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# Estimation of $PM_{2.5}$ mortality burden in China with new exposure estimation and local concentration-response function☆

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ABSTRACT

The estimation of $PM_{2.5}$-related mortality is becoming increasingly important. The accuracy of results is largely dependent on the selection of methods for $PM_{2.5}$ exposure assessment and Concentration-Response (C-R) function. In this study, $PM_{2.5}$ observed data from the China National Environmental Monitoring Center, satellite-derived estimation, widely collected geographic and socioeconomic information variables were applied to develop a national satellite-based Land Use Regression model and evaluate $PM_{2.5}$ exposure concentrations within 2013–2015 with the resolution of 1 km × 1 km. Population weighted concentration declined from 72.52 μg/m$^3$ in 2013 to 57.18 μg/m$^3$ in 2015. C-R function is another important section of health effect assessment, but most previous studies used the Integrated Exposure Regression (IER) function which may currently underestimate the excess relative risk of exceeding the exposure range in China. A new Shape Constrained Health Impact Function (SCHIF) method, which was developed from a national cohort of 189,793 Chinese men, was adopted to estimate the $PM_{2.5}$-related premature deaths in China. Results showed that 2.19 million (2013), 1.94 million (2014), 1.65 million (2015) premature deaths were attributed to $PM_{2.5}$ long-term exposure, different from previous understanding around 1.1–1.7 million. The top three provinces of the highest premature deaths were Henan, Shandong, Sichuan, while the least ones were Tibet, Hainan, Qinghai. The proportions of premature deaths caused by specific diseases were 53.2% for stroke, 20.5% for ischemic heart disease, 16.8% for chronic obstructive pulmonary disease and 9.5% for lung cancer. IER function was also used to calculate $PM_{2.5}$-related premature deaths with the same exposed level used in SCHIF method, and the comparison of results indicated that IER had made a much lower estimation with less annual amounts around 0.15–0.5 million premature deaths within 2013–2015.



## 1. Introduction

$PM_{2.5}$ pollution has caused people's growing concern about its potential adverse health effects. The Global Burden of Disease (GBD) 2015 assessment indicated that $PM_{2.5}$ contributed 4.24 million premature deaths around the world (Forouzanfar et al., 2016; IHME and HEI, 2017) and a number of epidemiological studies suggested that long-term exposure to high concentrations of $PM_{2.5}$ is positively associated with deaths from stroke, lung cancer (LC), chronic obstructive pulmonary disease (COPD) and ischemic heart disease (IHD) (Anderson et al., 2012; Dockery et al., 1993; Ren et al., 2006). For China, rapid economic development and urbanization over the past years have greatly damaged the environment (Lin et al., 2016b; Richter et al., 2005; Liu et al., 2016a, 2018) and Lin et al., 2016a revealed a significant association between $PM_{2.5}$ and mortality and estimated mortality burden of $PM_{2.5}$ in six Chinese cities. About 98% people are exposed to the severe $PM_{2.5}$ pollution that could not reach the World Health Organization (WHO) Air Quality Guidelines (AQG) of 10 μg/m$^3$ for annual mean (Apte et al., 2015). Therefore, it is extremely important to estimate China's premature deaths related by $PM_{2.5}$ for improving the

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control policy against air pollution.

Prior studies have suggested diverse results of premature deaths associated with $PM_{2.5}$ in China. For the year of 2010, 1.1 million (HEI, 2013), 1.28 million (OECD, 2014), 1.36 million (Lelieveld et al., 2015), 1.26 million (Xie et al., 2016) and 1.6 million (Rohde and Muller, 2015) of total premature deaths were estimated respectively. These distinctions are mainly caused by different selections of exposure assessment methods and concentration-response (C-R) functions, which are also the most significant parts of health effect assessments (Maji et al., 2017).

Most previous $PM_{2.5}$ ambient exposure estimation mainly depended on station monitoring interpolation method, satellite-based aerosol optical depth (AOD) model, air quality model and land use regression (LUR) model. However, each approach has somewhat limitations such as the mis-classification of interpolation method (Rohde and Muller, 2015), limited resolution of satellite device (Ma et al., 2016), huge computational cost of air quality model (Marshall et al., 2008) and space limitations of LUR (Johnson et al., 2010), restricting its application for predicting fine-scale air pollution concentrations over large geographical areas. More recently, a small number of LUR models combined with satellite estimation have been presented with broader spatial coverage, throughout United States (Novotny et al., 2011; Beckerman et al., 2013; Bechle et al., 2015), Europe (Beelen et al., 2009) and Australia (Knibbs et al., 2014). Knibbs et al., 2016 performed independent evaluations of national satellite-based LUR model in Australia and demonstrated its reliability. Combining the advantages of the satellite estimation and LUR models, national satellite-based LUR model for $PM_{2.5}$ is considered as a powerful tool to provide the accurate estimation of $PM_{2.5}$ exposure levels and its long-term health effect at a fine resolution in a large area (Novotny et al., 2011), but as far what we know, it has not been reported in China.

Another significant section of health effect assessment is the selection of C-R function. Previous studies have estimated premature mortality related by $PM_{2.5}$ with linear extrapolation of the C-R function which was announced in cohort studies (Lelieveld et al., 2013; Zhang et al., 2008). Later, the Integrated Exposure Regression (IER) was developed by Burnett et al. (2014) to produce a more reasonable prediction on relative risks by the integration of available information, including ambient air pollution (AAP) and second-hand smoke (SHS), and IER has been applied in a number of recent estimation of premature deaths related by PM2.5 (Liu et al., 2016b; Liu et al., 2017; Song et al., 2017; Xie et al., 2016; Wang et al., 2017). More recently, based on a large national cohort of 189,793 Chinese men, Yin et al., 2017 developed a Shape Constrained Health Impact Function (SCHIF), which could provide the exposure-response relationship between $PM_{2.5}$ and mortality over a much broader range of exposure than previously studied specifically at high PM2.5 concentrations. Yin et al., 2017 also showed that IER may underestimate the excess relative risk of $PM_{2.5}$ that exceeds the exposure range in China and other developing countries. Therefore, it is necessary to update the health burden attributable to $PM_{2.5}$ in China as well as quantitatively examine the impacts caused by various selections of C-R function.

