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Exploring sources of uncertainty in premature mortality estimates from fine particulate matter: the case of China

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Abstract

Atmospheric pollution from fine particulate matter (PM_{2.5}) is one of the major concerns in China because of its widespread and harmful impacts on human health. In recent years, multiple studies have sought to estimate the premature mortality burden from exposure to PM_{2.5} to inform policy decisions. However, different modeling choices have led to a wide array of results, with significant discrepancies both in the total mortality burden and in the confidence intervals. Here, we present a new comprehensive assessment of PM_{2.5}-related mortality for China, which includes quantification of the main sources of variability, as well as of age and province-specific premature mortality trends during 2015–2018. Our approach integrates PM_{2.5} observations from more than 1600 monitoring stations with the output of a high-resolution (8 km) regional simulation, to accurately estimate PM_{2.5} fields along with their uncertainty, which is generally neglected. We discuss the sensitivity of mortality estimates to the choice of the exposure-response functions (ERFs), by comparing the widely used integrated exposure response functions (IERs) to the recently developed Global Exposure Mortality Models (GEMMs). By propagating the uncertainty in baseline mortalities, PM_{2.5} and ERFs under a Monte Carlo framework, we show that the 95% confidence intervals of mortality estimates are considerably wider than previously reported. We thus highlight the need for more epidemiological studies to constrain ERFs and we argue that uncertainty related to PM_{2.5} estimate should be also incorporated in health impact assessment studies. Although the overall mortality burden remains vast in China (~1.6 million premature deaths, according to GEMMs), our results suggest that 200 000 premature deaths were avoided and 195 billion US dollars were saved in 2018 compared to 2015, bolstering the mounting evidence about the effectiveness of China's air quality policies.

1. Introduction

Exposure to high concentrations of air pollutants, including fine particulate matter (i.e. PM_{2.5}), leads to premature mortality related to cardiovascular and pulmonary diseases, as highlighted by a number of epidemiological studies (Kan *et al* 2012, Apte *et al* 2015, Zhang *et al* 2017). As a result of rapid economic growth, industrialization and urbanization, China is experiencing severely degraded air quality conditions (Li *et al* 2016a) and ranks first for premature mortality due to air pollution with more than

one million deaths per year (Lelieveld *et al* 2015, Gu *et al* 2018), followed by India with ~650 000 deaths (Lelieveld *et al* 2015). In response to this situation, since the year 2010, the Chinese government has implemented clean air policies to reduce air pollution levels (State-Council 2013) by imposing more stringent emission standards and by promoting transition to cleaner fuels and/or non-fossil energy sources for the industrial, power, and residential sectors (Zheng *et al* 2018). These actions will contribute to mitigate not only pollution impacts on air quality (Yang and Zhang 2018), but also those

on climate change (Li *et al* 2018). The recent decreasing trend in pollution concentrations (during 2015–2017) has been linked to the effective implementation of emissions reduction strategies by purely observation-based studies (Silver *et al* 2018) as well as by model simulations (Ding *et al* 2019).

Multiple studies have sought to estimate the nationwide mortality burden associated with PM_{2.5} in China, by employing different techniques. Most of these analyses rely on regional chemical transport model (CTMs) simulations, which, however, are subject to significant uncertainties that are often not quantified (Lelieveld *et al* 2015, Silva *et al* 2016, Butt *et al* 2017, Hu *et al* 2017, Zheng *et al* 2017b, Aunan *et al* 2018, Gao *et al* 2018). Depending on the simulation setup and the chosen exposure-response functions (ERFs), significantly different estimates of the total premature mortality burden were presented, ranging from 916 000 premature deaths in 2014 (Archer-Nicholls *et al* 2016) to 1331 000 in 2013 (Gao *et al* 2018). Much larger figures (up to ~2.47 million) are obtained when using the latest exposure response functions from Burnett *et al* (2018). Furthermore, the 95% confidence intervals (CI) reported in such studies can be as narrow as (1 090 000–1 190 000) (Aunan *et al* 2018) or as wide as (168 000–1 796 000) (Gu *et al* 2018). Besides model simulations and ERFs, other sources of uncertainty include baseline mortalities and exposed population estimates (Kodros *et al* 2018).

Mortality estimates have also been proposed based on a combination of ground and/or satellite observations (Zhou *et al* 2015, 2019, Jerrett *et al* 2017, Dong *et al* 2018, Huang *et al* 2018), despite being mostly focused on geographically limited areas. A few attempts to combine observations—either from monitoring stations or satellites—and CTM results were made to overcome limitations of prior studies, both worldwide (Ford and Heald 2016, Van Donkelaar *et al* 2016, Shaddick *et al* 2018) and in China (Liu *et al* 2016, Liang *et al* 2017, Bai *et al* 2019, Zou *et al* 2019). For instance, multiple studies used different geostatistical techniques to integrate PM_{2.5} ground measurements, CTMs output, satellite observations of AOD and other land use and meteorological factors to estimate PM_{2.5} exposure at different spatial scales, ranging from the city-level (Li *et al* 2016b, Tao *et al* 2020) to regional and national scales (Liang *et al* 2017, Bai *et al* 2019, Wei *et al* 2019). Although these works improved representation of PM_{2.5} spatial patterns, the uncertainty in PM_{2.5} concentrations—as well as its propagation to the mortality estimate—was not quantified. Using data assimilation techniques, Liu *et al* (2016) quantified the PM_{2.5} uncertainty from a CTM and observations, although the resolution of that simulation was rather coarse (45 km).

Building from previous attempts to combine observations and modeling results, here we propose a novel approach to quantify the PM_{2.5}-related

mortality burden and its uncertainty in China. Our methodology integrates ground-based observations from the recently established Chinese monitoring network (>1500 sites) with a high-resolution CTM simulation (8 km). This work aims to:

- (a) Provide a multi-year assessment of the mortality burden related to PM_{2.5} (2015–2018) by nudging model results to the observations.
- (b) Discuss the effect of using recently revised ERFs (Burnett *et al* 2018) in the recent mortality trends.
- (c) Overcome key limitations of previous studies on the uncertainty of such estimates.

Specifically, our methodology allows estimating the uncertainty in PM_{2.5} concentrations and its propagation to the final mortality results. We show that confidence intervals of mortality estimates previously reported are likely narrower than the true ones, as they do not take into account PM_{2.5} uncertainty (e.g. Reddington *et al* 2019, Zou *et al* 2019). We discuss nationwide and province-level mortality trends during 2015–2018 as well as the advantages of using our modeling framework compared to other approaches.

