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Assessment of short-term $PM_{2.5}$-related mortality due to different emission sources in the Yangtze River Delta, China

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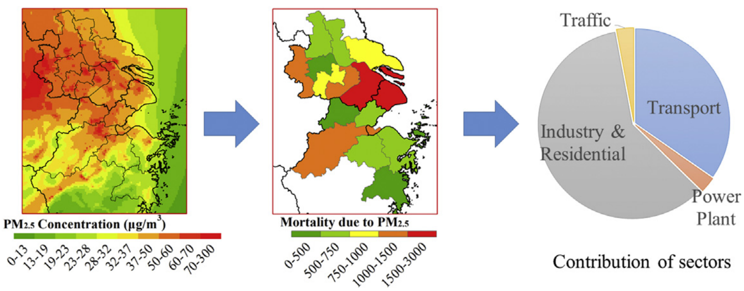
HIGHLIGHTS

- Short-term mortality due to $PM_{2.5}$ in YRD is estimated to be 13,162 in 2010.
- The economic loss due to $PM_{2.5}$ is 22.1 billion Chinese Yuan.
- The industry and residential sectors account for over 50% of the damages.
- The contribution of different air pollutant emissions varies with seasons.

GRAPHICAL ABSTRACT



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ABSTRACT

Air pollution is a major environmental risk to health. In this study, short-term premature mortality due to particulate matter equal to or less than 2.5 μm in aerodynamic diameter ($PM_{2.5}$) in the Yangtze River Delta (YRD) is estimated by using a PC-based human health benefits software. The economic loss is assessed by using the willingness to pay (WTP) method. The contributions of each region, sector and gaseous precursor are also determined by employing brute-force method. The results show that, in the YRD in 2010, the short-term premature deaths caused by $PM_{2.5}$ are estimated to be 13,162 (95% confidence interval (CI): 10,761-15,554), while the economic loss is 22.1 (95% CI: 18.1–26.1) billion Chinese Yuan. The industrial and residential sectors contributed the most, accounting for more than 50% of the total economic loss. Emissions of primary $PM_{2.5}$ and NH_3 are major contributors to the health-related loss in winter, while the contribution of gaseous precursors such as SO_2 and NO_x is higher than primary $PM_{2.5}$ in summer.

1. Introduction

Located in the eastern part of China, the Yangtze River Delta (YRD) is the largest estuary delta in China. It covers the municipality of Shanghai, southern Jiangsu Province, and eastern and northern Zhejiang Province. As one of the most densely populated

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regions of China, the YRD contains 107 million people with an area of 213 thousands square kilometers. Accompanied with a rapid economic development and a dramatic industrial expansion in the past two decades, the YRD has experienced a sharp increase in energy consumption and air pollutants emissions, which leads to serious air pollution problem. In 2010, the average emission intensity in the YRD for SO_2, NO_X, particulate matter equal to or less than 10 μm in aerodynamic diameter (PM_{10}), particulate matter equal to or less than 2.5 μm in aerodynamic diameter ($PM_{2.5}$), non-methane volatile organic compounds (NMVOCs) and NH_3 were 2–7 times higher than the national average (Fu et al., 2013). In 2013, the annual average concentrations of $PM_{2.5}$ in Shanghai, Nanjing and Hangzhou were 56 μg/m^3, 75 μg/m^3 and 64 μg/m^3, respectively (Wang et al., 2014c), which were 5–7 times of the World Health Organization Air Quality Guidelines (WHO, 2005) and far above the current annual air quality standards of 35 μg/m^3 (GB 3095-2012). A significant decrease in visibility was evident judging from historical record (Chang et al., 2009; Cheng et al., 2013a; Gao et al., 2011). Since 2010, the YRD has been identified as a key area for joint prevention and control of air pollution, which is an important air pollution control plan for China (Wang and Hao, 2012). The PM pollution is a key air pollution problem in the YRD, leading to adverse health impacts.

Several studies have evaluated the health loss due to PM on national or regional scale in China. The Global Burden of Disease 2010 used integrated exposure-response model and estimated that the number of deaths attributable to ambient $PM_{2.5}$ was 3.2 million worldwide and 1.2 million in China in 2010 (Burnett et al., 2014; Lim et al., 2012). Monitoring data are limited before the China National Urban Air Quality Real-time Publishing Platform was introduced publically in January, 2013 (Jiang et al., 2014). Most studies used PM_{10} and total suspended particles (TSPs) to estimate health impact on exposure to PM, or estimated effect of $PM_{2.5}$ with the derivative coefficient from the epidemiological reference linking the mortality and PM_{10}. On national scale, Cheng et al. (2013b) estimated the PM_{10}-related premature deaths increased from 418,000 to 514,000 from 2001 to 2011. The World Bank (2007) estimated the health cost of urban PM pollution in China in 2003 to be 157 billion Chinese Yuan by using adjusted human capital (AHC) method and 520 billion Chinese Yuan by using the value of statistical life (VSL) method. In addition to these national air pollution studies, the total health loss due to PM pollution on a region or city scale has also been estimated (Huang et al., 2012a; Tang et al., 2014; Zhang et al., 2007).

