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Nonlinear relationships between air pollutant emissions and $PM_{2.5}$-related health impacts in the Beijing-Tianjin-Hebei region

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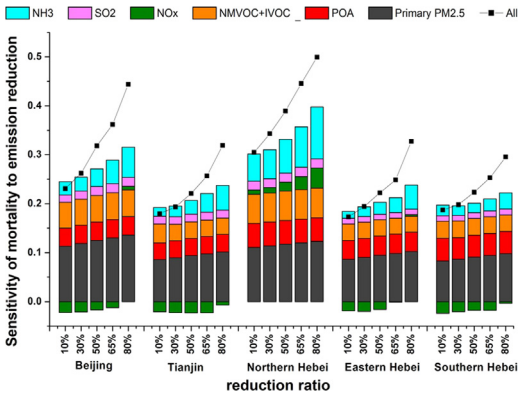
HIGHLIGHTS

- $PM_{2.5}$-related mortality is most sensitive to the emissions of primary $PM_{2.5}$.
- The mortality responds linearly to emission reduction of NMVOC, IVOC and SO_2.
- The mortality is more sensitive to primary $PM_{2.5}$, NH_3 and NO_x at larger reductions.
- Multi-pollutant and multi-region controls result in larger marginal health benefit.
- A nationwide control strategy of NO_x emissions is needed to enhance health benefit.

GRAPHICAL ABSTRACT



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ABSTRACT

A direct and quantitative linkage of air pollution-related health effects to emissions from different sources is critically important for decision-making. While a number of studies have attributed the $PM_{2.5}$-related health impacts to emission sources, they have seldom examined the complicated nonlinear relationships between them. Here we investigate the nonlinear relationships between $PM_{2.5}$-related premature mortality in the Beijing-Tianjin-Hebei (BTH) region, one of the most polluted regions in the world, and emissions of different pollutants from multiple sectors and regions, through a combination of chemical transport model (CTM), extended response surface model (ERSM), and concentration-response functions (CRFs). The mortalities due to both long-term and short-term exposures to $PM_{2.5}$ are most sensitive to the emission reductions of primary $PM_{2.5}$, followed by NH_3, nonmethane volatile organic compounds and intermediate volatility organic compounds (NMVOC +IVOC). The sensitivities of long-term mortality to emissions of primary organic aerosol (POA), NMVOC +IVOC and SO_2 do not change much with reduction ratio, whereas the sensitivities to primary inorganic $PM_{2.5}$ (defined as all chemical components of primary $PM_{2.5}$ other than POA), NH_3 and NO_x increase significantly

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China
extended response surface model (ERSM)

with the increase of reduction ratio. The emissions of primary $PM_{2.5}$, especially those from the residential and commercial sectors, contribute a larger fraction of mortality in winter (57–70%) than in other seasons (28-42%). When emissions of multiple pollutants or those from both local and regional emissions are controlled simultaneously, the overall sensitivity of long-term mortality is much larger than the arithmetic sum of the sensitivities to emissions of individual pollutants or from individual regions. This implies that a multi-pollutant, multi-sector and regional joint control strategy should be implemented to maximize the marginal health benefits. For NO_x emissions, we suggest a nationwide control strategy which significantly enhances the effectiveness for reducing mortality by avoiding possible side effects when only the emissions within the BTH region are reduced.

1. Introduction

Atmospheric fine particle ($PM_{2.5}$) pollution has adverse effects on human health (Shang et al., 2013; Lelieveld et al., 2015). The effects are especially important in the Beijing-Hebei-Tianjin (BTH) urban agglomeration region, one of the most heavily polluted regions in China (Wang et al., 2017; Cai et al., 2017). It's estimated that monetize health losses caused by $PM_{2.5}$ pollution in the BTH region can reach 134.3 billion RMB, which accounts for 2.16% of the GDP in this region (Lv and Li, 2016). Protecting public health is the ultimate goal of air pollution control. Therefore, it is crucially important for environmental decision making to quantitatively link the $PM_{2.5}$-related health impacts to air pollutant emissions from different sources.

$PM_{2.5}$ pollution is formed through complex physical and chemical processes. As a result, the relationships between $PM_{2.5}$ concentrations and emissions of precursors are nonlinear, and the nonlinearity is especially significant in urban agglomerations (Zhao et al., 2015; Zhao et al., 2017; Xing et al., 2017; Fu et al., 2012). In addition, some studies (Martenies et al., 2015; Burnett et al., 2014; Burnett et al., 2018) pointed out that the relationships between $PM_{2.5}$ concentrations and the resulting health effects are also nonlinear. When these two relationships are overlaid, we can expect highly nonlinear relationships between the $PM_{2.5}$-associated health effects and the emissions of precursors. A number of studies (GBD MAPS Working Group, 2016; Hu et al., 2017; Fann et al., 2012; Wang et al., 2015; Heo et al., 2016; Andersson et al., 2009) have attributed the $PM_{2.5}$-associated health effects to different emission sources, but to our best knowledge, none of them have explicitly considered the aforementioned nonlinear relationships, which brings inaccuracy to the source attribution results. Therefore, it's a meaningful and also challenging scientific issue to quantify the nonlinear responses of $PM_{2.5}$-related health effects to primary air pollutant emissions.

In this study, we quantitatively evaluate the nonlinear relationships between premature mortality caused by long-term/short-term exposures to $PM_{2.5}$ in the BTH region and emissions of different air pollutants from multiple sectors and regions, by combining a chemical transport model (CTM), concentration-response functions (CRFs), and an extended response surface model (ERSM). By providing the first quantitative assessment of the nonlinear relationships between emission reductions and health impacts, this study helps decision makers in the BTH region to develop specific and optimized control measures that best protect the public health. Also, the modeling framework developed in this study enables an instantaneous prediction of $PM_{2.5}$-related mortality for any given emission scenario. It can be readily adopted by researchers, policymakers, and advocates from public, private, and nonprofit sectors, and holds strong potential for replication in other regions in the world, further extending this study's reach and impact.

2. Methods

2.1. Health impact assessment methodology

In this study, we estimate the health effects due to both short-term and long-term exposures to ambient $PM_{2.5}$. For long-term health impact assessment, Burnett et al. (2014) proposed integrated expose-response (IER) function, which were constructed by combining risk estimates from studies of ambient and household air pollution, and active/second-hand smoking that cover a full range of $PM_{2.5}$ exposure up to about 30000 μg/m^3 (Cohen et al., 2017; Burnett et al., 2014). The IER function is given by Eq. (1), which has been proved to fit best to the actual risk estimates among a variety of equation forms (Cohen et al., 2017; Burnett et al., 2014).

