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Nasal filter reveal exposure risks of inhalable particulates and heavy metals in urban women

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ABSTRACT

Urban populations, especially women, are vunerable to exposure to airborne pollution, particularly inhalable particulates (PM_{10}). Thus, more accurate measurement of PM_{10} levels and evaluating their health effects is critical for guiding policy to improve human health. Previous studies obtained personal PM_{10} with time-weighted average by air filter-based sampling (AFS), which ignores individual differences and behavioral patterns. Here, we used nasal filters instead of AFS to obtain actual inhaled PM_{10} under short-term exposure for urban dwelling women during a severe haze event in Beijing in 2016. The levels of six heavy metals such as As, Cd, Ni, Cr, Pb, and Co in PM_{10} were investigated, and carcinogenic and non-carcinogenic risks evaluated based on an adjusted US EPA health risk assessment model. The health endpoints for urban dwelling women were further assessed through an exposure-reponse model. We found that the hourly inhaled dose of PM_{10} obtained through the nasal filter was about 2.5–17.6 times that obtained by AFS, which also resulted in greater exposure to particulate matter (>18.8 µg/kg·h) and heavy metals (>2.2 ng/kg·h), and these populations are therefore at greatest risk of developing non-cancer (HI = 4.16) and cancer ($R_t = 7.8 \times 10^{-3}$) related morbities.

1. Introduction

Particulate air pollution is a major global public health concern. Inhalable particulate matter (PM) may be defined as having a diameter below 10 μ m (PM₁₀), and are known as a key component of air pollution and an associated health hazard (Nel, 2005; Palacios et al., 2017). Because of their size these fine particles may be inhaled deeply into the lungs, and can reach the alveolus and blood supply resulting in a range of respiratory and cardiovascular diseases (Karakatsani et al., 2012; Orellano et al., 2020). There are also positive associations of these fine particles with cancers including lung, colorectal, breast, cervical and bladder cancer (Huo et al., 2013; Diaz-Sanchez, 2015; Turner et al., 2017). Moreover, pollutants such as organic compounds and heavy metals loaded on PM₁₀ can cause further harm to human health (Rylance et al., 2013; Sun et al., 2021). For example, arsenic (As), cadmium (Cd), cobalt (Co), chromium (Cr), nickel (Ni) and lead (Pb) are reported to be human carcinogens (IARC, 2006). Further, As and Cd can cause teratogenesis, Co and Ni can injure the lungs, and Pb has adverse effects on cognitive development in children (ATSDR, 2017). Thus, an accurate estimate of the concentration and toxic effects of PM_{10} for exposed populations is important for taking effective measures and informing policies on air pollution control and potential disease prevention.

Females are known to be more vulnerable to PM_{10} exposure due to differences in physiological structure (Yaghjyan et al., 2017). In particular, womens' lungs and airway diameters are narrower than in men, which may increase sensitivity to PM stress (Valavanidis et al., 2008; Zeng et al., 2017). Vrijens et al. (2016) found that there is greater gene expression associated with PM_{10} in females than in males, which confirms the enhanced vharmful effects of PM_{10} . Compared to females living in rural or suburban areas, urban dwelling women may be exposed to greater levels of PM_{10} due to more traffic related exposure, for example due to commuting or working outdoors (Khosrorad et al., 2022). In addition, air dispersion conditions in cities are significantly worse than in suburban areas, which makes the problems of urban exposure to air pollution more serious. A recent study confirmed that risks for lung cancer and leukemia with increased fine particle exposure

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are more significant for women living in urban areas (Wang et al., 2019). According to Beijing Cancer Registry Annual Report statistics from 2021, the top three female cancers were breast, lung and thyroid cancer, which are directly related to airborne PM pollution in city (Huo et al., 2013; Diaz-Sanchez, 2015; Turner et al., 2017). Although an increasing number of studies have investigated the relationship between breast cancer and personal PM₁₀ exposures in city dwelling females (Parikh and Wei, 2016; Guo et al., 2020; Hwang et al., 2020), few studies have focused on the exposure risk for urban dwellers based on work and lifestyle. The personal exposure data obtained from fixed-site ambient (outdoor) monitoring stations or time-integrated samples of point measurement (Lin et al., 2020) cannot accurately represent the exposure characteristics of urban dwelling women due to differences in occupational and time-activity patterns. For example, home workers are likely to spend most of their time indoors, while bus conductors and professional drivers are readily exposed to mobile traffic sources.

Epidemiological studies, including longitudinal cohort studies and timeseries studies of the health effects of air pollution, along with focused panel studies and toxicological evaluations, have developed approaches to establish national and state regulatory standards for the sake of public health (Huang et al., 2017; Morakinyo et al., 2017). These approaches are mostly based on air filter-based sampling devices from specific static monitoring sites to obtain and calculate the average concentration of PM₁₀ over a period time, and an exposure model based on theoretical inhalation dose to estimate the influence of particles on population in a large scale and region (Wu et al., 2016; Palacios et al., 2017; Li et al., 2020). However, existing studies did not provide information on individual physiological differences and personal behaviors in indoor or outdoor environments which may affect uptake, absorption, and metabolism of PM₁₀ (Allen et al., 2003; Hou et al., 2016). Taking into account variations in pollution sources and exposure scenarios, the complexity of pollutants, and the results of the above evaluations may under- or overestimate the risk of individual exposure to PM₁₀ pollution (Ozkaynak et al., 2013; Kioumourtzoglou et al., 2014; Hu et al., 2020). Therefore, for specific exposure-sensitive populations, more precise investigation is needed to answer questions such as how much particulate matter is ingested during the exposure period, which chemical components or elements are mainly involved, and how big is the toxic effect of inhaled PM10?