In this study, Chinese national air monitoring network, satellite-derived estimation, wide collection of geographic and socioeconomic information were utilized to develop a national satellite-based LUR model for $PM_{2.5}$. Exposed levels across China within 2013–2015 were quantified with the fine resolution of 1 km × 1 km. Based on this long-term exposure, the estimation on premature mortality attributable to $PM_{2.5}$ in China was updated by using new SCHIF method, whose differences were analyzed quantitatively with the widely used IER method.

## 2. Data and methods

### 2.1. Data

#### 2.1.1. Monitor data

The $PM_{2.5}$ monitoring datasets were available from the China National Environmental Monitoring Center, available at: http://106.37.208.233:20035/. The measurements and quality control observed the regulations of Chinese national standards Ambient Air Quality Standards (GB3095-2012) and Ambient Air Quality Index (AQI) technology (HJ633-2012). In this study, $PM_{2.5}$ concentrations from all available national sites on January 1, 2013 to December 31, 2015 were collected hourly from stations that more than 75% of data integrity. Fig. S1 and Table S1 show the distribution of monitoring stations and descriptive statistics of selected samples. In the period of 2013–2015, available stations increased and the $PM_{2.5}$ concentration decreased sharply.

#### 2.1.2. Predictor variables

Land Use Regression proposed by Briggs et al. (1997) is an empirical statistical method, which combines air quality monitoring measurements and surrounding relevant information to estimate pollutants exposure for locations without measurements. In this study, several types of predictor variables were collected, including satellite estimation, geographical position, emission inventory, road network, socioeconomic indicators, land use and meteorological condition, as shown in Table S2 and the details. The variables were chosen in accordance with available data in China and previous satellite LUR models (Kloog et al., 2014; Knibbs et al., 2014; Yang et al., 2017). A group of variables were extracted from station location (point type) and the others were derived from the buffers-centered monitoring site (buffer type) with the radius from 100 m to 10 km (buffer distances are listed in Table S1). As a result, 244 variables (10 × 22 buffer variables and 14 point variables) were selected to promote a national LUR model.

#### 2.1.3. Population data and baseline mortality

The Chinese population data with the 1 km resolution were derived from the 6th National Population Census and projected it to 2013–2015 based on the provincial population of National Bureau of Statistics of China (Yang et al., 2009; NBSC, 2013, 2014, 2015). Provincial baselines for 2013–2015 disease-specific mortality of stroke, IHD, COPD, LC were gained from Zhou et al. (2016) and the dataset of GBD study (http://vizhub.healthdata.org/gbd-compare/).

### 2.2. Methods

#### 2.2.1. LUR modelling approach

The two-procedure LUR building approach was adopted, consisting of Lasso variable selection (Tibshirani, 1996) and multiple stepwise forward regression (Su et al., 2009). This approach was successfully utilized by Knibbs et al. (2014) recently, and utilized to build national and global air pollution exposure models by Larkin et al. (2017).

Since the study time was selected as 2013–2015, the single-year and multi-year models were developed. For single-year model, Lasso regression was first opted for a subset of initial 212 generated variables for the next stage. It places a bound on the sum of absolute coefficient values, minimises the sum of squared errors and could preliminarily select the subset of the variables and reduce the subsequent computational complexity. In the second procedure, the independent variable, which was most correlated with the dependent variable, was added to the single-year model first; then the independent variable, which was most correlated with model residuals, was selected to join the model; and the procedure was repeated till the variable could not satisfy the mentioned conditions. The included variables in the final model must meet these criteria for improving model's accuracy and interpretability. (1) The statistical significance is below 0.05. (2) The variance inflation factor (VIF) of all variables included in the model should be less than 3 to avoid the collinearity. (3) The variable can increase adjust-$R^2$ of the model more than 1%. Models with incremental buffer sizes of the same land use characteristic were reduced to only include the smallest buffer size. For multi-year model, an time factor was added to model, which was defined by −1 for 2013 year, 0 for 2014 year and 1 for 2015 year. Lasso method was adopted for each year, once the variable was selected, it could enter the next step. The second procedure was similar to the foregoing process except for an extra time factor. In addition, 10-fold cross-validation was used to test model's robustness. 10% of the monitoring data was selected randomly into a testing data set, and the rest of 90% was used to constitute the training data set. The two steps were repeated for 1000 iterations to generate the cross-validation estimates of $R^2$, mean absolute error (MAE) and mean absolute bias (MAB). To analyze the performance improvement of LUR and satellite estimation, two auxiliary models were added, namely the model of forcibly excluding satellite estimation variable and the model of adding a single variable. The final national satellite-based LUR model would be applied to create $PM_{2.5}$ distribution mapping within 2013–2015 with 1 km spatial resolution to match gridded population, and in our study, spatial smoothing approach was not used for residuals because of the sufficient accuracy (indicated by $R^2$) for assessment requirements.

