Health Risk Assessment of PM_{2.5}-bound Components in Beijing, China during 2013–2015

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ABSTRACT

Risk assessment methods of the US Environment Protection Agency (U.S. EPA) were employed to estimate lifetime cancer risk in Beijing using the following fine particulate matter (PM_{2.5}) components: six elements and 16 U.S. EPA priority polycyclic aromatic hydrocarbons (PAHs), and lifetime non-cancer hazard quotients (HQ) using 11 elements, based on data collected in Beijing during 2013–2015. The three-year average PM_{2.5} mass concentration was 82 $\mu\text{g m}^{-3}$. Both lifetime cancer risk ($1.9\text{E-}4$) from exposure to ambient PM_{2.5}-bound elements and PAHs and non-cancer HQ (18) from exposure to ambient PM_{2.5}-bound elements in Beijing were much higher than the corresponding U.S. EPA acceptable levels. Cancer risks by source were, in descending order, road dust ($7.3\text{E-}5$), fossil fuel combustion ($4.4\text{E-}5$), vehicle exhaust ($3.8\text{E-}5$), soil dust ($8.4\text{E-}6$), metal processing ($8.2\text{E-}6$), secondary sulphur ($8.0\text{E-}6$), and biomass burning ($6.4\text{E-}6$). Cancer risks by PM_{2.5} component were, in descending order, As ($1.1\text{E-}4$), Cr(VI) ($3.4\text{E-}5$), total PAHs ($1.5\text{E-}5$), Pb ($1.1\text{E-}5$), Co ($8.4\text{E-}6$), Ni ($3.9\text{E-}6$), and Cd ($3.9\text{E-}6$). HQ by PM_{2.5}-bound elements were, in descending order, Cl (14), As (1.8), Pb (0.94), P (0.81), Cd (0.22), Mn (0.22), Ni (0.18), Ba (0.1), Cr(VI) (0.03), Co (0.01), and Se (0.002). Fossil fuel combustion and vehicle exhaust were the top two sources, accounting for 77% of total HQ. HQ by target organ were, in descending order, respiratory (15), reproductive (1.8), nervous (1.2), renal (0.22), fetus (0.1), and alimentary system (0.002). The seasonal variations in PM_{2.5} mass concentrations, risks, as well as source and element contributions were largely due to increased coal combustion in winter.

Keywords: Cancer risk; Non-cancer hazard quotient; PM_{2.5}; Element; Polycyclic aromatic hydrocarbons; Seasonal variation.

INTRODUCTION

Beijing is the capital city, and the political, economic, and cultural center of China. In 2017, the population of Beijing reached 22 million (MBS, 2018) with 5.6 million registered vehicles (MEE, 2018). Owing to rapid economic development and urbanization, Beijing and surrounding regions (i.e., Tianjin and Hebei province) have experienced serious air pollution in recent years, including fine particulate matter (PM_{2.5}) (Zhang *et al.*, 2016; Chang *et al.*, 2018). Although annual PM_{2.5} mass concentrations decreased from 90 $\mu\text{g m}^{-3}$ in 2013 to 80 $\mu\text{g m}^{-3}$ in 2015 in Beijing, the 2015 level is still two times higher than the Chinese National Ambient Air Quality Standard of 35 $\mu\text{g m}^{-3}$ and eight times higher than World Health Organization's guideline of 10 $\mu\text{g m}^{-3}$ (UNEP, 2019).

A number of scientific studies have investigated associations between exposure to high concentrations of PM_{2.5} and human health issues, including lung cancer, heart

attacks, and premature death (Li *et al.*, 2018; U.S. EPA, 2018). An exposure assessment study in China used the Shape Constrained Health Impact Function to estimate the PM_{2.5}-related premature deaths during 2013–2015. It was estimated that premature deaths due to long-term exposure to PM_{2.5} were 2.19, 1.94, and 1.65 million in 2013, 2014, and 2015, respectively. Over half of these premature deaths were caused by stroke, followed by ischemic heart disease (20.5%), chronic obstructive pulmonary disease (16.8%), and lung cancer (9.5%) (Li *et al.*, 2018). A PM_{2.5} health effects study in Beijing estimated that the premature mortality rate due to exposure to PM_{2.5} reached 22,000–30,000 per year during 2001–2012 (Zheng *et al.*, 2015).

Assessment of the health risk of ambient PM_{2.5} provides valuable information on the major sources and major PM_{2.5} components causing adverse health effects, and helps to identify environmental health issues and to set effective air pollution control targets (Hu *et al.*, 2017; Liu *et al.*, 2017). Health risk assessment of PM_{2.5} components has been carried out in some Chinese cities in recent years, for example, in the Pearl River Delta region using data collected in 2014 (Zhou *et al.*, 2018). This study used the concentrations of six elements (As, Cr, Ni, Mn, Pb, Si) in PM_{2.5} to estimate the cancer risks and hazard quotients (HQ) through the inhalation

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pathway. The cancer risk was $3.7\text{E-}3$ and the HQ was 2.1. Both cancer risks and HQ were high (Zhou *et al.*, 2018). In Beijing, risk assessment studies have been limited to short time periods. For example, Wan *et al.* (2016) used 12 elements in $\text{PM}_{2.5}$ to estimate cancer risk and HQ in Beijing during spring 2015. Given the large seasonal variability of $\text{PM}_{2.5}$ mass and component concentrations, a health risk assessment using multiple-year data is warranted.