To the best of our knowledge there are no studies which have measured actual inhaled PM10 doses in urban dwelling females as a result of air pollution. In order to gain more accurate insights into exposure risks of PM₁₀ in urban dwelling women with different work and lifestyles exposed to pollution this study, for the first time, makes use of nasal filters to investigate the characteristics of directly inhaled PM₁₀ for three different occupations i.e., bus conductor (BC), manual laborer (ML) and home worker (HW) under a short-term exposure to a severe haze event. Compared with traditional methods, this sampling method has the ability to better mimic human breathing patterns and exposure conditions, and is more accurate for assessing the health risks of exposed populations. Our study therefore helps better understand the direct risks of PM_{10} and heavy metal pollution exposure faced by urban dwelling women in different occupations during haze episodes. Our study may be particularly important in formulating reasonable air pollution prevention and control strategies for exposed population groups.

2. Materials and methods

2.1. Study area and sample collection

Beijing is a well-known mega-city in northern China, having 21.7 million inhabitants and 5.7 million motor vehicles in 2016 (Beijing Municipal Bureau of Statistics, 2017). The 'heating season', which is when centralized/district heating systems are switched on during the colder winter months in Beijing, normally lasts for five months starting

in early November and ending in early March. High population density, high levels of industrialization, a long heating period largely enabled by coal-burning, and typically adverse meteorological conditions make air pollution in Beijing amongst the most harzardous over the entire country (Chen et al., 2016). For example, recorded PM₁₀ concentrations in Beijing were 101.5 µg/m³ in 2015 (Beijing Municipal Environmental Protection Bureau, 2016), which was > 5 times higher than World Health Organization (WHO) annual guidelines ($20 \mu g/m^3$ for PM₁₀). Our study was carried out in the populous Chaoyang District in north-eastern Beijing, which is home to the Central Business District (CBD), has many diplomatic missions, as well as many commercial and residential associated activities such as cooking at home or restaurant (see Fig. 1). Transportation is a major contributor to air pollution at this area because it is located between the 3rd and 4th Ring Roads (Lin et al., 2020), the busiest traffic lines in Beijing. Atmospheric transmission of industrial pollution from surrounding areas is also an important source of air pollution in Chaoyang (Liu et al., 2023).

A total of six female volunteers living and working in this area were selected for personal exposure monitoring lasting about 10 days during Beijing's heating season, 2016-17. The volunteers were divided into 3 occupation-based groups, with two people in each group. In order to minimize individual effects volunteers had similar age, weight, height, body mass index (BMI), vital capacity and health status (see Table 1). For the three groups, BC had similar activity routines and behaviors, ML worked in the same area, and HW lived in same community. The exposure characteristics of each group are summarised in Table 1. The women working as BC and ML were often exposed to air pollution on busy urban roads, while HW usually carried out indoor activities such as housework and shopping and were less exposed. Bus conductors and drivers represented the highly exposed urban residential population, which had a higher exposure risk to secondary oxidized traffic particles (Baccarelli et al., 2014; Hou et al., 2016). Every volunteer wore a Woodyknows nasal filter (M&M Pure Air Systems LLC, Delaware, USA), which are mainly used for the prevention of pollen allergy, and filter PM and dust (Sigsgaard and Tovey, 2014; Kenney et al., 2016). The nasal filters comprise a filter frame, electrostatic filter and retainer ring, and the electrostatic filter can effectively attract and filter 0.1-10 µm PM (tested by Nelson Lab) with a thinner (0.1 mm) fiber layer (see Fig. 2). Considering that this filter may affect breathing and is not suitable for long term use, the wearing time selected in the study was for one hour between 8:00 am to 9:00 am. After wearing for an hour, the filter was removed and sent for analysis. During the test period of 1.5 months, each volunteer obtained a total of 10 PM₁₀ samples. This study was subjected to ethical review and approval by the Human Investigation Committee of the National Institute of Environmental Health in the University, and all participants signed informed consent.

2.2. The environmental measurements of PM and the inhaled dose calculation of PM_{10}

During the study period mass concentrations of PM_{10} were obtained by the Beijing Municipal Environmental Monitoring Center at the Olympic Sports Center site (https://zx.bjmemc.com.cn/), which is the nearest automatic monitoring site to the study area. At the same time, visible air pollution was recorded by taking photos in the same area (Fig. S1).

Generally, the inhaled route was the major route of exposure to PM and pollutants in the air (Deng et al., 2016). In the study, the actual inhaled PM₁₀ dose (DI_a) of the individual was obtained by weighing the nasal filters worn by the participant during each short-term exposure round. The recovered filters were equilibrated for 48 h at ambient temperature (21 ± 3 °C) and relative humidity (40 ± 3 %), and were then weighed at least three times. The mass difference of the filters before and after sampling was recorded for the calculation of particulate concentration. Filter blanks, laboratory blanks, and externally certified standard weights were used for all gravimetric analyses for quality



Fig. 1. The sampling area and main traffic networks in Beijing, including the 2nd – 6th ring roads (. Source: Bing Maps)

Table 1

Descriptive characteristics of the female participants (n = 6) in the study area in Beijing, China in 2016.