#### 2.2.2. $PM_{2.5}$-related premature deaths calculation

Disease-specific premature mortality for stroke, COPD, IHD and LC related by $PM_{2.5}$ was estimated in each 1 km grid on the basis of the widely used general equation (1) (Song et al., 2017; Liu et al., 2016b):

$$\Delta M = P \times \left(\frac{RR-1}{RR}\right) \times IN, \tag{1}$$

where ΔM is the mortality related by $PM_{2.5}$ which is caused by a given specific disease in each 1 km grid. P is the exposed population and IN represents the provincial baselines for disease-specific mortality. RR is the relative risk for each end point at a certain concentration decided by C-R function. As mentioned above, IER function developed by Burnett et al. (2014) is broadly used in recent estimation (Song et al., 2017) of premature mortality related by $PM_{2.5}$. Yin et al., 2017 used an integrated modelling framework SCHIF in which a class of flexible algebraic C-R function was fit survival models using standard computer software, and constructed the class by defining transportation of concentration as the product of either a linear or log-linear function of concentration multiplied by a logistic weighting function (Nasari et al., 2016). Equation (2) represents the form of IER function and equations (3) and (4) represent the form of SCHIF function. Here we have an equivalent assumption: The population is in a relatively stable exposure condition and the annual average concentration can reflect the state of the population throughout the exposure process.

$$R(\beta, z) = \begin{cases} 1, \text{ if } z < z_0 \text{ otherwise} \\ 1 + \beta_1 \times \left(1 - e^{-\beta_2 (z - z_0)^{\beta_3}}\right), \end{cases} \tag{2}$$

where z is the annual average ambient PM2.5 concentration; $z_0$ is threshold concentration from 5.8 to 8.0 $\mu g/m^3$, $\beta_1$, $\beta_2$ and $\beta_3$ are parameters used to describe the different shapes of the exposure-response curve among various diseases (Burnett et al., 2014)

$$R(t|z) = R_0(t)\exp\{\beta \times \omega(z|\mu, \tau) \times f(z)\}, \tag{3}$$

$$\omega(z|\mu, \tau, r) = \left\{1 + \exp\left(-\left(\frac{z-\mu}{\tau \times r}\right)\right)\right\}^{-1} \tag{4}$$

where $R_0(t)$ is the baseline hazard function of follow-up time t, f is a known parametric function of air pollution concentration z. $\omega(z|\mu, \tau, r)$ is a known weighting function indexed by scalar value μ,τ and r, with β an unknown parameter to be estimated from the survival data using standard computer software. Two forms of f were considered in the study: f(z) = log(z) and f(z) = z. (Nasari et al., 2016)

In this study, $PM_{2.5}$ exposure above and new SCHIF method were used to estimate the premature mortality attributable to $PM_{2.5}$ in China, whose results were also compared with IER method and other researches.

## 3. Results

### 3.1. National LUR model

Table S3 and Table 1 show the performance and structure of single-year and multi-year satellite-based model, whose variables are listed in the descending order of the importance by $R^2$ reduction without this variable. The similar values of $R^2$ ranging from 0.67 to 0.70 are included in the three single-year models, whose structures have certain similarities. For example, in each model, all the satellite estimation variable plays the most important roles and the wind speed variable is kept in the final model. Through the multi-year model instead of three single-year models, the performance of models is improved and a lot of computation in the subsequent application is saved. As for multi-year model, $R^2$ is 0.72, MAE is 9.17 $\mu g/m^3$, MAB is 15.7%. The p-value is less than 0.001, which reflects the statistical significance in this model, and all the VIFs are less than three, which reflects there is no multicollinearity problem. A total of 5 variables are selected into the final LUR model. The satellite-based estimation provides the most significant improvement in model performance as other previous national LUR studies (Beckerman et al., 2013). The structure and performance of models without satellite estimation are presented in Table S4, and a simple model with only satellite estimation in Table S5. The values of $R^2$ in these models, which are relatively lower, are 0.53 and 0.48 respectively, corroborating that satellite-based LUR model can provide more accurate estimation of PM2.5 estimation than single satellite estimation or ordinary LUR model (Knibbs et al., 2014). The meteorological factor of wind speed and precipitation variables both have a negative coefficient, probably arising from that increasing wind speed is a better condition for pollutants to diffuse and rainfall can scour particular matters in the air to decrease the $PM_{2.5}$ concentration (Meng et al., 2015). The factor of time which has a negative coefficient in the final model, is also taken into consideration. During this study period, the sharp decreases of $PM_{2.5}$ is partially caused by the strict control policy in that year. In addition, the unique variable of land use in this model is impervious land in proportion with 800 m buffer, and its negative coefficient likely demonstrates that more transportation or the other source of particulate precursors will increase $PM_{2.5}$ concentration. The resolutions of the variables retained in the model are 10 km (satellite estimates), 1 km (wind speed and precipitation) and 30 m (Impervious_600 m) (Table S2). Compared with the single satellite model, more high resolution data in our model can improve the resolution of prediction. Other variables such as road length, geographical position are not included in this model, probably because these

**Table 1**
Multi-year national satellite-based LUR model's structure and performance.

| Model Performance | Variable | Unit | B[a] | SE[b] | P | % $R^2$ reduction | VIF [c] |
|---|---|---|---|---|---|---|---|
| adjust $R^2 = 0.72$ | Intercept | $\mu g/m^3$ | 40.16 | 4.08 | <0.001 | | |
| MAE = 9.17 $\mu g/m^3$ | satellite estimation | $\mu g/m^3$ | 0.5 | 0.01 | <0.001 | 14.9 | 1.76 |
| MAB = 15.7% | Wind speed | m/s | −13.22 | 0.62 | <0.001 | 5.3 | 1.27 |
| CV[d] adjust $R^2 = 0.70$ | Year | | −5.61 | 0.32 | <0.001 | 3.9 | 1.27 |
| CV MAE = 9.88 $\mu g/m^3$ | Precipitation | mm | −3.55 | 0.26 | <0.001 | 2.5 | 2.03 |
| CV MAB = 16.9% | Impervious_800 m | % | 13.4 | 1.32 | <0.001 | 1.8 | 1.68 |

[a] Coefficient values of variables.
[b] Standard error of variables.
[c] Variance inflation factor.
[d] Cross validation.

factors don't have a more significant relationship with $PM_{2.5}$ throughout the whole China or our data are not accurate enough. The cross validation (CV) $R^2$, CV-MAE, CV-MAB are 0.70, 9.88 $\mu g/m^3$ and 16.9% separately. The cross validation, which is resulted from the relatively smaller reduction of model performance, may suggest the robustness of our model. The predicted and observed values of all samples are provided in Fig. 1, showing that most points are distributed around the Y = X line, but a small part of points have big residuals which mainly come from 2013 year dataset due to existences of some extremely high concentration points in that year. There is no effective way to fully simulate this highly polluted condition now (Ma et al., 2016), and subsequent calculation of premature deaths related by PM2.5 may not be greatly affected because both SCHIF and IER have a stable curve at this concentration interval.