2. Methods

2.1. PM_{2.5} observations

This investigation is based on hourly PM_{2.5} concentrations for Mainland China and Hong Kong. Data are retrieved from online repositories of the Ministry of Ecology and Environment for Mainland China (<http://beijingair.sinaapp.com/>) and of the Hong Kong Environmental Protection Department (www.epd.gov.hk/epd) for Hong Kong. The monitoring network comprises 1601 and 16 sites for Mainland China and Hong Kong, respectively. PM_{2.5} measurements are acquired and analyzed for four consecutive years, from 2015 to 2018, at hourly resolution. Data are pre-processed for use in the mortality assessment as described in the Supplementary Information.

2.2. WRF-Chem simulation

Hourly PM_{2.5} concentrations are simulated by the Weather Research and Forecasting model with Chemistry (WRF-Chem, version 3.9) (Grell *et al* 2005, Skamarock and Klemp 2008) at 8 km resolution. This resolution allows to capture important regional details in complex mountainous areas and resolve atmospheric processes at urban and regional scales including formation/transformation/removal of air pollutants, thus facilitating the direct comparison between model results and station measurements. Monthly varying anthropogenic emissions are based on the EDGAR-HTAP (Emission Database for Global Atmospheric Research for Hemispheric Transport of

Air Pollution, v2.2 (Janssens-Maenhout *et al* 2015) inventory which is available on a grid of $0.1^\circ \times 0.1^\circ$ for the year 2010 (latest year available). EDGAR-HTAPv2.2 uses the Model Intercomparison Study for Asia Phase III (MIX) mosaic Asian emission inventory version 1.0 at $0.25^\circ \times 0.25^\circ$ horizontal resolution (Li *et al* 2017), which incorporates data from the Multi-resolution Emission Inventory for China (MEIC, www.meicmodel.org). We select EDGAR-HTAPv2.2 for consistency with the simulation resolution (8 km). Other emission inventories, like ECLIPSE (Klimont *et al* 2017), provide more up-to-date emissions but at a lower resolution (~ 50 km), which would cause a mismatch with the resolution applied in our model simulation. The entire 2015 is simulated, with a spin up of 1 month (1–31 December 2014). Details on the simulation setup are provided in the Supplementary Information (table S2).

2.3. Integrating observations and WRF-Chem results

To account for the uncertainty of WRF-Chem results, we introduce a statistical methodology which integrates measured data in the analysis. The novelty of the proposed methodology is three-fold: (i) nudging the CTM results towards the observations, (ii) providing a better quantification of the mortality CI by accounting for $PM_{2.5}$ uncertainty and (iii) estimating a multi-year premature mortality trend using only one year of computationally-expensive WRF-Chem simulations.

The $PM_{2.5}$ concentration field is reconstructed by applying a linear regression model followed by kriging of the residuals (ε) generated by the model:

$$\log z = \beta_0 + \beta_1 x + \varepsilon \quad (1)$$

where z is the corrected annual mean concentration of $PM_{2.5}$ in each grid cell, x is the logarithmically transformed annual mean concentration simulated by WRF-Chem, β_0 and β_1 are parameters to be estimated. In other words, the observed data are first detrended using WRF-Chem results as the only covariate, and then ordinary kriging is performed on the detrended data to include the residual spatial dependence in the model. A logarithmic transformation is used in equation (1) to work under the Gaussianity assumption, as the original data are slightly skewed (figure S1 (stacks.iop.org/ERL/15/064027/mmedia)). Moreover, the logarithmic transformation avoids unphysically negative $PM_{2.5}$ concentrations that could arise from the regression. Since x and ε are Gaussian and the model is additive, we can calculate the variance of the predictions (i.e. the uncertainty in the $PM_{2.5}$ field) by adding the variance related to the linear regression part and the variance arising from the kriging predictions. Details on the calculation of prediction variances for linear regression and ordinary kriging are reported in the Supplementary Information for completeness.

This procedure is applied for each year in the 2015–2018 period, with time-varying coefficients (i.e. β_0, β_1 and the parameters of the variogram are estimated on an annual basis). Linear regression parameters are estimated by maximizing the likelihood of the data, which is equivalent to ordinary least squares for linear models, and the variogram is estimated using a spherical covariance function, which proved to be the most numerically stable covariance function across the years. The values of the estimated parameters can be found in the Supplementary Information (table S1). For comparison with other models, hereinafter we will denote model (1) as ‘Regression + Kriging’, whereas the same model without kriging on the residuals ε will be denoted as ‘Regression-only’ (i.e. model (1) where we do not model the spatial structure in ε and we assume that the residuals are instead normally distributed with zero mean and variance σ^2).

2.4. Population exposure and mortality estimates

Long-term premature mortality associated with $PM_{2.5}$ is calculated following a well-established approach (e.g. Burnett *et al* 2014, Apte *et al* 2015):

$$M_{adk} = P_{ak} \times B_{ad} \cdot \frac{RR_{adk} - 1}{RR_{adk}} \quad (2)$$

where M is the estimated premature mortality burden, P is the population count, B is the baseline mortality rate and RR is the relative risk due to exposure to annual mean concentration of $PM_{2.5}$. Subscripts a, d and k refer to the specific age interval, disease and grid cell, respectively. As in previous studies (Gu *et al* 2018), we consider premature mortality associated to lung cancer (LC), ischemic heart disease (IHD), stroke and chronic obstructive pulmonary diseases (COPD) for individuals above 25 years with 5-years groupings (e.g. 25–30, 31–35, etc). Spatially resolved population count from the 2017 LandScan High Resolution global Population Data (Rose *et al* 2018) at ~ 1 km resolution is regridded to the WRF-Chem grid to match the pollutant field. Population is evolved during 2015–2018 by rescaling Landscan data with the population structure from the Chinese statistical yearbook, as done in Ding *et al* (2019). China- and age-specific baseline mortality data for 2015, 2016 and 2017 are retrieved from the 2017 Global Burden of Disease (GBD) study (Dicker *et al* 2018). We linearly extrapolate baseline mortality data for 2018 as they were not available at the time of the analysis.

