Nested Air Quality Prediction Modeling System (NAQPMS) is used in this study. This is a 3-D regional Eulerian chemical transport model developed at the Institute of Atmospheric Physics, Chinese Academy of Sciences (Wang et al, 2006; Li et al 2011, 2013). NAQPMS includes a complete description of chemical reactions, advection, diffusion, and dry and wet deposition (Wang et al, 2001). It uses the CBM-Z chemical mechanism that includes 71 species and 176 reactions (Zaveri and Peters, 1999) to calculate gas-phase chemistry. An aerosol thermodynamic model (ISORROPIA1.7) is used to simulate inorganic aerosol chemistry (Nenes et al, 1998, 1999). Heterogeneous chemistry is included for 14 species with 28 reactions (Li et al, 2012). Dry deposition of gases and aerosols is based on the parameterization of Wesely (1989) and Zhang (2001). Wet deposition and aqueous phase chemistry are based on RADM2 (Chang et al, 1987). Dust and sea salt production are calculated following Luo et al (2006) and Athanasopoulou et al (2008). Six secondary organic aerosols are simulated using a two-product module (Odum et al, 1997; Pandis et al, 1991). A more detailed description of NAQPMS can be found in Li et al (2012).

2.2 Model configuration

Figure 1 shows the model domain used in this study, covering China, Mongolia, the Korean Peninsula and Southeast Asia. The model is run at 5 km × 5 km horizontal resolution and has 999 × 1069 grid points in latitude and longitude, respectively. Vertically, the model uses 20 terrain-following layers from the surface to 20 km, with the lowest 12 layers below 3 km.

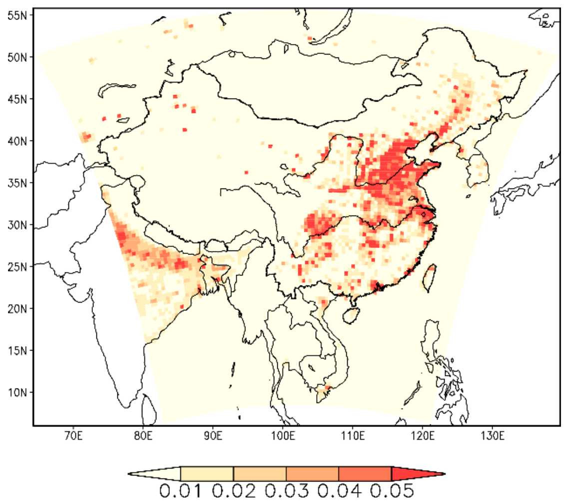

Fig 1 Model domain and annual mean emission rate of primary $PM_{2.5}$ (μg m^{-2} yr^{-1})

Hourly meteorological fields for NAQPMS are taken from the Weather Research and Forecasting (WRF) model v3.3 run at 5 km resolution and driven by the National Centers for Environmental Prediction (NCEP) Final Analysis (FNL) 6-hourly data. Comparison of observed and simulated daily mean temperature (T), relative humidity (RH) and wind speed (WS) at 83 sites across China shows normalized mean biases between -0.07 and -0.02 and high correlation coefficients, particularly for temperature (0.98) and relative humidity (0.86), see Table S1 in supplementary material. The model captures both the magnitude and temporal variation of these three meteorological variables well, as shown in Fig S1. The chemical initial and boundary conditions for NAQPMS are taken from simulations with the global model MOZARTv2.4 (Emmons et al., 2010).

Anthropogenic emissions are based on an updated version of the Multi Resolution Emission Inventory (MEIC, www.meicmodel.org) for 2013. MEIC provides emissions of 10 major air pollutants and greenhouse gases (SO_2, NO_x, CO, NMVOC, NH_3, CO_2, $PM_{2.5}$, PM_{10}, BC and OC) from anthropogenic sources. The original resolution of MEIC 2013 is 0.25° × 0.25° and the data is interpolated to the 5 km grid weighted by gridded area and location. Biogenic emissions are taken from off-line simulations of the Model for Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al, 2012) at an original resolution of 0.1° × 0.1° and re-gridded to 5 km × 5 km by the same approach. The major emissions in MEGAN are isoprene and monoterpenes, which account for nearly 70% of total biogenic volatile organic compound emissions. China land cover data for 2015 at 1 km resolution were used to distinguish urban and rural areas. This was re-gridded to 5 km x 5 km and grid squares with an urban ratio greater than 0.33 were defined as urban areas (Feng et al., 2018).

2.3 Long-term (chronic) premature mortality estimation

An integrated exposure risk (IER) model (Burnett et al., 2014) for $PM_{2.5}$ concentration–response functions was used to evaluate the premature mortality attributable to ambient $PM_{2.5}$ exposure over China from 2013 to 2017. The IER model is based on cohort studies of ambient $PM_{2.5}$ in the US and Europe, including tobacco smoke and the household burning of solid fuel, and provides concentration-response relationships for a wide range of ambient $PM_{2.5}$ concentrations. It has been employed previously in the GBD studies (Lim et al., 2010; GBD 2013; GBD 2015). We consider the premature mortality attributable to $PM_{2.5}$ for the four main health endpoints among adults (over 25 years old), including cerebrovascular disease (Stroke), ischemic heart disease (IHD), chronic obstructive pulmonary disease (COPD) and lung cancer (LC), and for one endpoint among young children, acute lower respiratory infection (LRI).

The relative risk (RR) for each health endpoint was calculated using equation (1)

$$RR = \begin{cases} 1 + \alpha\left(1 - e^{-\beta(C - C_0)^{\gamma}}\right), & C \geq C_0 \\ 1, & C < C_0 \end{cases} \tag{1}$$

where C is the annual average $PM_{2.5}$ concentration, C_0 is the counterfactual concentration below which no adverse health effect is observed, and α, β, γ are fitting parameters that reproduce the observed concentration-response curves for the different health endpoints. C_0, α, β and γ are updated from the values used in GBD 2015 based on Cohen et al., (2017). A distribution of 1000 sets of parameters is used in the

IER model to calculate the mean relative risk and its 95% confidence intervals (CIs).

The relative risk can be converted to a premature mortality attributable to ambient $PM_{2.5}$ exposure, for each endpoint for each age and sex (gender) subgroup in each region (subscripts e, a, s, r, respectively).

$$\Delta M_{e,a,s,r}(C_r) = Pop_{a,s,r} \times B_{e,a,s,r} \times \frac{RR_{e,a,s}(C_r) - 1}{RR_{e,a,s}(C_r)} \qquad (2)$$

where $\Delta M_{e,a,s,r}$ is the change in mortality attributable to long-term $PM_{2.5}$ exposure of a specific endpoint at a specific age, sex and region. $Pop_{a,s,r}$ represents the population exposed in a specific age-sex group at a region-level. Population data at 1 km × 1 km resolution for 2014 was obtained from LandScan (https://landscan.ornl.gov/) and re-gridded to 5 km × 5 km to match the model resolution. $B_{e,a,s,r}$ represents the baseline provincial mortality, i.e. the incidence of a specific endpoint at a specific age and sex in a particular region, and $RR_{e,a,s}$ reflects the relative risk of the endpoint at a specific age and sex. National baseline incidence rates of Stroke, IHD, COPD, LC and LRI were obtained from the online GBD database (https://vizhub.healthdata.org/gbd-compare), and provincial baseline incidence rates were estimated using the relationships between provincial and national rates given by Xie et al. (2016). Per capita mortality is calculated by dividing the number of premature deaths by the population.

2.4 Short-term (acute) premature mortality estimation

In this study, a Poisson regression model is used to estimate the acute risk attributable to high $PM_{2.5}$ concentrations during a pollution episode, and this approach has been applied widely in the epidemiology of air pollution (Wang and Mauzerall., 2006), based on the following:

$$\Delta E_r = Pop_r[1 - e^{-\varepsilon(C - C_0)}]E \qquad (3)$$

where ΔE is the estimated total non-accidental number of mortalities due to daily $PM_{2.5}$ concentrations at a regional level; Pop is the exposed population in that region; ε is the concentration-response coefficient for mortality, which was selected from a meta-analysis of the coefficients associating short-term $PM_{2.5}$ exposure and health responses in China (Lu et al., 2015); C is the simulated daily mean $PM_{2.5}$ concentration; C_0 is the threshold concentration, for which we use the WHO 24-hour mean $PM_{2.5}$ guideline value (25 μg/m³) as reference, and E is the national incidence rate of the total non-accidental mortality endpoints listed in Section 2.3, was taken from the 2013 China Statistical Yearbook of Public Health (NBSC, 2013).

3 Results

3.1 Evaluation of $PM_{2.5}$ concentrations

Observations of hourly surface $PM_{2.5}$ concentrations from 2013 to 2017 were obtained from the China National Environmental Monitoring Centre (www.cnemc.cn) and are used to evaluate the model performance. Monthly frequency distributions of observed and simulated concentrations of $PM_{2.5}$ from 1288 sites in China over five years are presented in Fig 2. Strong seasonal variations are observed, with higher $PM_{2.5}$ concentrations in winter and spring, and lower concentrations in summer and autumn. These patterns are reproduced well with the model; the distributions of the two data sets are close. In summertime, simulations are 7.6 μg/m³ higher than observations. This may be because there is more precipitation in this season and wet deposition in the model is underestimated (Wang et al., 2018). $PM_{2.5}$ concentrations are reproduced well by the model, particularly between 30 and 60 μg/m³, and are a little high between 70 and 90 μg/m³, with 93% of sites lying within a factor of two of the observations, see Fig S2 and Table S2 in supplementary material. The low model mean bias (0.9 μg/m³) and reasonably high correlation coefficient (r=0.74) indicate that at high resolution the NAQPMS model can capture the magnitude and temporal variation of daily $PM_{2.5}$ concentrations relatively well.

