Environmental Science & Technology

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Associations between Source-Specific Fine Particulate Matter and Mortality and Hospital Admissions in Beijing, China

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Cite This: *Environ. Sci. Technol.* 2022, 56, 1174−1182

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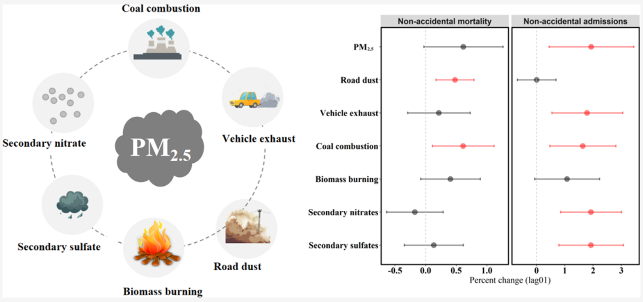
ABSTRACT: The health effects of $PM_{2.5}$ exposure have become a major public concern in developing countries. Identifying major $PM_{2.5}$ sources and quantifying the health effects at the population level are essential for controlling $PM_{2.5}$ pollution and formulating targeted emissions reduction policies. In the current study, we have obtained $PM_{2.5}$ mass data and used positive matrix factorization to identify the major sources of $PM_{2.5}$. We evaluated the relationship between short-term exposure to $PM_{2.5}$ sources and mortality or hospital admissions in Beijing, China, using 441 742 deaths and 9 420 305 hospital admissions from 2013 to 2018. We found positive associations for coal combustion and road dust sources with mortality. Increased hospital admission risks were significantly associated with sources of vehicle exhaust, coal combustion, secondary sulfates, and secondary nitrates. Compared to the cool season, excess mortality risk estimates of coal combustion source were significantly higher in the warm season. Our findings show that reducing more toxic sources of $PM_{2.5}$, especially coal emissions, and developing clean energy alternatives can have critical implications for improving air quality and protecting public health.



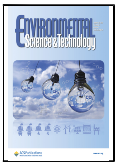
KEYWORDS: *fine particulate matter, source apportionment, mortality, hospital admissions, coal combustion*

INTRODUCTION

The health effects of $PM_{2.5}$ exposure have become a major public concern in China. The 2019 Global Burden of Disease Study (GBD) showed that ambient $PM_{2.5}$ pollution contributed to 1 420 000 deaths in China, accounting for 34.3% of the total disease burden worldwide.[1] With the acceleration of industrialization and urbanization, coal combustion, diesel and gasoline, and solid fuels have gradually become the major sources of increased $PM_{2.5}$ emissions in China, accounting for up to 70% of total emissions.[2,3] China is the world's largest coal consumer, and studies have shown that coal combustion can contribute to more than 60% of $PM_{2.5}$ concentrations in China.[4,5] In addition, motor vehicle ownership in China is second only to the United States.[6] Automobiles consume a lot of fossil fuels, thereby emitting high levels of sulfur and nitrogen oxides, and produce vehicle exhaust, accounting for 17.2−37.3% of $PM_{2.5}$ emissions.[7,8] Moreover, household biomass and solid fuel use are important sources of $PM_{2.5}$ emissions due to low efficient fuel sources and high consumption, resulting in nearly 33−47% of $PM_{2.5}$ in China.[9,10] Therefore, coal combustion, vehicle exhaust, and biomass burning are important sources of $PM_{2.5}$ in China. Identifying the major source of increased $PM_{2.5}$ emissions in China and quantifying the health effects to the population are essential for controlling $PM_{2.5}$ pollution and formulating targeted emission reduction policies.

A limited number of studies have been conducted in developed countries to explore the relationships between $PM_{2.5}$ sources and different critical endpoints.[11−13] However, although developing countries such as China require high energy consumption, which constitutes the primary cause of heavy $PM_{2.5}$ emissions, few studies have explored the health risks associated with specific $PM_{2.5}$ sources in developing countries.[14,15] The characteristics of energy composition, penetration rate of diesel vehicles, and population density of cities in developed and developing countries may lead to different $PM_{2.5}$ emission sources. Since source-specific $PM_{2.5}$ cannot be measured directly, methods such as source allocation models must be used for estimation.[11] Due to differences in chemical composition, $PM_{2.5}$ sources may vary across different areas. Existing studies from developed countries have mainly shown that vehicle exhaust and biomass burning may be major sources responsible for $PM_{2.5}$-related health impacts.[16,17] However, coal combustion as an emissions source is generally lacking in developed countries, while it is the major source of increased $PM_{2.5}$ in developing countries.