$$\Delta Y_i = y_{0,i} P \times \frac{RR_i - 1}{RR_i}$$
$$RR_i = \begin{cases} 1, & C < C_0 \\ 1 + \alpha_i \left[1 - e^{-\gamma_i (C - C_0)^{\delta_i}}\right], & C \geq C_0 \end{cases} \quad (1)$$

where ΔY_i refers to $PM_{2.5}$-induced mortality from endpoint i; $y_{0,i}$ refers to the actual mortality rate of endpoint i at the current $PM_{2.5}$ concentration (C); P refers to exposed population; C_0 refers to threshold $PM_{2.5}$ concentration below which no health impact is expected. α_i, γ_i, and δ_i are regression parameters for endpoint i. According to the IER function, the change in mortality due to a unit concentration change decreases significantly with the deterioration of $PM_{2.5}$ pollution. Recently, Burnett et al. (2018) developed new CRFs for long-term $PM_{2.5}$ exposures based on only cohort studies of ambient air pollution, which resulted in larger mortality estimates. However, this would not change our major conclusions about the nonlinear emission-mortality relationships, since the general shape of the functions retained. In this study, we adopt the same health endpoints and IER parameters (α_i, γ_i, and δ_i) as in Burnett et al. (2014). These endpoints include chronic obstructive pulmonary disease (COPD), ischemic heart disease (IHD), lung cancer, and stroke for adults > 25 years old, as well as acute lower respiratory infection (ALRI) for children < 5 years old. The disease specific baseline mortality rates by age and gender are obtained from the Institute of Health Metrics and Evaluation (Global Burden of Disease Collaborative Network, 2017).

For health impact assessment of short-term exposure to $PM_{2.5}$, the CRFs are developed by epidemiological studies based on time series analysis of $PM_{2.5}$ and health. In most studies (Shang et al., 2013; Wang et al., 2015; Dominici et al., 2002; Kan and Chen, 2004), the incidence of mortality caused by air pollution is considered to be subject to Poisson distribution. Subsequently, the relationship between mortality and $PM_{2.5}$ concentrations can be regressed in the following form by using Poisson regression (log-linear regression) or similar methods (Kan et al., 2008; Dominici et al., 2002; Shang et al., 2013; Kan and Chen, 2004).

$$\Delta Y_i = y_{0,i} P \left[1 - e^{\beta_i (C - C_0)}\right] \quad (2)$$

where ΔY_i, $y_{0,i}$, P, C, and C_0 have the same meaning as defined in Eq. (1). β_i is a regression coefficient derived from epidemiological studies, which refers to excess risk of mortality per each increase in 1 μg/m^3 of $PM_{2.5}$ concentration. Reasonable regression results have been obtained using Eq. (2) in over 25 epidemiological studies conducted in China (Shang et al., 2013), where the $PM_{2.5}$ concentrations range from very small to very large (mean concentration > 170 μg/m^3), indicating the

applicability of Eq. (2) to the Chinese environments. Based on Eq. (2), the relationship between ΔY_i and C presents a slightly convex function form, indicating subtly less marginal effect at larger concentration increase. The health endpoints considered in the short-term assessment include all-cause mortality, cardiovascular mortality, and respiratory mortality. The baseline mortality rates are derived from China Health and Family Planning Statistics Yearbook 2015 (National Health and Family Planning Commission of China, 2015) and Huang and Zhang (2013), and the parameter estimates are taken from Chen et al. (2011), a local study over the BTH region. It should be noted that, in addition to mortality, the short-term $PM_{2.5}$ exposure also leads to various types of morbidity (e.g., cardiovascular and respiratory hospital admissions or outpatients). We focus on mortality in the present study since it accounts about 80% of the total monetized health losses (Kan and Chen, 2004; Wu, 2016).

The age-specific population data at city level in 2014 are acquired from the statistical bureaus of Beijing, Tianjin and Hebei, and the spatial distribution of population at sub-city level is based on the LandScan dataset at 30″ × 30″ (approximately 1 km × 1 km) resolution (Oak Ridge National Laboratory, 2016). The spatial distribution of population is shown in Fig. S1. Annually average and monthly average $PM_{2.5}$ concentrations derived in Section 2.2 are fed into the CRFs to assess the health impacts due to long-term and short-term $PM_{2.5}$ exposures, respectively. To give the uncertainty of the health effects, we calculate the 95% confidence intervals (CIs) using 95% CIs of the parameters of the CRFs for both short-term and long-term assessments.

2.2. Determination of exposed concentrations

The exposed concentrations of $PM_{2.5}$ (C in Eqs. (1) and (2)) in the base case are simulated using the Community Multi-scale Air Quality model with two-dimensional Volatility Basis Set extension (CMAQ/2D-VBS; Zhao et al., 2016). The CMAQ/2D-VBS model was developed by incorporating the 2D-VBS framework in CMAQ, a widely used three-dimensional CTM, in order to improve the simulation of secondary organic aerosol (SOA) (Zhao et al., 2016). The Weather Research and Forecasting (WRF) model version 3.7 is used to simulate the meteorological field to provide input data for the CMAQ/2D-VBS model. The simulation periods are January, March, July and October in 2014, representing four seasons. We select these four months in 2014 because they agree best with the average meteorological conditions in winter, spring, summer, and fall during 2004–2014, ensuring the representativeness of meteorological conditions in the simulation periods (Zhao et al., 2017).

We apply the WRF/CMAQ/2D-VBS model to double-nesting simulation domains, with grid resolutions of 36 km × 36 km and 12 km × 12 km. The first domain covers China mainland and its surrounding areas, and the second domain covers the BTH region, as shown in Fig. 1. The configurations of WRF and CMAQ/2D-VBS and the emission inventory used in this paper are the same as Zhao et al. (2017). In brief, in the BTH region, we use a high-resolution anthropogenic emission inventory in 2014 developed by Zheng et al. (2018). The anthropogenic emission inventory in other provinces of China was developed in our previous studies (Wang et al., 2014; Zhao et al., 2018b). The emissions from open burning of agricultural residue have been included in both inventories. The emissions outside China are obtained from the MIX emission inventory (Li et al., 2015) for 2010, which is the latest year available. The biogenic emissions are calculated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN; Guenther et al., 2006). The simulated results from WRFv3.7 and CMAQ/2D-VBS model generally agree well with ground observations (Zhao et al., 2017).

The 3-D CTM is time-consuming and computationally expensive. In this study, we acquire the exposed $PM_{2.5}$ concentrations in various emission control scenarios using the ERSM technique (Zhao et al., 2015), which enables quick prediction of $PM_{2.5}$ concentrations in any given emission scenario. The conventional response surface model (RSM) builds the relationships between $PM_{2.5}$ concentrations and a set of control variables (i.e., emissions of specific precursors from specific sources) using a number of CTM simulation scenarios and advanced statistical techniques (Xing et al., 2011; Wang et al., 2011). The ERSM technique extends the applicability of conventional RSM to a much larger number of control variables and geographical regions (Zhao et al., 2015). It first quantifies the relationship between $PM_{2.5}$ concentrations and precursor emissions in each single region using the conventional RSM, and then assesses the effects of inter-regional transport of $PM_{2.5}$ and its precursors on $PM_{2.5}$ concentration in the target region. The ERSM technique has showed good performances in several recent studies (Zhao et al., 2015; Zhao et al., 2017; Xing et al., 2017).