As such, the current study provides information about long-term cancer risks and HQ associated with airborne $\text{PM}_{2.5}$ exposure in Beijing during 2013 to 2015. The specific objectives of this study are to 1) estimate lifetime cancer risk and HQ in Beijing using ambient $\text{PM}_{2.5}$ mass concentrations collected during 2013–2015, 2) identify which elements or sources had major contributions to risk or HQ, 3) identify which target organs are at greatest risk, and 4) investigate seasonal variability of cancer risk and HQ by element, by source, and by target organ. This information will inform health risk managers about which elements in $\text{PM}_{2.5}$ have the greatest impact on human health, from which sources, and in which seasons.

METHODOLOGY

Study Area and Period

The study area is the city of Beijing (Fig. 1). The study

period is from 2013 to 2015, because the proportions of $\text{PM}_{2.5}$ -bound elements and PAHs in total $\text{PM}_{2.5}$ mass concentration were estimated using data from previous studies in Beijing from 2006 to 2014. After 2015, there have been a number of air pollution control policies implemented in Beijing, such as, eliminating coal-fired boilers to phase out coal consumption, and eliminating 389,000 outdated vehicles (Clean Air Asia, 2016). Therefore, the proportions of elements and PAHs in the previous studies of 2006–2014 may not be representative of the proportions in 2016–2018.

Data Source

Hourly ambient $\text{PM}_{2.5}$ mass concentrations at 12 monitoring stations (Fig. 1) in Beijing during 2013–2015 were downloaded from the Qing Yue Open Environmental Data Center (Shanghai Qingyue, 2019). Eight of those stations were in urban areas close to the city center, and the other four were in suburban settings. Detailed information for the 12 monitoring stations can be found in Table S1. Proportions of $\text{PM}_{2.5}$ -bound elements and PAHs in total $\text{PM}_{2.5}$ mass concentrations were obtained from previous studies in Beijing from 2006 to 2014 (see supplemental Tables S2 and S3).

Daily, Seasonal, and Three-year $\text{PM}_{2.5}$ Mass Concentrations

Daily $\text{PM}_{2.5}$ concentrations were calculated by averaging

Fig. 1. The 12 monitoring stations in Beijing. Red star: urban stations; black star: suburban stations. (Base map from Google map, locations of the 12 stations from Shanghai Qingyue (2019))

hourly PM_{2.5} mass concentrations for each of the 12 stations in Beijing, when at least 20 hourly concentrations are available on that day/site during 2013–2015 (MEP and AQSIQ, 2012). City-wide daily PM_{2.5} mass concentrations were calculated by averaging daily PM_{2.5} mass concentrations from all stations with available daily PM_{2.5} mass concentrations. There were 331, 357, and 353 days with daily concentrations available in 2013, 2014, and 2015, respectively. Seasonal and three-year average PM_{2.5} mass concentrations were calculated using daily PM_{2.5} mass concentrations, with the seasons defined as, spring: March–May; summer: June–August; fall: September–November; winter: December–February.

Cancer Risk and HQ

The PM_{2.5} mass and component concentrations from previous studies (2006–2014) in Beijing (Tables S2 and S3) were used to calculate average proportions of each PM_{2.5}-bound element and PAHs in total PM_{2.5} mass on an annual basis and in each of the four seasons. This allowed us to estimate overall and seasonal cancer risk/HQ. The exposure concentrations of each element and PAH in PM_{2.5} were estimated using the average proportions of each element (Table S4) and PAH (Table S5) and the observed seasonal or three-year PM_{2.5} mass concentrations.

Six PM_{2.5}-bound elements (As, Cd, Co, Cr, Ni, Pb) and 16 priority PAHs defined by U.S. EPA (2002) were used to estimate the lifetime cancer risk based on data collected during 2013–2015. These six elements and 16 PAHs were selected because they are carcinogenic (U.S. EPA, 2019), and their proportions in PM_{2.5} mass concentrations are available from previous studies in Beijing from 2006 to 2014. The cancer risk for the six elements (CR_E) was calculated using Eq. (1) following the U.S. EPA method (U.S. EPA, 2005).

$$CR_E = \sum_{i=1}^N ECE_i \times UR_i \quad (1)$$

where ECE_i is the exposure concentration of the *i*th element (μg m⁻³), and UR_i is inhalation unit risk (μg m⁻³)⁻¹ for the *i*th element, as listed in Table S6. The UR for Cr(VI) is available instead of Cr (U.S. EPA, 2019). Therefore, Cr concentrations were used to estimate Cr(VI) concentration which is one seventh (1/7) of Cr concentration as in Perišić *et al.* (2017).