RRs are computed using the recent cause-specific Global Exposure Mortality Models (GEMMs), which were constructed based on cohort studies of outdoor air pollution only (Burnett *et al* 2018):

$$RR_k = \exp \left(\frac{\theta \log \left(\frac{\bar{z}_k}{\alpha} + 1 \right)}{1 + \exp \left(-\frac{\bar{z}_k - \mu}{\nu} \right)} \right) \quad (3)$$

where θ , α , μ , ν are age and disease-specific parameters, and \tilde{z}_k is the maximum between zero and the difference between the PM_{2.5} concentration in grid cell k and the counterfactual concentration ($z_{cf} = 2.4 \mu\text{g m}^{-3}$), i.e. the minimum concentration below which no impacts are assumed to occur. The parameters were estimated by Burnett *et al* (2018) using cohort studies that cover the global exposure range, including a Chinese male cohort. For comparison purposes, we also calculate RRs with the traditional IERs (Burnett *et al* 2014), which however include risk information from indoor air pollution and secondhand and active smoking:

$$RR_k = 1 + \alpha \left\{ 1 - e^{-\gamma(z_k - z_{cf})^\delta} \right\} \quad (4)$$

where α , γ and δ are age- and disease-specific parameters estimated in the GBD study (Dicker *et al* 2018) along with their uncertainty. Equation (4) is valid only if z_k is greater than z_{cf} , otherwise RR is set to 1. The counterfactual concentration for the IERs is uniformly distributed between 2.4 and 5.9 $\mu\text{g m}^{-3}$. Total mortality is finally computed as the sum for each health endpoint, age group and grid cell:

$$M = \sum_a \sum_d \sum_k M_{adk} \quad (5)$$

We also compute the monetary savings as the difference in mortality between two instances in time multiplied by the value of statistical life (VSL), which is assumed to be equal to 978 219 US dollars (in 2011 values), following previous literature (Zou *et al* 2019). A thorough discussion of the monetary evaluation uncertainty exceeds the scope of this work, but could be considered in future investigations.

2.5. Uncertainty quantification

We propagate the uncertainty in the input (i.e. PM_{2.5} fields, baseline mortality and ERFs parameters) to the mortality estimates with a numerical approach. PM_{2.5} fields, baseline mortality and ERFs are assumed to be Gaussian distributions, with parameters (i.e. mean and standard deviations) specified in the Supplementary Information. For each Monte Carlo simulation, we independently sample from such distributions and we apply equations (2) to (5) to calculate the mortality value for that specific simulation. To ensure robustness of our results (i.e. a coefficient of variation of the estimated total mean mortality less than 0.05%), we perform 108 000 simulations (i.e. 4500 simulations per processor distributed on 24 processors) which resulted in 108 000 total mortality values. The mortality central estimate is therefore the mean of the 108 000 mortality estimates, whereas the 95% confidence interval is calculated as the 0.025- and 0.975-quantiles of the empirical distribution obtained with the 108 000 simulations.

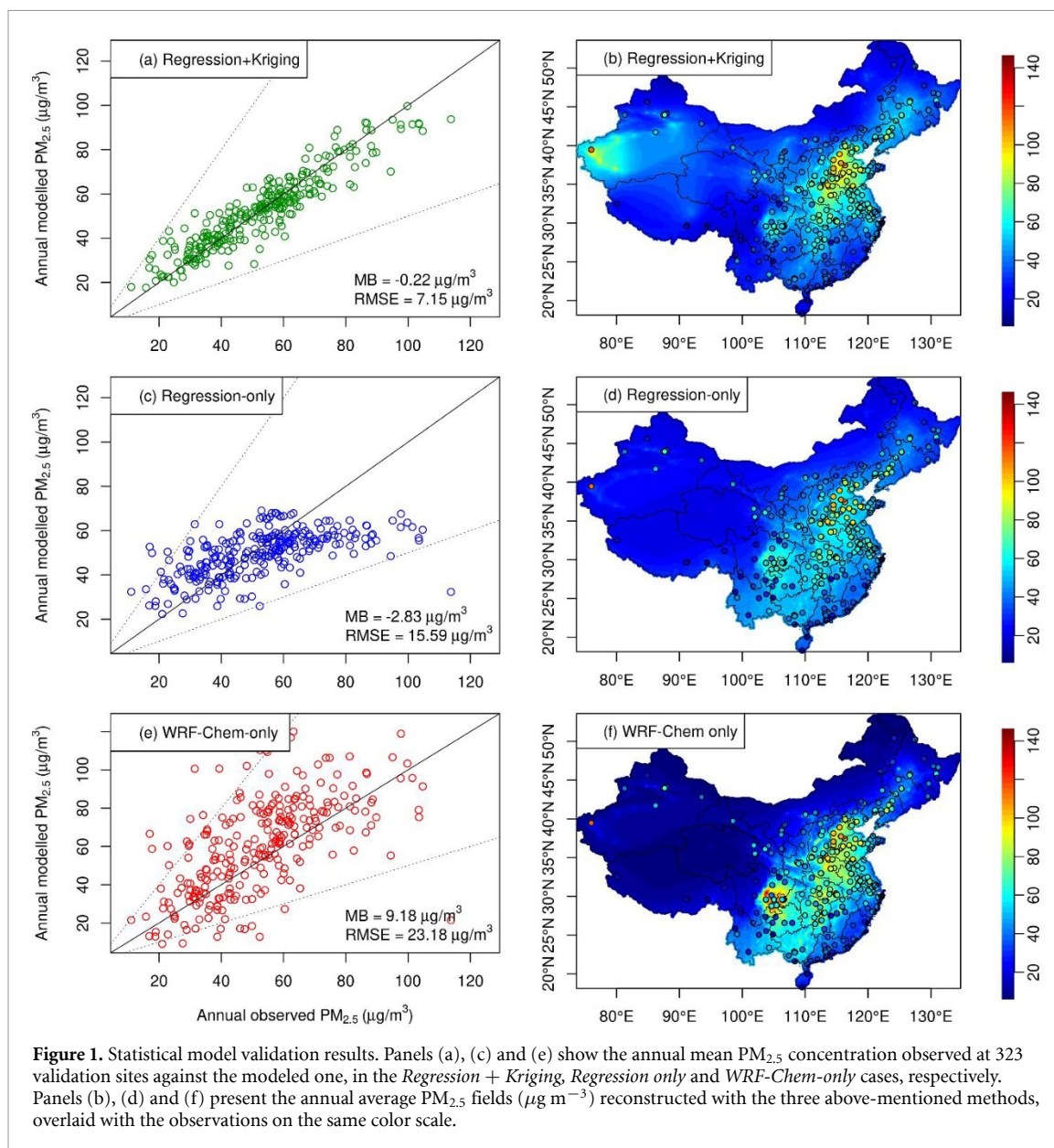
To assess the influence of each input (i.e. PM_{2.5}, ERFs parameters and baseline mortality) on the total uncertainty, we run four different cases by marginally assuming no uncertainty in some of the input features (i.e. treating that input feature as an exact value rather than a Gaussian random variable): (i) only ERFs parameters are uncertain (*ERFs-only*), as usually done in previous literature, (ii) only ERFs and baseline mortalities are uncertain (*ERFs + BM*), (iii) only ERFs and PM_{2.5} are uncertain (*ERFs + PM_{2.5}*) and (iv) all three input features are uncertain (*ERFs + BM + PM_{2.5}*).