### 3.2. $PM_{2.5}$ exposure in China

Fig. 2 shows the spatial pattern of $PM_{2.5}$ exposure concentration across China and the reduction from 2013 to 2015. Geographically weighted annual average concentrations are 55.72 $\mu g/m^3$, 47.15 $\mu g/m^3$, 40.24 $\mu g/m^3$ for 2013–2015 respectively. Concentrated areas of industrialization or urbanization with high $PM_{2.5}$ concentrations include Beijing-Tianjin-Hebei (BTH) region, the Pearl River Delta, the Yangtze River Delta and Sichuan Basin. Another polluted area of high $PM_{2.5}$ is Taklamakan Desert in Xinjiang, owing to a major dust source. Although the overall $PM_{2.5}$ concentration decreases significantly, there are some differences in air quality improvement

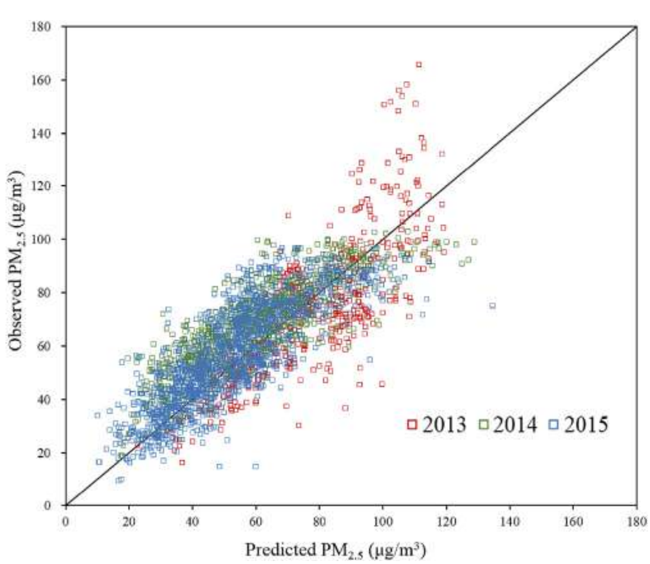


**Fig. 1.** The predicted and observed values of all $PM_{2.5}$ samples.

between regions. As shown in Fig. 2d, some regions like BTH and Shandong province have relatively less reduction of $PM_{2.5}$ concentrations than those in most other regions, which implies more strict pollution control measures should be taken in these regions and its surrounding areas.

Population weighed $PM_{2.5}$ concentrations are calculated to make a comparison with the study of other years conducted by Liu et al. (2017). Fig. 3 shows population weighted $PM_{2.5}$ concentration variety within 8 years and our study period is distinguished with a rapid annual reduction of 8.49 $\mu g/m^3$ and 6.85 $\mu g/m^3$. In contrast, it only reduces 3.13 $\mu g/m^3$ within five years (2008–2012). It is worth noting that the State Council of China released “Air Pollution Prevention and Control Action Plan (Action Plan)” in 2013 (Zheng et al., 2017) to improve air quality and protect public health. Thus a lot of strict air control measures have been employed to achieve this target. Moreover, population weighted concentrations are approximately 17 $\mu g/m^3$ higher than geographically weighted concentrations, reflecting that the pollution has a larger distribution in densely populated areas. As shown in Fig. 3, approximately 94.27%, 89.13%, 84.66% of population (Cumulative distribution is shown in Fig. S2) in 2013–2015 are separately exposed to $PM_{2.5}$ concentrations, exceeding the National Ambient Air Quality Standard (NAAQS) of 35 $\mu g/m^3$. This remarkable reduction may support the significant effect of the Action Plan.

### 3.3. Premature deaths related by $PM_{2.5}$ in China

Premature deaths related by $PM_{2.5}$ of four specific diseases (stroke, COPD, LC and IHD) are evaluated, and the spatial pattern of total deaths related by $PM_{2.5}$ is shown in Fig. 4, which follows the population density pattern with correlation coefficients of 0.96 in 2013, 0.94 in 2014 and 0.91 in 2015. The correlation coefficients between $PM_{2.5}$ exposures and premature deaths are 0.21, 0.22, 0.23 respectively, but the center of mortality is similarly distributed in industrialized urbanization area, including BTH region, the North China Plain, the Pearl River Delta, the Yangtze River Delta and Sichuan Basin. The data highlights the urgency of China's efforts to control air pollution in these regions. As for reduction of premature deaths owing to air pollution improvement (Fig. 4d), there also exist differences between different regions. For example, Sichuan Basin has a larger reduction of premature mortality than that in BTH where undertakes the similar high burden of diseases.