Cancer risk for the 16 PAHs (CR_P, referred to as risks by total PAHs hereafter) was calculated using Eq. (2) following the U.S. EPA method (U.S. EPA, 1993).

$$CR_P = \sum_{j=1}^N ECP_j \times TEF_j \times UR_{BaP} \quad (2)$$

where ECP_j is the exposure concentration of the *j*th PAH (μg m⁻³), and TEF_j is the toxic equivalency factor for the *j*th PAH, as listed in Table S7, where the higher TEF values indicate higher toxicity. UR_{BaP} is the inhalation unit risk of exposure to BaP at 1.1E-3 (μg m⁻³)⁻¹ (OEHHA, 2019).

The total cancer risk is the summation of the cancer risks for the six PM_{2.5}-bound elements (CR_E) and the cancer risks

for the PM_{2.5}-bound 16 U.S. EPA priority PAHs (CR_P). According to the U.S. EPA guideline, cancer risks within the range of 1E-4 to 1E-6 indicate an acceptable or tolerable risk for regulatory purposes; cancer risks above 1E-4 suggest high cancer risk and remedial actions are required; and cancer risks below the 1E-6 suggest cancer risks to human health are negligible (U.S. EPA, 2000).

Eleven PM_{2.5}-bound elements (As, Ba, Cd, Cl, Co, Cr, Mn, Ni, P, Pb, and Se) were used to estimate HQ in Beijing based on data collected during 2013–2015. These elements were selected because they pose significant non-carcinogenic health effects to humans (U.S. EPA, 2019), and their proportions in PM_{2.5} mass concentrations are available from previous studies in Beijing from 2006 to 2014. The HQ was calculated using Eq. (3) following the U.S. EPA method (U.S. EPA, 2015).

$$HQ = \sum_{i=1}^N \frac{ECE_i}{1000 \times RfC_i} \quad (3)$$

where RfC_i is the reference concentration (mg m⁻³) for the *i*th element, as listed in Table S8. The RfC for Cr(VI) is available instead of Cr (U.S. EPA, 2019). Therefore, Cr(VI) concentrations were estimated as one seventh of Cr concentrations as in Perišić *et al.* (2017). HQ equal to or less than 1 indicates that exposure to these elements has insignificant non-cancer health effects to humans, while HQ greater than 1 suggests exposure to these elements might pose significant non-cancer health effects, and remedial actions are needed (U.S. EPA, 2000).

Risks and HQ by each of the PM_{2.5}-bound elements and total PAHs were estimated in Beijing during 2013–2015. Similarly, lifetime risks and HQ by season from each of the components were estimated.

Lifetime risks and HQ from each of the seven sources were estimated using the source profiles (Table S9) from the study of Yu *et al.* (2013). The source profiles contain percentage contributions by each of the 11 elements to each of the seven sources. The seven sources are secondary sulphur, vehicle exhaust, soil dust, road dust, biomass burning, fossil fuel combustion, and metal processing. For example, cancer risk from soil dust is the summation of 100% of cancer risk of Co, 21% of cancer risk of P, and 19% of cancer risk of Se. HQ by target organ was estimated using Table S8. For example, lifetime HQ to respiratory system is the summation of lifetime HQ from all elements involved, i.e., Cl, Co, Cr(VI), Ni, and P.

Assumptions

As in almost all risk assessment studies, lifetime personal exposure levels are not available in this study. Therefore assumptions were made, including 1) three-year average PM_{2.5} concentration represents lifetime average exposure concentration; 2) ambient concentrations represent personal exposure concentration; 3) proportions of elements and PAHs in PM_{2.5} from previous studies (2006–2014) in Beijing (Tables S4 and S5) represent proportions of elements and PAHs in PM_{2.5} in Beijing during 2013–2015; 4) source

profiles from a previous study in Beijing (Table S9) represent source profiles in 2013–2015, and 5) independence of effect by the compounds involved, i.e., there are no synergistic or antagonistic chemical interactions (U.S. EPA, 1993).

RESULTS AND DISCUSSION

PM_{2.5} Mass Concentrations, Lifetime Cancer Risk and HQ

The PM_{2.5} mass concentrations decreased slightly from 84 $\mu\text{g m}^{-3}$ in 2013 to 81 $\mu\text{g m}^{-3}$ in 2015, with a three-year average of 82 $\mu\text{g m}^{-3}$ (Fig. 2), much higher than the Chinese National Ambient Air Quality Standard of 35 $\mu\text{g m}^{-3}$. The lifetime cancer risk from exposure to PM_{2.5}-bound elements and PAHs in Beijing during 2013–2015 was 1.9E-4, which is nearly twice as high as the U.S. EPA acceptable level of 1E-4, suggesting the risk is sufficiently large that some sort of remediation is desirable (U.S. EPA, 2000). Similar values of cancer risks have been reported in the nearby city of Baoding (141 km southwest of Beijing), China in 2016 (Liu *et al.*, 2018). The PM_{2.5} samples were collected from September 25th to December 26th. The lifetime cancer risks for men and women were 1.8E-4 and 1.03E-4, respectively, during the non-heating period (September 25th to November 14th), and 2.2E-4 and 1.23E-4, respectively, during the heating period (November 15th to December 26th). Thus, the cancer risks in Baoding exceeded the U.S. EPA acceptable level of 1E-4 for both periods and both genders.