Item	Bus conductor (BC)	Manual laborer (ML)	Home worker (HW)
Female, n	2	2	2
Age, years, mean \pm SD	36.5 ± 0.7	$\textbf{38.0} \pm \textbf{1.4}$	38.5 ± 2.1
Height, m, mean \pm SD	1.59 ± 0.01	1.62 ± 0.01	1.61 ± 0.02
Weight, kg, mean \pm SD	61.5 ± 1.34	63.8 ± 0.64	$\textbf{62.6} \pm \textbf{1.48}$
BMI, kg/m ² , mean \pm SD	21.0 ± 0.28	22.1 ± 0.35	$\textbf{22.2} \pm \textbf{0.78}$
Vital capacity, mL, mean \pm SD	2540 ± 85	2700 ± 70	2655 ± 50
Health status	Good	Good	Good
Activity area at 8:00–9:00 am	Bus	Outdoor space	Home
Distance to the major road, m	Near, 0–5	Near, 10–20	Far, 40–50



Fig. 2. Nasal filter components and wearing style (Refer to the website: http s://woodyknows.com/).

assurance. BCR-723 (road dust collected in Austria with a particle size fraction of $<90~\mu m$ and a median value of 14.6 μm) was used as the certified standard of particulate matter. The actual inhalation dose of PM_{10} was obtained as follows:

$$DI_a = MPM/(ET \times BW) \tag{1}$$

where DI_a is the actual hourly dose for PM₁₀ inhalation (µg/kg·h), *MPM* is the mass of PM₁₀ by nasal filter measurement (µg), and *ET* is the exposure time (h).

In order to compare DI_{a} , the theoretical inhalation dose (DI_{t}) of PM₁₀ was estimated by the measured data of PM₁₀ concentration and exposure parameter values based on the acute exposure model (Dean et al., 2017; Morakinyo et al., 2017). DI_{t} of PM₁₀ is defined as:

$$DI_{t} = (C_{\rm PM} \times TR \times IR)/BW$$
⁽²⁾

where DI_t is the theoretical hourly dose for PM_{10} inhalation (µg/kg·h); C_{PM} is the monitoring concentration of PM_{10} in the surrounding environment (µg/m³); *TR* is the tracheobronchial retention, expressed as a fraction (0.70 was recommend by Sturm (2007)); *IR* is the inhalation rate (the recommended US EPA default value was 1.2 m³/h for adults under acute exposure (US EPA, 1997); and *BW* is body weight (kg).

2.3. The analysis of heavy metals in PM_{10}

In our study some of the more harmful trace metals in PM_{10} were investigated, of which As, Cd, Ni, Cr, Pb, and Co were categorized as known carcinogens or probable carcinogens according to the International Agency for Research on Cancer (IARC). Filter subsamples were introduced into clean Teflon tubes with 3 mL of concentrated HF (40 %), 3 mL of concentrated HCl (37 %) and 1 mL of concentrated HNO₃ (65 %) added. The resulting solution was heated to 70 °C overnight before

cooling, and 20 mL of H_3BO_3 (4 %) was added. This solution was once again heated to 70 °C for 3 h before cooling and transfer to PE vessels and further dilution for analysis. To validate the process, a certified reference material BCR-723 was treated in the same way as the samples. In the final solutions, trace metals were determined using High Resolution-Inductively Coupled Plasma Mass Spectrometry (HR-ICP-MS, Thermo Finnigan Element II). Calibration was carried out with appropriate dilutions of an acidified multi-element stock solution (Merck, ICP-MS standard XIII), and indium (1 ng/mL) was used as an internal standard. Limits of detection (LOD) were around 0.002 µg/L for Cd, 0.01 µg/L for Pb, 0.005 µg/L for Cr, 0.001 µg/L for Co, 0.005 µg/L for Ni, and 0.01 µg/L for As. The relative standard deviation (RSD) was about 10 % for the certified reference material.

2.4. The health effect endpoints evalution of urban women

The health effect endpoints due to PM₁₀ include both mortality and morbidity (Liu et al., 2023). Morbidity mainly include acute and chronic bronchitis, asthma, and hospital admissions (cardiovascular and respiratory). To further illustrate how many urban women are at risk of illness and death after inhaling particulate matter in a severe haze event, the health effect endpoints for BC, ML, and HW groups in Beijing were estimated according to the exposure-response model. In the study, six health effect endpoints of individual mortality, chronic bronchitis, acute bronchitis, asthma, respiratory hospitalization, and cardiovascular hospitalization were evaluated based on an exposure-response coefficient and baseline incidence rate for PM₁₀ (Burnett et al., 2014; Yin et al., 2015; Madaniyazi et al., 2016; Liu et al., 2023). The model includes two parts: (1) there is an approximately linear relationship between the inhaled PM₁₀ dose of the surveyed population and the related health effects (E_i) (equation (3)); and (2) the incidence rate of the health endpoint (Ni) follows a Poisson distribution in a given population. Therefore, the PM₁₀ inhalation dose can be linked to the incidence rate of the health endpoint through Poisson regression (equation (5)).