Received: October 26, 2021
Revised: December 9, 2021
Accepted: December 10, 2021
Published: December 23, 2021



ACS Publications

https://doi.org/10.1021/acs.est.1c07290
Environ. Sci. Technol. 2022, 56, 1174−1182

As a typical developing country, China has unique characteristics of energy composition. Limited studies have explored the effects of ambient $PM_{2.5}$ sources in China.[14,18] However, there are no studies with a sufficiently large sample size that explore the associations between $PM_{2.5}$ sources and different critical health outcomes in China. Only one study, conducted in Beijing, reported that different sources of $PM_{2.5}$ contribute to increased risk of respiratory emergency room visits (ERVs) to different extents.[14] However, that study only focused on ERVs and had a small sample size, without quantifying the relative risk of mortality and hospital admissions. The exposure data only comes from a monitor on the roof of a building within 500 meters of Peking University Third Hospital, which is a poor representation of the exposure levels in all of Beijing.

As one of the largest cities in China, Beijing has experienced some of the most serious air pollution seen globally in recent years. In the current study, we considered the most serious health outcomes and included close to all mortality and hospital admissions data in Beijing from 2013 to 2018. We evaluated short-term associations between $PM_{2.5}$ sources and mortality or hospital admissions with a large sample size in Beijing, China. We also examined seasonal differences and performed stratified analyses by age and sex to explore which subgroup was the most vulnerable to $PM_{2.5}$ sources.

■ METHODS

Exposure for $PM_{2.5}$ and Components. Daily $PM_{2.5}$ mass was obtained from the Beijing Municipal Environmental Monitoring Center, based on 35 environmental protection fixed monitoring stations in Beijing (2013−2018). The locations of the study monitoring stations are shown in Figure S1. We calculated the daily averages of monitoring data over stations to obtain the city-level $PM_{2.5}$ mass concentrations. $PM_{2.5}$ components were obtained from the $PM_{2.5}$ chemical components monitoring station in the Chaoyang district of Beijing. We chose the main components of $PM_{2.5}$ in Beijing, including organic carbon (OC), elemental carbon (EC), ammonium (NH_4^+), nitrate (NO_3^-), sulfate (SO_4^{2-}), sodium (Na^+), magnesium (Mg^{2+}), potassium (K^+), calcium (Ca^{2+}), and chloride (Cl^-). These components had been identified as potentially harmful in previous epidemiologic studies.[18,19] More details about the air monitoring stations and component stations are provided in the Supporting Information.

Source Apportionment. We used positive matrix factorization (PMF version 5.0) on the $PM_{2.5}$ mass data of 35 environmental monitoring stations to identify the major sources of $PM_{2.5}$. PMF is a factor-based receptor model, which can identify potential pollution source categories and contributions to $PM_{2.5}$ mass.[20] It is a source-unknown model that does not need source profiles as input information to identify the possible source. The principle of PMF is detailed in the Supporting Information. PMF minimizes a "Q" function, which takes into account the uncertainty associated with the daily measurements of each element and imposes the restriction that both source profiles and contributions are non-negative.[21] We also calculated $PM_{2.5}$ sources on the chemical component data of the $PM_{2.5}$ component station in Chaoyang district and $PM_{2.5}$ mass data of the monitoring station nearest the $PM_{2.5}$ component station. For this study, $PM_{2.5}$ sources data from the $PM_{2.5}$ mass at 35 environmental monitoring stations were used as the main result of source appointment. Based on the PMF model, we identified six source categories: road dust, vehicle exhaust, coal combustion, biomass burning, secondary nitrates, and secondary sulfates.

Information on source tracers is required for accurate manual source categories. The details on source profiles are shown in Figure S2. Ca^{2+} and Mg^{2+} are the characteristic markers for road dust;[22] OC, EC, SO_4^{2-}, and Cl^- can be used as tracers to identify coal combustion;[23] NO_3^- and NH_4^+ are tracers of secondary nitrate; SO_4^{2-} and NH_4^+ are tracers of secondary sulfate;[24] OC, EC, and Mg^{2+} are good indicators of vehicle exhaust;[25] and biomass burning is an important source for Na^+, K^+, and Cl^-.[26]

Health Data. We obtained daily mortality and hospital admissions data between 2013 and 2018 from the China's Disease Surveillance Points system (DSPs) and Beijing Municipal Health Commission Information Center. According to diagnosis codes in the International Classification of Diseases, 10th Revision (ICD-10), nonaccidental mortality and hospital admissions (ICD-10: A00−R99), cardiovascular mortality and hospital admissions (ICD-10: I00−I99), and respiratory mortality and hospital admissions (ICD-10: J00−J99) were considered in this study. For cause-specific mortality data, we divided into subgroups by sex (male and female) and age (<65, 65−74, and >74 years). For hospital admissions, we also analyzed subgroups and classified cause-specific hospital admissions by sex (male and female) and age (<15, 15−64, 65−74, and >74 years).

Meteorological Data. We obtained daily temperature and relative humidity data from the Chinese Meteorological Bureau, based on Beijing station no. 54511. The location of the weather station is shown in Figure S1.

Statistical Analysis. We performed a descriptive analysis of $PM_{2.5}$ mass and sources, daily mortality and hospital admissions, and meteorological data.