The lifetime HQ from exposure to PM_{2.5}-bound elements in Beijing during 2013–2015 was 18, which is much higher than the U.S. EPA acceptable level of 1, suggesting that a non-carcinogenic effect is likely to appear (U.S. EPA, 2000). A similarly high HQ value of 11 was reported in Xi'an, China during winter in 2008 and summer in 2009 (Liu *et al.*, 2017). Concentrations of the eight elements (As, Cd, Cr, Cu, Mn, Ni, Pb, and Zn) were used in the study to estimate HQ through inhalation exposure.

Seasonal PM_{2.5} Mass Concentrations, and Contributions to Cancer Risk and HQ

Seasonal PM_{2.5} mass concentrations, and their contributions to cancer risks and HQ in Beijing during 2013–2015 are shown in Fig. 3 and Table 1. PM_{2.5} mass concentrations and cancer risks had similar seasonal variations: i.e., higher in

Fig. 2. Annual and three-year PM_{2.5} mass concentrations in Beijing (average of 12 monitoring stations).

winter (109 $\mu\text{g m}^{-3}$, 30% of risk), medium in spring and fall (80 $\mu\text{g m}^{-3}$, 25%), and lower in summer (68 $\mu\text{g m}^{-3}$, 20%). High PM_{2.5} mass concentrations in winter were because of increased local emission due to space heating and a decreased mixing height (Li *et al.*, 2015). HQ showed a different seasonal pattern in comparison with those of PM_{2.5} mass concentrations and cancer risk. HQ in winter alone contributed more than half (51%) of lifetime HQ, followed by spring (22%) and fall (19%), with a small contribution (8%) in summer. This is mainly attributable to the higher proportion of Cl in PM_{2.5} mass in winter (3.2%) than those in other seasons (0.36%–1.4%) (Table S4), and the relatively high RfC value of Cl (Table S8). Similar results were reported in a health risk assessment study in Baoding, China, from January to December 2015 (Liang *et al.*, 2019). Cancer risks and HQ for children, male adults, and female adults were all higher in the heating period (November 15–March 15) than those in the non-heating period (March 16–November 14).

By normalizing the seasonal cancer risks with corresponding PM_{2.5} mass concentrations, the normalized CR in winter became lower than in the other seasons, while the normalized CR for spring, summer, and fall were similar to one another (Table 1(a)). This indicates that higher PM_{2.5} mass concentrations in winter would have increased CR, however there is diminishment in elements and PAH species with high unit risks (Table S6). The net effect is an increased winter CR but to a lesser degree.

(a) PM_{2.5} mass concentration

(b) Cancer risk

(c) Non-cancer HQ

Fig. 3. (a) PM_{2.5} mass concentrations, (b) lifetime cancer risk, and (c) HQ by season in Beijing.

Table 1. Seasonal PM_{2.5} mass concentrations, cancer risks (CR), and HQ.(a) Normalized cancer risk and HQ by PM_{2.5} mass concentrations

Season	PM _{2.5} mass ($\mu\text{g m}^{-3}$)	CR	Normalized CR per ($\mu\text{g m}^{-3}$)	HQ	Normalized HQ per ($\mu\text{g m}^{-3}$)
Spring	79.94	1.89E-4	2.4E-6	12.54	0.16
Summer	67.75	1.56E-4	2.3E-6	4.36	0.06
Fall	79.56	1.90E-4	2.4E-6	11.09	0.14
Winter	108.7	2.31E-4	2.1E-6	28.91	0.27

(b) Normalized PM_{2.5} mass concentration, cancer risk, and HQ by corresponding summer values

Season	PM _{2.5} mass	CR	Normalized CR	HQ	Normalized HQ
Spring	1.18	1.21	1.04	2.88	2.67
Summer	1.00	1.00	1.00	1.00	1.00
Fall	1.17	1.22	1.04	2.54	2.33
Winter	1.61	1.48	0.91	6.63	4.50

The normalized HQs exhibited greater seasonality with a much higher value in winter and much lower value in summer (Table 1(a)), suggesting more enrichment in winter but diminishment in summer in elements with higher reference concentrations. The winter/summer ratio of HQ was 6.6, higher than that of the normalized HQ of 4.5, indicating that the winter-high summer-low pattern of HQ is attributable to both higher winter PM_{2.5} mass and enrichment in elements with high reference concentrations (Table S8) in winter.