The establishment and validation of ERSM in the BTH region have been described in Zhao et al. (2017), so we only summarize several key points. First, we defined 5 target regions in the BTH region,

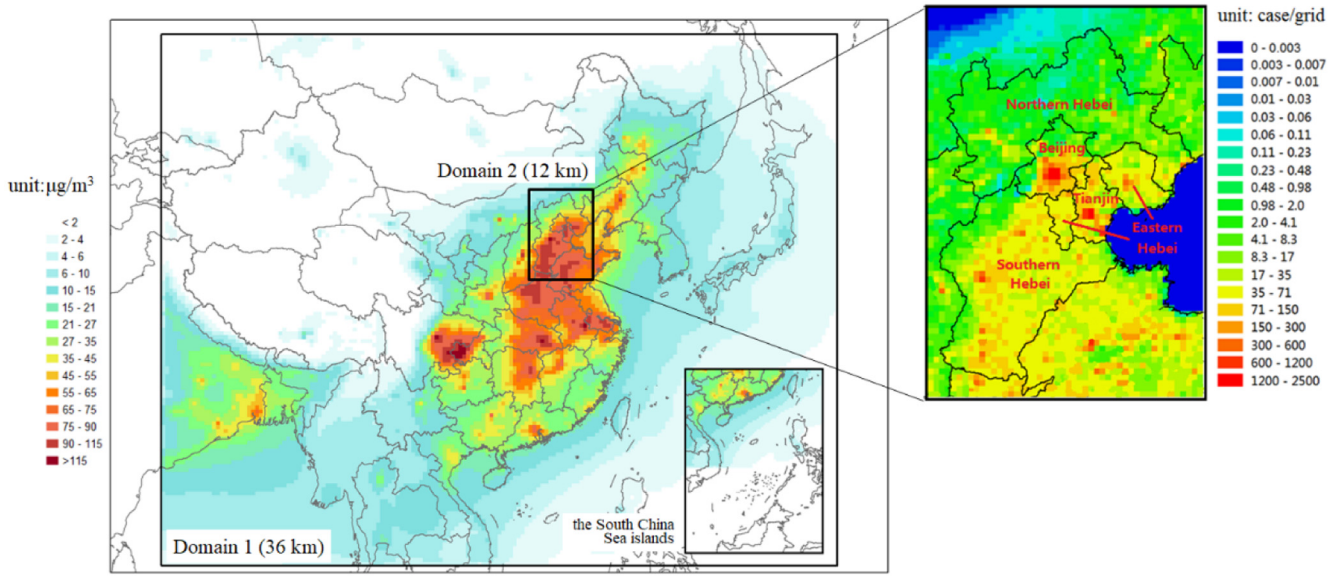

Fig. 1. The simulation domains in this study and simulated annual $PM_{2.5}$ concentrations (left), as well as the spatial distribution of premature mortality by stroke due to long-term exposure to $PM_{2.5}$ pollution in the BTH region (right).

i.e., Beijing, Tianjin, Northern Hebei, Eastern Hebei, and Southern Hebei (see Fig. 1). Next, we used 1121 scenarios simulated by CMAQ/2D-VBS to establish the ERSM prediction system, which maps atmospheric $PM_{2.5}$ concentrations versus emissions of 55 combinations of regions, sectors, and pollutants. We assessed the prediction capability of the ERSM prediction system by employing the "out-of-sample" and 2D-isopleths validation methods, and showed satisfying accuracy and stability.

2.3. Sensitivity of mortality to emission reductions

We calculate the sensitivity of mortality to emission reductions of various sources with the following formula:

$$S_{i,j} = \frac{(H_{i,j} - H_{i,0})/H_{i,0}}{R_j - 1}, (0 \le R_j \le 1) \quad (3)$$

where $S_{i,j}$ refers to the sensitivity of health endpoint i to emission source j; R_j refers to the ratio of emissions from source j to the base-case emissions; $H_{i,j}$ is the health effect of endpoint i when the emission ratio of source j is R_j, which is calculated using the ERSM prediction system developed above; $H_{i,0}$ refers to the health effect in the base case. For each emission source, we calculate $S_{i,j}$ for reduction ratios $(1 - R_j)$ of 10%, 30%, 50%, 65%, 80% to assess the nonlinear relationships between health effects and emission reductions.

There are also other methods to attribute $PM_{2.5}$-related mortality to different emission sources. Global Road Safety Facility et al. (2014) summarized three methods: 1) direct proportion of burden, 2) top of curve, and 3) average risk. This study aims to calculate the changes in mortality in response to emission reductions from various sources. We used Method 2 (top of curve), because this is the only method that mimics how the mortality would actually change if the emissions from certain sources were to be reduced. Global Road Safety Facility et al. (2014) also suggested that "Method 2 is the most appropriate when one is predicting the future change in risk if certain emissions sources were eliminated". Method 1 (direct proportion of burden) or Method 3 (average risk) both introduced certain assumptions so that the health burdens attributed to individual emission sources are independent of the sequence in which they are removed. These two methods are suitable to attribute the current health burden to different sources, but are not applicable to the prediction of change in mortality when emissions are controlled. If Method 1 or Method 3 were applied in this study, the sensitivities of mortality to emission reduction would be quite similar to those of $PM_{2.5}$ concentrations presented in Zhao et al. (2017).

3. Results and discussion

3.1. $PM_{2.5}$-related premature mortality in the BTH region

The premature mortalities by different diseases due to long-term $PM_{2.5}$ exposure in the BTH region are shown in Fig. 2a. According to our assessment, 17.42 (95% CI, 9.45–24.40) thousand, 36.29 (27.24–48.48) thousand, 13.53 (5.19–18.19) thousand, 61.91 (27.71–79.93) thousand, and 0.91 (0.62–1.14) thousand people die of COPD, IHD, lung cancer, stroke, and ALRI due to long-term exposure to $PM_{2.5}$ in the BTH region annually. Stroke is the most important health endpoint, accounting for 48% of the total premature deaths due to long-term exposure. Beijing, Tianjin, and cities in Southern Hebei are areas where people most severely suffer from $PM_{2.5}$-related mortality, and cities in Northern Hebei have the least mortality.

The short-term exposure to $PM_{2.5}$ raises more concerns when heavy pollution episode occurs. As Fig. 2b shows, 18.72 (11.20–25.65) thousand premature mortality is caused by the short-term exposure to $PM_{2.5}$ in the BTH region annually, among which 59% and 16% are by cardiovascular and respiratory diseases, respectively. Similar to long-term effects, the majority of short-term health effects come from Beijing, Tianjin, and cities in Southern Hebei.

We also assess the spatial distribution of health effects due to $PM_{2.5}$ pollution, and the long-term mortality by stroke is shown in Fig. 1. The spatial distribution characteristics of other health endpoints due to either long-term or short-term $PM_{2.5}$ exposures are highly similar (Fig. S2). Comparing the spatial distribution of the health effects with population and $PM_{2.5}$ concentrations (Fig. S1), we can conclude that the health effects are strongly correlated with population intensity. For this reason, high mortality appears in urban areas.