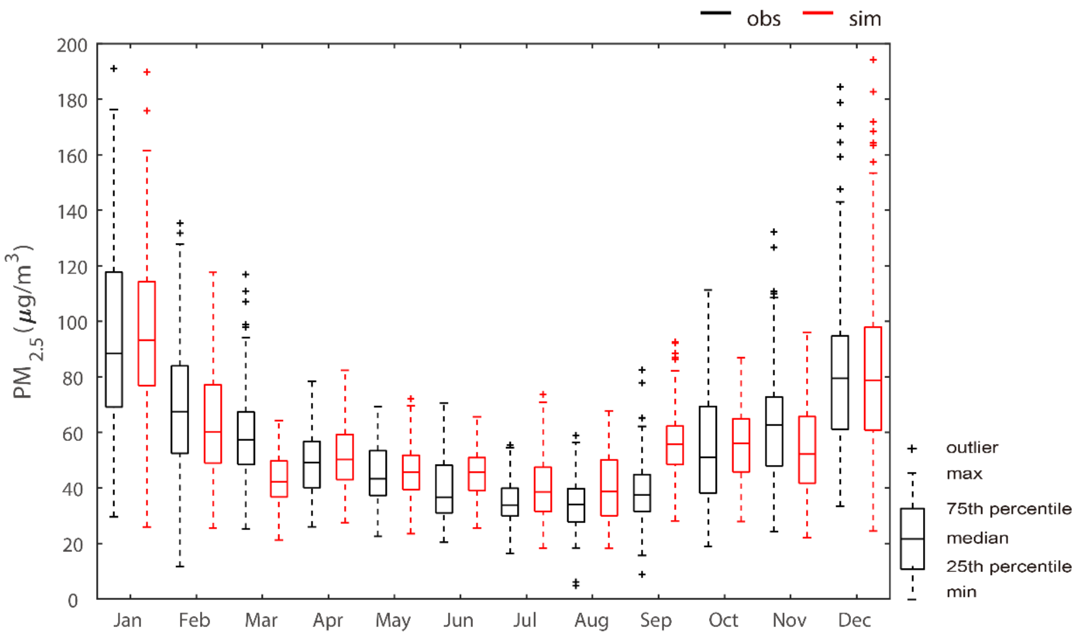

Fig 2 Observed (black) and simulated (red) monthly frequency distributions of daily mean $PM_{2.5}$ concentrations averaged across 1288 sites in China.

NAQPMS also reproduces the spatial distribution of $PM_{2.5}$ concentration well. Fig 3 shows a comparison of the 5-year annual mean simulated concentration of $PM_{2.5}$ against observed average concentrations in 31 provincial capital cities across China. Since the highest urbanization and industrialization occur in the Beijing-Tianjin-Hebei, Yangtze River Delta, Pearl River Delta and Sichuan basin regions, high concentrations of $PM_{2.5}$ are found in these regions. The highest 5-year mean concentration is greater than 100 μg/m³, greatly exceeding the Chinese annual mean $PM_{2.5}$ concentration standard of 35 μg/m³, and a factor of ten higher than the WHO limit. In contrast, less populated regions in western China show much lower $PM_{2.5}$ concentrations (<25μg/m³). Differences from observations at some locations are most likely caused by uncertainties in emissions, changing weather conditions, and weaknesses in process representation in the model. Overall, the temporal variation and spatial distribution of surface $PM_{2.5}$ concentration are captured very well at 5 km resolution, providing confidence in the suitability of the results for analysis of the health impacts attributable to ambient $PM_{2.5}$.

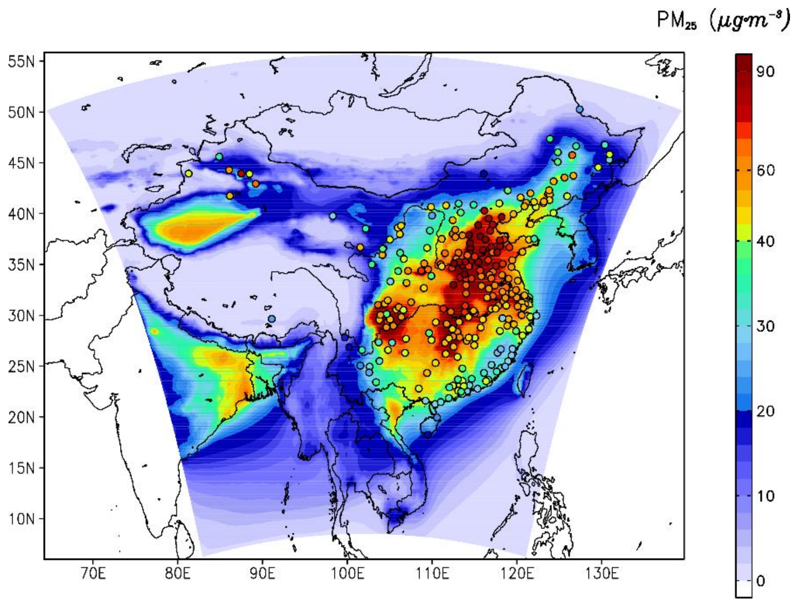

Fig 3 Spatial distribution of 5-year annual mean $PM_{2.5}$ concentration across China; circles show average observed concentration in 259 cities.

3.2 Premature mortality attributable to chronic exposure to $PM_{2.5}$

Population exposure to $PM_{2.5}$ was calculated using simulated $PM_{2.5}$ concentrations in combination with high-resolution population data. Fig 4 shows the number of people exposed to different 5-year mean $PM_{2.5}$ concentrations and the cumulative exposure. About 1.2 billion people (87% of the total population) are exposed to 5-year mean $PM_{2.5}$

concentrations of between 30 and 100 μg/m³. The number of people exposed to 5-year mean concentrations above 100 μg/m³ is about 16.6 million, 1.2% of the population. About 235 million people (17% of the population) experience levels ≤35 μg/m³, meeting Chinese annual mean $PM_{2.5}$ concentration standards, but only 14.4 million (1.1% of the population) do not exceed the WHO annual mean exposure limit of ≤10 μg/m³.

Fig 5 shows how the 5-year mean $PM_{2.5}$ concentration varies with population density across China. $PM_{2.5}$ concentrations increase with population density up to about 300 people/km^2, and then stabilize at around 65 μg/m³. It is clear that $PM_{2.5}$ is lowest in rural and remote districts where the population density is less, and is highest in urban districts with large populations. The similar concentrations for population densities greater than 300 people/km^2 likely reflects the timescales for transport and mixing and thus the regional nature of $PM_{2.5}$ pollution.

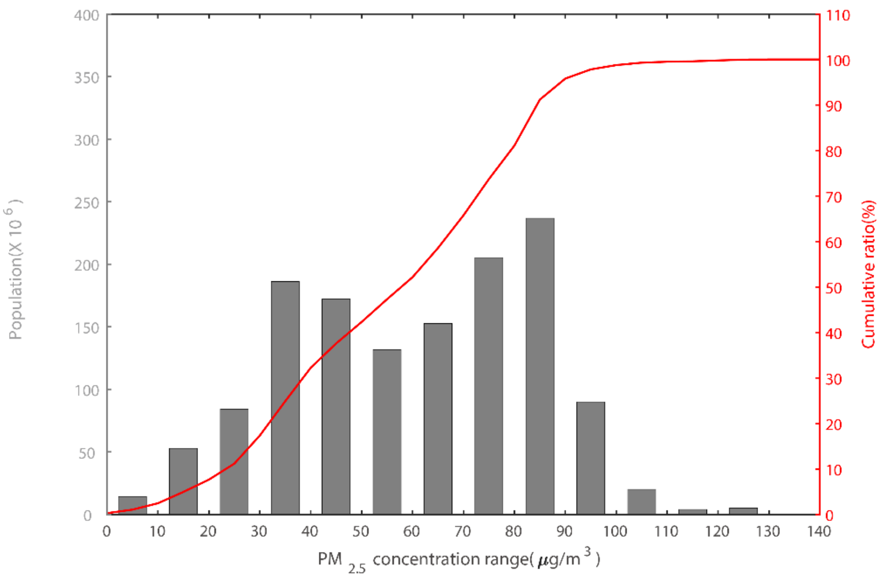

Fig 4 Population exposure to 5-year mean $PM_{2.5}$; bars show the number of people exposed in each 10 μg/m³ concentration range, and the curve shows the cumulative exposure.