Epidemiologic studies in the United States and worldwide have demonstrated more robust associations with fine particular matters. Pope and Dockery (2006) reviewed the progress in the evaluation of PM health effects. In the United States (US), $PM_{2.5}$ air pollution and mortality were linked in the often cited Six Cities Study, where an association was reported with lung cancer- and cardiopulmonary disease-related mortality (Dockery et al., 1993). Pope et al. (1995) reported an association between $PM_{2.5}$ and all cause, cardiopulmonary, and lung cancer mortality. In China, substantial progress has been made on epidemiological studies since 2005. Several short-term studies were done in recent years (Cao et al., 2012; Guo et al., 2009; Huang et al., 2012b; Kan et al., 2007; Shang et al., 2013; Venners et al., 2003; Yang et al., 2012), including the systematic multi-cities research, “China Air Pollution and Health Effects Study” (CAPES) (Chen et al., 2011). Impacts of long-term exposure to $PM_{2.5}$ are limited in China. Together with the limit of observational data, there are still only a few studies assessing the premature mortality caused by $PM_{2.5}$ with Chinese local concentration-response (C-R) coefficient.

Emission reductions in different sectors may have different levels of effectiveness on reducing human exposure (Li et al., 2004; Streets et al., 1999). Besides, the purpose of establishment and revision of ambient air quality standards is to protect public health and welfare. Therefore, evaluation of health impact and the economic loss due to emissions of various pollutants and different sectors could potentially help for future pollution control. Fann et al. (2009, 2012) assessed the human health impacts and monetized benefits due to different emission sectors by applying the Response Surface Model (RSM) and CAMx. In China, Zhou et al. (2010, 2014) discussed the contribution of and NOx emissions of different sectors under different emission control scenarios. However, the researches on contribution of different gaseous precursors and total contribution of one sector are still lacked.

In this study, Environmental Benefits Mapping and Analysis Program Community Edition (BenMAP CE) with China's local epidemiological studies and health data is employed to estimate the premature mortality caused by $PM_{2.5}$ in the YRD region in 2010. The spatial and temporal distribution of $PM_{2.5}$ concentrations are simulated by Community Multi-scale Air Quality (CMAQ) modeling system. Source apportionment was conducted on the causes of $PM_{2.5}$–related premature mortality. The contributions of different regions were evaluated and the contribution of different sectors and gaseous precursors were also assessed. The results will help prioritize future pollution control strategies among the different regions, sectors and gaseous precursors in the YRD.

2. Materials and methods

2.1. $PM_{2.5}$ modeling system and emission inventory

To get the $PM_{2.5}$ concentration for health assessment, Weather Research & Forecasting Model (WRF) - CMAQ modeling system is utilized in this study. WRF version 3.3 is applied to generate the meteorological fields for CMAQ. The spatial distributions and temporal variations of $PM_{2.5}$ are simulated by the CMAQ of version 4.7.1. Triple nesting simulation domains are employed in this study, as shown in Fig. 1. Domain 1 covers most of China with 36 km*36 km resolution, Domain 2 covers eastern China with 12 km*12 km resolution, and Domain 3 covers the YRD region with 4 km*4 km resolution, respectively. The vertical resolution includes 14 layers from the surface to the tropopause, consistent with our previous work (Wang et al., 2010b). A combination of a few emission inventories developed by Tsinghua University is used as the input of the modeling system. The anthropogenic emissions inventory for Domain 1 and Domain 2 is from Wang et al. (2014b), Zhao et al. (2013a, 2013c). A high-resolution anthropogenic emission inventory for the YRD region from Fu et al. (2013) is used in Domain 3. The CMAQ model, which is developed by U.S. Environmental Protection Agency (EPA), has been tested, evaluated, and applied in China (Wang et al., 2010b; Zhao et al., 2011, 2013a). The simulation periods are selected as January, May, August and November, representing the average concentration in each season. The annual average $PM_{2.5}$ is represented by the average concentration of these four months. The configurations of chemical initial conditions and boundary conditions are consistent with our previous papers (Wang et al., 2014a; Zhao et al., 2014) and summarized in SI. The simulated meteorological parameters and PM concentrations have been evaluated by comparison with observational data, which indicated that the model can well capture the temporal trend and spatial distribution. The detailed methods and results of validation were reported in our previous work (Zhao et al., 2014) and shown in Supplemental Information (SI, Table S1 to S4 and Figure S1).

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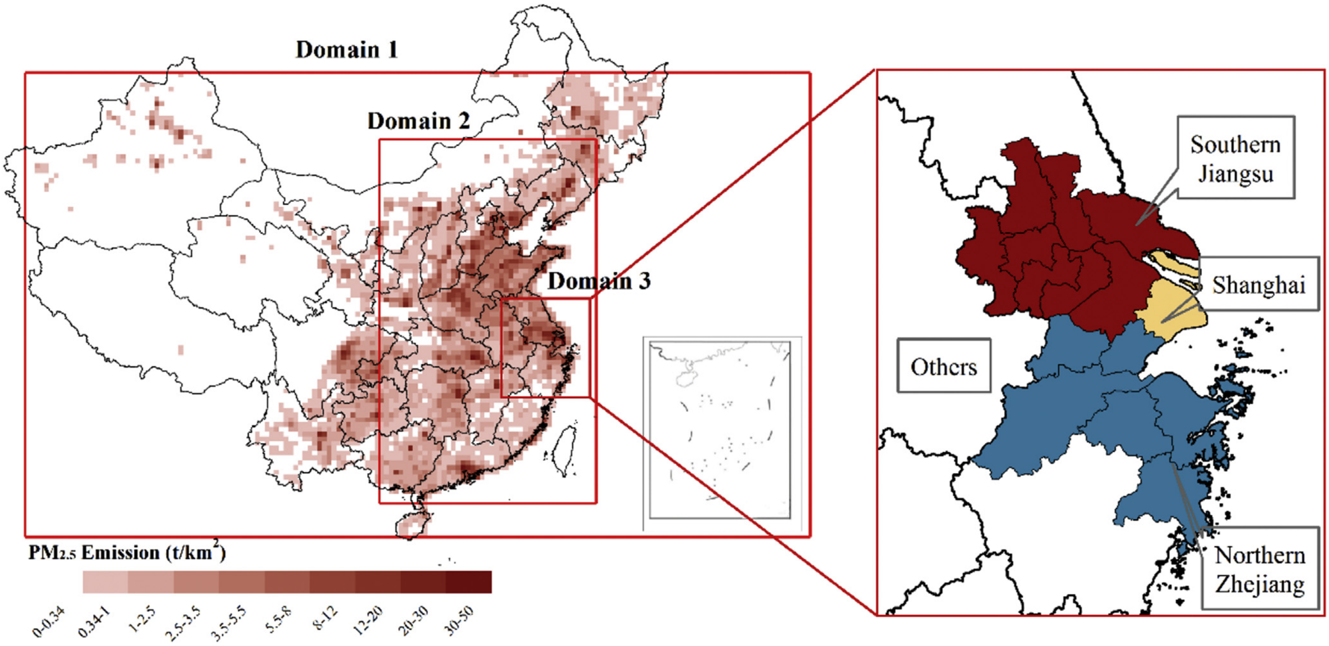