3.2. Nonlinear relationships between air pollutant emissions and mortality

We assess the nonlinearity between air pollutant emission control and $PM_{2.5}$-related mortality using the ERSM technique and the sensitivity analysis method described in Sections 2.1 and 2.2. For long-term exposure, to simplify the analysis, we sum up the premature mortality by COPD, IHD, lung cancer, stroke, and ALRI to approximately represent the total $PM_{2.5}$-related mortality, following previous studies (Cohen et al., 2017; Apte et al., 2015). As shown in Fig. 3, among all pollutants, the annual mortality due to long-term exposure to $PM_{2.5}$ is most sensitive to emissions of primary inorganic $PM_{2.5}$, which is defined as all chemical components of primary $PM_{2.5}$ other than organics, including black carbon, metals, crustal elements, etc. Primary organic aerosol (POA) is treated separately because it experiences chemical oxidations and forms SOA in the CMAQ/2D-VBS model, whereas primary inorganic $PM_{2.5}$ is assumed to be chemically inert. It's worth noting that the estimation methods of the mortality assume that the health effects depend only on the inhaled amount of $PM_{2.5}$ and are independent of the chemical composition, which appears reasonable in view of the available quantitative epidemiological studies. However, some studies have reported that some aerosol species, such as the carbonaceous aerosols (black carbon and organic aerosols), could be significantly more toxic than others (Tuomisto et al., 2008; Lelieveld et al., 2015). The relative toxicity of different aerosol species may affect the relative contributions of primary inorganic $PM_{2.5}$ emissions to premature mortality, which warrants further in-depth study.

Among all sources of primary inorganic $PM_{2.5}$, the industry sector is estimated to make the largest contribution (45–70%), followed by the residential and commercial sectors (20–45%), while the contributions from power plants and transportation are quite small (<15%). The sensitivity of long-term mortality to primary inorganic $PM_{2.5}$ emissions increases gradually by ~20% when the emission reduction ratio increases from 10% to 80%. In contrast, the sensitivity of $PM_{2.5}$ concentrations remains constant regardless of reduction ratio according to our previous study (Zhao et al., 2017). The difference is explained by the fact that the mortality reduction due to a unit drop of $PM_{2.5}$ concentration is larger at lower $PM_{2.5}$ concentration range, according to the curvilinear shape of the IER function. For this reason, emission of primary inorganic $PM_{2.5}$ is expected to be controlled as stringent as possible in the long run to efficiently prevent $PM_{2.5}$-related mortality.

Among the precursors, the long-term mortality is primarily sensitive to the emissions of NH_3, nonmethane volatile organic compounds and intermediate volatility organic compounds (NMVOC+IVOC), and POA. Their relative importance differs by region and reduction ratio (Fig. 3). The large contributions of NMVOC and IVOC are different from the results in many modeling studies (Wang et al., 2015; Wang et al., 2011), primarily because of an improved treatment of SOA formation from these precursors in the CMAQ/2D-VBS. In the present study, we have combined NMVOC and IVOC emissions into an individual control variable. Nevertheless, the O_3 and $PM_{2.5}$ formation potentials of NMVOC and IVOC are quite different (Zhao et al., 2016; Wu et al., 2017). In addition, different control strategies are required to control NMVOC and IVOC since the control technologies designed for NMVOC may not effectively reduce IVOC emissions (Gordon et al., 2014; Jathar et al., 2014). Considering the important contributions of NMVOC and IVOC, further

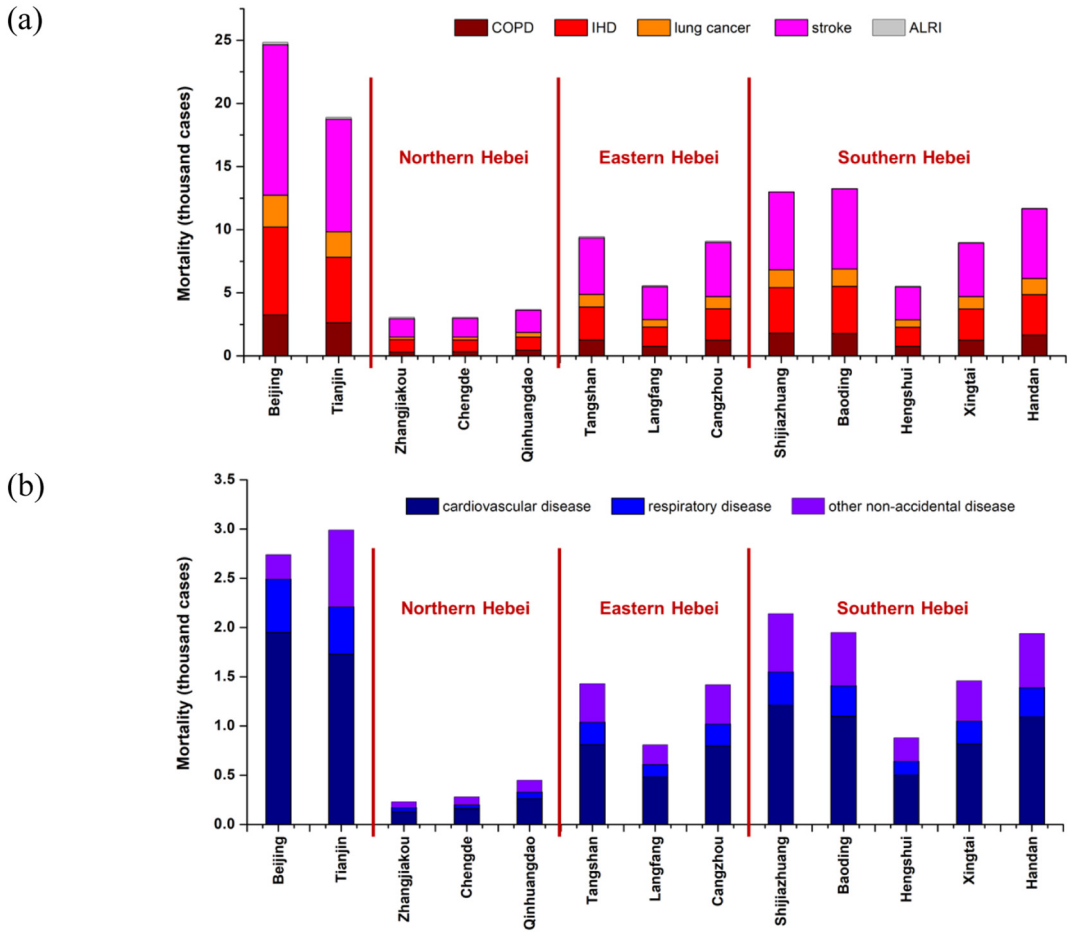

Fig. 2. Annual premature mortality due to long-term (a) and short-term (b) exposures to $PM_{2.5}$ concentrations in the BTH region. Only median values of the mortality estimates are displayed in this figure.

study is needed to separately assess the effects of NMVOC and IVOC emissions from various sectors on premature mortality.