3. Results

3.1. Model evaluation

Annual PM_{2.5} concentrations are generally well reproduced by the high-resolution WRF-Chem simulation, with 93.5% of the modeled values being within a factor of 2 of the measured concentrations (figure S2). On average, the model overestimates observations, with a mean bias of 10.16 $\mu\text{g m}^{-3}$ (figure S2(b)), which is likely related to the mismatch between the simulated year and the emission inventory year. Such overestimation mainly occurs during the summer, when the overall difference between the model and the observations is significant (figure S2(b)). Conversely, the model is negatively biased at most sites measuring extremely high annual average concentrations (i.e. above 100 $\mu\text{g m}^{-3}$). As clear from figure S3, the most misrepresented areas are the Sichuan province (e.g. Chengdu, which is highly overestimated) and the westernmost cities of China (e.g. Kashgar, which is underestimated). Despite the relatively fine spatial resolution, limitations remain in the model's ability to capture local scale PM_{2.5} concentrations, particularly over megacities where urban measurements are highly impacted by local emissions and sub-grid scale dispersion processes not resolved by a CTM.

A more accurate PM_{2.5} field can be obtained by integrating observations with the model results, as described in section 2.3. According to the calibration/validation procedure described in the Supplementary Information, the 'Regression + Kriging' statistical model is the most suitable to reproduce the actual observations (figure 1 and table S3). Major improvements to the WRF-Chem results are thus achieved when integrated with observations, even with a simple linear regression only (the mean bias decreases from 10.2 $\mu\text{g m}^{-3}$ to $-2.50 \mu\text{g m}^{-3}$ and Root Mean Square Error (RMSE, defined in the Supplementary Information) from 24.87 $\mu\text{g m}^{-3}$ to 15.62 $\mu\text{g m}^{-3}$). However, by also relaxing the assumption of *iid* residuals (ε in equation 1) and modeling the spatial dependence via a kriging procedure, we can accomplish almost zero bias with a threefold decrease in RMSE compared to the WRF-Chem model results. More complex statistical models were



also investigated but did not provide significantly more accurate results despite the additional complexity (Supplementary Information).

3.2. Premature mortality estimates

The long-term premature mortality burden during 2015–2018 is reported in table 1. Age- and disease-specific premature mortality estimates indicate that IHD and stroke are responsible for most premature deaths attributable to $PM_{2.5}$ (36.3% and 32.0% of total $PM_{2.5}$ related deaths, respectively), especially among the young population (figure 2). More than 50% premature deaths due to LC, IHD, COPD and stroke are attributable to $PM_{2.5}$ exposure (i.e. $RR \geq 2$, using GEMMs) for adults less than 40 years old. This percentage decreases with age, as other medical conditions leading to those diseases may dominate for the elderly population (figure 2). Given the rather short timeframe considered in this work, the premature mortality distribution by age groups remains almost

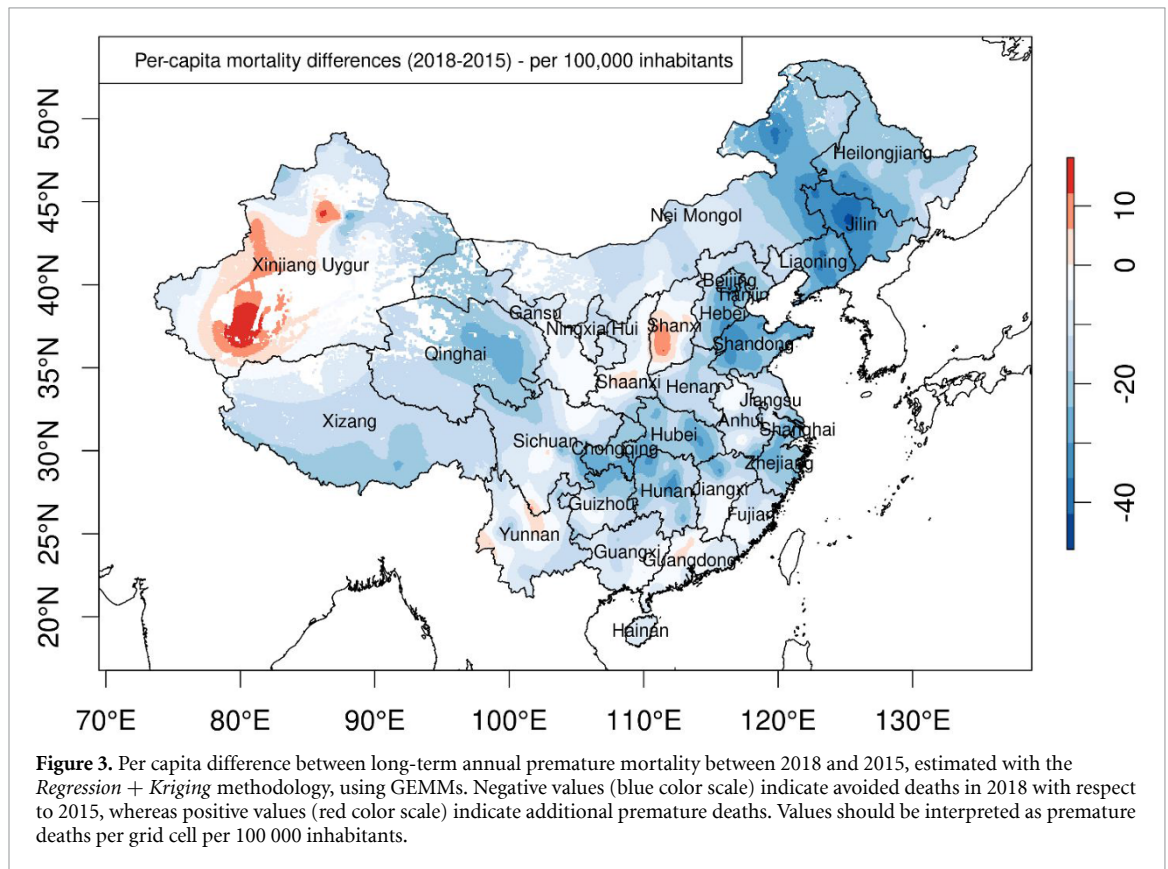
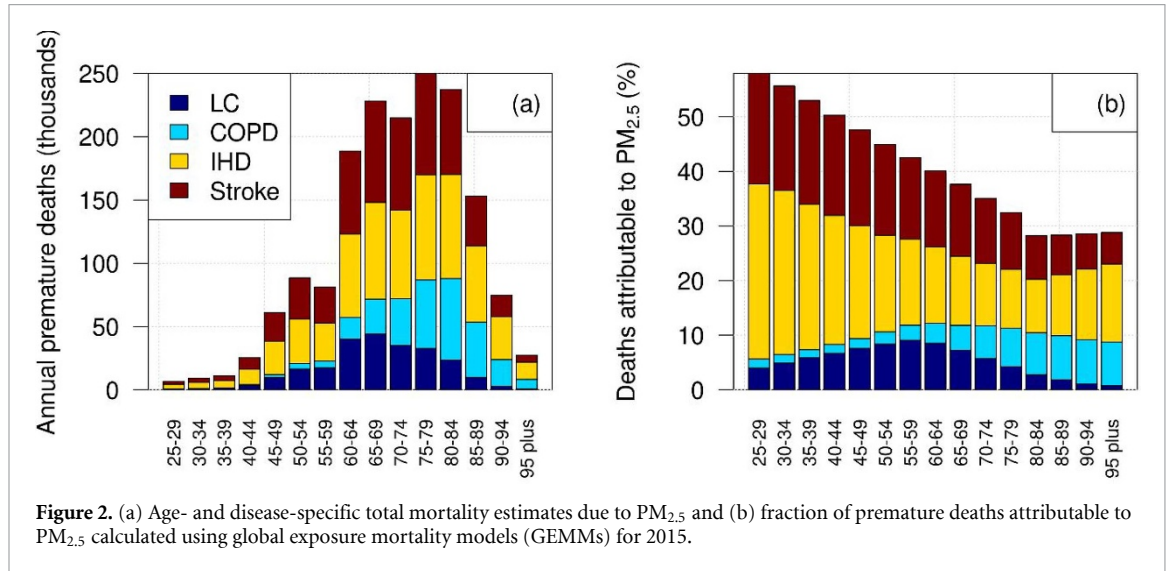
constant in time (figure S5), i.e. both the baseline mortality and the total population distributions do not vary considerably during the years of study.