Fig. 1. Triple nesting domains utilized in CMAQ simulation (left) and the definition of four regions in Domain 3 (right). The black lines represent the provincial boundaries and national boundaries. The red frames show the modeling domains. The colors in left part show the emission intensity of $PM_{2.5}$. The red, yellow, blue and white colors in right part show the region of Southern Jiangsu, Shanghai, Northern Zhejiang, and others, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2.2. *Health impact assessment and economic valuation estimates*

2.2.1. *C-R function*

As discussed in Voorhees et al. (2014), although health impact functions could be identified for several specific morbidity endpoints (e.g., cardiovascular and respiratory hospital admissions, cardiovascular and respiratory outpatients), we were only able to identify non-specific population morbidity incidence rates (total outpatient visits, total emergency visits, total hospital admissions). This prevented an estimation of avoided cases of any specific morbidity endpoints. Therefore, only short-term mortality was analyzed in this study. For most epidemiological short-term exposure studies, Poisson distribution is used to express the relationship of health effect and $PM_{2.5}$ concentration, referring to C-R functions. Therefore, C-R functions in each grid cell, can be formulized as Eq. (1).

$$Y = E_0 \times Pop \times \left(1 - e^{-\beta\times(C-C_0)}\right) \tag{1}$$

In Eq. (1), Y is the number of excess deaths caused by $PM_{2.5}$ concentration. E_0 (%) is the actual mortality rate at $PM_{2.5}$ concentration C. Pop is the exposed population. The unit less C-R function coefficient β is derived from the relative risk (RR) reported in epidemiological references. C is the actual $PM_{2.5}$ concentration. C_0 is the threshold concentration.

The references were surveyed for studies linking ambient $PM_{2.5}$ concentration to premature mortality. A literature of meta-analysis was selected (Shang et al., 2013), which reports a combined estimation from 9 references. Shang et al. (2013) shows an increase of 10 $\mu g/m^3$ annual average of $PM_{2.5}$ would cause a 0.38% (95% Confidence intervals (CI): 0.31%–0.45%) increase in mortality rate. Therefore, C-R function coefficient β is calculated by $\beta = \ln(RR)/\Delta C$ and equals 3.8×10^{-4} (95% CI: 3.1×10^{-4}–4.5×10^{-4}).

BenMAP CE was applied to estimate the premature mortality due to $PM_{2.5}$. BenMAP CE, released by the United States Environmental Protection Agency (U.S. EPA), is a flexible tool for systematically analyzing the impacts of changes in air quality and adequately address uncertainty and variability. The parameters used in the software is described as below.

2.2.2. *Baseline incidence rate, threshold concentration and population*

Baseline incidence rate was collected from China Health Statistics Yearbook 2011 (Ministry of Health (2011)). Incidence of mortality for all causes in urban area was selected here due to the high urbanization level in YRD region. Threshold concentration refers to that below which $PM_{2.5}$ had no adverse effect on mortality. The threshold concentration of PM is still inconclusive and no evidence of obvious threshold concentrations was found (Cao et al., 2012). Thus, in this study, we assumed there is no threshold concentration.

The population data were extracted from Landscan (Bright et al., 2011), which used spatial data and imagery analysis technologies and a multi-variable dasymetric modeling approach to disaggregate census counts within an administrative boundary. The original resolution is approximately 1 km*1 km resolution (30″*30″). It represents an average population over 24 h. We aggregate the population data into 4 km*4 km grids, consistent with the grids of air quality simulation. To validate the gridded population, the gridded data were aggregated to each county and compared with the China 2010 Census data. The results are shown as figure S2 in SI. The pearson correlation coefficient is 0.87, indicating that the gridded population data were consistent with China 2010 Census data.

2.2.3. *Economic assessment methodologies*

Welfare economics assumes that life (or health) has values like other goods and the values can be compared. Given that health and life are irreplaceable and have no market price, indirect approaches, the value of statistical life (VSL) (Xie, 2011), has usually been employed to assess the value. It mainly includes two methods, the willingness to pay (WTP) method and human capital (HC) method (Huang et al., 2012a). In this study, WTP was selected because HC

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does not account some potential economic loss, such as spiritual damage and the cost of pain and suffering, which may lead to an underestimation of the health loss.