We conducted a two-stage time series study using generalized additive models with a quasi-Poisson regression. Temperature, relative humidity, day of the week, and temporal trend were controlled in the main model. We used a natural cubic spline with 5 degrees of freedom per year to control for long-term and seasonal trends, and 3 degrees of freedom to control temperature and relative humidity for each. The model can be expressed in the formula as

$$\begin{aligned}\log E(Y_t) = \text{intercept} + \beta Z_t + \text{ns}(\text{time}, df)\\ + \text{ns}(\text{temperature}, df) + \text{ns}(\text{humidity}, df)\\ + \text{dow} + \text{holiday}\end{aligned}$$

where $E(Y_t)$ is the number of deaths and hospital admissions on day t, β is the regression coefficient of a certain $PM_{2.5}$ source estimated by the model, Z_t represents the concentrations of $PM_{2.5}$ sources on day t, ns is a natural-spline function of the meteorological indicators and temporal trend, dow is an indicator variable for day of the week, and holiday represents an indicator variable for major federal holidays.

Based on the results of previous studies, stronger associations between mortality and hospital admissions and $PM_{2.5}$ sources have been reported for current and previous day (lag 01) or current and previous 2 days (lag 02),[16,27] so we considered the lag structure of $PM_{2.5}$ sources for single-day lags (lag 0, lag 1, lag 2, and lag 3) and for cumulative lags (lag 01, lag 02, and lag 03). We also stratified by age, sex, and seasons (warm (April−September) and cool (October−March)). A Z-test was used to examine whether the risk of $PM_{2.5}$ sources in

Table 1. Descriptive Statistics of Daily Mortality, Daily Hospital Admissions (counts), $PM_{2.5}$ Mass, $PM_{2.5}$ Sources, and Meteorological Data in Beijing from 2013 to 2018

variable	mean	SD	min	median	max	IQR
health data						
nonaccidental mortality	202	29	131	198	313	36
cardiovascular mortality	93	18	48	91	172	24
respiratory mortality	22	7	4	21	56	8
nonaccidental admissions	4300	1893	874	4632	11109	3554
cardiovascular admissions	947	396	217	1019	2477	737
respiratory admissions	511	191	151	539	1189	342
$PM_{2.5}$ mass and sources (daily concentration, $\mu g/m^3$)						
$PM_{2.5}$	74.86	63.21	4.29	57.22	423.19	67.28
road dust	3.16	4.50	0.00	2.09	64.23	3.05
vehicle exhaust	11.76	9.77	0.00	9.62	69.90	8.56
coal combustion	7.22	6.44	0.00	5.81	62.51	5.93
biomass burning	4.55	6.70	0.00	1.98	78.92	6.61
secondary nitrate	21.57	26.24	0.00	14.39	235.09	22.94
secondary sulfate	24.20	26.43	0.00	16.60	207.84	23.26
meteorological indicators						
temperature (°C)	13.76	11.22	−14.3	15.35	32.6	21.2
relative humidity (%)	52.06	19.99	8	52	99	32

the subgroups was statistically different. The statistical tests were one-sided, and p-values <0.05 were considered statistically significant. A separate analysis of the "key" tracer of the identified source was conducted to verify the health impact of the corresponding source.[27]

We performed a series of sensitivity analyses. First, we changed the df of time (df = 6, 7) and meteorological parameters (df = 5) in the natural cubic regression spline to assess model stability. Second, to investigate possible exposure misclassification, we estimated the risk of $PM_{2.5}$ sources calculated based on $PM_{2.5}$ component data and $PM_{2.5}$ mass data of the nearest monitoring station.

All statistical analyses were performed using the R Statistical Software, version 4.0.2.

■ RESULTS

During the study period from 2013 to 2018 in Beijing, there were 441 742 deaths and 9 420 305 hospital admissions from nonaccidental causes, of which 203 648 deaths and 2 075 898 hospital admissions were related to cardiovascular disease, and 47 943 deaths and 1 120 353 hospital admissions were related to respiratory disease, respectively. Descriptive statistics in terms of $PM_{2.5}$ mass and sources, cause-specific mortality and hospital admissions, and meteorological variables are shown in Table 1. The average daily $PM_{2.5}$ concentration from 2013 to 2018 was 74.86 $\mu g/m^3$, and the main sources were secondary sulfates (32.19% of the total), secondary nitrates (28.69%), vehicle exhaust (15.64%), coal combustion (9.6%), biomass burning (6.05%), and road dust (4.2%). The daily average concentration of each $PM_{2.5}$ component is shown in Table S1. Table S2 compares the concentration and contribution of three sources data. There is a negligible change in the source contribution. The correlation coefficients between $PM_{2.5}$ sources are shown in Figure S3.