Provincial disease-specific deaths information is provided in Table S6. Fig. 5 shows the total mortality quantity by descending order. The total numbers of premature mortality are 2.19 million in 2013, 1.94 million in 2014 and 1.65 million in 2015, respectively. It is noteworthy that total premature deaths decreases 0.24 million from 2013 to 2014 and 0.29 million from 2014 to 2015, and corresponding reductions of $PM_{2.5}$ population weighted exposure are 8.49 $\mu g/m^3$ and 6.85 $\mu g/m^3$ respectively. The phenomenon that less exposure reduction brings about more premature deaths reduction

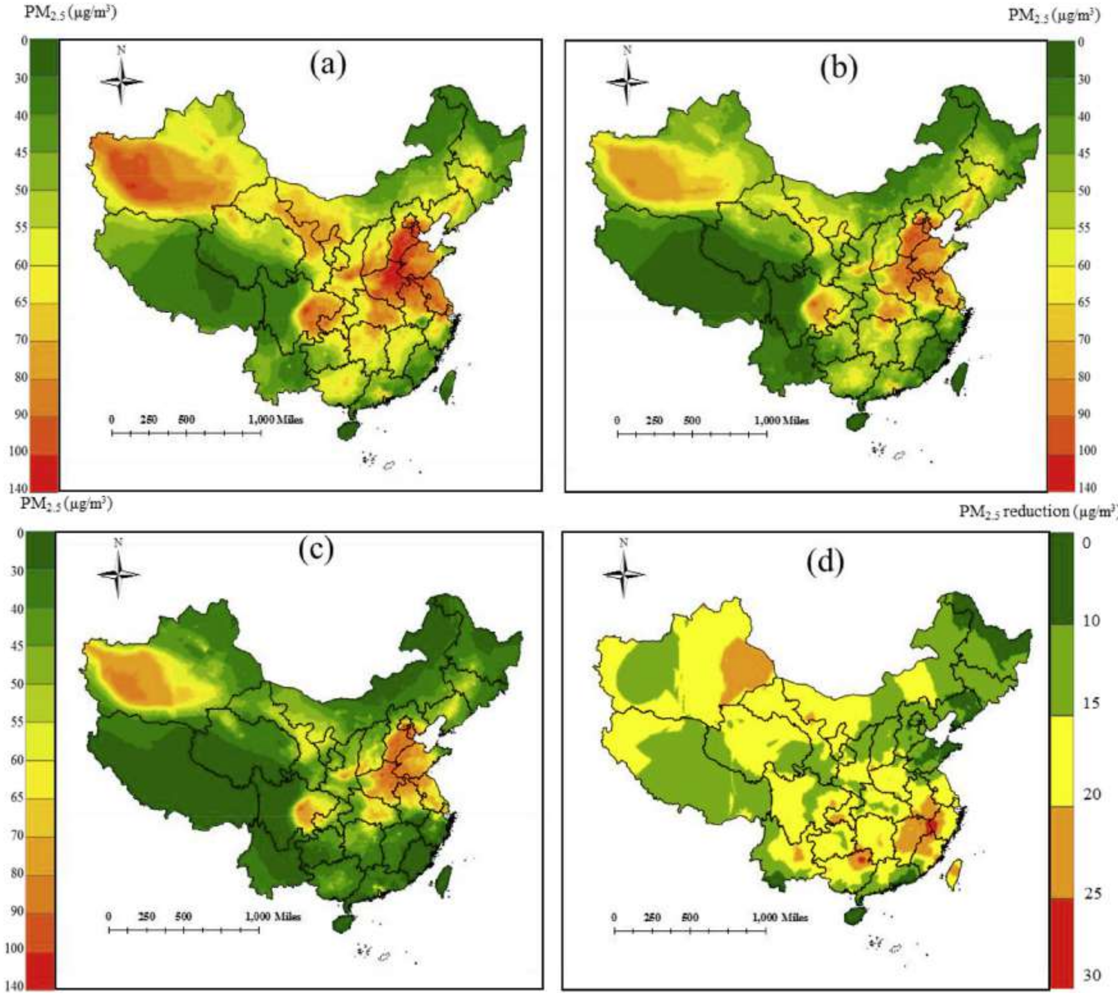


**Fig. 2.** Spatial pattern of $PM_{2.5}$ exposure concentration across China in 2013 (a), 2014 (b), 2015 (c) and reduction from 2013 to 2015 (d).

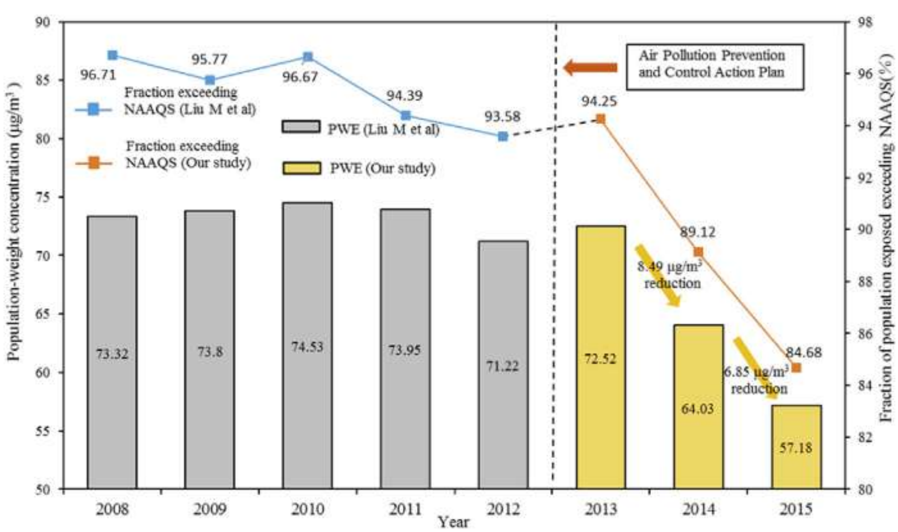


**Fig. 3.** $PM_{2.5}$ population-weight concentration and population fraction of living in areas that exceed NAAQS in China from 2008 to 2012 (Liu M et al.) and 2013–2015 (our study).