A literature search for WTP studies was done to identify China-specific valuation published over the past 10 years (Voorhees et al., 2014), which includes eleven studies (Hammitt and Zhou, 2006; Kan and Chen, 2004). Most of the studies utilized stated preference method in WTP research, however, the respondents are not sensitive to the relative size of health risks presented to them. Considering the shortcoming of the stated preference method, Xie (2011) employed the choice experiment method, in which the respondents make choice of the willingness to pay from many hypothetical scenarios, to investigate the WTP for improvement of air quality in Beijing in 2010. The VSL estimated is 1.68 million Chinese Yuan. This value is adopted in this study considering the similar economic level of Beijing and YRD.

2.3. Source apportionment of premature mortality

To estimate the contribution for the health loss of different regions, sectors and pollutants, the brute-force method (Russell et al., 1995; Seigneur et al., 1981; Zhang et al., 2009; Zhao et al., 2013b) is employed in this study. January and August are selected to represent winter and summer situation. The detailed set of scenarios is shown in Table 1. First, we classify the health impact of $PM_{2.5}$ as due to combined contributions of both outside and local emissions, which refers to the contribution of emissions out of Domain 3 and in Domain 3, respectively. The emissions out of Domain 3 include both primary $PM_{2.5}$ and $PM_{2.5}$ precursors. Therefore, two scenarios are designed. For scenario A, the boundary condition of Domain 3 is generated without anthropogenic PM emissions in Domain 1 and Domain 2. For scenario B, the boundary condition of Domain 3 is generated without anthropogenic gaseous precursors' emissions in Domain 1 and Domain 2. We designed scenario C and D by turning off the emission of anthropogenic primary PM emissions and all anthropogenic gaseous precursors' emissions in Domain 3, respectively. Then, the anthropogenic NO_X, SO_2, VOC and NH_3 emissions in Domain 3 are turned off in turn for Scenario E to H. To understand the contribution of different regions in YRD, different anthropogenic emission of regions were turned off in turn in Scenario I to L. To further understand the contribution of different sectors in YRD, power plants' emissions were turned off in Scenario M. Industrial and residential emissions were turned off in Scenario N. Traffic emissions were turned off in Scenario O.

3. Results and discussions

3.1. Population-weighted average $PM_{2.5}$ concentrations in 2010

Fig. 2 (a) shows the annual average surface $PM_{2.5}$ concentration given by the first layer of CMAQ simulation in the YRD domain in 2010. The most polluted regions include Nanjing, Zhenjiang, eastern part of Hangzhou and northern part of Shanghai. $PM_{2.5}$ concentration distribution shows a gradient from northwest to southeast. The urban areas are the most polluted regions, consistent with the most densely populated areas. To further quantify the $PM_{2.5}$ concentration people are exposed to, annual average $PM_{2.5}$ concentration of grids belonging to one city and population-weighted annual average $PM_{2.5}$ concentration of one city are shown in Table 2.

As shown in Table 2, the five cities with highest average $PM_{2.5}$ concentrations are Nanjing, Changzhou, Wuxi, Zhenjiang and Suzhou. They are consistent with the cities with highest population-weighted $PM_{2.5}$ concentration. The ratio of population-weighted average $PM_{2.5}$ and average $PM_{2.5}$ is not consistent among different cities. Higher ratio indicates that $PM_{2.5}$ tends to be concentrated in urban area with dense population. The city with highest ratio is Hangzhou. We notice that the average $PM_{2.5}$ concentration of Hangzhou is lower than most cities in Southern Jiangsu. However, its population-weighted $PM_{2.5}$ concentration is on the similar level with Suzhou and Zhenjiang. Besides Hangzhou, Shanghai, Ningbo and Shaoxing all have a higher ratio of population-weighted average $PM_{2.5}$ to average $PM_{2.5}$. The different ratio is mainly due to the pollutant from different sources. The $PM_{2.5}$ comes from long-range transport and elevated sources have a uniform distribution, while $PM_{2.5}$ from low-stack emission sources

Table 1
Description of the scenarios designed to assess the contribution of different emission sources to premature mortality.

Scenario	Description of the scenarios	Objective of the scenarios
Base case	The CMAQ base case.	Assess premature mortality due to $PM_{2.5}$ in YRD.
A	Turn off anthropogenic primary $PM_{2.5}$ emission in Domain 1 and Domain 2.	Assess premature mortality due to anthropogenic $PM_{2.5}$ in YRD from transport/local primary $PM_{2.5}$ or its gaseous precursors.
B	Turn off all anthropogenic gaseous precursors' emission in Domain 1 and Domain 2.	
C	Turn off anthropogenic primary $PM_{2.5}$ emission in Domain 3.	
D	Turn off all anthropogenic gaseous precursors' emission in Domain 3.	
E	Turn off all anthropogenic NO_x emission in Domain 3.	Assess premature mortality due to anthropogenic $PM_{2.5}$ in YRD from local different gaseous precursors.
F	Turn off all anthropogenic SO_2 emission in Domain 3.	
G	Turn off all anthropogenic VOC emission in Domain 3.	
H	Turn off all anthropogenic NH_3 emission in Domain 3.	
I	Turn off all anthropogenic primary $PM_{2.5}$ and gaseous precursors' emission in Shanghai.	Assess premature mortality due to anthropogenic $PM_{2.5}$ in YRD from local different regions.
J	Turn off all anthropogenic primary $PM_{2.5}$ and gaseous precursors' emission in Southern Jiangsu.	
K	Turn off all anthropogenic primary $PM_{2.5}$ and gaseous precursors' emission in Northern Zhejiang.	
L	Turn off all anthropogenic primary $PM_{2.5}$ and gaseous precursors' emission in other area in Domain 3.	
M	Turn off all anthropogenic primary $PM_{2.5}$ and gaseous precursors' emissions of power plant.	Assess premature mortality due to anthropogenic $PM_{2.5}$ in YRD from local different sectors.
N	Turn off all anthropogenic primary $PM_{2.5}$ and gaseous precursors' industrial and residential emissions.	
O	Turn off all anthropogenic primary $PM_{2.5}$ and gaseous precursors' emission of traffic.	