Full results for all lags are provided in the Supporting Information (Figures S4 and S5), and the strongest estimates were observed at lag 0−1 day. Figure 1 shows the estimates of $PM_{2.5}$ mass and sources on mortality or hospital admissions at lag 0−1 day. Briefly, significant associations were observed for road dust and nonaccidental mortality (0.48% (95% CI: 0.17, 0.79)) and coal combustion and nonaccidental mortality (0.61% (95% CI: 0.11, 1.12)). Only road dust was associated with respiratory mortality (1.45% (95% CI: 0.63, 2.27)). For hospital admissions, per interquartile range (IQR) increases in $PM_{2.5}$, vehicle exhaust, coal combustion, secondary sulfates, and secondary nitrates were associated with 1.93% (95% CI: 0.44, 3.44), 1.79% (95% CI: 0.54, 3.05), 1.63% (95% CI: 0.47, 2.81), 1.92% (95% CI: 0.79, 3.07), and 1.92% (95% CI: 0.85, 3.01) increase in nonaccidental admissions at lag 0−1 day. $PM_{2.5}$, vehicle exhaust, coal combustion, secondary sulfates, and secondary nitrates were associated with cardiovascular admissions: 2.41% (95% CI: 0.94, 3.9), 2.08% (95% CI: 0.85, 3.32), 1.99% (95% CI: 0.83, 3.15), 2.2% (95% CI: 1.07, 3.33), and 2.16% (95% CI: 1.1, 3.22), respectively. Coal combustion was positively associated with respiratory admissions (1.11% (95% CI: 0.15, 2.07)).

In the seasonal stratified analyses (Figure 2), we observed that excess mortality risk estimates of road dust were mostly greater in the cool season (0.61% (95% CI: 0.16, 1.07)). Vehicle exhaust, coal combustion, and biomass burning sources were associated with higher risk estimates of nonaccidental mortality in the warm season: 0.52% (95% CI: −0.98, 2.05), 1.42% (95% CI: 0.22, 2.63), and 0.99% (95% CI: −0.62, 2.64), respectively. Stronger risks of nonaccidental admissions were observed in the cool season: an IQR increase in vehicle exhaust, coal combustion, secondary sulfates, and secondary nitrates were associated with 1.75% (95% CI: 0.18, 3.35), 1.5% (95% CI: 0.01, 3.02), 2.1% (95% CI: 0.53, 3.69), and 1.85% (95% CI: 0.39, 3.33) increase, respectively. Biomass burning was associated with higher risk estimates of nonaccidental admissions in the cool season (0.87% (95% CI: −0.54, 2.3)).

The results of the stratified analysis by age and sex are shown in Table 2. For the sex-stratified analysis, the estimated risks of $PM_{2.5}$ sources with mortality were frequently higher in females than in males, while the estimated risks of $PM_{2.5}$ sources with hospital admissions were higher in males than in females, although the difference is not significant. We also found that the 65−74 years age group presented a greater risk