is mainly caused by nonlinear C-R function with a gentle curve at high concentration interval and steep curve in the middle concentration interval. It also implicates that if China continues its stringent emission control strategy, the more significant health benefits could be achieved than the past years. The top three provinces of the highest premature deaths are Henan, Shandong, Sichuan, while the least ones are Tibet, Hainan, Qinghai. Fig. 5a shows that total premature deaths of different provinces. There are

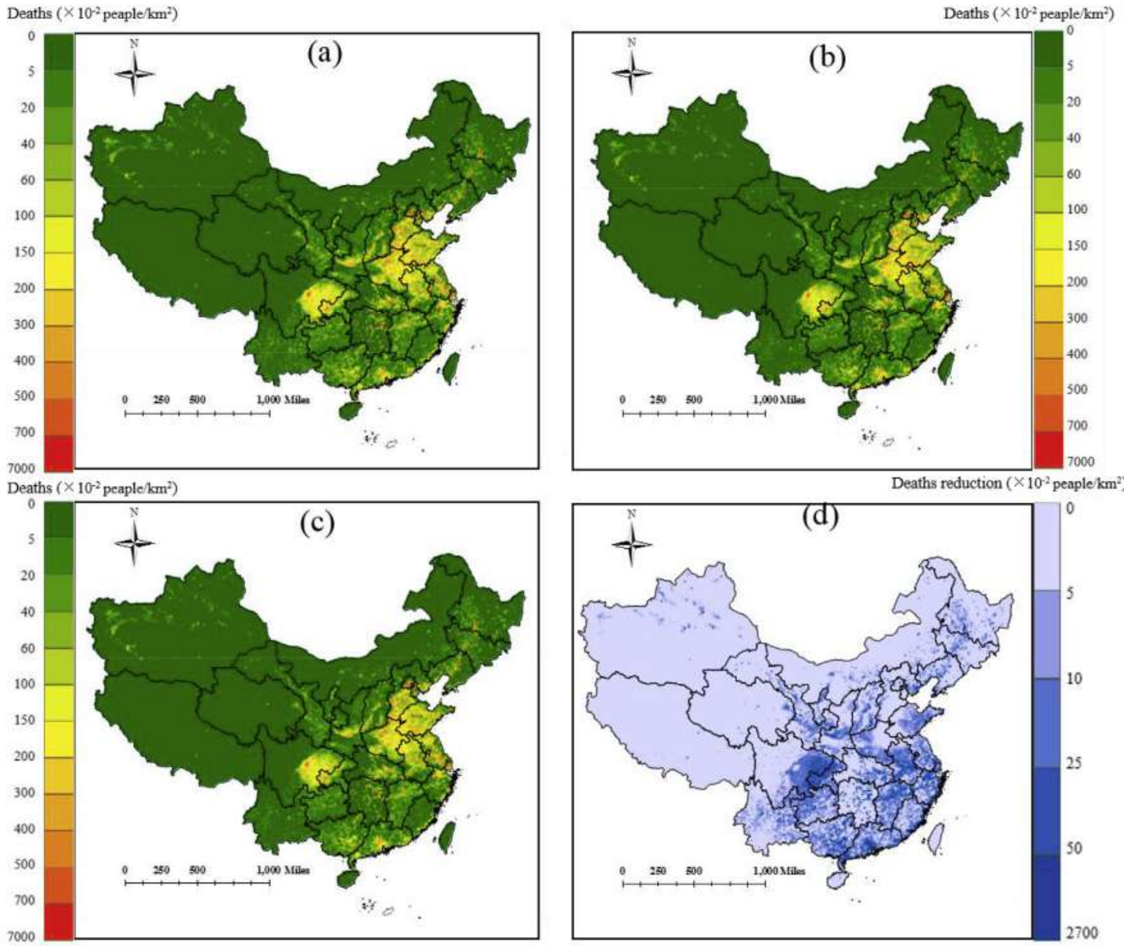


**Fig. 4.** Spatial pattern of $PM_{2.5}$-related premature deaths across China in 2013 (a), 2014 (b), 2015 (c) and reduction from 2013 to 2015 (d).

diverse variation trends in study period with only 1.7% reduction in Tianjin and 3.6% reduction in Beijing, but 84% reduction in Tibet and 73% in Fujian, indicating that the situation of mortality attributable to $PM_{2.5}$ in some economically developed areas had not been improved effectively as the above analysis. As for different diseases (Fig. 5b), the average annual premature deaths during the three-year period are 1.02 million (53.2%) for stroke, 0.39 million (20.5%) for IHD, 0.32 million (16.8%) for COPD and 0.18 million (9.5%) for LC. Fig. 5b also shows that stroke is the first factor of premature mortality in all provinces, while the three following factors have some differences. IHD has a larger proportion of total deaths than COPD in most provinces, while these conclusion is quite contrary in Sichuan, Chongqing, Guangxi and etc. Analogously, LC has the minimum proportion of total deaths in most provinces, while it is opposite in Liaoning and Heilongjiang. This distinction may be caused by the difference of $PM_{2.5}$ exposure and C-R function equation for each disease.