$$E_i = E_{0i} \times \left[1 + \beta_i \times f \times (DI_a - DI_0)\right]$$
(3)

$$f = \frac{BW}{1000 \times TR \times IR} \tag{4}$$

$$N_i = P \times E_i \times \{1 - \exp[-\beta_i \times f \times (DI_a - DI_0)]\}$$
(5)

where E_i represents the relative risk of different health endpoints caused by increase of 1 mg/m³ PM₁₀; E_{0i} represents the baseline incidence of certain health endpoints, β_i represents the exposure–response coefficient for a particular health endpoint; DI_a (µg/kg·h) is the amount of PM₁₀ inhaled per hour by the surveyed population; DI_0 (µg/kg·h) is the inhalation concentration threshold below which there is no health impact, which is set to the WHO annual guidance value ($PM_{10} = 0.02$ mg/m³); $DI_0 = 0.267 \ \mu g/kg \cdot h$; f represents the transfer coefficient of PM_{10} between the environment and the human body; *P* represents the target exposed population in the study area; and N_i represents the population of the health endpoint. To estimate the PM₁₀ health impacts, the exposure–response coefficients β_i and baseline frequencies E_{0i} were mainly collected from previous research data from Beijing (Table S1) (Yin et al., 2015). The target exposed population P was based on the number (about 430,000) of women working outdoors in the region in 2016 (Beijing Municipal Bureau of Statistics, 2017).

2.5. Health risk assessment of direct inhalable heavy metals in PM_{10}

There have been few studies using direct exposure to PM_{10} on human subjects in their everyday lives to assess the health risk of heavy metals. For the first time our study has applied actual exposure factors in the US EPA (2007) model to estimate the potential exposure to heavy metals in PM_{10} and their carcinogen/non-carcinogen risk within a 1 h exposure during a heavy environmental haze.

2.5.1. The exposure assessment of PM_{10} inhalation

$$HDI = C \times DI_a \tag{6}$$

where *HDI* represents hourly exposure of heavy metals $(ng/kg \cdot h)$ through inhalation; and *C* is the vconcentration of heavy metals $(ng/\mu g)$ in PM₁₀.

2.5.2. Non-cancer risk

Non-carcinogenic risk is defined as all adverse impacts on human health caused by exposure factors excluding cancer (Broomandi et al., 2023). In accordance with US EPA (2007), non-cancer risk was evaluated by means of the hazard quotient (*HQ*). A *HQ* < 1 indicates an allowable non-cancer risk, whereas HQ > 1 suggests non-carcinogenic effects may occur (US EPA, 1989). The health risk values of non-cancer for inhaled heavy metals may be estimated by the following equation:

$$HQ = (HDI \times BW) / (TR \times IR \times RfD \times 1000)$$
(7)

where *R*fD is the inhalation reference concentration (μ g/m³). The US EPA (2016) provides information concerning values of *R*fD for As (0.015 μ g/m³), Cd (0.01 μ g/m³), Co (0.006 μ g/m³), Cr (0.1 μ g/m³), and Ni (0.02 μ g/m³). As no data was available for Pb, a value of 0.5 μ g/m³ for *R*fD from the World Health Organization Air Quality Guidelines (WHO, 2005) was used.

2.5.3. Cancer risk

According to the US EPA (1989), cancer risk is estimated as "the incremental probability of an individual developing cancer over a lifetime as a result of exposure to the potential carcinogen". A cancer risk (*R*) value lower than $1.0 \times 10^{-6} - 1.0 \times 10^{-4}$ indicates a carcinogenic risk which may be acceptable (Deng et al., 2016), whereas the most tolerable risk rate is 1.0×10^{-6} . The cancer risk *via* inhaled heavy metals was estimated by the following equation (US EPA, 2007):

$$R = (HDI \times BW \times IUR) / (TR \times IR \times 1000)$$
(8)

where *IUR* is the inhalation unit risk (m³/µg). The US EPA (2016) provides information concerning values of *IUR* for: As $(4.3 \times 10^{-3} \text{ m}^3/\mu\text{g})$, Cd $(1.8 \times 10^{-3} \text{ m}^3/\mu\text{g})$, Co $(9.0 \times 10^{-3} \text{ m}^3/\mu\text{g})$, Cr $(8.5 \times 10^{-2} \text{ m}^3/\mu\text{g})$, Ni $(2.6 \times 10^{-4} \text{ m}^3/\mu\text{g})$, and Pb $(1.2 \times 10^{-5} \text{ m}^3/\mu\text{g})$.

2.5.4. Combined non-cancer and cancer risk rates

Equation (6) was used to estimate the total non-cancer risk (*HI*) for the inhalation of heavy metals at the same time (US EPA, 2007):

$$HI = HQ_1 + HQ_2 + \dots + HQ_n \tag{9}$$

Also, equation (7) was used to estimate the total cancer risk (R) for the inhalation of heavy metals at the same time (US EPA, 2007):

$$R_{\rm t} = R_1 + R_2 + \dots + R_{\rm n} \tag{10}$$

where 1-n: specified the number of air pollutants such as heavy metals.