The sensitivities of mortality to emission reductions of POA and NMVOC+IVOC roughly remain unchanged at various reduction ratios, and the sensitivities to SO_2 increase marginally with increasing emission reduction. This is a combined effect of the emission-concentration relationships and the CRFs. First, the sensitivities of $PM_{2.5}$ to emission reductions of NMVOC+IVOC and POA decrease slightly with increasing reduction ratio, and the sensitivities to SO_2 roughly remain invariant, according to Zhao et al. (2017). Second, due to the curvilinear IER function, the sensitivities of mortality to $PM_{2.5}$ concentration increase with decreasing $PM_{2.5}$ concentration. In contrast, the sensitivities to NH_3 emissions increase substantially with the increase of reduction ratio because of 1) an increased sensitivity of $PM_{2.5}$ concentrations due to a transition from NH_3-rich to NH_3-poor conditions (Wang et al., 2011) and 2) an increased marginal mortality benefit at larger reductions. The sensitivities of mortality to NO_x emissions can be either negative or positive, depending on region and reduction ratio — they tend to be negative at small reduction ratio and positive at large reductions, since a small reduction in NO_x emissions is likely to produce more oxidants due to a NMVOC-limited photochemical regime, thus leading to the formation of more secondary inorganic and organic aerosols (Dong et al., 2014; Zhao et al., 2015; Cai et al., 2017). However, the results would be quite different if the NO_x emissions outside the BTH region are jointly controlled. We design a number of sensitivity scenarios in which the NO_x emissions in the whole China are uniformly reduced by 10%–80% (depending on scenario), and re-run the CMAQ/2D-VBS model for two nested domains. The difference between the baseline and sensitivity simulations represents the influence of NO_x emission reductions in the whole China. As shown in Fig. 4, the sensitivities of mortality in the BTH to uniform NO_x reduction in China are positive even at a small reduction ratio. The reason is that NO_x emission reductions in farther regions are more likely to lead to a net decrease in $PM_{2.5}$ concentration than local emission reductions, since the photochemical regime usually changes from NMVOC-limited in local urban areas at ground level to NO_x-limited in downwind regions or higher altitudes where regional transport occurs (Xing et al., 2011). Therefore, it is critically important to enforce stringent NO_x emission controls on national scale.

Regarding different emission sectors, the contributions of SO_2 and NO_x emissions are dominated by non-point sources (Fig. 3b). If all pollutants and sectors are jointly controlled, the overall sensitivity of long-term mortality dramatically increases with reduction ratio, and this sensitivity is remarkably larger than the sum of the sensitivities to emissions of individual pollutants and sectors, mainly because of the curvilinear shape of the IER functions. This is in contrast to the sensitivities of $PM_{2.5}$ concentrations shown in Zhao et al. (2017), where the overall sensitivity is generally similar to the sum of the sensitivities to individual pollutants/sectors. The nonlinearity indicates that extra health benefits will be achieved if a multi-pollutant multi-sector joint control strategy is implemented.

As for short-term exposure, we assess the sensitivities of all-cause mortality to the emissions of various pollutants from various sectors (Fig. 5). The relative contributions of different pollutants and sectors are largely similar to those of long-term exposure. However, the patterns of the nonlinear relationships are quite different. Specifically, when the emission reduction ratio increases from 10% to 80%, the sensitivity of mortality to primary inorganic $PM_{2.5}$ emissions increases so slightly that it seems to be constant. This can be explained by the fact

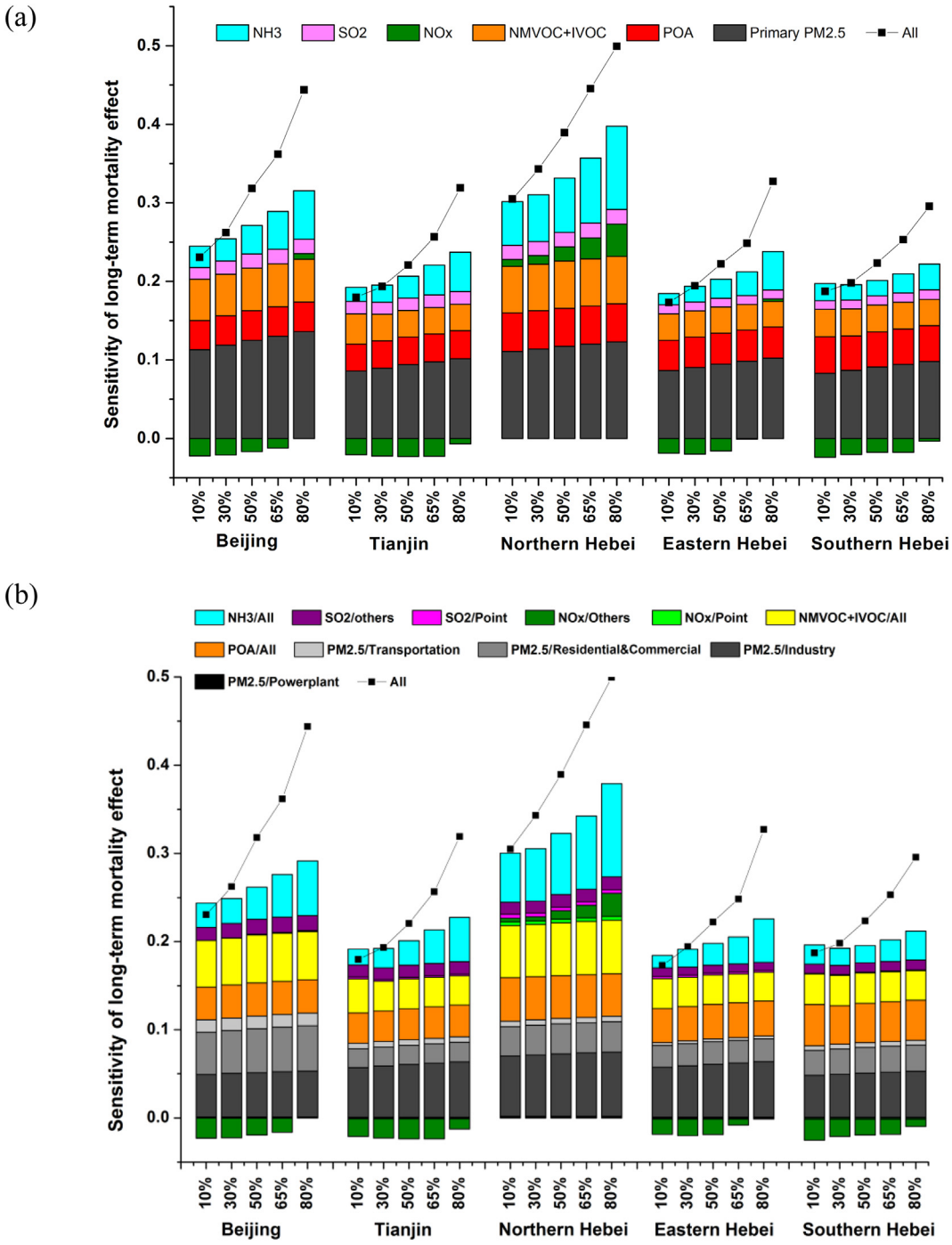

Fig. 3. Sensitivity of annual mortality due to long-term exposure to $PM_{2.5}$ to stepped control of each pollutant (a) and each pollutant-sector combination (b) within the BTH region. The X-axis represents the reduction ratio (i.e., 1 – emission ratio). The Y-axis represents the sensitivity of mortality, as defined by Eq. (3). The colored bars denote the sensitivity of mortality when a specific emission source is controlled while the others remain the base-case level. The black dotted line is the sensitivity of mortality when all emission sources are jointly controlled.

that the nonlinearity of the CRFs for short-term assessment (Eq. (2)) is much weaker than that of the IER function (Eq. (1)). If all pollutants and sectors are jointly controlled, the sensitivity of mortality is similar to or less than the sum of the sensitivities to emissions of individual pollutants and sectors, and the sensitivity only increases slightly with reduction ratio. The finding implies that, to minimize the mortality in the long run, multi-pollutant multi-sector control strategies should be implemented, but this strategy may not be vital for temporary control measures of severe pollution events.