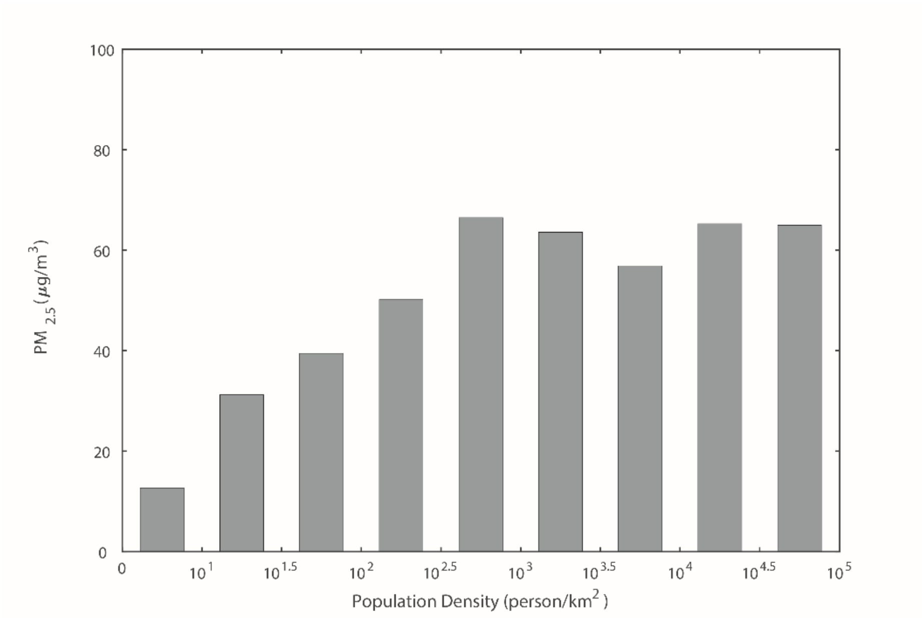

Fig 5 Relationship between population density and 5-year mean $PM_{2.5}$

Fig 6 shows the spatial distribution of total (all-factor) premature mortality attributable to chronic exposure to ambient $PM_{2.5}$ across China. Consistent with the spatial distributions of $PM_{2.5}$ and population, high levels of mortality occur in central-eastern China and the Sichuan Basin, with approximately 1,210,000 (95% Confidence Interval: 720,000–1,750,000) premature deaths attributable to chronic exposure to $PM_{2.5}$ in China on a 5-year average basis. The major causes of deaths are stroke, IHD and COPD, which contribute 330,000 (180,000–500,000), 350,000 (200,000–550,000) and 320,000 (200,000–440,000) deaths, accounting for 27%, 29% and 26%, respectively. Because of lower relative risks in IER model and provincial incidence rates, contributions from LC and LRI are comparatively small, with 130,000 (90,000–170,000) and 70,000 (50,000–90,000) premature deaths, 11% and 6%, respectively (Fig 7).

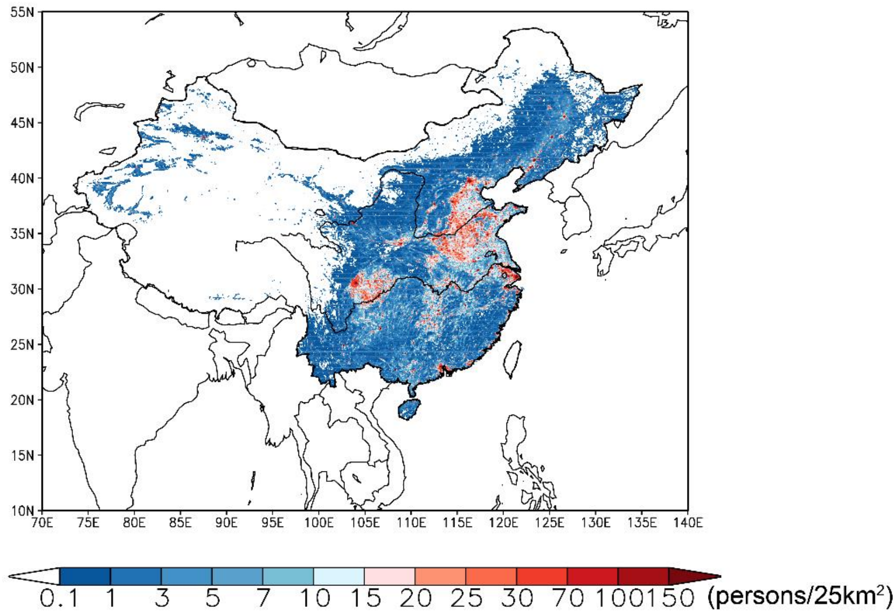

Fig 6 Spatial distribution of premature deaths per year attributable to 5-year mean $PM_{2.5}$ exposure in mainland China.

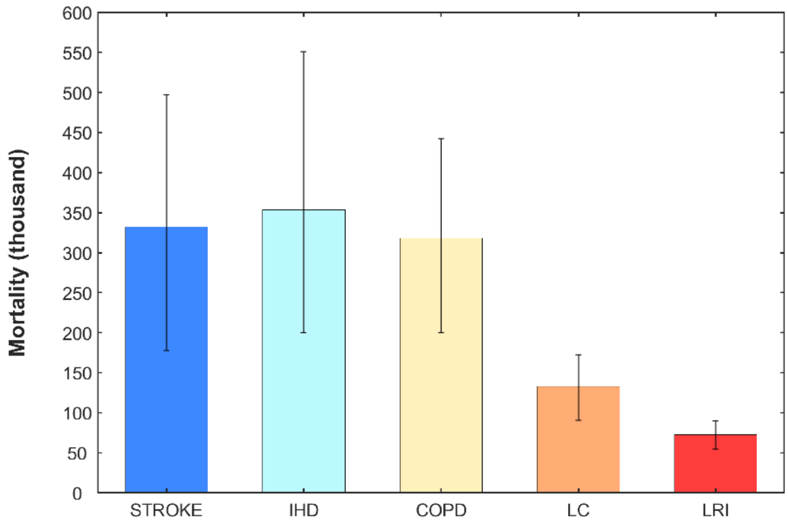

Fig 7 Total deaths in China attributable to chronic $PM_{2.5}$ exposure for different health endpoints: Stroke, ischemic heart disease (IHD), chronic obstructive pulmonary disease (COPD), lung cancer (LC), and lower respiratory infection (LRI). Error bars represent 95% CI.

Chronic per capita deaths and corresponding total premature deaths in 31 provinces are presented in Fig 8. There are more than 0.5‰ per capita deaths per year across the country except in the remote provinces of Tibet and Qinghai. Due to high $PM_{2.5}$ concentrations in a heavily urbanized region, Chongqing has the highest per capita mortality (1.4‰) among all provinces (Fig 8a), followed by Sichuan (1.3‰) and Shandong (1.2‰). The largest totals of premature deaths due to ambient $PM_{2.5}$ occur in Henan (110,000, 95% CI: 69,000–159,000), Shandong (109,000; 68,000–157,000), Sichuan (107,000; 68,000–151,240), Hunan (74,000; 46,000–106,000) and Jiangsu (71,000; 44,000–102,000), which together account for 39% of China's total premature deaths. The high number of deaths in these provinces reflect high emissions of air pollutants and high population, combined with unfavorable geographical conditions such as their location in basins or downwind of major populated areas, which permit accumulation or transport of air pollutants. It is worth noting that deaths from COPD are relatively greater in southwestern provinces including Sichuan, Chongqing, Yunnan and Guizhou as there is a greater rural population in these regions, and the incidence rate of COPD is higher in rural areas (Zhong et al., 2011).

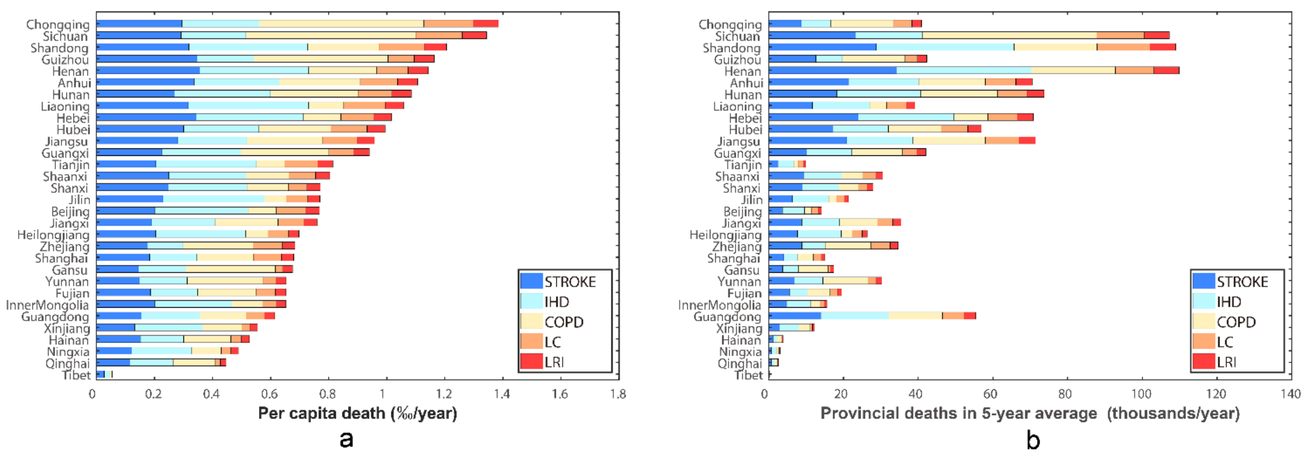

Fig 8 (a) Per capita and (b) total deaths per year due to chronic exposure to $PM_{2.5}$ by province.

3.3 The difference in premature deaths between urban and rural areas

Based on 1 km resolution China land cover data, approximately 44% of the Chinese population live in urban districts. The estimated number of premature deaths in these areas is 520,000 (95% CI: 320,000–760,000). Fig 9 shows the per capita mortality and total premature deaths in urban and rural areas for each province. Per capita deaths are slightly higher in urban (0.1–1.5‰) than in rural (0.05–1.4‰) areas for most provinces, but the differences are not large and the provincial rankings are quite similar. This highlights the regional nature of $PM_{2.5}$ pollution, but also suggests that much finer scale models resolving local sources may be needed to distinguish the characteristics of urban and rural air pollution.