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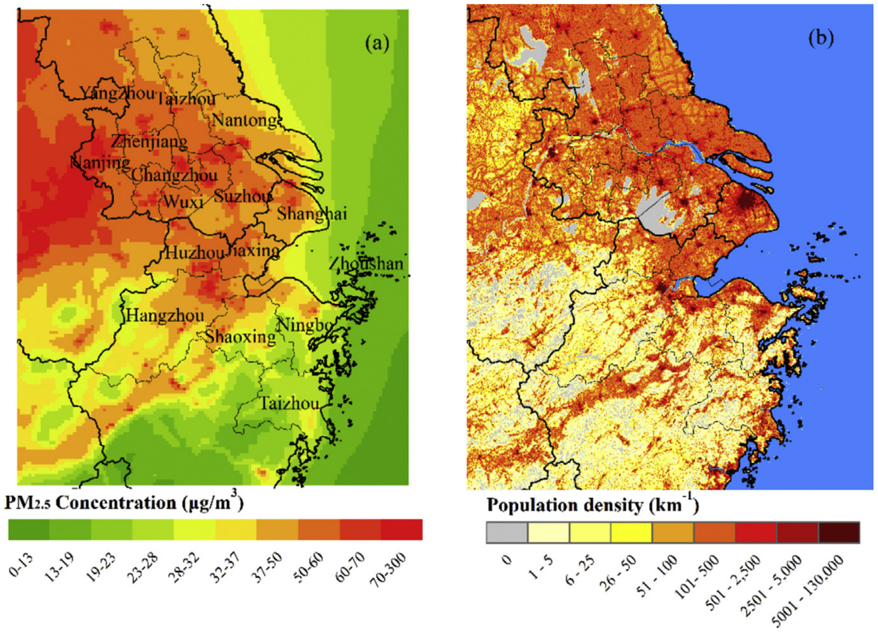

Fig. 2. Annual average surface $PM_{2.5}$ concentration given by the first layer of CMAQ simulation (a) and population density (b) in YRD, 2010. The thick and thin black lines represent provincial boundaries and city boundaries, respectively. The colors in figure (a) show the ambient concentration of $PM_{2.5}$. The colors in figure (b) show the population density. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

shows a scattered pattern. A higher ratio of population-weighted average $PM_{2.5}$ to average $PM_{2.5}$ implies that the local and low-stack emissions contribute more in this area, especially for the emissions from traffic and residential emission. The seasonal variance of annual average and population-weighted annual average $PM_{2.5}$ concentration are shown in Table S5 in SI. The premature mortality caused by exposure to $PM_{2.5}$ is assessed in Section 3.2. To further estimate the reason for the different ratio and its health impact, the contribution of different sources are analyzed in Section 3.3.

3.2. Premature mortality due to $PM_{2.5}$ and its economic loss assessment

The estimated annual premature death due to $PM_{2.5}$ and its economic loss for each city is shown in Table 3. It is shown that short-term premature mortality due to $PM_{2.5}$ are 13,162 (95% CI: 10,761–15,554) in YRD in 2010. The total economic loss is 22.1 (95% CI: 18.1–26.1) billion yuan. Shanghai, Nanjing, Hangzhou and Suzhou suffers the most serious health impact, accounted for about half of the total economic loss in YRD. The seasonal variance of premature mortality due to $PM_{2.5}$ for each city is shown in Table S6 in SI.

The distribution varies with months, as shown in Figure S3 in SI. The distribution of $PM_{2.5}$ concentration distribution could be grouped into two categories. In January and November, the heavily polluted areas are concentrated in the northeastern part of the map. In May and August, heavily contaminated areas on the map are scattered. It implies the $PM_{2.5}$ pollution in different seasons is from different source categories. Therefore, the sensitivity analysis was done for January and August.

Table 2
Annual average $PM_{2.5}$ concentration and population-weighted annual average $PM_{2.5}$ concentration.

Province	City	Annual average $PM_{2.5}$ concentration	Population-weighted average $PM_{2.5}$ concentration	Ratio
Shanghai	Shanghai	42.7	52.1	1.22
Jiangsu Province	Nanjing	61.2	71.1	1.16
	Wuxi	57.2	63.1	1.10
	Changzhou	58.3	63.9	1.10
	Suzhou	53.7	59.0	1.10
	Nantong	42.5	44.5	1.04
	Yangzhou	53.5	57.3	1.07
	Zhenjiang	57.0	60.0	1.05
	Taizhou	51.7	54.1	1.05
Zhejiang Province	Hangzhou	39.9	58.8	1.47
	Ningbo	29.6	37.4	1.27
	Jiaxing	49.6	50.4	1.02
	Huzhou	50.6	54.1	1.07
	Shaoxing	35.4	43.7	1.23
	Zhoushan	19.4	19.0	0.98
	Taizhou	24.3	27.0	1.11
YRD_total		44.3	52.0	1.17

Table 3
Premature mortality due to $PM_{2.5}$ for each city and its economic loss.