We further analyze the seasonal feature of sensitivity of short-term mortality to emissions of various pollutants and sectors. We choose Beijing and Northern Hebei as examples, as shown in Fig. 5b. The sensitivities show significant variations among different seasons. The emissions of primary $PM_{2.5}$ (including primary inorganic $PM_{2.5}$ and POA) contribute a much larger fraction of mortality in winter (57–70%) than in other seasons (28–42%), mainly as a result of weaker vertical mixing and slower reactions of gaseous precursors (Zhao et al., 2018a; Wang et al., 2011). Among different sources of primary inorganic $PM_{2.5}$, the residential and commercial sector has relatively larger contribution in winter because 1) the use of coal and biomass for heating enlarges the residential emissions in winter, and 2) the weaker vertical mixing in winter favors the accumulation of emissions from low-level sources,

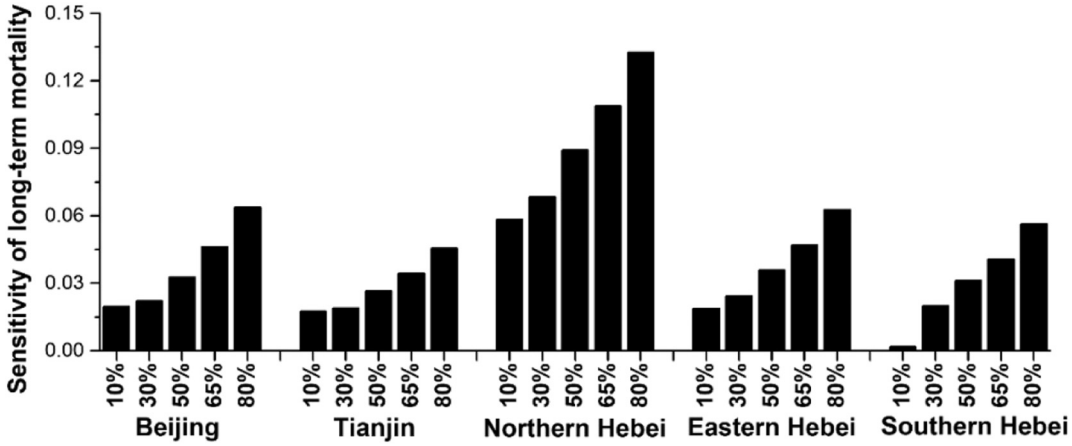

Fig. 4. Sensitivity of annual mortality due to long-term exposure to $PM_{2.5}$ in the BTH region to stepped control of NO_x emissions across the whole China. The X-axis shows the reduction ratio (=1 − emission ratio). The meanings of two axes are the same as in Fig. 3.

resulting in a relatively larger contribution from residential sources. The sensitivities of mortality to gaseous precursors are generally larger in summer than in winter primarily because of accelerated chemical reactions due to stronger radiation and higher temperature. The sensitivities to NO_x are more complicated — they are mostly negative in winter and positive in summer, and change from negative at small reduction to positive at large reduction in spring and autumn, because the photochemistry is prone to be NMVOC-limited in winter and NO_x-limited in summer (Zhao et al., 2013; Zhao et al., 2015; Xing et al., 2017). Therefore, a NO_x emission control strategy that lays emphasis on summer could maximize the public health benefits. A potential strategy is to strengthen the on-site inspection of the operation of NO_x control facilities in summer, especially the denitrification facilities in power plants and industrial sectors. This helps to ensure the normal operation of control equipment and minimize NO_x emissions without permission in summer.

3.3. Nonlinear relationships between region-specific emission control and mortality

We examine the nonlinear relationships between region-specific emission control and health effects by using the same method applied in Section 3.2. Here we select long-term mortality in Beijing as an example, as shown in Fig. 6. The annual mortality in Beijing is most sensitive to local emissions among the five regions, but the relative contributions of local and non-local sources differ significantly by pollutants. For primary inorganic $PM_{2.5}$, POA, and SO_2, local emissions account for more than 60% of the total contributions from all regions. With respect to NH_3 and NMVOC+IVOC, local emissions account for about 50% and one third, respectively. The sensitivity of mortality to local primary inorganic $PM_{2.5}$ emissions increases by ~20% when the reduction ratio increases from 10% to 80%, while the sensitivity to local NH_3 emissions nearly doubles in response to such increase in the reduction ratio. For SO_2, POA, and NMVOC+IVOC, no obvious change in sensitivities with the reduction ratio is noted. For NO_x, the sensitivity to local emissions is negative, whereas the sensitivities to emissions from other regions, especially Southern Hebei from which a large amount of air pollutants are transported to Beijing (Chang et al., 2018), could change from negative to positive when the reduction ratio increases from 10% to 80%.

For each individual pollutant, when emissions from all regions except Beijing are controlled together, the overall sensitivity of mortality nearly equals the arithmetic sum of the sensitivities to each single region, indicating quasi-linear effects of emission control among region outside Beijing. Nevertheless, when emissions from all regions including Beijing are controlled together, the overall sensitivity of mortality exceeds the arithmetic sum of the sensitivities to emissions from each single region, and the differences are more pronounced when the reduction ratio is large and/or multiple pollutants are controlled simultaneously (see the rightmost columns in Fig. 6). The enhanced sensitivity of mortality, which is an integrated effect of the nonlinearity in both atmospheric chemical reactions and the IER functions, implies that additional health benefits will be achieved when multi-pollutant and regional joint control strategies are implemented. Although we use Beijing as an example in the preceding analysis, similar increment in sensitivity due to multi-region and multi-pollutant controls is also found for other regions in the BTH.

3.4. Comparison with previous studies

We compared our results with previous studies which attributed the $PM_{2.5}$-associated health effects to different emission sources (GBD MAPS Working Group, 2016; Hu et al., 2017; Fann et al., 2012; Wang et al., 2015; Heo et al., 2016; Andersson et al., 2009). GBD MAPS Working Group (2016) and Hu et al. (2017) quantified the contributions of different economic sectors to mortality due to long-term $PM_{2.5}$ exposure in China. The two studies both showed that industrial and domestic sectors were the most important emission sources for $PM_{2.5}$-related mortality in the BTH region, which agrees with the results of this study. Nevertheless, this study also assessed the relationships between $PM_{2.5}$-associated mortality in the BTH and emissions of different pollutants from different regions and characterized the nonlinearity of the relationships, which was not investigated in GBD MAPS Working Group (2016) and Hu et al. (2017).