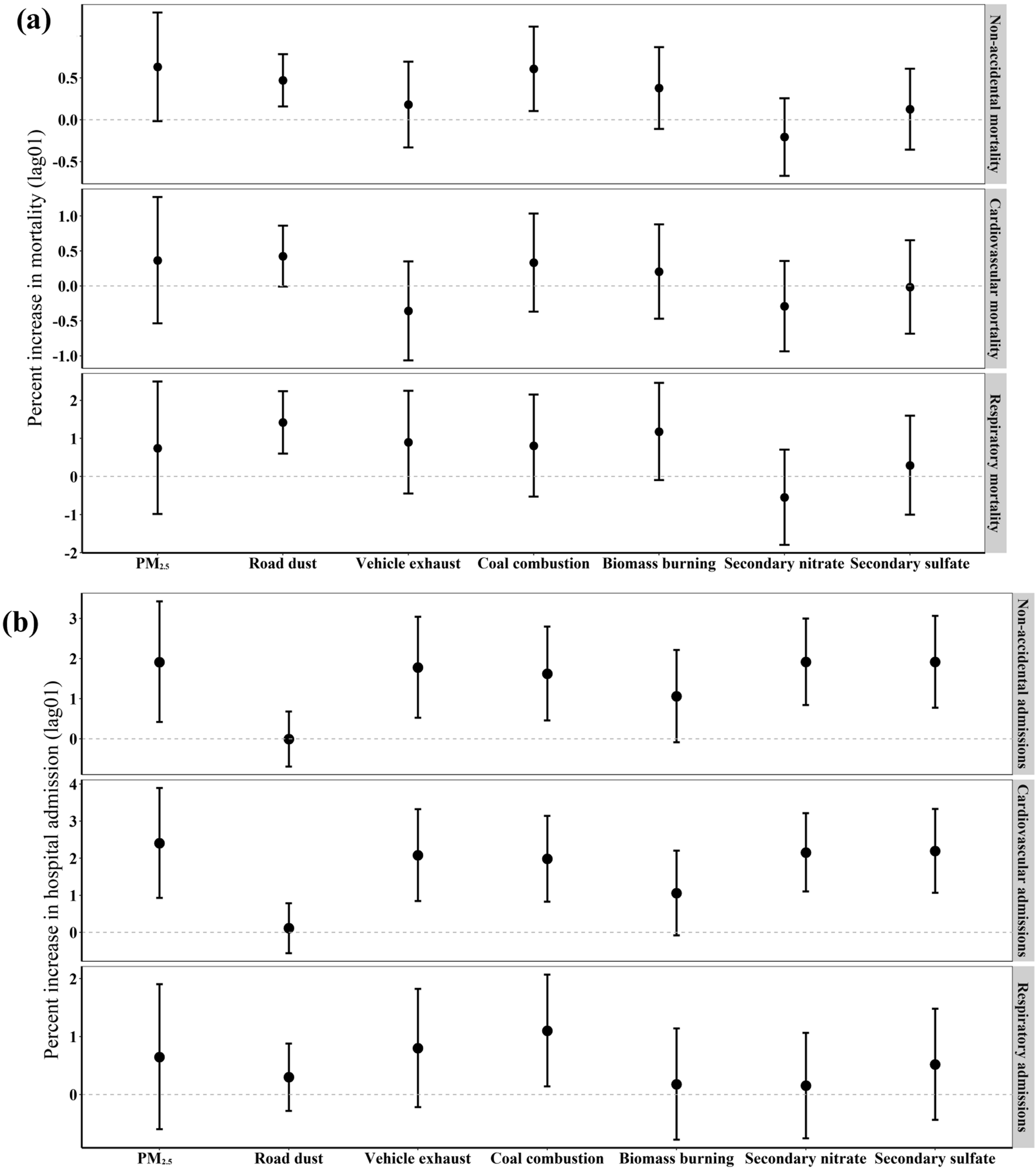


Figure 1. Estimated risks of $PM_{2.5}$ sources (per IQR increase) for (a) mortality and (b) hospital admissions (95% CI) in Beijing from 2013 to 2018 (lag 01).

with vehicle exhaust, coal combustion, secondary nitrates, and secondary sulfates than that of younger age groups.

Based on the details of source profiles, we selected key source markers to correspond to the identified sources, including Ca^{2+} for road dust, OC for vehicle exhaust, EC for coal combustion, Cl^- for biomass burning, NO_3^- for secondary nitrate, and SO_4^{2-} for secondary sulfate. Although none of these components are produced solely from a specified source, they can be used to help validate and interpret the results of specific sources. We yielded similar findings using these key element tracers (Table 3).

As a sensitivity analysis, we examined the contribution and the risks of $PM_{2.5}$ sources calculated by $PM_{2.5}$ components and $PM_{2.5}$ mass in the Chaoyang district of Beijing. Relative to the main results, there is a negligible change in the source contribution (Table S2), and the health risks of $PM_{2.5}$ sources were relatively robust (Table S3). We also changed the degrees of freedom of the time trend and meteorological indicators in the model. These results (Table S4) show that when the degree of freedom changes, the estimated effects of $PM_{2.5}$ sources were almost identical.

■ DISCUSSION

This study explores the health effects of $PM_{2.5}$ sources in Beijing, China. We found that coal combustion and road dust sources were significantly associated with increases in mortality risks. Increased hospital admission risks were significantly associated with sources of vehicle exhaust, coal combustion, secondary sulfates, and secondary nitrates. We also found that the increased risks associated with $PM_{2.5}$ sources under different seasonal patterns were different. The excess mortality risk estimates of coal combustion were significantly higher in

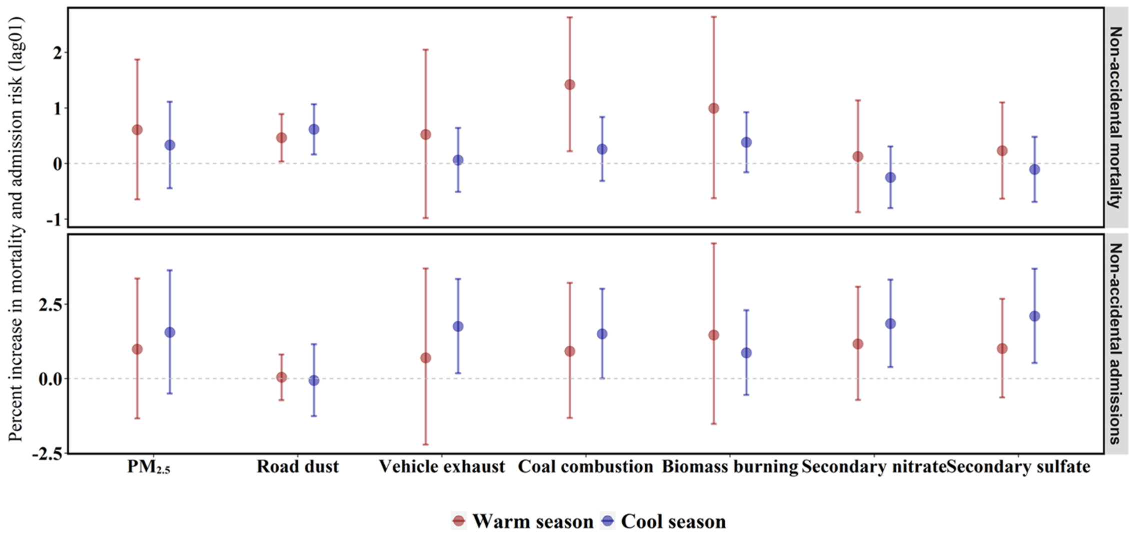


Figure 2. Estimated risks (per IQR increase) of season stratified sources of $PM_{2.5}$ in Beijing from 2013 to 2018 (lag 01).