3. Results

3.1. Inhaled PM mass

The range of hourly inhaled dose of PM_{10} experienced by each occupation type for the study volunteers during the haze event is summarized in Fig. 3. Compared to HW, females working on buses and as outdoor laborors inhaled more particulate matter within one hour of exposure during the morning peak due to more direct exposure to traffic sources. The corresponding average hourly inhaled doses were $11.82\pm4.47~\mu\text{g/kg}\cdot\text{h}$ for HW, $18.86\pm7.94~\mu\text{g/kg}\cdot\text{h}$ for BC and $17.40\pm6.75~\mu\text{g/kg}\cdot\text{h}$ for ML, respectively. The actual hourly inhaled dose (6.39–35.77 $\mu\text{g/kg}\cdot\text{h}$) of PM_{10} was significantly higher (between 2.5–17.6 times



Fig. 3. Comparison between the hourly inhaled dose (*DI*) of PM₁₀ derived from theoretical calculation, and actual exposure for three different occupation types undertaken by females. Here, DI_t is the theoretical hourly inhaled dose; DI_a -BC is the actual hourly inhaled dose of bus conductor; DI_a -ML is the actual hourly inhaled dose of manual laborer; and DI_a -HW is the actual hourly inhaled dose of home worker.

higher) than the theoretical calculation values (0.93–3.85 μ g/kg·h). The above results show the inhaled mass of PM₁₀ into the human body based on the acute exposure model are significantly underestimated. A previous study also found that the use of ambient outdoor PM exposure could have attenuated the estimates towards the null compared to estimates that would have been expected with personal PM monitoring (Kioumourtzoglou et al., 2014).

3.2. Inhaled heavy metal levels

The hourly inhaled mass of heavy metals in PM₁₀ during the haze event for the three occupation types are presented in Fig. 4. The inhaled mass of heavy metals in PM₁₀ for As, Cd, Co, Cr, Ni and Pb were in the range of 0-0.65, 0-0.079, 0-0.17, 0-2.74, 0-0.82, and 0-1.56 ng/kg·h, respectively. The inhaled mass of heavy metals in PM₁₀ showed the same trend as that presented in PM pollution over time (Fig. S1). The highest inhaled dose of heavy metals in PM₁₀ corresponded to the heaviest PM pollution (287 μ g/m³ on 5th November 2016), and the inhaled dose for the BC, ML and HW groups was 5.75, 2.61 and 0.99 ng/kg-h. On average, the BC group inhaled the highest mass of heavy metals, reaching 2.26 ng/kg·h, with Cr ranked as the main heavy metal contributor (54.2 %). This was mainly attributed to traffic derived oil combustion (Chen et al., 2016), and atmospheric pollution from surrounding steel metallurgy works (Liu et al., 2018). Overall, the ML group inhaled the greatest amount of heavy metals (1.12 ng/kg·h), then the HW group (0.38 ng/kg·h). Compared to the BC group, ML and HW inhaled more As, accounting for 38.7 % and 54.1 % of the inhaled heavy metals mass, respectively. For ML and HW exposure was mainly from atmospheric deposition originating from surrounding pollution sources. During the annual heating period in Beijing coal combustion is widely used for suburban household heating (Zhao et al., 2021), which resulted in greater exposure to As (Tian et al., 2015).

3.3. Health effect endpoints of urban dwelling women

The estimated numbers of six health effect endpoints for the BC, ML and HW groups in Beijing are summarised in Fig. 5. The results indicate that the predicted health effect endpoint incidence caused by actual inhaled PM_{10} is significantly underestimated (p < 0.05), especially for women exposed to traffic sources i.e., BC group. The estimated numbers



Fig. 4. Actual inhaled heavy metals under short-term exposure during the 2016–17 haze event for the three occupation types occupied by women, including a) bus conductor (BC), b) manual laborer (ML), and c) home worker (HW).

of individual mortality in the BC, ML and HW groups by actual inhaled PM_{10} were 17,000–54,000, 14,000–39,000 and 11,000–31,000, which were 9.57, 7.12 and 5.65 times greater than that obtained by traditional monitoring methods, respectively. Asthma had the highest number of expected cases amongst the six diseases, with the BC, ML and HW groups having 85,000–270,000, 69,000–198,000 and 56,000–156,000 cases, which were 9.71, 7.23 and 5.75 times greater than that calculated by traditional methods, respectively. The estimated number of chronic bronchitis cases in the BC, ML and HW groups was 30,000–97,000, 24,000–70,000 and 20,000–54,000, which was 9.20, 6.82 and 5.41



Fig. 5. Six chronic diseases endpoints from the exposure–response model (solid lines) and 95% confidence intervals (dashed lines) due to actual PM₁₀ inhalation during the November 2016 haze event for urban dwelling women including bus conductor (BC), manual laborer (ML), and home worker (HW); Theoretical calculations (TC).

times more than the traditionally derived values, respectively. The estimated numbers of acute bronchitis cases in the BC, ML and HW groups was 78,000–250,000, 64,000–182,000 and 53,000–143,000, which was 9.25, 6.86 and 5.44 times higher than traditionally calculated values, respectively. Although the estimated numbers of respiratory and cardiovascular hospitalization cases were lower amongst the six chronic diseases (BC group: 1,400–17,000, ML group: 1,100–12,000, and HW group: 1,000–9600), they exhibited higher excess cases, which were 12.30, 8.85 and 6.76 times the traditionally derived values, respectively.