Since limited studies have apportioned the sources of mortality in the BTH region, we also compare with a number of studies for other polluted regions in the world. Wang et al. (2015) evaluated the source contributions of mortality due to short-term $PM_{2.5}$ exposure in the Yangtze River Delta (YRD) region of China. They also found that industrial and domestic sectors were two largest emission sources for $PM_{2.5}$-associated mortality, consistent with the results of this study for the BTH region. Besides, Wang et al. (2015) found that the $PM_{2.5}$-related mortality in YRD was most sensitive to emissions of primary $PM_{2.5}$, followed by NH_3, in agreement with our results for the BTH. However, Wang et al. (2015) reported a negligibly small contribution from NMVOC, while our study reveals that the contributions from NMVOC and IVOC are comparable to those from other gaseous precursors, probably because the CMAQ/2D-VBS model applied in our study significantly improved the simulation of SOA formation. Regarding regional contributions, Wang et al. (2015) showed that the mortality in a specific region was most sensitive to local emissions among all regions within the YRD, which is similar to our finding for Beijing.

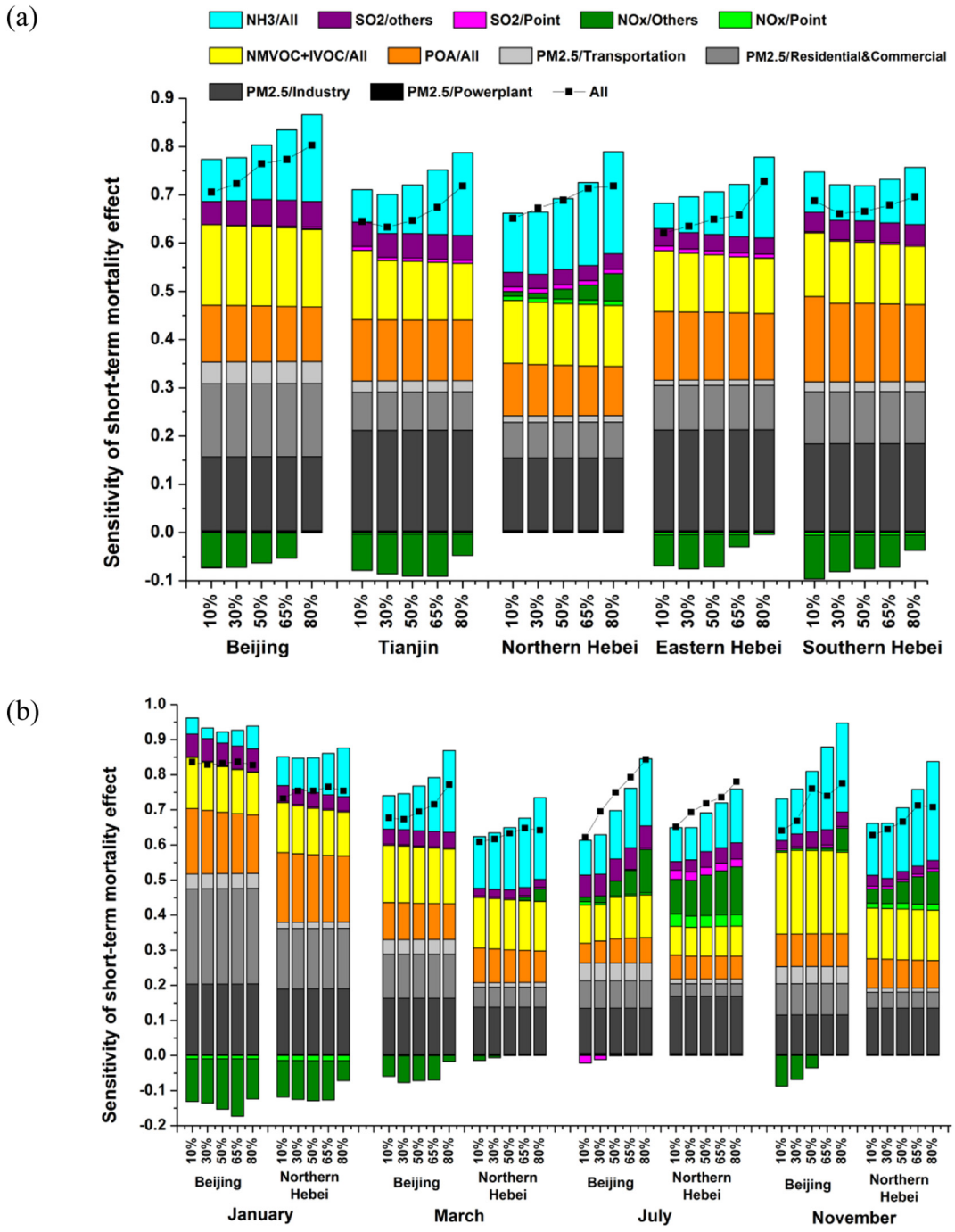

Fig. 5. Sensitivity of annual (a) and monthly (b) short-term $PM_{2.5}$-related mortality to stepped control of each pollutant-sector combination within the BTH region. The meanings of two axes, black dotted lines, and colored bars are the same as in Fig. 3.

Fann et al. (2012) and Heo et al. (2016) estimated the sensitivity of long-term $PM_{2.5}$-related mortality in the United States to emission reductions. They found that the mortality was most sensitive to emissions of primary $PM_{2.5}$, followed by SO_2 and NO_x, and NH_3. This is partially different from our results that the mortality is more sensitive to NH_3 than to SO_2 and NO_x, which can be explained by the differences in emission inventory and photochemical regimes between United States and the BTH region. Heo et al. (2016) further reported a 7% increase in marginal health benefits of SO_2 control and a 26% increase for NO_x from 2005 to 2016 (differences in population and income level between two years have been adjusted), because of significantly reduced SO_2 and NO_x emissions during this period. This is consistent with our finding that the sensitivities of mortality to NO_x/SO_2 emissions increase sharply/marginally with increasing reduction ratio of NO_x/SO_2. Andersson et al. (2009) evaluated the sources of premature deaths in Europe, and concluded that primary $PM_{2.5}$ was associated with more premature deaths than secondary inorganic aerosol (SIA), indicating that it might be more efficient for protecting public health to decrease primary PM emissions than to decrease precursors of SIA. This is in line with our conclusion that primary $PM_{2.5}$ emissions make the largest contribution to $PM_{2.5}$-related mortality and thus should be stringently controlled.