the warm season. The risk of vehicle exhaust was stronger in the cool season than in the warm season. Coal combustion had higher risk estimates for hospital admissions in the cool season, although the difference is not significant. Our findings provide initial evidence that consumption of coal in developing countries may be a major source of increased $PM_{2.5}$ emissions and that the development of clean energy may help formulate targeted pollution controlling strategies to reduce $PM_{2.5}$ emissions in the atmosphere.

Our findings have been well supported in the literature.[11,14] Limited studies have quantified the relationship between $PM_{2.5}$ sources and mortality,[17,28] while numerous studies have explored the associations between $PM_{2.5}$ components and mortality,[19] which were related to secondary sulfates, secondary nitrates, and traffic sources. A study of short-term exposure to $PM_{2.5}$ sources found that fuel oil combustion, road dust, vehicle exhaust, minerals, and traffic were significantly associated with nonaccidental mortality, and associations were also observed between secondary sulfate sources and cardiovascular mortality.[28] Our study found that coal combustion and road dust sources were significantly associated with nonaccidental mortality. The importance of these sources in Beijing may be especially attributable to greater population density and exposure to coal combustion and traffic in the winter. Previous studies between $PM_{2.5}$ components and mortality suggested that mortality was associated with SO_4^{2-}, NO_3^-, OC, and EC, which is consistent with the findings of our tracer components analysis.[19,29]

Recent studies have examined $PM_{2.5}$ components or sources and hospital admissions.[27,30,31] A study of hospital emergency department (ED) visits conducted in Atlanta, Georgia found associations between biomass burning and mobile sources with both cardiovascular and respiratory ED visits, and between secondary sulfate source and respiratory visits.[30] In the present study, we consistently observed significant associations between sources of coal combustion, secondary sulfates, and secondary nitrates with hospital admissions, consistent with previous studies.[13,27,31] The sources of secondary sulfates and secondary nitrates are gaseous pollutants (including SO_x, NO_x, and volatile organic compounds, VOCs, which are all combustion products related to vehicle exhaust emissions) formed by chemical reactions in the atmosphere.[32] A study in Boston and New York also found associations between cardiovascular admissions and traffic-related $PM_{2.5}$ in the elderly.[33] Vehicular emissions were also associated with cardiovascular emergency room visits in California.[31] Further, $PM_{2.5}$ source markers provided similar associations with hospital admissions. We found that exposures to OC, EC, NO_3^-, and SO_4^{2-} were related to hospital admissions, which were the tracer species of coal combustion, vehicle exhaust, secondary sulfates, and secondary nitrates. The health risks of these tracer components to hospital admissions are consistent with the percent change of the source, indicating that the health risk of the source may be driven by these components. In this analysis, we can find that the effects of different $PM_{2.5}$ sources on mortality and hospital admissions are synergistic. However, the estimated risks of $PM_{2.5}$ sources on hospital admissions are slightly higher than the estimated risks of mortality. This may be because hospital admission, as a relatively mild health outcome, is a more sensitive measurement of health.

We found positive associations between coal combustion, vehicle exhaust, secondary sulfates, and secondary nitrates and admissions, which were significant in the sex-stratified and age-stratified analyses. Ebisu et al. explored the relationship between $PM_{2.5}$ sources and age-specific hospital admissions, which found that biomass burning and vehicular emissions of $PM_{2.5}$ were in associations with hospital admissions of children and the elderly.[31] Different from Ebisu et al., we only found that the estimated risks of road dust with nonaccidental hospital admissions were frequently greater in the 0−15 years age group than in other groups, while the risks of other sources, such as sources of vehicle exhaust, coal combustion, secondary nitrates, and secondary sulfates with nonaccidental hospital admissions, were higher in the 65−74 years age group than in other age groups. This may be partly due to the fact that children are more susceptible to road dust, particularly hospital admissions, while the elderly are less susceptible. Second, children play more often outdoors, which increases their exposure to road dust relative to other age groups. In addition, elderly people are more susceptible and may have various chronic diseases, and the risk of mortality related to $PM_{2.5}$ sources may be the dominant health impact.