Since on average our WRF-Chem simulation overestimates $PM_{2.5}$ concentrations, likely because of the mismatch in the emission inventory year, among other reasons, the use of model output only—as done in prior studies with non-zero biases (e.g. Reddington *et al* 2019)—would overestimate the total annual premature mortality burden by almost 140 000 deaths in 2015 in our work (table 1), compared to the bias-correction methodology (*Regression + Kriging*).

According to the *Regression + Kriging* methodology, we estimate that in 2018 around 200 000 premature deaths were avoided compared to 2015. Several factors have contributed to this trend. During 2015–2018, population growth (+1.4% during 2015–2018) and ageing of the Chinese population (+1 year of median age) have slightly offset the reduction in premature deaths due to better air quality conditions,

Table 1. Central estimate (mean) of long-term premature mortality associated with PM_{2.5} during 2015–2018 for Mainland China and Hong Kong, using GEMMs.

	2015	2016	2017	2018
<i>Regression + Kriging</i>	1 656 500	1 579 600	1 527 100	1 424 800
<i>Regression-only</i>	1 641 400	1 552 800	1 520 800	1 438 600
<i>WRF-Chem-only</i>	1 785 800	—	—	—



whereas the decrease in baseline mortality rates positively reinforced such trend. Without any variations in the population age distribution and in baseline mortality, 20 000 additional deaths would have been avoided in 2018 compared to 2015, i.e. changes in

population structure outweighed the benefits deriving from improved health conditions.

The major improvements in terms of avoided premature deaths are concentrated in major urban areas (e.g. Beijing, Shanghai, Changchun, figure 3).