4. Conclusion and implications

In this study, we assessed the nonlinear relationships between $PM_{2.5}$-related mortality and air pollutant emissions in the BTH region

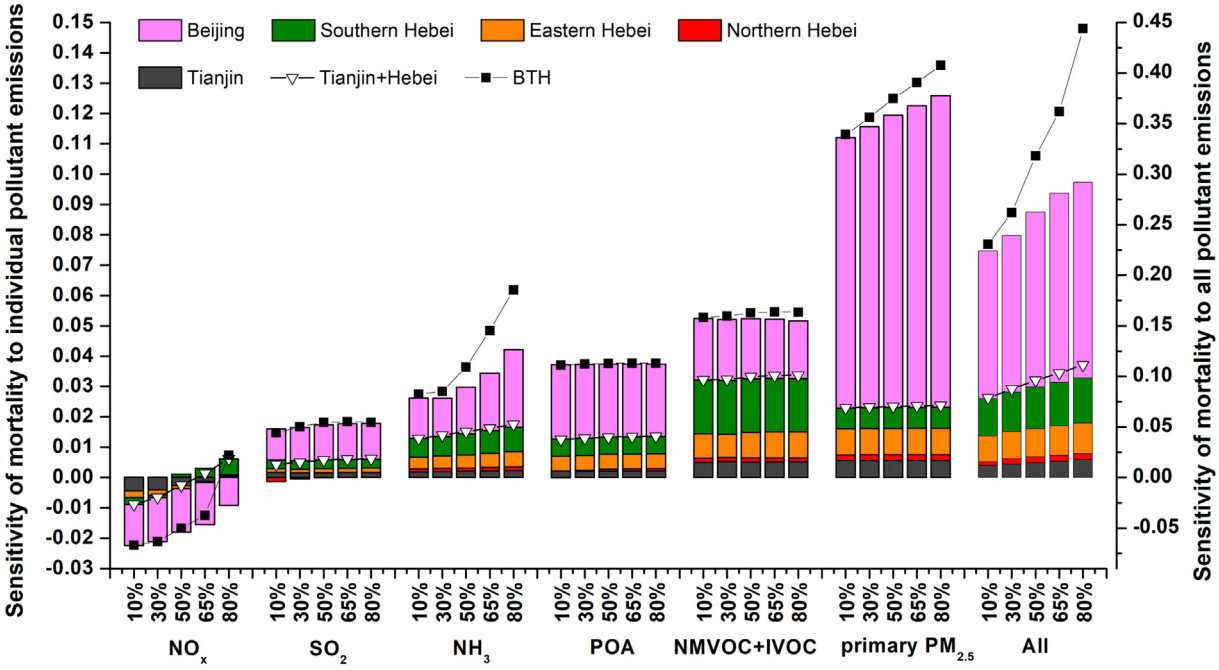

Fig. 6. Sensitivity of annual mortality caused by long-term exposure to $PM_{2.5}$ in Beijing to stepped control of individual/all air pollutants from various regions within the BTH. The meanings of two axes and colored bars are the same as in Fig. 3. The black dotted line denotes the sensitivity of mortality when emissions from all five regions are controlled simultaneously, while the hollow triangle dotted line denotes the sensitivity when emissions from regions except Beijing are controlled simultaneously.

by integrating the CMAQ/2D-VBS model, the ERSM technique, and the CRFs.

The annual premature mortalities attributable to long-term and short-term exposures to $PM_{2.5}$ are estimated at 130.1 thousand and 18.7 thousand, respectively, in the BTH region. The mortalities due to both long-term and short-term $PM_{2.5}$ exposures are most sensitive to the reduction of primary inorganic $PM_{2.5}$ emissions. The sensitivity of long-term mortality to primary inorganic $PM_{2.5}$ increases significantly with reduction ratio, as a result of the curvilinear IER function. In winter, the primary inorganic $PM_{2.5}$ emissions, especially those from residential and commercial sources, contribute much more to short-term mortality than in other seasons. We suggest a sharp reduction in the emissions of primary $PM_{2.5}$ over the BTH region, with a particular focus placed on those from the residential and commercial sectors in winter, which have been largely neglected in the country's control policies until recently.

Among the precursors of $PM_{2.5}$, the $PM_{2.5}$-related mortality is primarily sensitive to emissions of NH_3, NMVOC+IVOC, and POA. Their relative importance differs by region and reduction ratio. The sensitivities of mortality to POA, NMVOC+IVOC and SO_2 do not change much with reduction ratio, whereas the sensitivities to NH_3 and NO_x increase significantly with the increase of reduction ratio. The control of NO_x emissions in BTH tends to increase mortality at small reduction ratio and reduce mortality at large reductions; however, a nationwide NO_x emission control always reduces $PM_{2.5}$-related mortality in the BTH region. The sensitivities of short-term mortality to gaseous precursors are generally larger in summer than in winter. Substantial control policies should be implemented for NO_x and NH_3 emissions in the BTH region. For NO_x, stringent controls on national scale with an emphasis on summertime emissions are suggested to avoid possible side effects and to achieve maximal health benefits.

Regarding the emissions from different regions, the annual $PM_{2.5}$-related mortality in Beijing is most sensitive to local emissions. Over 60% of the contributions from emissions of primary inorganic $PM_{2.5}$, POA, and SO_2 to long-term mortality in Beijing are attributed to local emissions. For NH_3 and NMVOC+IVOC, local emissions account for 50% and one third, respectively. Therefore, the control of local emissions is of primary importance for the reduction of $PM_{2.5}$-related mortality in a specific region. When emissions from local and nonlocal emissions or those of multiple pollutants are controlled simultaneously, the overall sensitivity of mortality due to long-term $PM_{2.5}$ exposure exceeds the arithmetic sum of the sensitivities to emissions of each individual region/pollutant, and the differences are more pronounced for larger reduction ratios. Therefore, in the long run, a multi-pollutant, multi-sector and regional joint control strategy is recommended wherever possible in order to maximize the marginal benefits of public health. In contrast, with respect to short-term mortality, the overall sensitivity resembles the sum of the sensitivities to individual pollutants or regions. For this reason, the jointly control strategy may not be vital for temporary control measures to reduce the mortality induced by severe pollution events; instead, more precisely targeted measures should be developed according to the characteristics of each individual event.

Competing interest statement

The authors declare no conflict of interests.

CRediT authorship contribution statement

Bin Zhao: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Validation, Visualization, Writing - original draft, Writing - review & editing. **Shuxiao Wang:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Writing - original draft, Writing - review & editing. **Dian Ding:** Validation, Visualization, Writing - original draft, Writing - review & editing. **Wenjing Wu:** Formal analysis, Methodology, Validation, Visualization, Writing - original draft, Writing - review & editing. **Xing Chang:** Validation, Visualization, Writing - original draft, Writing - review & editing. **Jiandong Wang:** Investigation. **Jia Xing:** Validation, Visualization, Writing - original draft, Writing - review & editing. **Carey Jang:** Software, Writing - original draft, Writing - review & editing. **Joshua S. Fu:** Software, Writing - original draft, Writing - review & editing. **Yun Zhu:** Writing - original draft, Writing - review & editing. **Mei Zheng:** Validation,

Writing - original draft, Writing - review & editing. **Yu Gu:** Validation, Writing - original draft, Writing - review & editing.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.01.169.

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