## 4. Discussion

In this study, $PM_{2.5}$ exposure and relevant premature deaths were estimated across China within 2013–2015 with 1 km resolution, by the use of national satellite-based LUR model and new SCHIF function based on a large national cohort of 189,793 Chinese men. There are several important distinctions between this analysis and previous studies:

As far what we know, national satellite-based LUR model for China's $PM_{2.5}$ of high pollutant concentration has not been reported so far. Recent national LUR studies have paid more attention to $NO_2$, which is a strong marker of traffic and other combustion derived pollution (Richter et al., 2005). However, the relevant model for $PM_{2.5}$ has not performed an ideal effect relatively, probably because secondary pollutants were generated through a complex chemical reaction without direct emission resource or other geographical information. For example, Hystad et al., 2011 developed a national LUR model in Canada, which combined with literature-derived assumptions of near-roadway trends and adjust-$R^2$ for $PM_{2.5}$ was 0.46 in their model. Beckerman et al., 2013 used a hybrid model combined with LUR, Bayesian maximum entropy interpolation and normalized $R^2$ of their LUR model with remote sensing of 0.46. These studies both used supplemented method to improve the model performance, and Beckerman et al., 2013 demonstrated that the single satellite-based LUR model is insufficient to describe the complex spatio-temporal variability in ambient $PM_{2.5}$ at the national scale. In this study, however, the great model performance with CV-adjust $R^2$ 0.70 may prove that this national LUR model could be used to assess $PM_{2.5}$ exposure variability in China. These improvements come from several aspects: First, since 2015, there has been at least 1457 national monitoring stations available in China, and this development may effectively improve national LUR model performance. Secondly, previous research demonstrated that there is a critical need to establish an extensive monitoring network to obtain a robust LUR (Johnson et al., 2010) and Hystad et al., 2011 illustrated that Canadian ambient monitors (177

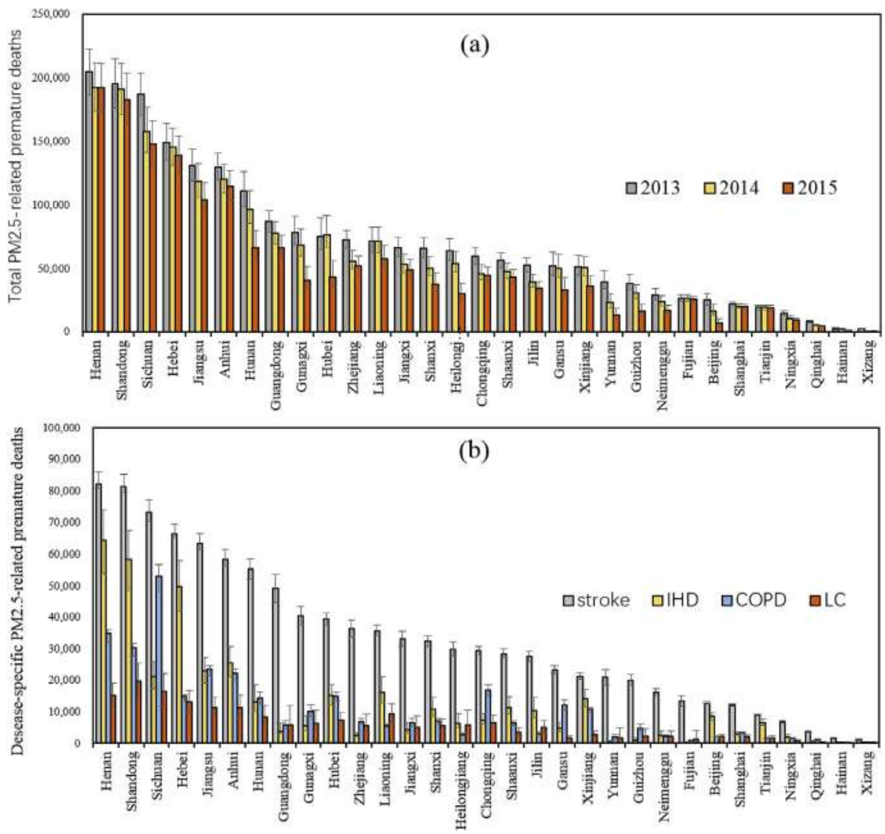


**Fig. 5.** $PM_{2.5}$-related premature deaths of each province (a) Total number in 2013–2015. (b) Average disease-specific number.

monitors for $PM_{2.5}$) are too sparse to derive a straightforward national-scale LUR model. Secondly, the wide selection of predictor variables includes various factors of meteorological information and the time. The value of $R^2$ of this LUR model without satellite was 0.53 (Table S4) and five variables were selected in the final model. However, LUR model without remote sensing in the contiguous United States (Beckerman et al., 2013) just contained two variables and its $R^2$ in the training data set was only 0.08.

As mentioned above, selections of exposure assessment methods and C-R functions are the most important sections in health effect assessment. In order to effectively evaluate the impact of these two elements, IER method was also used to calculate premature deaths related by $PM_{2.5}$, and estimations by SCHIF and IER method were compared with other studies that adopted IER function in the same study year (Fig. 6a). For different selections of C-R function, the results of total numbers by IER were 1.70 million for 2013, 1.59 million for 2014 and 1.49 million for 2015, which were significantly lower than that calculated by SCHIF with less amounts of 0.52 million, 0.34 million and 0.15 million within 2013–2015 respectively. It indicates that IER function has a more underestimation at higher $PM_{2.5}$ concentration range. As air quality improving with implementation of the stricter policy in China, the difference between using IER and SCHIF will become smaller, but this new function SCHIF will play an important role in the long term for other developing regions in the world. Another advantage of SCHIF is the smaller uncertainty and the 95% confidence interval (CI) of total premature deaths using IER method is almost two times wider than that using SCHIF. As for the influence of different $PM_{2.5}$ exposure methods, researches on 2014 and 2015 conducted by Rohde and Muller, 2015 and Song et al., 2017 respectively showed the similar results with this IER-based estimation, but the result