Table 2. Percent Increase (95% CI) in Nonaccidental Mortality and Nonaccidental Hospital Admissions by Sex and Age Group Associated with an IQR Increase at lag 01

	nonaccidental mortality					nonaccidental hospital admissions					
	sex		age (years)			sex		age (years)			
$PM_{2.5}$ source	male	female	<65	65−74	>74	male	female	<15	15−64	65−74	>74
$PM_{2.5}$	0.2 (−0.58, 0.98)	1.16 (0.25, 2.08)	−0.53 (−1.7, 0.65)	1.8 (0.45, 3.16)[a]	0.71 (−0.09, 1.52)[a]	2.14 (0.5, 3.81)	1.77 (0.38, 3.18)	−0.17 (−1.14, 0.81)	1.87 (0.35, 3.42)[a]	2.94 (1.12, 4.8)[a]	1.95 (0.43, 3.5)[a]
road dust	0.41 (0.04, 0.79)	0.56 (0.13, 1)	0.29 (−0.28, 0.86)	0.29 (−0.36, 0.93)	0.6 (0.22, 0.99)	0.04 (−0.71, 0.8)	−0.03 (−0.67, 0.62)	0.5 (0.04, 0.96)	−0.04 (−0.75, 0.66)	−0.1 (−0.93, 0.73)	0.06 (−0.63, 0.75)
vehicle exhaust	0.01 (−0.6, 0.63)	0.48 (−0.24, 1.2)	−0.3 (−1.24, 0.64)	0.34 (−0.72, 1.42)	0.37 (−0.26, 1)	1.99 (0.61, 3.39)	1.64 (0.47, 2.82)	−0.1 (−0.9, 0.71)	1.72 (0.44, 3.01)[a]	2.53 (0.98, 4.09)[a]	2.03 (0.75, 3.32)[a]
coal combustion	0.3 (−0.31, 0.91)	1.02 (0.31, 1.73)	0.06 (−0.86, 0.99)	1.33 (0.28, 2.4)[a]	0.62 (0, 1.24)	1.9 (0.61, 3.2)	1.43 (0.35, 2.53)	0.6 (−0.15, 1.36)	1.54 (0.35, 2.74)	2.28 (0.85, 3.72)[a]	1.73 (0.54, 2.94)
biomass burning	0.39 (−0.2, 0.98)	0.42 (−0.26, 1.11)	−0.08 (−0.97, 0.82)	0.72 (−0.28, 1.74)	0.49 (−0.11, 1.1)	1.22 (−0.05, 2.49)	0.97 (−0.09, 2.05)	0.19 (−0.58, 0.96)	1.12 (−0.04, 2.29)	1.4 (−0.01, 2.83)	0.94 (−0.23, 2.13)
secondary nitrates	−0.35 (−0.9, 0.21)	0.04 (−0.61, 0.7)	−0.64 (−1.48, 0.21)	0.54 (−0.41, 1.51)[a]	−0.21 (−0.78, 0.37)	2.18 (0.99, 3.38)	1.74 (0.74, 2.75)	0.17 (−0.54, 0.89)	2 (0.91, 3.11)[a]	2.57 (1.27, 3.9)[a]	1.71 (0.62, 2.82)[a]
secondary sulfates	−0.11 (−0.69, 0.48)	0.44 (−0.24, 1.12)	−0.16 (−1.03, 0.72)	0.99 (0, 2)[a]	0 (−0.6, 0.6)	2.13 (0.88, 3.41)	1.77 (0.7, 2.84)	−0.5 (−1.25, 0.25)	2 (0.84, 3.17)[a]	2.74 (1.34, 4.16)[a]	1.77 (0.6, 2.95)[a]

[a] Statistically significant differences, compared with <65 or <15 subgroup.

Table 3. Percent Increase (95% CI) for Mortality and Hospital Admissions per IQR Increment for Trace Element $PM_{2.5}$ Components

trace element $PM_{2.5}$ components	pollution sources	nonaccidental mortality	cardiovascular mortality	respiratory mortality	nonaccidental admissions	cardiovascular admissions	respiratory admissions
Ca^{2+}	road dust	0.29 (0.05, 0.53)	0.18 (−0.17, 0.52)	1.15 (0.51, 1.8)	−0.14 (−0.65, 0.37)	−0.08 (−0.58, 0.43)	0.15 (−0.3, 0.59)
OC	vehicle exhaust	0.37 (−0.18, 0.92)	−0.17 (−0.93, 0.6)	1.15 (−0.3, 2.62)	2.08 (0.75, 3.43)	2.48 (1.17, 3.81)	1.07 (0, 2.16)
EC	coal combustion	0.54 (0.03, 1.05)	0.25 (−0.47, 0.97)	0.68 (−0.67, 2.05)	1.51 (0.35, 2.69)	1.94 (0.77, 3.11)	1.08 (0.12, 2.05)
Cl^-	biomass burning	0.09 (−0.33, 0.52)	−0.13 (−0.72, 0.46)	0.74 (−0.37, 1.87)	1.12 (0.13, 2.11)	1.16 (0.18, 2.14)	0.09 (−0.74, 0.92)
NO_3^-	secondary nitrates	−0.07 (−0.56, 0.42)	−0.21 (−0.9, 0.48)	−0.24 (−1.57, 1.1)	1.9 (0.82, 2.99)	2.16 (1.09, 3.25)	−0.07 (−1.01, 0.89)
SO_4^{2-}	secondary sulfates	0.09 (−0.46, 0.65)	0.01 (−0.76, 0.78)	0.02 (−1.48, 1.54)	2.24 (0.97, 3.53)	2.67 (1.4, 3.95)	0.39 (−0.69, 1.5)