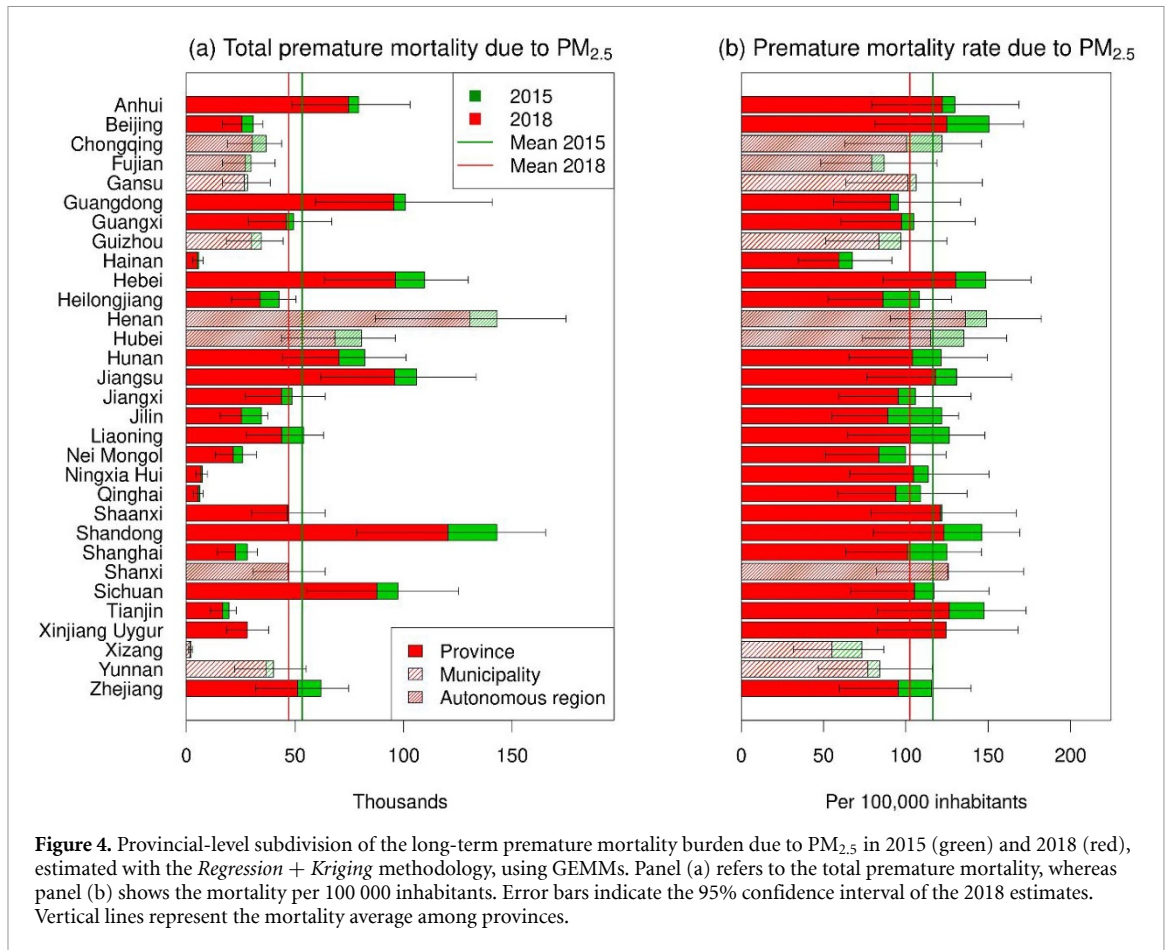


Figure 4. Provincial-level subdivision of the long-term premature mortality burden due to PM_{2.5} in 2015 (green) and 2018 (red), estimated with the *Regression + Kriging* methodology, using GEMMs. Panel (a) refers to the total premature mortality, whereas panel (b) shows the mortality per 100 000 inhabitants. Error bars indicate the 95% confidence interval of the 2018 estimates. Vertical lines represent the mortality average among provinces.

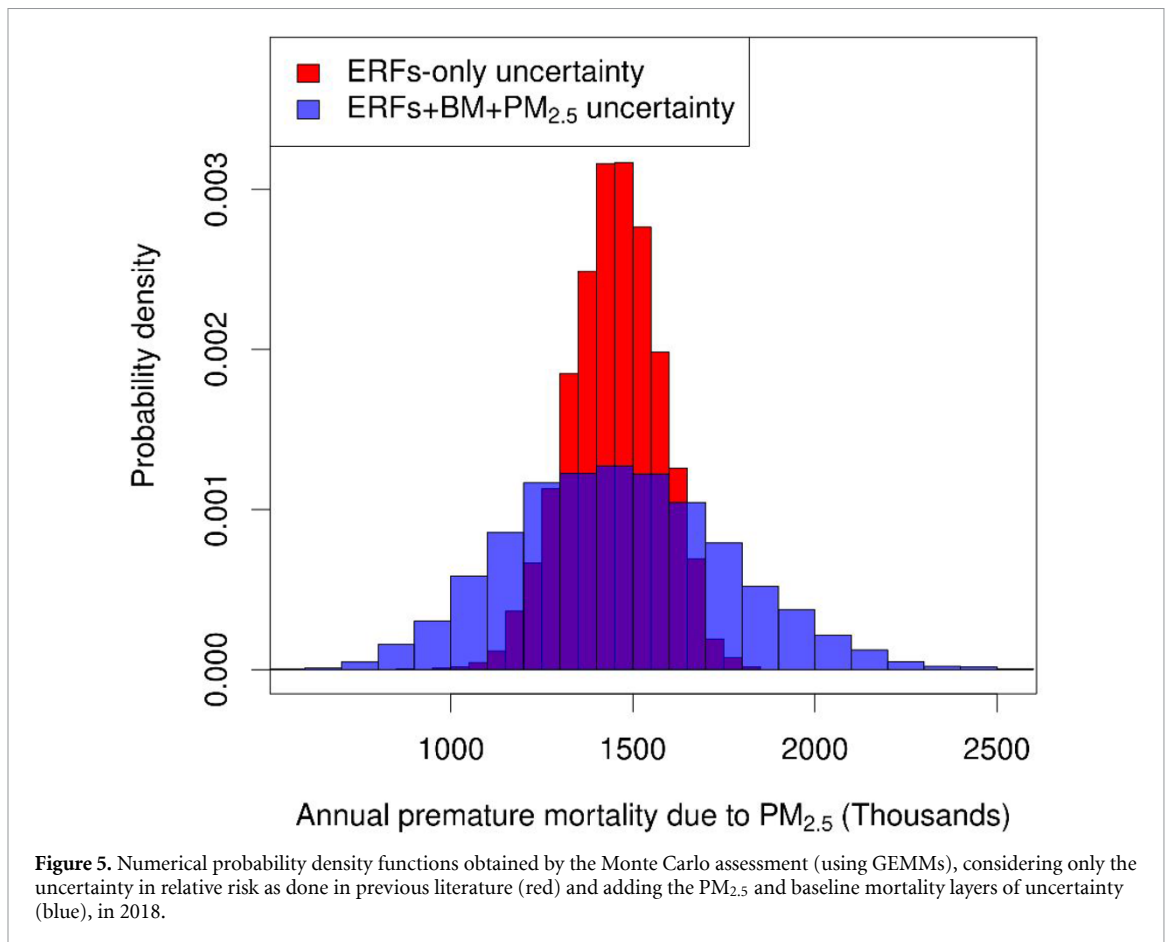
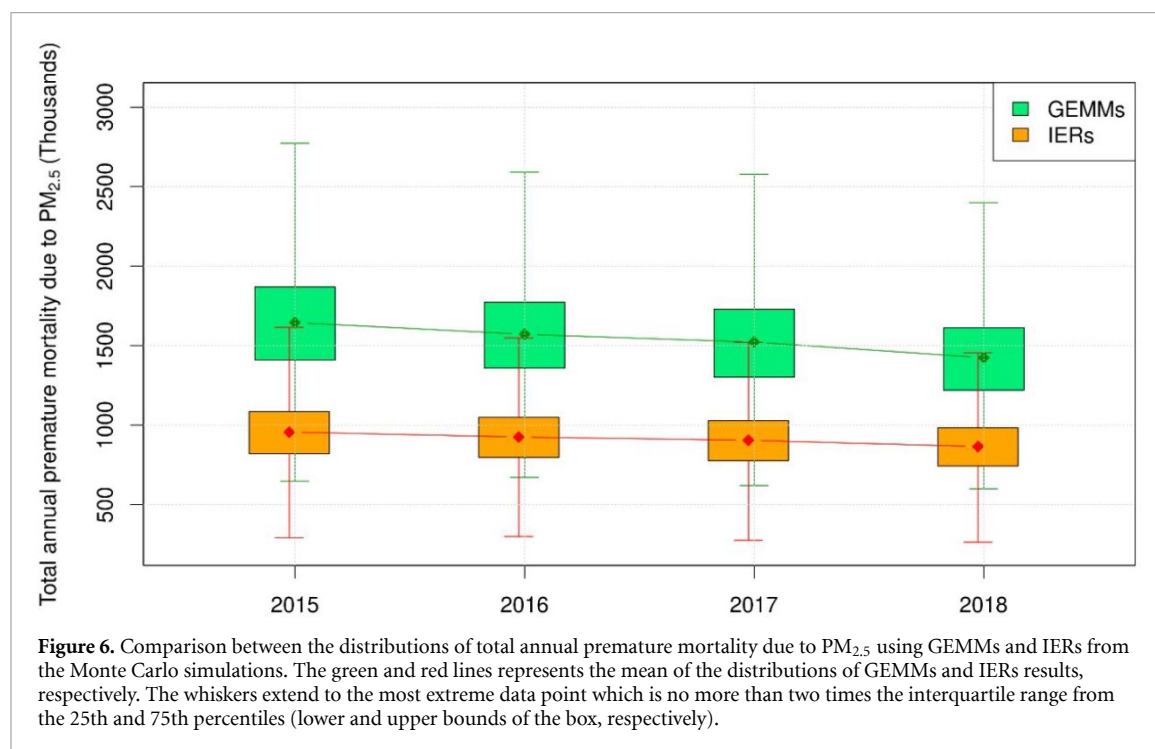