Henan province has the greatest number of urban premature deaths (70,000, 95% CI: 40,000–100,000). This province in central China has a large population, high levels of urbanization, and is strongly affected by regional transport of pollution. Sichuan province in southwestern China is dominated by mountains and has a large rural population, and this region sees the greatest number of rural deaths (86,000, 95% CI: 50,000–120,000). In remote provinces, such as Tibet, Qinghai, Ningxia and Hainan, premature deaths attributable to ambient $PM_{2.5}$ are relatively low in both urban and rural populations.

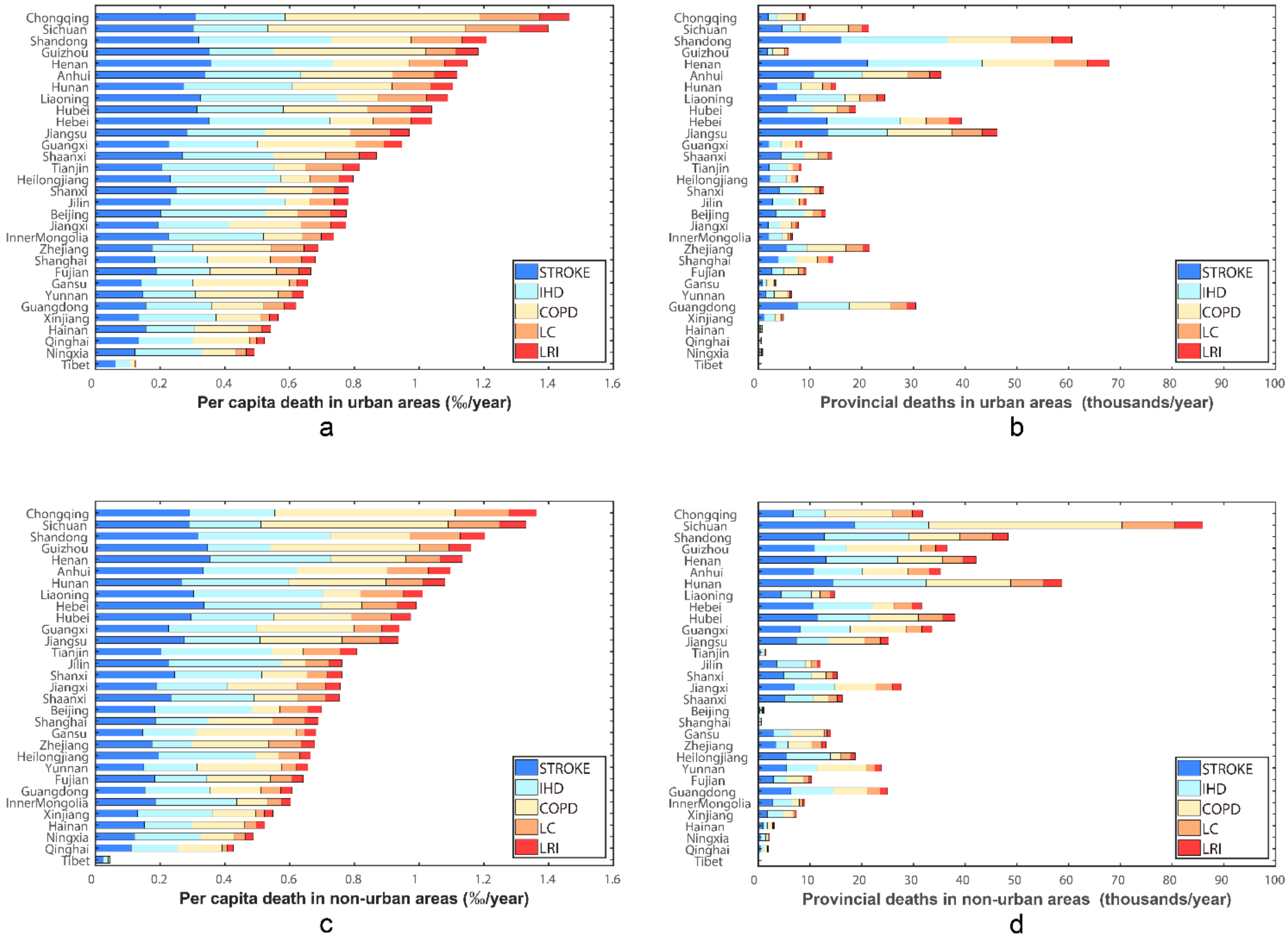

Fig 9 Per capita and total premature deaths per year due to 5-year mean $PM_{2.5}$ in urban (a, b) and rural (c, d) areas.

3.4 Effects of interannual meteorological variations on premature mortality

Several studies have evaluated the air quality and health benefits associated with emission reduction policies over the 2013-2017 period (Zheng et al., 2017; Huang et al., 2018). However, the impact of meteorological conditions on air quality and health is also important and few studies have explored this. Fig 10 shows the relative variability in annual mean $PM_{2.5}$ concentration and total premature deaths in 31 provinces of China from 2013 to 2017. During this period, the interannual variations in $PM_{2.5}$ concentration and $PM_{2.5}$-attibutable mortality due to meteorology are quite small,

±5% and ±1%, respectively. However, at a provincial level annual mean $PM_{2.5}$ concentrations vary by as much as 20%, and $PM_{2.5}$-attributable deaths by up to ±8%, except in Tibet (where concentrations vary by ±20% but health impacts by ±75%), reflecting the nonlinear relationship between $PM_{2.5}$ concentration and its health impacts. The total number of premature deaths in most Northern provinces was lower in 2016 and 2017 than in 2014 and 2015. Although these variations arise from meteorology alone, they supplement the improvement in air quality caused by reducing emissions over the period. The pattern in coastal provinces such as Shanghai and Guangdong is the reverse of that in inland provinces like Anhui and Jiangxi, (i.e. the number of premature deaths increases from 2013-2017) reflecting the influence of marine flow. A strong El Niño affected atmospheric circulation in 2015, leading to weaker winds and greater air pollution over most of China (Chang et al., 2016). The intensity of the Siberian high (I_{SH}) and the East Asia winter monsoon (I_{EAWM}) can also affect air quality in China (Chin et al., 2017; Zhang et al., 2010; Cheng et al., 2016; Li et al., 2016). For example, in 2014 there was a very clear gradient in pollution from northwest to southeast because of the decreased I_{SH} and I_{EAWM} (Ding et al., 2017). Strong winds in 2016 and 2017 associated with increasing I_{SH} and I_{EAWM} led to better air quality in these years (Zhang et al., 2018). In Tibet, where emissions are low and air pollution is driven mainly by regional transport, weather conditions have a big impact on air quality. For example, strong winds caused by increased I_{SH} and I_{EAWM} transported more air pollutants to Tibet in 2016. Thus, although interannual variations in meteorology have little influence on $PM_{2.5}$ concentration and premature deaths at a national level, the impacts at a provincial level are substantially larger.

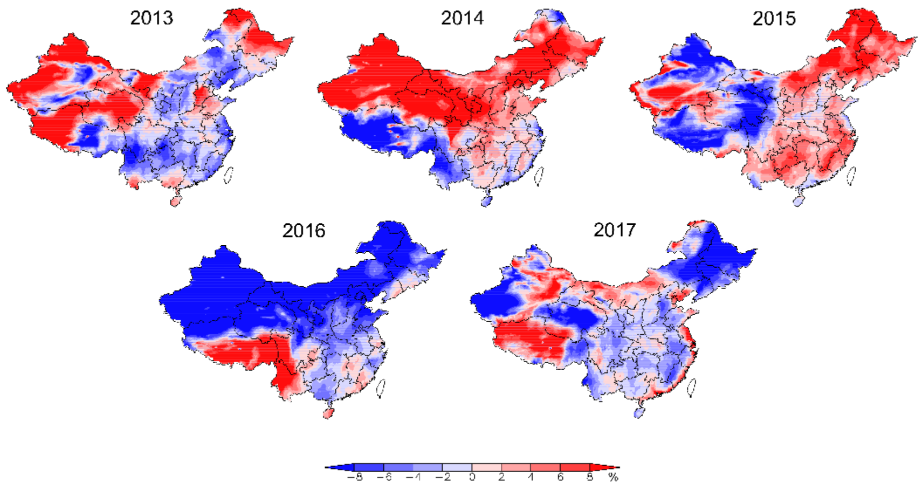

Fig 10 (a) Interannual anomaly in gridded $PM_{2.5}$ concentration relative to the 5-year mean for mainland China.

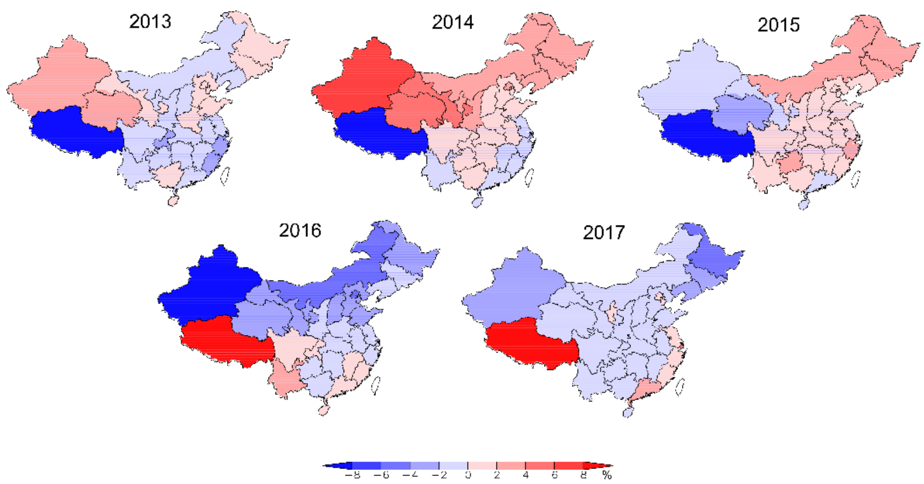

Fig 10 (b) Interannual anomaly in premature deaths attributable to chronic exposure to $PM_{2.5}$ relative to the 5-year mean for mainland China.