Seasonal patterns in the association with $PM_{2.5}$ sources and mortality or admissions were observed in our study. Particulate composition varies seasonally, toxic components appear in specific time periods, and the sources of $PM_{2.5}$ vary depending on the composition of $PM_{2.5.}$[34,35] Our study shows that road dust had a stronger association with mortality in the cool season. Severe weather conditions in the cool season may lead to increased concentrations of some sources. In Beijing, Ca^{2+}, the key tracer of road dust, was mainly from soil dust and building materials.[36] In addition, we also found that sources of vehicle exhaust, coal combustion, secondary sulfates, secondary nitrates, and biomass burning were more strongly associated with hospital admission in warm seasons, while these sources were more associated with mortality in cool seasons. Several hypotheses may explain these results. Warm seasons may promote people to spend more time outdoors, which may lead to less exposures misclassifications than in cool seasons. Also, warm season may encourage people to open windows for longer periods of ventilation, making it easier for $PM_{2.5}$ to penetrate into indoor settings.[37]

This study has several limitations. The $PM_{2.5}$ component concentrations in this study were based on the ambient fixed monitoring site to represent population-level exposure, which might result in exposure misclassification. A study reported that personal exposure only accounts for 68% of $PM_{2.5}$, which indicates that studies using ambient concentrations may overestimate personal exposure to air pollutants, which in turn may lead to biased estimates of health effects.[38] The lack of information on individual time activities in this study is another potential source of misclassification. For example, people who work in a heavy traffic environment may have higher exposure to vehicle emissions. However, this is a known limitation for most time series studies of air pollution and health that is challenging to address.[13,38] Changes in the chemical compositions of $PM_{2.5}$ transported from outdoor to indoor may change the relative contribution of different $PM_{2.5}$ sources to indoor $PM_{2.5}$. Study has shown that air infiltration could affect the source contribution of the $PM_{2.5}$ exposure. However, the difference between the contribution of outdoor sources and the contribution of indoor sources is less than 3%, which may slightly alter health effects.[39] In addition, there is no gold standard source apportionment method in existing studies. However, results from epidemiologic studies show that no matter which method is used to identify the source of $PM_{2.5}$, the associations are consistent.[37] Although there is a problem of collinearity among $PM_{2.5}$ sources, in the current air pollution epidemiology, the generalized additive model is widely used in exploring the health risks of $PM_{2.5}$ sources and components, such as Kim et al. and Halonen et al.[40,41] Collinearity between pollutants is still a challenge in the field of environmental health, and researchers are currently working on mixed exposure models to address this issue.

Our study helps clarify which $PM_{2.5}$ sources lead to greater health risks and which sources should be further regulated. This study indicates that specific $PM_{2.5}$ sources, such as coal combustion, vehicle exhaust, and road dust, can play a vital role in the increasing risks of mortality and admissions, while these sources of $PM_{2.5}$ are almost completely produced by dirty energy consumption. Our findings suggest that different sources lead to different health risks in different seasons and some sources may lead to greater risks for children and the elderly. Our study indicates that reducing coal emissions in developing countries and developing clean energy have critical implications for improving air quality and protecting public health. The findings of this study suggest that stricter regulations on more toxic sources of $PM_{2.5}$ will be a more effective way of protecting the public's health and can provide a useful reference for other developing countries.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.1c07290.

> Details of air monitoring stations and source apportionment methods; map of study area and locations of fixed-site monitors (Figure S1); factor profile of each source (Figure S2); correlation coefficients between $PM_{2.5}$ sources (Figure S3); estimated effects of nonaccidental mortality (Figure S4); estimated effects of nonaccidental hospital admissions (Figure S5); summary of $PM_{2.5}$ constituents included in PMF source apportionment analysis (Table S1); source concentrations and contribution of each $PM_{2.5}$ source (Table S2); estimated risk for nonaccidental mortality and hospital admissions (Table S3); and percent increase in mortality and hospital admissions (Table S4) (PDF)

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Author Contributions

T.L. made contributions to the concept and design of the study. H.D. performed data analysis and manuscript writing. Y.L. performed data cleaning and preprocessing. F.W. performed the source apportionment. Y.L., F.W., and M.Z.H. conducted writing—review and editing. G.S. and T.L. guided analysis methods and writing—review and editing. All authors contributed to the interpretation of the results and critical revisions.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This study was supported by the National Natural Science Foundation of China (Grant Numbers 92143202, 92043301, and 42077191).

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https://doi.org/10.1021/acs.est.1c07290