Figure 5. Numerical probability density functions obtained by the Monte Carlo assessment (using GEMMs), considering only the uncertainty in relative risk as done in previous literature (red) and adding the PM_{2.5} and baseline mortality layers of uncertainty (blue), in 2018.

Table 2. Central estimates and 95% confidence intervals (CI) for the long-term premature mortality burden due to PM_{2.5} during 2015–2018, for the four different cases presented in section 2.5, using GEMMs.

Central estimate (mean)	2015	2016	2017	2018
<i>All cases</i>	1 656 500	1 579 600	1 527 100	1 424 800
95% Confidence Interval	2015	2016	2017	2018
<i>ERFs-only</i>	1 359 300–1899 400	1 289 900–1804 400	1 257 700–1763 800	1 195 500–1680 100
<i>ERFs + BM</i>	1 357 700–1911 900	1 284 500–1815 500	1 252 000–1774 500	1 187 900–1689 900
<i>ERFs + PM_{2.5}</i>	1 025 700–2307 500	1 009 300–2172 000	961 100–2165 600	928 700–2070 100
<i>ERFs + BM + PM_{2.5}</i>	1 021 200–2322 100	1 002 300–2181 100	956 000–2174 200	924 420–2078 700

**Figure 6.** Comparison between the distributions of total annual premature mortality due to PM_{2.5} using GEMMs and IERs from the Monte Carlo simulations. The green and red lines represents the mean of the distributions of GEMMs and IERs results, respectively. The whiskers extend to the most extreme data point which is no more than two times the interquartile range from the 25th and 75th percentiles (lower and upper bounds of the box, respectively).

Nonetheless, some other areas have experienced a slight worsening of their mortality burden in the last few years. The most notable examples are the urban area of Xi'an and the westernmost cities of China, coherently with the findings (in terms of air quality) reported in Silver *et al* (2018). The spatial distribution of the difference in terms of total mortality values can be found in the Supplementary Information (figure S6).

The provincial-level partitioning of the total mortality burden indicates that the most populated provinces of Shandong and Henan are characterized by the highest premature mortality due to PM_{2.5} (figure 4), although Shandong province achieved the largest improvements between 2015 and 2018 (25 000 avoided premature deaths, –18%). When normalizing by mortality, the highest reduction was around –28%, accomplished in the Jilin province in only four years. The leading coal producing regions of Shanxi and Shaanxi instead rank last for health improvements, with almost a zero net change during the study period, as also reported by Zou *et al* (2019). On the other hand, major steel-related industrial provinces (e.g. Hebei and Jiangsu, Reddington *et al*

2019) achieved important reductions of 13% and 10%, respectively. The northern, western and central provinces where the residential sector is a major contributor to poor air quality (Tao *et al* 2020) also showed a considerable decrease (e.g. Beijing, 21%, Sichuan, 11% and Hubei, 16%).

3.3. Uncertainty quantification

All the results presented above are related to the estimated mean of the mortality burden. However, such estimates are affected by three layers of uncertainty, associated with the PM_{2.5} concentration fields, the RR values and the baseline mortality. Here, we identify the full probability distribution of the mortality estimates for the four different cases presented in section 2.5. Although all the numerical probability distributions are approximately Gaussian (even if some skewness is present, figure 5), the standard deviations are remarkably different. This implies that the traditional 95% confidence interval, computed only with the ERFs uncertainty, is narrower of about 700 000 premature deaths with respect to the case where the uncertainty in PM_{2.5} fields, baseline mortality and ERFs is considered (table 2 and figure

S8). Because of a combination of the large sensitivity of GEMMs at high $PM_{2.5}$ concentrations and the log-normality of $PM_{2.5}$ concentrations, both the lower bound (2.5% percentile) and the upper bound (97.5% percentile) are significantly sensitive to the addition of the $PM_{2.5}$ layer of uncertainty. On the other hand, the baseline mortality does not play a major role in the premature mortality confidence intervals as it is known with higher accuracy (the average coefficient of variation of age- and disease-specific baseline mortality rates is approximately 3%).

Finally, figure 6 shows the comparison between the mortality estimates using GEMMs and the traditional IERs. The discrepancy between the two methods is significant, with the upper bound of IERs mortality estimates being approximately the mean of the GEMMs mortality distribution. Because the GEMMs relationship between $PM_{2.5}$ and mortality is much stronger at high $PM_{2.5}$ concentrations, the difference is more pronounced in the regions where $PM_{2.5}$ concentrations are particularly large (e.g. urban areas).