3.5 Acute deaths attributable to heavy pollution episodes

The number of acute deaths across China was calculated using daily mean $PM_{2.5}$ concentrations with a Poisson regression function, concentration-response coefficients and incidence rates of mortality as described in Section 2. We find an average annual total of 116,000 (95% CI: 64,000–170,000) premature deaths over the 5-year period. The highest acute per capita mortality (0.15‰) is in Hubei province, while the greatest total number occur in Henan province (14,100, 95% CI: 7,800–20,600) (Fig 11). Both experienced heavy pollution episodes but Henan has the larger population. Li et al (2019) estimated that there were 33,000 deaths attributable to acute exposure to $PM_{2.5}$ in 2015 using daily observations from 104 counties across China. Our results are much higher because we have considered heavy air pollution episodes across all 2862 counties in China.

We estimate a total of 55,000 (3,000-81,000) acute deaths in urban areas, and the acute per capita deaths are highest in Henan and Hubei provinces, most likely due to higher concentrations, terrain and the regional nature of $PM_{2.5.}$ Interannual variability in meteorology has greater impacts on acute risks than chronic, particularly at a provincial level. Acute deaths vary by ±20% in most provinces, while in some remote places variations can be much larger. This highlights that meteorological conditions have a greater influence on daily than annual mean $PM_{2.5}$ concentrations, and this leads to larger variations in risks. There are substantially fewer acute than chronic deaths, but as acute risks are associated with air pollution episodes, there is a much greater range in per capita acute deaths from $PM_{2.5}$ across provinces (0.001–0.15‰) than chronic deaths (0.4–1.4‰), except in Tibet where per capita deaths are much lower for both acute (insignificant) and chronic (0.6‰) exposure.

Our results demonstrate that the long-term health impacts attributable to chronic exposure to $PM_{2.5}$ are more important for the overall public health burden than acute risks (Pope III, 2000), even though heavy pollution episodes and their related health impacts attract more public attention. It should be noted however, that understanding of the mortality burden attributable to short-term exposure needs to be improved and more specific exposure-response models for different populations and end-points should be developed in future.

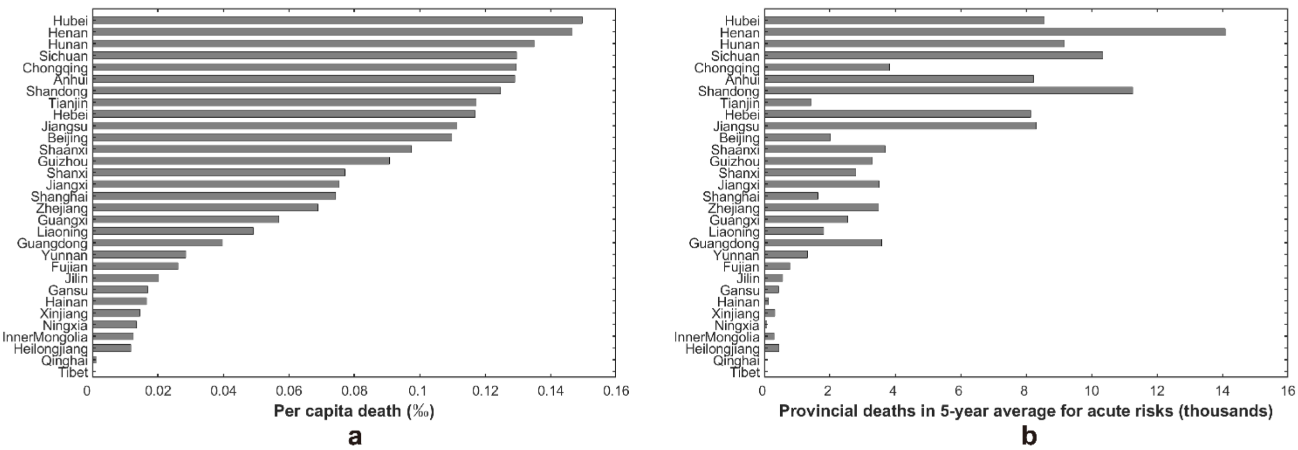

Fig 11 (a) Per capita and (b) total deaths due to the acute risks of $PM_{2.5}$ for each province.

4 Discussion

A comparison of studies of premature deaths attributable to chronic exposure to $PM_{2.5}$ across China is shown in Table 1. Our estimate of 1.19–1.22 million from 2013 to 2017, is consistent with other studies (Lim et al., 2012; Liu et al., 2016; Cohen et al., 2017; Zheng et al., 2016; Ding et al., 2019). Here we used emissions for 2013 for all five years to allow exploration of the effect of meteorological variability on concentrations and health impacts. The effect of emissions reductions was neglected and our estimate for 2015 is therefore a little higher than the GBD evaluation (Cohen et al., 2017), even though the same exposure-response coefficients were adopted. The variations we find due to meteorology are much smaller than the differences between previous studies shown in Table 1, highlighting that these differences arise from the analysis approach, exposure-response coefficients, emissions and model resolution rather than differences in meteorology. Coarse spatial resolution is likely to lead to greater discrepancies, as seen in the study of Lelieveld et al., 2015. Our model simulations performed at 5 km resolution over the whole of China are at higher resolution than any previous published studies. Higher resolution distributions of surface $PM_{2.5}$ are now becoming available from satellite products (e.g. Wei et al., 2019) although these are derived from aerosol optical depth retrievals using downscaled meteorological variables and interpolation/scaling approaches, and thus the reliable information content is less than the resolution suggests. In contrast, our studies are based on sound understanding of the underlying physical processes, and have information content that is as high as these alternative products.

Table 1 Comparison of studies for premature deaths attributable to $PM_{2.5}$ in China

Year	**Exposure-Response coefficients**	**Observation or Model**	**Premature deaths (million)**	**Reference**
2010	GBD2010	TM5+satellite+obs 0.1°×0.1°	1.23	Lim et al. (2012)
2010	GBD2010	EMAC, 1.1°×1.1°	1.36	Lelieveld et al. (2015)
2004-2012	GBD2010	Satellite, 10 km	0.80-1.20	Liu et al (2016)
2013	GBD2015	WRF-Chem, 60 km	1.33	Gao et al. (2018)

2013	GBD2010	Assimilation+obs	1.37	Liu et al. (2016)
2015	GBD2015	TM5+satellite+obs 0.1°×0.1°	1.10	Cohen et al. (2017)
2013-2015	GBD2010	CMAQ+satellite+obs, 36 km	1.10-1.22	Zheng et al. (2016)
2013-2015	GBD2015	CMAQ+obs, 27 km	1.10-1.39	Ding et al. (2019)
2013-2017	GBD2015	NAQPMS, 5 km	1.19-1.22	This study

To determine the benefits of using fine model resolution, we estimated the chronic premature deaths in 2013 at 5 km resolution and compared them with results aggregated to coarser resolutions from 10 km to 200 km. The number of premature deaths remains similar between 5 and 45 km resolution, and then decreases gradually, so that it is about 6% lower at 200 km resolution. One reason for the relative insensitivity of mortality estimates to model resolution is that the lifetime of $PM_{2.5}$ is relatively long, allowing transport over regional scales. However, we may underestimate this sensitivity as our estimates are based on aggregation from 5 km resolution rather than native model simulations at coarse resolution where the use of coarse resolution meteorology and emissions will lead to larger differences.

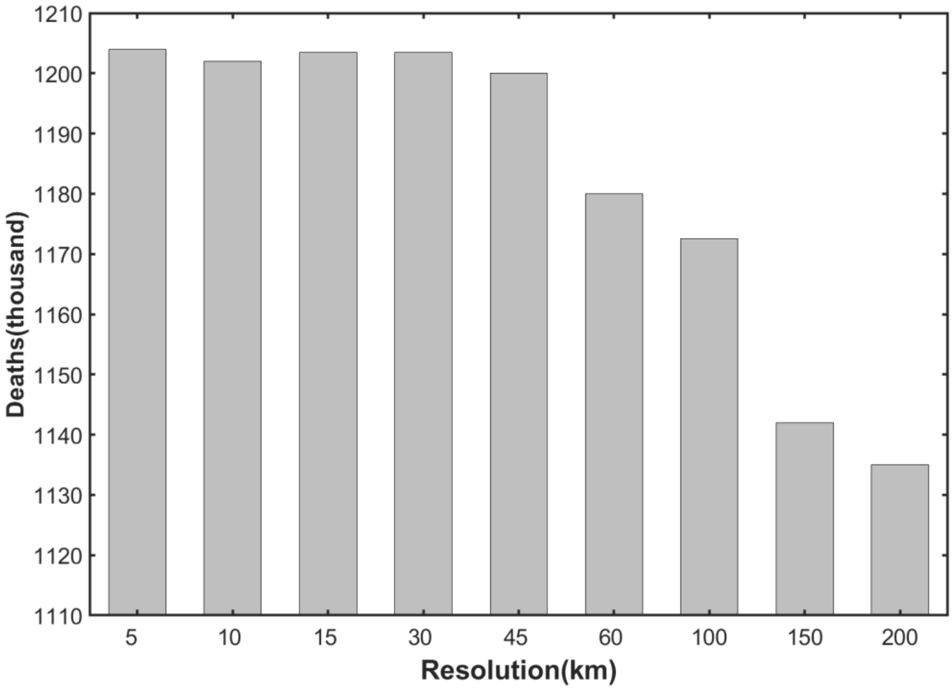

Fig 12 The premature deaths due to $PM_{2.5}$ at different resolutions.