The ratios of seasonal HQ to summer HQ were much higher than the ratios of seasonal CR (Table 1(b)); this further demonstrates that HQ values are much more sensitive to the seasonal compositions of elements in PM_{2.5}. For both CR and HQ, the seasonal variability is primarily due to changes in PM_{2.5} compositions, i.e., diminishment in elements with high cancer risks but enrichment in elements with high reference concentrations in winter, while changes in PM_{2.5} mass played a lesser role. More analysis and discussion on the seasonal variation of CR and HQ are presented in the following sections.

Cancer Risk by Element

Fig. 4 and Table 2 depict the individual contribution of six PM_{2.5}-bound elements and total PAHs to lifetime cancer risk, and in summer and winter in Beijing during 2013–2015. Lifetime risks by PM_{2.5} components were, in descending order, As, Cr(VI), total PAHs, Pb, Co, Ni, and Cd. Among these PM_{2.5} components, As had a risk of 1.1E-4, being the only element where the cancer risk is above 1E-4. Similar results were reported by a health risk assessment study in Jinan (377 km south of Beijing), China (Du *et al.*, 2019). The measurements of elements in PM_{2.5} were conducted from January 2014 to January 2015. The concentrations of five elements (As, Cd, Cr, Ni, and Pb) in PM_{2.5} were used to estimate the lifetime cancer risks through the inhalation pathway, and the results showed that the cancer risks of Cr contributed 72% of total cancer risk, followed by As (22%), Ni (3%), Cd (2%), and Pb (1%).

Fig. 5 demonstrates proportions of PAHs and each of the six elements in PM_{2.5} mass. For all elements, the proportions are similar in spring and fall. Cr(VI) had significantly higher proportion in summer than in winter (winter/summer ratio

of 0.1, Table S10), which had the highest unit risk among all components considered (Fig. 5). All other components had higher proportions in winter (winter/summer ratios of 1.2–4, Table S10). Overall, the winter profile is diminished in elements with higher unit risks, as evidenced by a lower normalized CR than in other seasons (Table 1(a)).

Contributions to cancer risk by PM_{2.5}-bound elements and total PAHs were overall similar in spring and fall (Fig. S1), but different between summer and winter (Fig. 4). The seasonal variability in contributions of PM_{2.5}-bound elements and total PAHs are consistent with changes in the proportions of the elements to PM_{2.5} mass (Fig. 5). Contributions of Cr(VI) were much lower in winter (6%) than in summer (44%). Higher contributions in winter were observed for As (72% in winter vs. 43% in summer), PAHs (15% vs. 5%), Pb (8% vs. 5%), and Co (5% vs. 1%). Contributions of Ni and Cd were similar between summer and winter.

Cancer Risk by Source

Fig. 6 and Table 3 show the contributions by each of the seven PM_{2.5} emission sources to lifetime, summer, and winter cancer risks from exposure to PM_{2.5}-bound elements and PAHs in Beijing during 2013–2015. Among the seven sources considered, road dust contributed the highest lifetime cancer risk (39%). This is because nearly 40% of As and 70% of Cr(VI) originated from road dust (Table S9) and the unit risks from As and Cr(VI) were high, 4.3E-3 per ($\mu\text{g m}^{-3}$) and 1.2E-2 per ($\mu\text{g m}^{-3}$), respectively (Table S6). Fossil fuel combustion contributed the second-highest cancer risk (24%), followed by vehicle exhaust (21%), because 35% of As originated from fossil fuel combustions and 20% of As was from vehicle exhaust (Table S9). The remaining four sources (soil dust, secondary sulphur, metal processing, and biomass burning) each contributed no more than 10% of cancer risks. Somewhat similar contributions to cancer risks by source were reported in the study in Jinan, China in 2014 (Du *et al.*, 2019). The result showed that vehicle exhaust contributed 55% of total cancer risks, followed by coal combustion (27%), the smelting industry (11%), and the soil dust (7%).

The contributions of emission sources to cancer risks are somewhat similar in the four seasons in terms of rank order of contributions (Fig. S2). However, there is a significant

Fig. 4. Lifetime cancer risk by element and in summer and winter.

Table 2. Lifetime cancer risk by element.

As	Cr(VI)	PAHs	Pb	Co	Ni	Cd
1.1E-4	3.4E-5	1.5E-5	1.1E-5	8.4E-6	3.9E-6	3.9E-6

Fig. 5. Seasonal proportions of the six elements and total PAHs in total PM_{2.5} mass (#s in parentheses are unit risks).

seasonality with more contrast between summer and winter, as shown in Fig. 6. Road dust had high contributions in summer (49%) and low contributions in winter (33%) because of higher proportion of Cr(VI) (0.008% in summer vs. 0.001% in winter, Table S4). Contribution of metal processing was high in summer (10%) and low in winter (1%), because of higher proportion of Cr(VI) (0.008% in summer vs. 0.001% in winter, Table S4). Contributions of biomass burning were high in summer (4%) and low in winter (3%). This is not unexpected because the North China Plain experienced more agricultural residual burning emissions in harvest seasons (Yin *et al.*, 2019). Winter-high and summer-low contributions were observed for fossil fuel combustion (26% vs. 16%), vehicle exhaust (28% vs. 16%), soil dust

(5.1% vs. 1.2%), and secondary sulphur (4.6% vs. 3.1%). For fossil fuel combustion and vehicle exhaust, this seasonal pattern was attributable to higher proportion of As in winter (0.031% vs. 0.023, Table S4). For soil dust, the seasonal pattern was due to higher proportions of Co in winter (0.001% vs. 2.9E-6%, Table S4). Beijing experienced higher relative humidity (70%) in summer than that in winter (45%) (Time and Date, 2020), which increased soil particle cohesion (Csavina *et al.*, 2019) resulting in less soil dust.