Province	City	Mortality due to $PM_{2.5}$	Economic loss (million yuan)
Shanghai	Shanghai	2415 (1974,2854)	4058 (3317,4795)
Jiangsu Province	Nanjing	1303 (1066,1539)	2189 (1791,2585)
	Wuxi	919 (752,1086)	1544 (1263,1825)
	Changzhou	689 (563,814)	1157 (946,1367)
	Suzhou	1146 (937,1354)	1926 (1574,2275)
	Nantong	981 (802,1160)	1649 (1347,1949)
	Yangzhou	753 (616,890)	1266 (1035,1496)
	Zhenjiang	490 (400,578)	822 (672,972)
	Taizhou	763 (624,902)	1282 (1048,1515)
Zhejiang Province	Hangzhou	1159 (947,1369)	1946 (1592,2300)
	Ningbo	664 (542,785)	1115 (911,1318)
	Jiaxing	524 (428,620)	881 (720,1041)
	Huzhou	410 (335,484)	688 (563,814)
	Shaoxing	547 (447,647)	920 (751,1087)
	Zhoushan	1 (1,1)	2 (2,2)
	Taizhou	398 (325,471)	669 (546,791)
YRD_total	Total	13,162 (10,761,15,554)	22,113 (18,078,26130)

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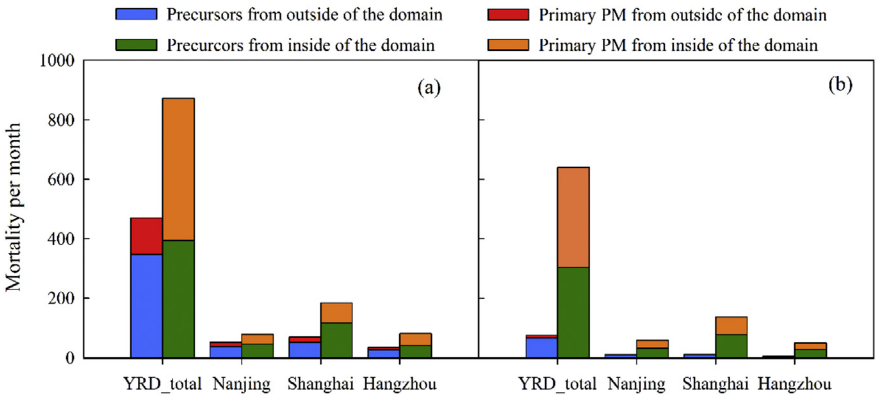

Fig. 3. Mortality per month caused by transport from outside of boundary of domain 3 and local emission. The left bar in each group shows the contribution of transport from outside of boundary and the right bar shows the contribution of local emission. Figure (a) represents the result of January and Figure (b) represents the result of August.

3.3. *Contribution of different emission sources to mortality*

The differences of $PM_{2.5}$ concentration between control scenarios and base case are shown in Figure S4 to Figure S7 in SI. The contributions of local emissions and emissions from outside of Domain 3 to premature deaths are shown in Fig. 3. It shows that in January, the $PM_{2.5}$ and its precursors transported from outside of Domain 3 lead to 470 premature deaths in YRD, accounting for 35% of deaths caused by $PM_{2.5}$. In August, the premature deaths due to transported pollutants are less than 100, accounting for only 10%. It indicates that the long-range transport is more significant in winter time. It is mainly because that in winter, the wind direction is from northwest. The emissions in northern China increase by coal consumption due to heating. In January, the emissions of SO_2, NOx, VOC, and primary $PM_{2.5}$ from residential sector in North China (including Beijing, Tianjin, Hebei, Shandong, and Henan Provinces) are 118.1, 47.3, 236.9, and 152.6 kton, respectively, much higher than that in August when the corresponding emissions are 26.0, 10.5, 54.3, and 34.2 kton, respectively (Wang et al., 2010b; Zhao et al., 2013b). In summer, the wind direction is from southeast. The wind comes from the ocean and brings clean air to the YRD. Local primary $PM_{2.5}$ and local gaseous precursors show a similar contribution both in January and August. However, the transported gaseous precursors contribute more than transported primary $PM_{2.5}$. It indicates that regional joint prevention and control for the gaseous precursors is important for air quality improvement.

For Shanghai and Hangzhou, the contributions of emission from outside of domain 3 are 27% and 30% to the total premature deaths in January, respectively. This is in line with the high ratio of population-weighted average $PM_{2.5}$ to average $PM_{2.5}$ and indicates that the control of emissions from urban area should be the focus of air quality management. In contrast, for Nanjing, located in the southwest of Jiangsu and affected by the transport of air pollutants, the contribution of emission from outside of domain 3 accounts for 40% and 15% of the total premature deaths in January and August, respectively. Therefore, regional joint prevention and control with neighboring provinces should also be emphasized.

To further explore the response of premature deaths with $PM_{2.5}$ exposure, we selected the key gas precursors inside the domain, including NO_X, SO_2, NH_3 and VOCs in this study. As discussed above, the total contribution of local gas precursors does not vary much. However, Fig. 4 shows that the contributions of individual gas precursors have obvious seasonal difference. In January, premature mortality is most sensitive to emissions of NH_3 and NO_x, and relatively insensitive to SO_2 and VOCs; In August, NH_3, SO_2, and NO_X have approximately equal contributions. The results indicate that based on protecting public health, the control of NH_3 emissions should receive more attention. However, it should be noted that the contribution of VOC may be underestimated due to the uncertainties in secondary organic aerosol mechanism in CMAQ (Jaemeen et al., 2011).