There are a number of uncertainties in our analysis of the health impacts of $PM_{2.5}$ at a provincial level. The IER model integrates data from cohort studies in western countries, based on active and secondhand smoking and household air pollution, and these factors may be significantly different in China. Additionally, the health impact of $PM_{2.5}$ are likely influenced by chemical composition, size distribution and source (Ostro et al., 2015) which can also be expected to differ between world regions and even different provinces. These suggest that in order to improve evaluation of the long-term health impacts in China, researchers should develop and update location- and population-specific exposure-response functions or coefficients. In the current study we have considered the health impact attributable to outdoor $PM_{2.5}$ concentration only, and ignored the impacts of exposure to indoor pollution. Household fuel burning, decoration materials and household and personal care products can release gaseous and particulate pollutants, and people spend about 90% of their time in a public or private indoor environment (Cincinelli et al., 2017). Therefore, it is important to develop indoor exposure-response functions and include estimates of indoor health impacts in China in future studies (Pan X, 2005). There are model biases in heavily polluted and densely populated areas because of uncertainties in emissions, meteorology and model process representation, but NAQPMS reproduce the temporal and spatial distribution of annual average $PM_{2.5}$ concentration reasonably well. In future work, however, higher resolution models with finer scale emissions would be useful to quantity health impacts at smaller scales.

5 Conclusions

We have estimated chronic and acute premature deaths attributable to ambient $PM_{2.5}$ over China for 2013-2017 using the NAQPMS model at 5 km resolution combined with integrated exposure-response methods. An estimated average of 1,210,000 (95% CI: 720,000–1,750,000) premature deaths per year are caused by chronic exposure to ambient $PM_{2.5}$ in China. Stroke, ischemic heart disease and chronic obstructive pulmonary disease each account for about 25% of this total. The polluted and highly populated provinces of Henan, Shandong, Sichuan, Hunan and Jiangsu account for 39% of the premature deaths due to chronic exposure to $PM_{2.5}$ in China. However, the inland municipality of Chongqing has the biggest chronic per capita mortality (1.4‰) among all provinces. These results are based on use of a model at 5 km resolution, and we demonstrate that this provides a modest improvement over the use of a coarser resolution assessment, although the benefits would be greater for short-lived pollutants.

Combining concentration data from NAQPMS simulations with high-resolution land cover data, we differentiate the chronic health impacts of $PM_{2.5}$ in urban and rural districts. Approximately 44% of the Chinese population live in urban districts, and these areas account for 520,000 (320,000–760,000) deaths, i.e. around 43% of the total. Per capita deaths are marginally higher in urban areas (0.1–1.5‰) than in rural areas (0.05–1.4‰) for most provinces. These results highlight the regional nature of $PM_{2.5}$ pollution, and hence the need for high resolution concentration data, but we note that much finer scale models may be needed to capture the different features of air pollution and health impacts in urban and rural areas.

At a national level the impacts of interannual variability in meteorological conditions on $PM_{2.5}$ concentrations and consequently chronic premature deaths are quite small, ±5% and ±1%, respectively. However, the interannual variation in annual mean $PM_{2.5}$ concentration and chronic mortality are much more variable and differ from year to year in different provinces. Annual mean $PM_{2.5}$ concentration varies by ±20% at a provincial level, and the provincial deaths attributable to $PM_{2.5}$ varies by ±8% except in Tibet, where the variability is as high as ±75%. Large-scale weather systems such as El Niño, the Siberian high and the East Asian winter monsoon are the main causes of these variations. These systems typically affect circulation, including wind speed and direction, and lead to differences in the transport and accumulation of air pollutants at a provincial level.

We also quantify the acute health impacts of $PM_{2.5}$ exposure during high pollution episodes. There are 116,000 (64,000–170,000) acute deaths in China attributable to episodes of high $PM_{2.5}$ concentration each year, and 47% of these occur in urban areas. Interannual variability of meteorological conditions has larger impacts on acute risks than chronic exposure, acute deaths vary by ±20% in most provinces. This highlights that meteorological conditions have more substantial influence on daily mean $PM_{2.5}$ concentrations, leading to larger variations in acute risks. There is a much greater range in acute per capita deaths (0.00–0.15‰) than chronic (0.4–1.4‰). The shortcomings in current analyses of acute deaths suggests that the current understanding of the mortality burden attributable to short-term $PM_{2.5}$ exposure should be improved and better exposure-response models need to be developed in future. Although heavy pollution episodes and their related health impacts attract substantial public attention, we find that the long-term health impacts caused by ambient $PM_{2.5}$ concentration are more important for the overall public health burden. These results suggest that policy makers need not only to control and prevent heavy pollution episodes, but also design effective policies for the long-term improvement of regional air quality to reduce adverse health impacts.

Acknowledgements

We would like to acknowledge Dr Kirsti Ashworth at Lancaster Environment Centre for insightful discussion and helpful comments. We are grateful Dr Aaron J. Cohen for providing the latest version IER parameters. This work was supported by the National Natural Science Foundation of China (Grant No 41620104008). Yuanlin Wang is supported by a scholarship from the China Scholarship Council.

Data Availability Statement

All the main research data can be found at Mendeley Data.

References

Anenberg, S.C., Horowitz, L.W., Tong, D.Q. and West, J.J., 2010. An estimate of the global burden of anthropogenic ozone and fine particulate matter on premature human mortality using atmospheric modeling. *Environmental health perspectives*, *118*(9), pp.1189-1195. http://doi.org/10.1289/ehp.0901220

Apte, J.S., Marshall, J.D., Cohen, A.J. and Brauer, M., 2015. Addressing global mortality from ambient PM2. 5. *Environmental science & technology*, *49*(13), pp.8057-8066. http://doi.org/10.1021/acs.est.5b01236

Athanasopoulou, E., Tombrou, M., Pandis, S.N. and Russell, A.G., 2008. The role of sea-salt emissions and heterogeneous chemistry in the air quality of polluted coastal areas. http://doi.org/10.5194/acpd-8-3807-2008

Beelen, R., Raaschou-Nielsen, O., Stafoggia, M., Andersen, Z.J., Weinmayr, G., Hoffmann, B., Wolf, K., Samoli, E., Fischer, P., Nieuwenhuijsen, M. and Vineis, P., 2014. Effects of long-term exposure to air pollution on natural-cause mortality: an analysis of 22 European cohorts within the multicentre ESCAPE project. *The Lancet*, *383*(9919), pp.785-795. http://doi.org/10.1016/S0140-6736(13)62158-3

Burnett, R.T., Pope III, C.A., Ezzati, M., Olives, C., Lim, S.S., Mehta, S., Shin, H.H., Singh, G., Hubbell, B., Brauer, M. and Anderson, H.R., 2014. An integrated risk function for estimating the global burden of disease attributable to ambient fine particulate matter exposure. *Environmental health perspectives*, *122*(4), pp.397-403. http://doi.org/10.1289/ehp.1307049

Byun, D.W. and Dennis, R., 1995. Design artifacts in Eulerian air quality models: Evaluation of the effects of layer thickness and vertical profile correction on surface ozone concentrations. *Atmospheric Environment*, *29*(1), pp.105-126. http://doi.org/10.1016/1352-2310(94)00225-a

Chan, C.K. and Yao, X., 2008. Air pollution in mega cities in China. *Atmospheric environment*, *42*(1), pp.1-42. http://doi.org/10.1016/j,atmosenv.2007.09.003

Chang, J.S., Brost, R.A., Isaksen, I.S.A., Madronich, S., Middleton, P., Stockwell, W.R. and Walcek, C.J., 1987. A three-dimensional Eulerian acid deposition model: Physical concepts and formulation. Journal of Geophysical Research: Atmospheres, 92(D12), pp.14681-14700. http://doi.org/10.1029/jd092id12p14681

Chang, L., Xu, J., Tie, X. and Wu, J., 2016. Impact of the 2015 El Nino event on winter air quality in China. *Scientific reports*, *6*, p.34275. http://doi.org/10.1038/srep34275

Chemel, C., Sokhi, R.S., Yu, Y., Hayman, G.D., Vincent, K.J., Dore, A.J., Tang, Y.S., Prain, H.D. and Fisher, B.E.A., 2010. Evaluation of a CMAQ simulation at high resolution over the UK for the calendar year 2003. *Atmospheric Environment*, *44*(24), pp.2927-2939. http://doi.org/10.1016/j,atmosenv.2010.03.029

Cheng, J., Su, J., Cui, T., Li, X., Dong, X., Sun, F., Yang, Y., Tong, D., Zheng, Y., Li, Y. and Li, J., 2019. Dominant role of emission reduction in PM 2.5 air quality improvement in Beijing during 2013–2017: a model-based decomposition analysis. *Atmospheric Chemistry and Physics*, *19*(9), pp.6125-6146. http://doi.orrg/10.5194/acp-2018-1145