HQ by Element

Fig. 7 and Table 4 illustrate the contributions of each of the 11 PM_{2.5}-bound elements to lifetime HQ, and in summer and winter in Beijing during 2013–2015. The breakdown of

Fig. 6. Lifetime cancer risks by source and in summer and winter.

Table 3. Lifetime cancer risk by source.

Road dust	Fossil fuel combustion	Vehicle exhaust	Soil dust	Metal Processing	Secondary sulphur	Biomass burning
7.3E-5	4.4E-5	3.8E-5	8.4E-6	8.2E-6	8.0E-6	6.4E-6

Fig. 7. Lifetime non-cancer HQ by PM_{2.5}-bound element and in summer and winter.

Table 4. Lifetime HQ by element.

Cl	As	Pb	P	Cd	Mn	Ni	Ba	Cr(VI)	Co	Se
13.9	1.8	0.94	0.81	0.22	0.22	0.18	0.10	0.03	0.01	0.002

lifetime HQ by PM_{2.5}-bound elements were, in descending order, Cl, As, Pb, P, Cd, Mn, Ni, Ba, Cr(VI), Co, and Se. Co and Se are not shown in Fig. 7, because the contributions were less than 0.1% each. Among the 11 elements, Cl (HQ = 14) and As (1.8) were the only two elements where the HQ was greater than 1. The contribution of Cl to HQ was 76%.

In comparison, the health risk assessment study in Jinan, China, in 2014 used five elements (As, Cd, Cr, Mn, and Ni) in PM_{2.5} to estimate HQ via the inhalation pathway (Du *et al.*, 2019). Results showed that As was the largest contributor to HQ (38%), followed by Mn (35%), Ni (13%), Cr (11%), and Cd (2%).

Fig. 8 demonstrates proportions of each of the 11 elements in PM_{2.5} mass by season. Ba and P had the same proportions in the four seasons. For the other nine elements, the proportions were overall similar in spring and fall. Cr (VI) had significantly higher proportions in summer than in winter (winter/summer ratio of 0.1, Table S11), which had medium-high reference concentrations among all elements considered (Fig. 8). The other eight elements had lower proportions in summer (winter/summer ratios of 1.2–9, Table S11). In comparison with the transition seasons of spring and fall, the winter profile was greatly enriched while the summer profile was greatly diminished in elements with higher reference concentrations (Fig. 8), evident by the normalized HQ being much higher in winter and lower in summer (0.27 per $\mu\text{g m}^{-3}$ in winter vs. 0.06 per $\mu\text{g m}^{-3}$ in summer, Table 1(a)). The wider range of winter/summer ratios in HQ profiles than in CR profiles (0.1–9 vs 0.1–4, Tables S10 and S11) suggests more distinct seasonality.

HQ in spring and fall showed similar contributions by the 11 PM_{2.5}-bound elements (Fig. S3). The seasonality was more pronounced between summer and winter (Fig. 7). Contributions of Cl were high in winter (81%) and low in summer (37%). Contributions of all other elements were high in summer and low in winter, inconsistent with lower proportions of those elements in summer (Fig. 8). This is because HQ values of Cl reduced by 14 times in summer (1.6 in summer vs. 23 in winter), leading to increased contributions in summer of As (24% in summer vs. 7.9% in winter), Pb (14% vs. 5.1%), P (15% vs. 3.7%), Mn (2.3% vs. 0.8%), Cd (2.1% vs. 1.0%), Ni (0.45% vs. 0.19%), Ba (2.0% vs. 0.47%), and Cr(VI) (1.3% vs. 0.04%).

HQ by Source

The contributions to lifetime HQ by source, and in summer and winter in Beijing during 2013–2015 are shown in Fig. 9 and Table 5. Among the seven sources considered, fossil fuel combustion and vehicle exhaust were the top two sources contributing to lifetime HQ at 67% and 10%,

respectively, and the remaining five sources (road dust, metal processing, biomass burning, secondary sulphur, and soil dust) combined contributed 23% of HQ. Somewhat similar results were reported by the health risk assessment study in Jinan, China, in 2014, where coal combustion contributed 34% of HQ, followed by the smelting industry (25%), vehicle exhaust (22%), and soil dust (19%) (Du et al., 2019).