4. Discussion and conclusions

In this study, we quantify the long-term health burden associated with $PM_{2.5}$ exposure in China during 2015–2018. To our knowledge, this is the one of the first studies that integrates ground-based observations and high-resolution model output to accurately estimate the trend of nationwide premature mortality due to $PM_{2.5}$ for China. Our approach improves fidelity of premature mortality estimates by accounting and correcting for potential model biases in representing $PM_{2.5}$ concentrations. In addition, we present a novel methodology both to account for the uncertainty in CTM results (Solazzo and Galmarini 2016) and to properly quantify the confidence intervals of the premature mortality estimates. Despite many attempts to validate and improve CTM results (Vautard *et al* 2007), inaccuracies in CTM output systematically occur due to improper spatial resolution (Crippa *et al* 2017), lack of accurate input data (e.g. emissions estimates (Zheng *et al* 2017a)), uncertain initial and boundary conditions (Georgiou *et al* 2018), and incomplete understanding of some physical-chemical processes (e.g. formation of secondary organic aerosol (Giani *et al* 2019)). These issues can lead to potentially large biases in mortality estimates and thus we stress the importance of data assimilation techniques in calculating the mortality burden.

The statistical methodology presented in this work also allows for an improved characterization of the confidence intervals of mortality estimates, by considering and propagating multiple sources of uncertainty. We show that most of the uncertainty in mortality estimates is associated with the uncertainty in $PM_{2.5}$ fields and ERFs parameters, whereas baseline mortality is known with higher accuracy and

therefore entails a minor impact on mortality confidence intervals. Hence, we argue that (i) uncertainty related to $PM_{2.5}$ estimate should be considered in mortality assessments, (ii) improving CTM results can considerably narrow the confidence intervals in mortality estimates and (iii) that some of the variability in mortality estimates presented in previous literature is likely coming from the neglected $PM_{2.5}$ uncertainty.

Results from our analyses suggest that 1 656 500 people prematurely died in 2015 due to $PM_{2.5}$ exposure, although a steady decline is estimated during 2015–2018 (~200 000 avoided deaths in 2018 with respect to 2015 over the entire China, which corresponds to an estimated saved monetary value of 195 billion US dollars). Using GEMMs instead of IERs leads to a significantly larger premature mortality burden compared to previous estimates (which usually range between 900 000 and 1 300 000, Reddington *et al* (2019)), as also highlighted by other studies, e.g. Burnett *et al* (2018) and Xue *et al* (2019b). The uncertainty related to the total premature deaths attributable to $PM_{2.5}$ remains vast, especially when taking into account different exposure response functions as well as the uncertainty related to $PM_{2.5}$ levels. If we consider both the results obtained with IERs and GEMMs together (i.e. assigning an uninformative prior probability of 50% to each of the two models), the range of our estimate (95% confidence interval) would be 574 000–2078 700 in 2018, which is much wider than previously reported. Although GEMMs are likely to be more representative of the health response to ambient $PM_{2.5}$ concentrations than IERs, as the former only include cohort studies of outdoor air pollution, there is no clear evidence of GEMMs providing more reliable results than IERs yet (Xue *et al* 2019b). More research and epidemiological studies are thus needed to constrain and evaluate the different exposure response functions as well as to narrow the current confidence intervals. Nonetheless, the trend of avoided deaths in recent years (~200 000 avoided deaths in 2018 compared to 2015, or 50 000 avoided deaths per year) is coherent with recent findings (42 800 avoided deaths per year, between 2013 and 2017, Zou *et al* (2019)), where estimates were derived from the use of a corrected version of the IERs to take into account exposure-related human activity patterns. Despite a considerably different total mortality burden, a similar trend in recent years was also found also in Ding *et al* (2019) (57 400 avoided deaths per year, between 2013 and 2017), which however relied on CTM simulations only, without any observations. The similarity of these three results, obtained with three independent methodologies, bolsters the robustness of the estimated trends.

Multi-year assessments of premature mortality usually require (i) large computational power to run expensive multi-year simulations and (ii) accurate and up-to-date emission inventories at fine

resolution. Whilst computational power has been increasingly more accessible in recent years, CTM-ready official emission inventories are often available for discontinuous years and released belatedly. The main advantage of our approach is that it partly compensates the need for large computational power (thus allowing for very high resolution in space) and it does not require yearly varying emission inventories which generally require considerable processing before utilization as input to CTMs. Moreover, the presented methodology allows the calculation of the uncertainty in $PM_{2.5}$ spatio-temporal fields and its propagation to the mortality estimates, which overcomes the limitations of deterministic CTM output. However, we acknowledge that our methodology should be used for limited and consecutive years (e.g. 4 years, in our work), to ensure that the base case simulation provides reasonable results for different meteorological and emission conditions. This procedure can thus be intended to fill the gaps for those years where official emission inventories are not available, provided that observations of $PM_{2.5}$ are accessible. Future investigations should be devoted to the refinement of the presented methodology, for example using non-parametric approaches to estimate the $PM_{2.5}$ field and uncertainty (e.g. with machine learning techniques (Bai *et al* 2019, Xue *et al* 2019a)). Population uncertainty, estimated with different global products (e.g. the Gridded Population of the World, v4), can also be included in the Monte Carlo assessment, even though we anticipate that its impact would be of the same order of the baseline mortality. Furthermore, this methodology could prove useful to quantify mortality trends in highly populated regions where the health impacts of air quality have been only partially quantified.

Although we find substantial reductions in the premature mortality rates associated with $PM_{2.5}$ during 2015–2018, and this trend has been reported since 2013 (Zheng *et al* 2017b), the health burden is still vast and multiple challenges await China in the near future. Climate change is expected to lead to more intense and frequent stagnation events and heat waves, which can exacerbate the $PM_{2.5}$ related premature mortality (Hong *et al* 2019). Even though emission reduction policies might be strengthened in the near future (Peng *et al* 2017), the premature mortality burden could still be negatively affected by the predicted population growth and ageing (Madaniyazi *et al* 2015, Cohen *et al* 2017), i.e. a slight reduction in $PM_{2.5}$ concentration could be more than compensated by an increase in the exposed population. Furthermore, socioeconomic factors like economic growth, industrialization and urbanization will strongly influence $PM_{2.5}$ concentrations (Li *et al* 2016a). As the economic growth is a vital goal in China's 13th five-year plan (Aglietta and Bai 2016), the government is urged to find a way to decouple the effects of economic growth on $PM_{2.5}$ pollution.

In light of our findings and other recent studies (Silver *et al* 2018), we conclude that China is headed in the right direction towards the decoupling of these two vital aspects, even though more stringent emission reduction policies will be needed to cope with all the challenges highlighted above.

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Data Availability Statement

The data that support the findings of this study are available upon request from the authors.

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