3.4. The health risk of inhaled heavy metals

According to the directly obtained hourly inhalation mass of As, Cd, Co, Cr, Ni and Pb in PM_{10} , the health risks to the three occupation groups were assessed (Fig. 6). The average non-cancer *HI* of BC (4.16), ML (3.49) and HW (1.48) were all greater than 1, indicating that these women all faced a harardous but non-cancer related health risk (US EPA, 1989). Among the metals these groups were exposed to As made the greatest contribution to non-cancer *HI*, with risks (contributions) of 1.91 (45.9 %), 2.19 (62.9 %) and 1.02 (68.8 %), respectively. Moreover, other metals showed increased non-cancer *HQ*, such as Cr (0.03–2.0), Ni



Fig. 6. The health risk of actual inhaled heavy metals under short-term exposure during the November 2016 haze event for the three occupational groups of women including a) bus conductor (BC), b) manual laborer (ML), and c) home worker (HW).

(0.09–2.92) and Co (0.08–2.09) for the BC group, Ni (0.04–3.11) for the ML group, and Ni (0–0.7) for the HW group. The non-cancer *HQ* of metals in descending order was As (1.71) > Ni (0.61) > Cr (0.35) > Cd (0.17) > Co (0.16) > Pb (0.03). The average cancer R_t of the BC group, ML group and HW group was 7.77×10^{-3} , 1.23×10^{-3} , and 4.26×10^{-4} respectively, indicating that there was cancer risk for the BC and ML groups (Deng et al., 2016). Among these metals Cr made the greatest contribution to cancer R_t , with risks (contributions) of 7.61×10^{-3} (98.0%), 1.07×10^{-3} (87.4%) and 3.55×10^{-4} (83.3%), respectively. The cancer *R* of Cr for the BC and ML groups was higher than the acceptable risk level ($1.0 \times 10^{-6} - 1.0 \times 10^{-4}$) (Megido et al., 2017). The element As also showed a higher cancer *R*, such as $5.11 \times 10^{-5} - 2.05 \times 10^{-4}$ for the BC group, $2.14 \times 10^{-5} - 1.91 \times 10^{-4}$ for the ML group, and

 $0{-}1.07\times10^{-4}$ for the HW group. The cancer R of metals in descending order was Cr (3.01 $\times10^{-3})$ > As (1.1 $\times10^{-4})$ > Co (9.88 $\times10^{-4})$ > Ni (3.15 $\times10^{-6})$ > Cd (3.09 $\times10^{-6})$ > Pb (2.03 $\times10^{-7})$. For non-cancer risk, excessive inhalation of As may increase cardiovascular risk in women (Wu et al., 2013). For cancer related risk, long-term inhalation of Cr and As may increase lung cancer risk in women (Coyle et al., 2006; Yoshikawa et al., 2013).

4. Discussion

4.1. Underestimated risk of PM_{10} and heavy metals exposure

In the study, the actual PM₁₀ intake in the exposure groups in Beijing

during a severe haze event was firstly measured by nasal filters (Fig. 3). We find that, the hourly inhaled dose of PM₁₀ obtained by nasal filters was 2.5-17.6 times that obtained by traditional PM sampling methods with a volume sampler and filters from static monitoring sites (Megido et al., 2017; Ramirez et al., 2020; Broomandi et al., 2023). These data collection methods were generally undertaken at variable sample flow rates, heights, and durations such as 1.7-500 L/min, 1-1.5 m and 1-24 h (Cores et al., 2016; Zhang et al., 2018; Puangprasert and Pueksasit, 2019). Obviously, the control condition for PM_{10} sampling utilizes passive sampling of a pumped flow at a defined suction pressure. The actual suction pressure may be expected to decrease as the filter membrane becomes saturated and blocked by PM₁₀, thereby limiting further collection. In addition the sampling height, distance and time of PM exposure may also be quite different from that of real exposure levels due to air pollution sources and contamination diffusion conditions (Chen et al., 2019; Bendl et al., 2023). The presence of personal clouds in relatively enclosed spaces, such as offices, confirmed that the actual concentrations of PM₁₀ exposed to individuals were much greater than ambient PM₁₀ measured by fixed sampling sites in the area (Yang et al., 2023). In actual exposure scenarios, people may be closer to the emitting source and are continuously adjusting inhaled and exhaled air volume possibly in response to perceived pollution or exertion levels (Patel et al., 2016). Such psysiological responses serve to increase the level of inhaled PM, exposing deeper parts of the lung tissue and greater capture. Thus, traditional sampling methods underestimated actual PM₁₀ intake for exposure groups in Beijing. As a result, among urban dwelling women who work in Beijing and are exposed to PM₁₀, the mortality and morbidity due to PM inhalation are underestimated by 17,000-32,000 and 200,000-380,000, respectively (Fig. 5).