Fig. 5 shows the response of premature mortality to emissions of different sectors. The premature mortality due to emissions from power plant, industrial and residential sectors, and traffic are approximately 36, 809 and 40 in January, and 107, 528 and 63 in August. Industrial and residential emissions are the leading contributors, which accounting for 91% and 75% in January and August, respectively. In part due to the active manufacturing industry in the YRD and the lower penetration rate for pollution control technologies compared with the power sector, the impacts of the industrial and residential sectors contribute most. It accounts for almost half of the total premature mortality due to $PM_{2.5}$. Fu et al. (2013) demonstrated that industrial processes and industrial combustion account for approximately 50% and 43.4% of anthropogenic primary $PM_{2.5}$ and SO_2 emissions in the YRD. It indicates that emission control in industrial sectors is the most urgent need. Figure S7 (a) and (d) show that the $PM_{2.5}$ pollution due to emissions from power plants is uniform, especially in August. The emissions of power

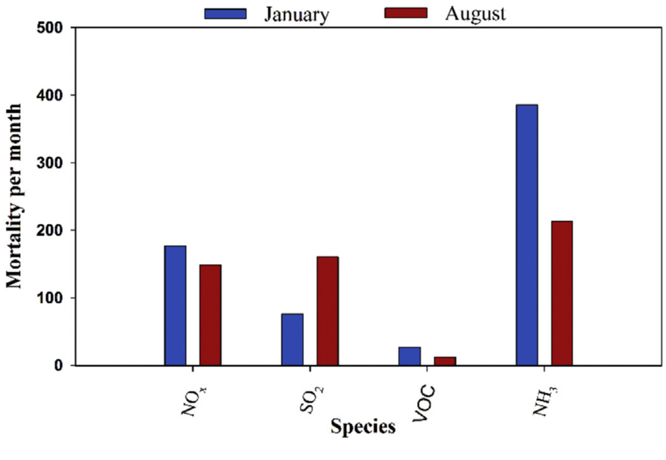

Fig. 4. Mortality per month caused by emissions of gaseous precursors in the YRD domain. The red color and blue color show the contribution of precursors in January and August, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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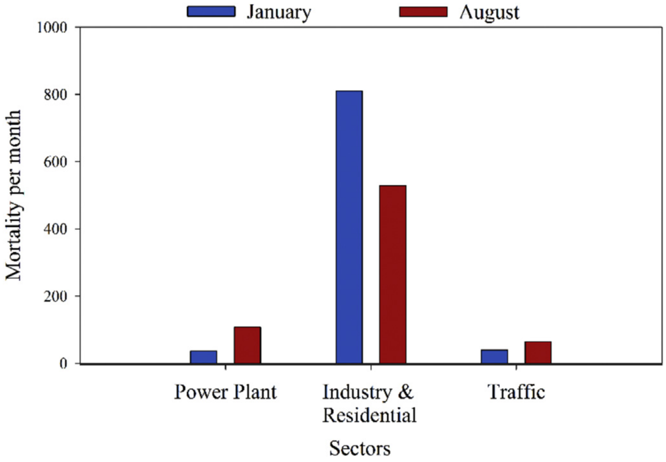

Fig. 5. Mortality per month caused by emissions of different sectors in the YRD domain. The red color and blue color show the contribution of sectors in January and August, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

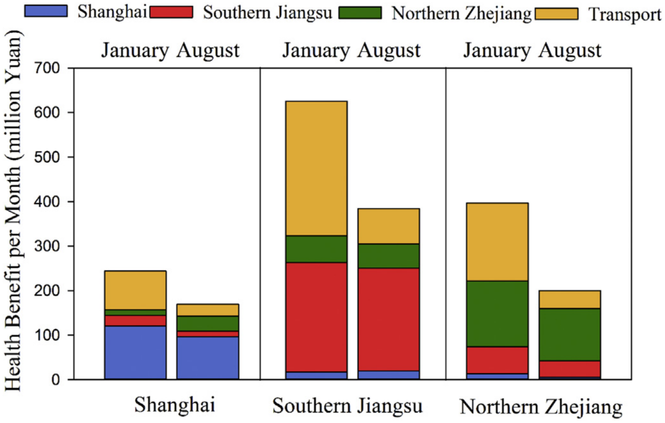

Fig. 6. Contribution of emissions from different regions to premature mortality. The left and right bar represent the result of January and August, respectively. The blue, red, green and yellow color show the contribution of emissions from Shanghai, southern Jiangsu, northern Zhejiang and areas outside of the YRD domain. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

plant are released at elevated heights. Therefore, it influences the surface $PM_{2.5}$ concentration in larger areas. Contrasted to that of power plants, the impact of traffic is dispersed (as shown in Figure S7 (c) and (f)).

Fig. 6 shows contribution of emissions from individual regions to premature mortality due to $PM_{2.5}$. We define emissions from all other regions except YRD as pollutant transport. The contributions of local emission in Shanghai, southern Jiangsu, and northern Zhejiang are 49.4%, 39.4% and 37.1% in January, and 57.0%, 60.0% and 59.0% in August, respectively. The contributions of pollutant transport from outside of YRD are 35.4%, 48.3% and 44.2% in January, and 15.8%, 20.7% and 19.9% in August, respectively. For Shanghai, the contribution of emissions from other regions is impacted by the predominant wind direction. In January, the contribution of southern Jiangsu emissions to premature deaths caused by $PM_{2.5}$ is larger than northern Zhejiang. But in August, emissions from northern Zhejiang contribute more. Northern Zhejiang shows a similar trend with Shanghai and impacted by the predominant wind directions.