Cheng, X., Zhao, T., Gong, S., Xu, X., Han, Y., Yin, Y., Tang, L., He, H. and He, J., 2016. Implications of East Asian summer and winter monsoons for interannual aerosol variations over central-eastern China. *Atmospheric environment*, *129*, pp.218-228. http://doi.org/10.1016/j,atmosenv.2016.01.037

Chin, M., Bian, H., Kucsera, T., Diehl, T., Tao, Z., Kim, D. and Pan, X., 2017. Connection Between East Asian Air Pollution and Monsoon System. In *Air Pollution in Eastern Asia: An Integrated Perspective* (pp. 87-103). Springer, Cham. http://doi.org/10.1007/978-3-319-59489-7_5

China National Environmental Monitoring Centre. www.cnemc.cn

Cincinelli, A. and Martellini, T., 2017. Indoor air quality and health. http://doi.org/10.3390/ijerph14111286

Cohen, A.J., Brauer, M., Burnett, R., Anderson, H.R., Frostad, J., Estep, K., Balakrishnan, K., Brunekreef, B., Dandona, L., Dandona, R. and Feigin, V., 2017. 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. *The Lancet*, *389*(10082), pp.1907-1918. http://doi.org/10.1016/s0140-6736(17)30505-6

Crippa, P., Sullivan, R.C., Thota, A. and Pryor, S.C., 2016. Evaluating the skill of high-resolution WRF-Chem simulations in describing drivers of aerosol direct climate forcing on the regional scale. *Atmospheric Chemistry and Physics*, *16*(1), pp.397-416. http://doi.org/10.5194/acp-16-397-2016

Ding, D., Xing, J., Wang, S., Liu, K. and Hao, J., 2019. Estimated Contributions of Emissions Controls, Meteorological Factors, Population Growth, and Changes in Baseline Mortality to Reductions in Ambient PM 2.5 and PM 2.5-Related Mortality in China, 2013–2017. *Environmental health perspectives*, *127*(6), p.067009. http://doi.org/10.1289/ehp4157

Ding, T., Wang, Y., KE, Z., Wang, Z. 2017. Northern Hemisphere Atmospheric Circulation in Winter

2016/2017 and Its Impact on Temperature in China. Meteorology, 43 (7), 887-893 (Chinese). doi:10.7519/j.issn.1000-0526.2017.07.013

Dockery, D.W., 2001. Epidemiologic evidence of cardiovascular effects of particulate air pollution. *Environmental health perspectives*, *109*(suppl 4), pp.483-486. http://doi.org/10.1289/ehp.01109s4483

Emmons, L.K., Walters, S., Hess, P.G., Lamarque, J.F., Pfister, G.G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T. and Orlando, J., 2010. Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4). http://doi.org/10.5194/gmd-3-43-2010

Feng, Y. and Qi, Y., 2018. Modeling Patterns of Land Use in Chinese Cities Using an Integrated Cellular Automata Model. *ISPRS International Journal of Geo-Information*, *7*(10), p.403. http://doi.org/10.3390/ijgi7100403

Fu, H. and Chen, J., 2017. Formation, features and controlling strategies of severe haze-fog pollutions in China. *Science of the Total Environment*, *578*, pp.121-138. http://doi.org/10.1016/j.scitotenv.2016.10.201

Gao, M., Guttikunda, S.K., Carmichael, G.R., Wang, Y., Liu, Z., Stanier, C.O., Saide, P.E. and Yu, M., 2015. Health impacts and economic losses assessment of the 2013 severe haze event in Beijing area. *Science of the Total Environment*, *511*, pp.553-561. http://doi.org/10.1016/j.scitotenv.2015.01.005

Gao, M., Beig, G., Song, S., Zhang, H., Hu, J., Ying, Q., Liang, F., Liu, Y., Wang, H., Lu, X. and Zhu, T., 2018. The impact of power generation emissions on ambient PM2. 5 pollution and human health in China and India. *Environment international*, *121*, pp.250-259.

http://doi.org/10.1016/j.envint.2018.09.015

GBD 2013 Risk Factors Collaborators. 2015. Global, regional, and national comparative risk assessment of 79 behavioural, environmental and occupational, and metabolic risks or clusters of risks in 188 countries, 1990-2013: a systematic analysis for the Global Burden of Disease Study 2013. Lancet 386(10010): 2287-2323, http://doi.org/10.1016/s0140-6736(15)00128-2.

GBD 2015 Risk Factors Collaborators. 2016. Global, regional, and national comparative risk assessment of 79 behavioural, environmental and occupational, and metabolic risks or clusters of risks, 1990-2015: a systematic analysis for the Global Burden of Disease Study 2015. Lancet 388(10053):1659-1724, http://doi.org/10.1016/S0140-6736(16)31679-8 .

Ghude, S.D., Chate, D.M., Jena, C., Beig, G., Kumar, R., Barth, M.C., Pfister, G.G., Fadnavis, S. and Pithani, P., 2016. Premature mortality in India due to PM2. 5 and ozone exposure. *Geophysical Research Letters*, *43*(9), pp.4650-4658. http://doi.org/10.1002/2016gl068949

Global Burden Disease Database. https://vizhub.healthdata.org/gbd-compare

Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P.I. and Geron, C., 2006. Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature). *Atmospheric Chemistry and Physics*, *6*(11), pp.3181-3210.http://doi.org/10.5194/acp-7-4327-2007

Gordian, M.E., Ozkaynak, H., Xue, J., Morris, S.S. and Spengler, J.D., 1996. Particulate air pollution and respiratory disease in Anchorage, Alaska. *Environmental Health Perspectives*, *104*(3), pp.290-297. http://doi.org/10.2307/3432887

Hauglustaine, D.A., Brasseur, G.P., Walters, S., Rasch, P.J., Müller, J.F., Emmons, L.K. and Carroll, M.A., 1998. MOZART, a global chemical transport model for ozone and related chemical tracers: 2. Model results and evaluation. *Journal of Geophysical Research: Atmospheres*, *103*(D21), pp.28291-28335.http://doi.org/10.1029/98jd02398

Hu, J., Zhang, H., Ying, Q., Chen, S.H., Vandenberghe, F. and Kleeman, M.J., 2015. Long-term particulate matter modeling for health effect studies in California–Part 1: Model performance on temporal and spatial variations. *Atmospheric Chemistry and Physics*, *15*(6), pp.3445-3461. http://doi.org/10.5194/acp-15-3445-2015

Huang, J., Pan, X., Guo, X. and Li, G., 2018. Health impact of China's Air Pollution Prevention and Control Action Plan: an analysis of national air quality monitoring and mortality data. *The Lancet Planetary Health*, *2*(7), pp.e313-e323. http://doi.org/10.1016/s2542-5196(18)30141-4

LandScan. https://landscan.ornl.gov/

Lelieveld, J., Evans, J.S., Fnais, M., Giannadaki, D. and Pozzer, A., 2015. The contribution of outdoor air pollution sources to premature mortality on a global scale. *Nature*, *525*(7569), p.367. http://doi.org/10.1038/nature15371

Li, J., Wang, Z., Wang, X., Yamaji, K., Takigawa, M., Kanaya, Y., Pochanart, P., Liu, Y., Irie, H., Hu, B. and Tanimoto, H., 2011. Impacts of aerosols on summertime tropospheric photolysis frequencies and photochemistry over Central Eastern China. *Atmospheric environment*, *45*(10), pp.1817-1829. http://doi.org/10.1016/j.atmosenv.2011.01.016

Li, J., Wang, Z., Zhuang, G., Luo, G., Sun, Y. and Wang, Q., 2012. Mixing of Asian mineral dust with anthropogenic pollutants over East Asia: a model case study of a super-duststorm in March 2010. *Atmospheric Chemistry and Physics*, *12*(16), pp.7591-7607. http://doi.org/10.5194/acp-12-7591-2012

Li, J., Wang, Z., Huang, H., Hu, M., Meng, F., Sun, Y., Wang, X., Wang, Y. and Wang, Q., 2013. Assessing the effects of trans-boundary aerosol transport between various city clusters on regional haze episodes in spring over East China. *Tellus B: Chemical and Physical Meteorology*, *65*(1), p.20052. http://doi.org/10.3402/tellusb.v65i0.20052

Li, T. T., Du, Y. J., Mo, Y., Xue, W. B., Xu, D. Q., Wang, J. N. (2013). Assessment of haze-related human health risks for four Chinese cities during extreme haze in January 2013, 93(34), 2699-2702. (Chinese) doi:10.3760/cma.j.issn.0376-2491.2013.34.004

Li, T., Guo, Y., Liu, Y., Wang, J., Wang, Q., Sun, Z., He, M.Z. and Shi, X., 2019. Estimating mortality burden attributable to short-term PM2. 5 exposure: A national observational study in China. *Environment international*, *125*, pp.245-251. http://doi.org/10.1016/j.envint.2019.01.073

Li, Q., Zhang, R. and Wang, Y., 2016. Interannual variation of the wintertime fog–haze days across central and eastern China and its relation with East Asian winter monsoon. *International Journal of Climatology*, *36*(1), pp.346-354. http://doi.org/10.1002/joc.4350

Lim, S.S., Vos, T., Flaxman, A.D., Danaei, G., Shibuya, K., Adair-Rohani, H., AlMazroa, M.A., Amann, M., Anderson, H.R., Andrews, K.G. and Aryee, M., 2012. A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990–2010: a systematic analysis for the Global Burden of Disease Study 2010. *The lancet*, *380*(9859), pp.2224-2260. http://doi.org/10.1016/S0140-6736(12)61766-8