Contributions to HQ by source were similar in spring and fall (Fig. S4), but different between summer and winter (Fig. 9). Fossil fuel combustion had higher contributions in winter (69%) and lower contributions in summer (46%), due to higher proportions of Cl (3.2% vs. 0.36%, Table S4). Higher contributions of vehicle exhaust (17% in summer vs. 10% in winter), road dust (17% vs. 5.5%), secondary sulphur (7.2% vs. 4%), and soil dust (3.4% vs. 0.8%) were observed in summer, in spite of lower HQ values in summer (Table S12). This is because the proportion of Cl reduced greatly from winter to summer (3.2% vs. 0.36%, Table S4), as discussed in Section HQ by Element. Lower contributions were observed in summer for metal processing due to lower proportions of Cl (0.36% in summer vs. 3.2% in winter, Table S4). Contributions of biomass burning were high in summer and low in winter, as expected.

HQ by Target Organ

Fig. 10 and Table 6 show the contributions to lifetime, summer, and winter HQ by target organ in Beijing during 2013–2015. Among the six target organs considered, respiration was the most vulnerable system, contributing 82% of lifetime HQ. This is because the respiratory system was affected by five elements (Cl, Co, Cr(VI), Ni, and P), and Cl alone contributed 76% of lifetime HQ (Fig. 7), followed by reproductive (10%), and nervous system (6%), and the remaining three target organs (renal, fetus, and alimentary) together contributed 2% of HQ. Alimentary system is not shown in Fig. 10, because the contributions were less than 0.001%.

Fig. 8. Seasonal proportions of 11 elements in total PM_{2.5} mass (#s in parentheses are reference concentrations).

Fig. 9. Lifetime non-cancer HQ by source and in summer and winter.

Table 5. Lifetime HQ by source.

Fossil fuel combustion	Vehicle exhaust	Metal Processing	Road dust	Secondary sulphur	Biomass burning	Soil dust
12.0	1.9	1.3	1.2	0.77	0.71	0.18

Fig. 10. Lifetime non-cancer HQ by target organ and in summer and winter.

Table 6. Lifetime HQ by organ.

Respiratory	Reproductive	Nervous	Renal	Fetus	Alimentary
14.9	1.8	1.2	0.22	0.10	0.002

Among the four seasons, the contributions by organ were similar in the transition seasons of spring and fall (Fig. S5), but dissimilar in summer and winter (Fig. 10). The contribution of respiratory system was high in winter (85%) and low in summer (54%). This is because Cl was the largest contributor

to HQ (Fig. 7) and the proportions of Cl were much higher in winter than in summer (3.2% vs. 0.36%, Table S4). The HQ of the nervous system depends on Pb and Mn. For the reproductive, renal, fetus, and alimentary system, the HQ values depend on As, Mn, Cd, Ba, and Se, respectively.

Among the six elements, five had higher proportions in winter (As: 0.031% in winter vs. 0.023% in summer; Mn: 0.064% vs. 0.044%; Cd: 0.003% vs. 0.002%; and Se: 0.05% vs. 0.007%; Pb: 0.204% vs. 0.138%, Table S4). Although Ba had the same proportions in the four seasons (Table S4), PM_{2.5} mass concentration was higher in winter (Fig. 3), leading to higher Ba concentration. However, the contribution by each of these five organs were lower in winter (Fig. 10). This is because the proportion of Cl reduced greatly from winter to summer (3.2% vs. 0.36%, Table S4), leading to increased contributions of other organs in summer, as discussed in Section HQ by Element.

Uncertainties

The health risk assessment methods developed by the US EPA were employed in this study for the estimation of cancer risks and non-cancer HQ from PM_{2.5}-bound elements and PAHs in Beijing. The results reveal seasonality and identify sources with large contributions. However, there are several possibilities for uncertainty.

Proportions of PM_{2.5}-bound elements and PAHs estimated from prior locally-conducted studies (2006–2014) may not represent the PM_{2.5} compositions in the current study (2013–2015) very well. This is because proportions of PM_{2.5}-bound elements and PAHs in prior studies were averaged over the relatively long period of 2006–2014, during which the study region experienced rapid industrialization and economic growth.

The uncertainty in PM_{2.5} mass determination is considered small (< 5%, e.g., Zheng *et al.*, 2005). By using estimated proportions of PM_{2.5} compositions from previous studies (2006–2014), the cancer risks and HQ results in this study are likely biased high. This is because, by switching to cleaner fuels and enforcing emission controls in recent years (UNEP, 2019), the proportions of fossil fuel combustion related-elements (i.e., As and Cl) in PM_{2.5} mass should be lower than before. These two elements were the largest contributors to CR (60%) and HQ (76%), respectively.

The risks and HQ results in this study were calculated based on the 11 PM_{2.5}-bound elements and 16 PAHs. In reality, exposures are not limited to these species. If considering other toxic elements/PAHs, such as mercury and benzo(e)pyrene, the health risks of toxic components in PM_{2.5} will increase.