3.4. Uncertainty analysis

Comparisons among Chinese studies on health impact of particulate matter are difficult. Voorhees et al. (2014) summarized the estimation in previous study of premature deaths for single city in China is ranging from 290 to 24,000 because the population data, short-term or long-term estimation, and threshold concentrations are different in different studies. Only a few studies estimating the mortality of Shanghai with similar method are compared. Kan and Chen (2004) estimated the PM_{10}-related deaths are 17,840 (95% CI: 11,010, 24,660) with no threshold concentration. Voorhees et al. (2014) estimated that the avoided impact on all-cause mortality of a year exposure to the annual or monthly mean 24-h $PM_{2.5}$ concentration was estimated to range from 39 to 1400 per year using short term health functions with a uniform threshold concentration of 35 $\mu g/m^3$. In our study, the annual premature mortality caused by $PM_{2.5}$ in Shanghai is 2415 (95% CI: 1974, 2854), in between of the two studies above.

There are two types of uncertainties in estimation of contribution of different emission sources on premature mortality due to $PM_{2.5}$. The first type is the uncertainty of epidemiological references, incidence and population data. The impacts of epidemiological references are shown as 95% CI. However, the uncertainty of incidence and population data is difficult to quantify. The second is the uncertainty due to the limit of brute-force method. The brute-force method could not capture the non-linear effects of emission on $PM_{2.5}$ ambient concentration. Thus, the sum of contribution of each kind of gaseous precursors may not equal to the total contribution of all gaseous precursors. To address this issue, future work is needed.

The estimation of economic loss due to premature mortality may also introduce uncertainty. The VSL of 1.68 million Chinese Yuan, approximately equaling to 0.27 million US dollar. The US EPA recommends a VSL of $7.9 million (2008 US dollars) (US EPA, 2010). Lindhjem et al. (2011) estimated the VSL to be $7.4 million (2005 U.S. dollars) by compiling and analyzing studies globally. The VSL in China is quite small comparing with the studies above. It could be a

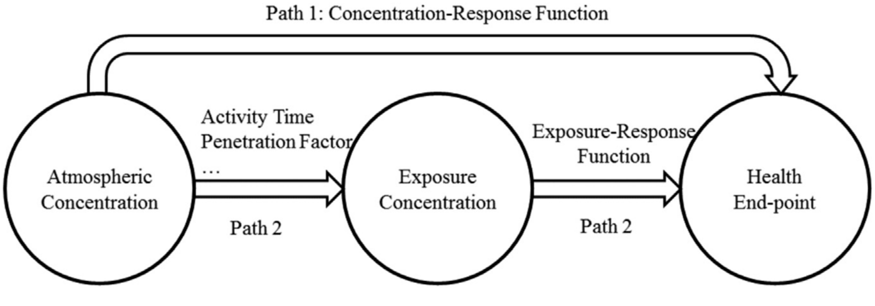

Fig. 7. Mapping from atmospheric $PM_{2.5}$ concentration to health end-point.

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key factor to impact on the uncertainty to assessment the economic loss due to premature mortality. More research on this aspect is still needed in the future.

To be noticed, the impact of indoor exposure was incorporated indirectly in this study. The indoor and outdoor $PM_{2.5}$ concentration are highly correlated, rather than independent variables. The ratio between them is presented as penetration factor or infiltration factor (Wu et al., 2003; Wang et al., 2008, 2010a). Therefore, when epidemiological studies established directly from ambient $PM_{2.5}$ concentration to health endpoint, shown as path 1 in Fig. 7, the contribution of indoor $PM_{2.5}$ was inevitably considered. Thus, estimating the health impact with both outdoor and indoor exposure with this kind of concentration-response function causes double counting for the indoor impact. However, the indoor and outdoor $PM_{2.5}$ concentration are not fully correlated, because the indoor/outdoor ratio of activity time and the penetration factor of $PM_{2.5}$ vary due to weather and resident behaviors, such as ventilation and air conditioning use. The indoor impacts are not fully estimated by path 1, which may produce biased results (Chen et al., 2012; Zhou et al., 2013). A better way is to consider the exposure rather than atmospheric $PM_{2.5}$ concentration, shown as path 2 in Fig. 7. The obstacle of doing this is the lack of epidemiological studies on exposure-response function considering both outdoor and indoor $PM_{2.5}$ exposure. Therefore, future research in this area is still needed.

4. Conclusions

In this study, Environmental Benefits Mapping and Analysis Program Community Edition (BenMAP CE) is employed to assess the premature mortality of $PM_{2.5}$ in the YRD region in 2010. It is found that the $PM_{2.5}$-related annual premature mortality is 13,162 (CI, 10761–15,554) in 2010, resulting in an economic loss of 22.1 (95% CI: 18.1–26.1) billion yuan. Shanghai, Nanjing, Hangzhou and Suzhou suffers the most serious health damages, accounting for about half of the total economic loss in YRD.

To provide scientific basis for air pollution control policies, the contribution of different regions, precursors and sectors are evaluated in January and August 2010. In January, the transported $PM_{2.5}$ and its precursors from outside of the boundary lead to 35% of premature deaths caused by $PM_{2.5}$. It indicates that the air pollutant transport is severe in winter. The contribution of pollutant transport is larger in western part of YRD, such as Nanjing. It indicates regional joint prevention and control with neighboring provinces shall be implemented in this area. For Shanghai and Hangzhou, $PM_{2.5}$ tends to be concentrated in urban area with dense population. The control of emissions from urban area and reducing population exposure should be emphasized.

Industrial and residential emissions are the leading contributors among different sectors, both in summer or winter. The control of emissions of the industrial and residential sources should be enhanced considering their large contribution to $PM_{2.5}$ concentrations. In January, premature mortality is most sensitive to emissions of NH_3 and NO_x, and relatively insensitive to VOCs and SO_2. The control of NH_3 emissions should also receive more attention.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2015.05.060.

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