Liu, J., Han, Y., Tang, X., Zhu, J. and Zhu, T., 2016. Estimating adult mortality attributable to PM2. 5 exposure in China with assimilated $PM_{2.5}$ concentrations based on a ground monitoring network. *Science of the Total Environment*, *568*, pp.1253-1262. http://doi.org/10.1016/j,scitotenv.2016.05.165

Liu, M., Huang, Y., Ma, Z., Jin, Z., Liu, X., Wang, H., Liu, Y., Wang, J., Jantunen, M., Bi, J. and Kinney, P.L., 2017. Spatial and temporal trends in the mortality burden of air pollution in China: 2004–2012. *Environment International*, *98*, pp.75-81. http://doi.org/10.1016/j.envint.2016.10.003

Lu, F., Xu, D., Cheng, Y., Dong, S., Guo, C., Jiang, X. and Zheng, X., 2015. Systematic review and meta-analysis of the adverse health effects of ambient PM2. 5 and PM10 pollution in the Chinese population. *Environmental research*, *136*, pp.196-204. http://doi.org/10.1016/j.envres.2014.06.029

Luo, G., Wang, Z. 2006. Establishment and verification of the global environmental atmospheric transport model (GEATM). *Chinese J Atmos Sci*, *3*, 504-518. (Chinese). Doi:10.3878/j,issn.1006-9895.2006.03.13

Maji, K.J., Dikshit, A.K., Arora, M. and Deshpande, A., 2018. Estimating premature mortality attributable to $PM_{2.5}$ exposure and benefit of air pollution control policies in China for 2020. *Science of The Total Environment*, *612*, pp.683-693. http://doi.org/10.1016/j.scitotenv.2017.08.254

Ministry of Ecology and Environment of the People's Republic of China. www.mee.gov.cn

Multi-resolution emission inventory for China. www.meicmodel.org

National Bureau of Statistics of China. 2013. China Health Statistical Yearbook, China Statistics Press, Beijing.

Nenes, A., Pandis, S.N. and Pilinis, C., 1999. Continued development and testing of a new thermodynamic aerosol module for urban and regional air quality models. *Atmospheric Environment*, *33*(10), pp.1553-1560. http://doi.org/10.1016/s1352-2310(98)00352-5

Odum, J.R., Jungkamp, T.P.W., Griffin, R.J., Flagan, R.C. and Seinfeld, J.H., 1997. The atmospheric aerosol-forming potential of whole gasoline vapor. *Science*, *276*(5309), pp.96-99. http://doi.org/10.1126/science.276.5309.96

Ostro, B., Hu, J., Goldberg, D., Reynolds, P., Hertz, A., Bernstein, L. and Kleeman, M.J., 2015. Associations of mortality with long-term exposures to fine and ultrafine particles, species and sources: results from the California Teachers Study Cohort. *Environmental health perspectives*, *123*(6), pp.549-556. http://doi.org/10.1289/ehp.1408565

Pan, X. C.,2005. Effects of indoor air quality on human health. China Science Foundation, 4:205-208. (Chinese) doi:10.16262/j.cnki.1000-8217.2005.04.005

Pandis, S.N., Harley, R.A., Cass, G.R. and Seinfeld, J.H., 1992. Secondary organic aerosol formation and transport. *Atmospheric Environment. Part A. General Topics*, *26*(13), pp.2269-2282. http://doi.org/10.1016/0960-1686(92)90358-R

Pope III, C.A., 2000. Epidemiological basis for particulate air pollution health standards. *Aerosol Science & Technology*, *32*(1), pp.4-14. http://doi.org/10.1080/027868200303885

Pope III, C.A. and Dockery, D.W., 2006. Health effects of fine particulate air pollution: lines that connect. *Journal of the air & waste management association*, *56*(6), pp.709-742. http://doi.org/10.1080/10473289.2006.10464485

Pope III, C.A., Lefler, J.S., Ezzati, M., Higbee, J.D., Marshall, J.D., Kim, S.Y., Bechle, M., Gilliat, K.S., Vernon, S.E., Robinson, A.L. and Burnett, R.T., 2019. Mortality risk and fine particulate air pollution in a large, representative cohort of US adults. *Environmental health perspectives, 127(7), p.077007*. http:// doi: 10.1289/EHP4438

Song, C., He, J., Wu, L., Jin, T., Chen, X., Li, R., Ren, P., Zhang, L. and Mao, H., 2017. Health burden attributable to ambient PM2. 5 in China. *Environmental pollution*, *223*, pp.575-586. http://doi.org/10.1016/j.envpol.2017.01.060

Walcek, C.J. and Aleksic, N.M., 1998. A simple but accurate mass conservative, peak-preserving, mixing ratio bounded advection algorithm with FORTRAN code. *Atmospheric Environment*, *32*(22), pp.3863-3880. http://doi.org/10.1016/s1352-2310(98)00099-5

Wang, X. and Mauzerall, D.L., 2006. Evaluating impacts of air pollution in China on public health: implications for future air pollution and energy policies. *Atmospheric Environment*, *40*(9), pp.1706-1721. http://doi.org/10.1016/j.atmosenv.2005.10.006

Wang, Z., Maeda, T., Hayashi, M., Hsiao, L.F. and Liu, K.Y., 2001. A nested air quality prediction modeling system for urban and regional scales: Application for high-ozone episode in Taiwan. *Water, Air, and Soil Pollution*, *130*(1-4), pp.391-396. http://doi.org/10.1023/A:1013833217916

Wang, Z. F., Xie, F. Y., Wang, X. Q., An, J., Zhu, J. 2006. Development and application of nested air quality prediction modeling system. Chinese Journal of Atmospheric Sciences *30*(5), 778. (Chinese). doi:10.3878/j.issn.1006-9895.2006.05.07

Wang, Y., Chen, H., Wu, Q., Chen, X., Wang, H., Gbaguidi, A., Wang, W. and Wang, Z., 2018. Three-year, 5 km resolution China $PM_{2.5}$ simulation: Model Performance evaluation. Atmospheric Research 207, pp 1-13. http://doi.org/10.1016/j.atmosres.2018.02.016

Wei, J., Li, Z., Huang, W., Xue, W., Sun, L., Guo, J., Peng, Y., Li, J., Lyapustin, A., Liu, L., Wu, H., and Song, Y.: Improved 1-km-resolution PM2.5 estimates across China using the space-time extremely randomized trees, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-815, in review, 2019.

Wesely, M.L., 1989. Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models. *Atmospheric Environment (1967)*, *23*(6), pp.1293-1304. http://doi.org/10.1016/0004-6981(89)90153-4

Xie, R., Sabel, C.E., Lu, X., Zhu, W., Kan, H., Nielsen, C.P. and Wang, H., 2016. Long-term trend and spatial pattern of PM2. 5 induced premature mortality in China. *Environment international*, *97*, pp.180-186. http://doi.org/10.1016/j.envint.2016.09.003

Xie, Y. B., Chen, J., Li, W. 2014. An Assessment of $PM_{2.5}$ Related Health Risks and Impaired Values of Beijing Residents in a Consecutive High-level Exposure During Heavy Haze Days. Environmental Science, 35(1), 1-8. (Chinese). doi: 0250-3301(2014)01-0001-08

Yang, G., Wang, Y., Zeng, Y., Gao, G.F., Liang, X., Zhou, M., Wan, X., Yu, S., Jiang, Y., Naghavi, M. and Vos, T., 2013. Rapid health transition in China, 1990–2010: findings from the Global Burden of Disease Study 2010. *The lancet*, *381*(9882), pp.1987-2015. http://doi.org/10.1016/s0140-6736(13)6109-1

Zaveri, R.A. and Peters, L.K., 1999. A new lumped structure photochemical mechanism for large-scale applications. Journal of Geophysical Research: Atmospheres, 104(D23), pp.30387-30415. http://doi.org/10.1029/1999jd900876

Zhang, D. Q., Song, W. L. 2018. Northern Hemisphere Atmospheric Circulation Characteristics in 2017/2018 Winter and Its Impact on Weather and Climate in China. Meteorology. 44(7): 969-976. (Chinese). doi:10.7519/j.issn.1000-0526.2018.07.013

Zhang, J.K., Sun, Y., Liu, Z.R., Ji, D.S., Hu, B., Liu, Q. and Wang, Y.S., 2013. Characterization of submicron aerosols during a serious pollution month in Beijing (2013) using an aerodyne high-resolution aerosol mass spectrometer. *Atmospheric Chemistry & Physics Discussions*, *13*(7).

http://doi.org/10.5194/acpd-13-19009-2013

Zhang, L., Gong, S., Padro, J. and Barrie, L., 2001. A size-segregated particle dry deposition scheme for an atmospheric aerosol module. *Atmospheric Environment*, *35*(3), pp.549-560. http://doi.org/10.1016/s1352-2310(00)00326-5

Zhang, L., Liao, H. and Li, J., 2010. Impacts of Asian summer monsoon on seasonal and interannual variations of aerosols over eastern China. *Journal of Geophysical Research: Atmospheres*, *115*(D7). http://doi.org/10.1029/2009jd012299

Zheng, Y., Xue, T., Zhang, Q., Geng, G., Tong, D., Li, X. and He, K., 2017. Air quality improvements and health benefits from China's clean air action since 2013. *Environmental Research Letters*, *12*(11), p.114020.. http://doi.org/10.1088/1748-9326/aa8a32

Zhong N. 2011. Chronic obstructive pulmonary disease in China. Chinese Journal of Practical Internal Medicine. 31(5), 321-322. (Chinese) doi:10.3760/cma.j.ssn.1673-436X.2011.007.004