Greater inherent uncertainty accrues by assigning source-based risk/HQ estimates based on metals composition in the source profiles from a source apportionment study (Yu *et al.*, 2013). Our study focuses on city-wide PM_{2.5} concentrations in Beijing and the associated health risks. However, there exists intra-city variability in PM_{2.5} mass and component concentrations and, therefore, in risk levels. A study of PM_{2.5} spatial and temporal variability during 2013–2018 in Beijing reported that the 6-year PM_{2.5} mass concentrations varied from 59 to 77 $\mu\text{g m}^{-3}$ among the 12 monitoring stations (Xu and Zhang, 2020). The proportions of PM_{2.5}-bound elements in total PM_{2.5} mass also differed significantly between urban and suburban stations, especially for As and Cr(VI), which are the major contributors to cancer risks (Chen, 2017).

CONCLUSIONS

Lifetime cancer risks and HQ due to inhalation exposure to elements and PAHs in PM_{2.5} were estimated in Beijing, China, based on data collected during 2013–2015. The three-year PM_{2.5} mass concentration was 82 $\mu\text{g m}^{-3}$, much higher than the Chinese National Ambient Air Quality Standard of 35 $\mu\text{g m}^{-3}$.

During 2013–2015, the lifetime cancer risk in Beijing from exposure to ambient PM_{2.5}-bound elements and ambient PM_{2.5}-bound PAHs was 1.9E-4, nearly twice the U.S. EPA acceptable level of 1E-4; while the lifetime HQ from exposure to ambient PM_{2.5}-bound elements was 18, much higher than the U.S. EPA acceptable level of 1. Among the six target organs considered, the respiratory system was the most vulnerable, contributing 82% to HQ. Fossil fuel combustion (risk: 24%, HQ: 67%), road dust (risk: 39%, HQ: 7%) and vehicle exhaust (risk: 21%, HQ: 10%) were the top three sources. Consequently, element As from coal combustion and road dust and Cr(VI) from road dust are the largest contributors to cancer risk (totaling 78%), while Cl from coal combustion and As accounted for 86% of HQ.

Among the four seasons, PM_{2.5} mass concentrations were highest in winter (109 $\mu\text{g m}^{-3}$) and lowest in summer (68 $\mu\text{g m}^{-3}$). Winter also accounted for the largest share of total lifetime cancer risk (30%) and total HQ (51%), with less contributions in summer (risk: 20%, HQ: 8%). Contributions of cancer risks or HQ by PM_{2.5}-bound elements or by source were similar in spring and fall but dissimilar in winter and summer primary. The seasonal variability is primarily driven by changes in PM_{2.5} compositions, especially As and Cl which are mainly from coal combustion and less coal was used in summer.

One of the limitations of this study is the use of proportions of elements and PAHs in PM_{2.5} mass from the literature to estimate health risks due to exposure to ambient PM_{2.5}. Long-term monitoring of PM_{2.5} components should be conducted at government stations in all major Chinese cities, and the data should be made available. This would allow the evaluation of health benefits of emission control and changes in energy sourcing, and facilitate investigations of the spatial distribution of source impacts and risk levels throughout the city.

This study identifies major PM_{2.5} sources and PM_{2.5}-bound elements that have the greatest impact on human health in Beijing, as well as their seasonality. Our findings may help develop mitigation measures in the region. Since 2016, coal combustion and industrial emission have been largely eliminated within the capital. However, those two types of sources still emit a large quantity of PM_{2.5} in regions surrounding the city. An analysis of fall 2016 field samples collected in Beijing indicated that secondary aerosols, dust, vehicle, and coal combustion were the major sources of PM (Zhang *et al.*, 2018). In order to reduce the health burden on Beijing residences due to inhalation exposure to ambient PM_{2.5}, the government should take measures to lower PM_{2.5} mass concentrations. Particularly, emissions from road dust, fossil fuel combustion, and vehicle exhaust should be reduced significantly because they contributed the most to

cancer risks and HQ. Control measures should be taken to reduce As and Cl emissions released to the atmosphere. This is because As and Cl ranked number one in cancer risk and HQ, respectively, during the study period, and resulted in cancer risk or HQ values greater than the U.S. EPA's acceptable levels. Residents of Beijing are encouraged to wear PM_{2.5} filtration face masks especially in winter to reduce inhalation exposure. There is a wide range of face masks on the market, and only some of these are effective at filtering the PM_{2.5} size fraction and thus having a notable effect on the CR and HQ risks estimated in this study. A field test of nine types of face masks indicated that the filtration efficiency of PM_{2.5} ranged from 14–96%, with a median value of 48% (Pacitto et al., 2019). People with respiratory diseases should be cautious because the respiratory system was the most vulnerable.

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CONFLICTS OF INTEREST

The authors declare no conflict of interest. The founding sponsors had no role in the design of the study, in the collection, analyses, or interpretation of data, in the writing of the manuscript, or in the decision to publish the results.

SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <https://aaqr.org/>

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