Based on the actual inhalation of PM obtained by nasal filters, the direct inhaled mass of heavy metals for urban dwelling women in Beijing was also obtained, which was 23.0 ng/m³ for As, 1.57 ng/m³ for Cd, 0.83 ng/m^3 for Co, 27.6 ng/m³ for Cr, 10.4 ng/m³ for Ni and 13.2 ng/m³ for Pb, respectively (Table 2). These values are significantly higher than those obtained using a medium-volume filter sampler during winter 2015 in Beijing (Gao and Ji, 2018); the multiple is between 1.0 times for Pb and 12.7 times for Ni. In addition, the exposure values of heavy metals (except Pb) obtained in this study were significantly higher than those found in other cities around the world and obtained by traditional sampling methods (Table 2). For example, Langreo in Spain (Megido et al., 2017), Acerrra in Italy (Vaio et al., 2018), Ulsan in Korea (Hieu and Lee, 2010) and Bogota in Colombia (Ramirez et al., 2020). The relatively low Pb inhalation rates may be attributed to China's energy consumption which is dominated by coal (Xue et al., 2016) with more released As (Tian et al., 2015). Pb is mainly derived from vehicle exhausts and tyre-abrasion materials (Das et al., 2020; Broomandi et al., 2023). The higher heavy metal inhalation rates observed in this study also led to elevated estimates of cancer and non-cancer morbidity risk. Both non-cancer (44.3 times) and cancer (13.0 times) risk were significantly higher than those obtained using a medium-volume filter sampler in winter 2015 in Beijing (0.06 and 1.0 \times 10⁻⁴) (Gao and Ji, 2018). Compared to other cities, the non-cancer and cancer risk found in this study were 2.9-6.5 times and 15.1-447.3 times higher, respectively (Table 2).

The high exposure levels of PM_{10} can increase cancer risks and cardiopulmonary injuries in humans (Ren et al., 2021; WHO, 2021). Therefore, in order to better prevent and control the problems caused by PM_{10} pollutionn to exposure human in urban scale, more accurate PM_{10} measurement is very important. For exposed population living or working in fixed areas, measurement of surrounding fixed stations can be used, but it is recommended to use smaller geographic units such as commercial area, industrial area, communities, campus and street to deploy monitoring instruments (Huang et al., 2018; Moniruzzaman et al., 2022; Rienda et al., 2023). For exposed population who work or spend a lot of time on vehicles such as bus and subway, PM_{10} monitoring devices can be installed on the vehicles, or volunteers in the exposed

Table 2 Comparison of	airhorne DM inh:	aled heavy metals concentrations and health rick	in Reiiing with thos	e renorted for other citie	e around i	he worl	Ţ						
City	Site feature	acutation sources concentrations and accutation task	Sampling time	PM ₁₀ mean	The mea	in mass o	of inhale	l heavy n	netals ng,	/m ³	Hazard in	dex of	References
				concentration in air							nealun rist		
				µg/m³	As	Cd	Co	ų	Ni	Pb	non- cancer	cancer	
Langreo,	Industrial area	A high-volume sampler with filters (MCV CAV-A/	2013 October-	37.9	3.10	0.30	0.10	2.20	1.80	13.6	0.91	3.82E-	Megido et al.,
Spain		MSb)	2014 October									05	2017
Acerra, Italy	Traffic and	A low-volume sampler with filters (Skypost, TCR	2013, Winter	39.6	2.74	4.77	3.80	19.7	16.7	50.6	0.41	1.40E-	Vaio et al.,
	industrial area	Tecora, Italy)										05	2018
Ulsan, Korea		An ambient cascade impactor with filters (Model	2008, Spring	60.3	I	1.10	I	3.70	10.9	49.5	I	3.51E-	Hieu and Lee,
		20–800, Tisch Environmental, Inc.)										05	2010
Bogota,		A high-volume sampler with filters (Hi-Vol Tisch	2016, January	55.0	1.20	0.44	0.51	4.43	1.79	19.5	I	5.50E-	Ramirez et al.,
Colombia		Environmental Inc.)										06	2020
Beijing,	Traffic area	A medium-volume sampler with filters (PM ₁₀ -	2015, Winter	195	I	0.51	T	5.2	0.82	13.2	0.06	1.90E-	Gao and Ji,
China		Laoying, China)										04	2018
Beijing,	Tracffic area	A Woodyknows nasal filter (M&M Pure Air Systems	2016, October-	184	23.0	1.57	0.83	27.6	10.4	13.2	2.66	2.46E-	In the study
China		LLC, Delaware, USA)	November									03	

population wear mobile monitoring devices (Hou et al., 2016; Adrian et al., 2023; Bendl et al., 2023). Using mobile measurements, a high spatio-temporal variability of PM10 such as dynamics and trajectories of PM₁₀ in exposed populations can be obtained. In addition, using spatial regression techniques and machine-learning algorithms to analyze the exposure data can further allow a reduction in measurement error and better identifcation of air pollution variability within these areas (Gouveia et al., 2022; Lyu et al., 2023). However, the above campaigns can have high requirements on organization and equipment installation, which may be one reason why only a limited studies around the world have been carried out. Although these methods are more accurate in obtaining the actual PM10 exposure concentration, they still do not reflect the actual amount of PM10 inhaled by the exposed population during air pollution. The use of nasal filters can help us solve this problem. Still, this method has some disadvantages in the practical application process, such as the need for a large number of volunteers, the need to consider the cost of nasal filters consumption, and the fact that it is not suitable for risk prediction of larger regions and larger populations. However, this method may be more effective for some sensitive minority groups to assess their exposure to PM₁₀, and can more accurately assess their risk during PM₁₀ pollution.