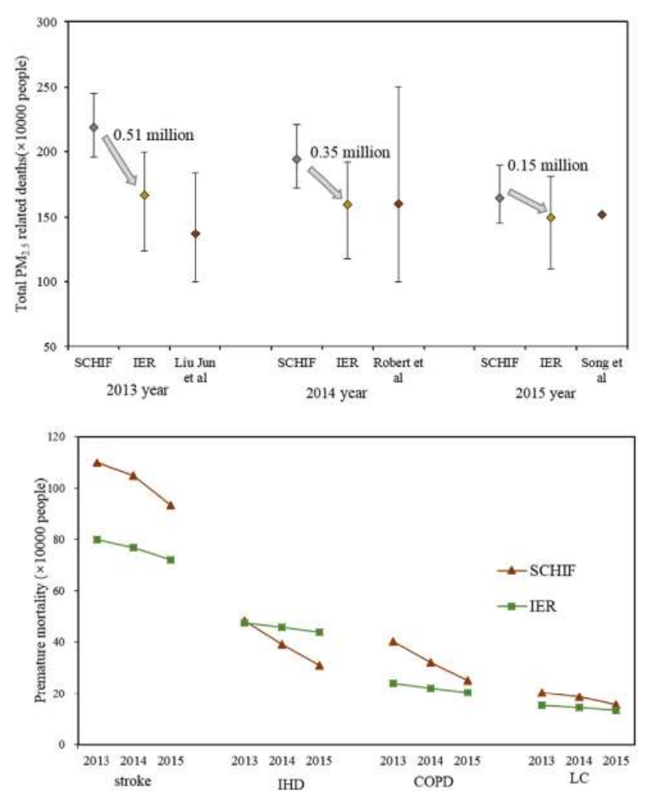


**Fig. 6.** Comparison of premature deaths by different methods. (a) Total numbers of this SCHIF and IER as well as other studies. (b) Disease-specific results of this SCHIF and IER.

from Liu et al. (2016b) in 2013 (1.37 million, 95% CI: 0.89–1.74 million) is lower than this result (1.70 million, 95% CI: 0.75–2.25million). The distinction can be explained by different $PM_{2.5}$ exposure estimations, in which 83% (Liu et al., 2016b) and 94% (this study) of the population in 2013 were separately exposed to $PM_{2.5}$ concentrations exceeding the NAAQS. It is widely recognized that existing studies of Chinese ambient $PM_{2.5}$ concentration do not always agree with each other (Xie et al., 2016). For example, according to Ma et al. (2016), van Donkelaar et al., 2015 might underestimate the $PM_{2.5}$ concentration across China, especially in heavily polluted and high populated regions, and the population weighted concentrations in China for 2010 are 56.5 $\mu g/m^3$ (van Donkelaar et al., 2015)and 74.53 $\mu g/m^3$ (Ma et al., 2016; Liu et al., 2016b). Our result agrees better with Ma et al., 2016, which implies that national model may be more suitable to estimate China's $PM_{2.5}$ exposure. In addition, the resolution of Liu et al. (2016b) is 45 km $\times$ 45 km and it is too coarse to capture $PM_{2.5}$ variation in the urban area, which may lead to the possible misclassification. Resolution of most recent research was $0.1^\circ \times 0.1^\circ$ or larger (Xie et al., 2016; Apte et al., 2015), suggesting the 1 km fine resolution has a certain breakthrough in this study. Disease-specific total deaths by two methods are shown in Fig. 6b and Table S7, indicating certain differences in disease attribution. The average annual premature moralities by IER method are 0.76 million (48.13%) for stroke, 0.46 million (28.82%) for IHD, 0.22 million (13.99%) for COPD and 0.14 million (9.06%) for LC respectively, which shows similar distribution with Wang et al., 2017 (54.90% for stroke, 30.77% for IHD, 9.40% for COPD and 6.62% for LC). Nevertheless, the estimation used by SCHIF shows a bigger proportion for COPD (16.80%) and a smaller proportion for IHD (20.47%). The underestimation by IER mainly comes from stroke and COPD, but as for IHD-caused premature deaths, the result calculated by IER is even bigger (Fig. 6b). But it's worth noting that the SCHIF was based on a cohort study of 189,793 men 40 years old or older during 1990-90 from 45 areas in China and the IER was fitted by integrating available RR information from many cohort studies in western Europe and North American. Although SCHIF seems to be closer to the actual exposure situation in China, it is hard to say that SCHIF can more accurately calculate the PM2.5-related premature deaths in China, because the PM2.5 long-term health effect of different individuals are different.

Although certain innovations have been made in this research, there are still several limitations in ecological analysis, which should be addressed in the further study. First, although $PM_{2.5}$ exposure and population distribution with the fine resolution of 1 km $\times$ 1 km were used, the baseline mortality data in this study was provincial-level and more elaborate data has not yet been available in China. While the mortality baseline is quite different from city to city (Maji et al., 2017), it is necessary for China to systematically collect and make more city-level mortality baseline available. Besides, the effect of $PM_{2.5}$ chemical composition, size distribution and sources were neglected in this paper, which may bias the estimates of mortality burdens (Ostro et al., 2015). For instance, IHD risk for coal combustion $PM_{2.5}$ is roughly five times higher than that for $PM_{2.5}$ mass in general (Thurston et al., 2016). However, current scientific evidence is unable to accurately estimate the relative risk of various constituents, chemical forms or sources of ambient $PM_{2.5}$ (Arnold, 2014). Besides, current epidemiologic studies are still based on long-term exposure to $PM_{2.5}$ total mass. Moreover, Yin et al., 2017 included only male participants for investigating the effect of smoking, and did not adjust for multiple pollutants and some area-level socioeconomic status (SES) variables, and the C-R function based on it only provides the overall statistical results instead of classification results such as race, age, gender, geographical locations, exposure assessment approaches and years, which should be improved by more classified cohort studies of long-term adverse effects of ambient $PM_{2.5}$ in China or other developing regions. Additionally, due to data availability, we did not assess the exposure change caused by the flow between populations. Lastly, annual premature deaths within three years were calculated in this study only, but this adverse effect could be accumulated for a longer period of time. Therefore, the long-term cumulative effects of $PM_{2.5}$ should be further estimated in follow-up studies.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2018.09.089.

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