4.2. Effect of different exposure scenarios on inhaled PM_{10} and health risk

In cities, women in different occupations have different exposure routes during air pollution due to different living and lifestyles, which also leads to different exposure risks for them (Ozkaynak et al., 2013; Broomandi et al., 2023). In the study, we used nasal filters to obtain actual PM10 levels for three groups of professional urban women including BC, ML and HW during a severe haze event, and accordingly asssed their health risk. The actual value of inhaled PM_{10} was 18.9 μ g/ kg·h for a BC group, 17.4 µg/kg·h for a ML group and 11.8 µg/kg·h for a HW group, respectively. The ratio of the BC, ML and HW to get diseases from PM₁₀ pollution was 1.74:1.27:1.00. BC was predicted to have the highest numbers of health endpoints the six chronic diseases (Fig. 5). At the same time, BC also faced the greatest non-cancer and cancer risks, and the corresponding hazard indexes reached 4.16 and 7.8 \times 10^{-3} (Fig. 6). Women who are exposed to outdoor traffic inhaled more PM and faced higher health risks. This finding is consistent with previous research that road traffic is a major contributor to people's short-term high exposure to PM₁₀ in urban areas (Pan et al., 2022; Rienda et al., 2023). Although HW spends a lot of time indoors, it also exposes HW to high PM₁₀ during severe haze due to cooking and opening windows for ventilation (Xiong et al., 2018; Ji et al., 2021). It was reported that indoor-outdoor concentration ratios of PM pollution are often over 50 % due to leakage in the building and windows (Shi et al., 2015). In addition, another reason for HW exposure to high concentrations PM₁₀ may be attributed to the fact that we used nasal filters to obtain actual mass of PM₁₀ inhalation, which is significantly higher than the fixed site monitoring value such as 0.95 μ g/kg·h (Patel et al., 2016).

The occupational characteristics of urban women make them different in distance and contact time with PM pollution sources, which brings different exposure risks. Therefore, it is necessary to take targeted protective measures for women in different professions to reduce the risk of PM pollution during severe air pollution. For women like BC who work in urban transport and its hubs, such as buses, taxis and subways, they spent about 8 h a day outdoors and stay in these places with traffic pollution sources. We recommend that they wear N95 respirators or surgical masks with higher filtration efficiency because of their long exposure time (Ji et al., 2021). Meanwhile, we propose to install air purifiers (ideally equipped with high-efficiency particulate air filters) in the vehicles and subways, and improve ventilation at hubs and platforms (Bendl et al., 2023; Peng et al., 2023). For women like ML who work outdoors, such as urban street cleaning and urban greening care, they also typically spend a lot of time (about 3–5 h per day) outdoors and

stay the place close to the traffic, but they are in areas with relatively good air diffusion conditions. We recommend that they wear dust mask or cotton masks to reduce their exposure to dust and vehicle emissions (Chen et al., 2019; Ejohwomu et al., 2022). For women like HW who stay indoors for a long time, including medical workers, office ladies and shopping mall sales, the improvement of the indoor environment will be especially important. Therefore, one of the effective measures is the capture of PM particles using high-performance air purification filters in the ventilation and air conditioning systems of buildings during hazy days (Sabirova et al., 2021). In addition, effectively closing doors and windows is also important to reduce indoor PM pollution (Xiong et al., 2018). In addition to the above-mentioned measures, a number of measures from management to technology should also be considered. For example, minimizing activities and working outdoors can effectively reduce the risks from PM pollution (Sun et al., 2021). Limiting the number of vehicles on the road, upgrading the public transport system, and encouraging green mobility such as walking and cycling can effectively reduce the level of PM pollution (Gouveia et al., 2022). Additional air pollution control options, such as fuel switches, energy efficiency optimization and improving vehicle emissions may also be useful (Patel et al., 2016; Rao et al., 2021).

4.3. Uncertainties and limitations

Although the above-mentioned modifications were adopted to improve the accuracy and reliability of exposure results and assessment model, appreciable uncertainties are inevitably introduced into the risk assessment. These uncertainties and limitations may be identified in at least four aspects. Firstly, the exposure data available were limited to a small sample size of participants (n = 6), and within a relatively short sampling duration (10 samples in 45 days) and a small-scale study area. These small numbers contributed to uncertainty which may not ensure statistical robustness in our data and estimates. Also, the limited representativeness of data may potentially prohibit data extrapolation across exposed populations and local calibration of exposure model parameters. Secondly, despite actual exposure measurements of individuals which may be regarded as preferable compared with traditional sampling methods (Kioumourtzoglou et al., 2014) and acceptable LOD and RSD values in this study, there were still instrumental and methodological errors in PM sampling and heavy metals analysis. Further, using data from the sampling time of day (8:00 am to 9:00 am) may overestimate the whole-day-average risk, partiuclarly for the bus conductor group exposed to elevated levels of mobile traffic sources of PM due to rush hour commuting (Zhao et al., 2009). Thirdly, although we adjusted the traditional exposure assessment model based on actual individual parameters, exposure parameters such as TR and IR used in the model may exhibit significant regional or individual differences. Similarly, toxic-related parameters such as RfD and IUR were recommended by the US EPA, which were obtained based on a different geographical population which may therefore differ to residents in Beijing; local parameters had not yet been developed. Finally, the toxic effects of metal elements are related to their valence and availability. For example, Cr(III) is generally regarded as an essential trace element in the human body, whereas, Cr(VI) has been determined as a human carcinogen. In this study, total metal concentrations were used in the risk assessment, which may cause an overestimation of risk levels. The joint toxicity of metal cocktails (or organometallic mixtures) and the physical-chemical properties (e.g., oxidative potential) of PM may also contribute to an underestimation of health risks (Lin and Yu, 2019; Xu et al., 2020). Even though there are uncertainties and limitations in risk assessment, our study directly obtains data on PM₁₀ and heavy metals inhaled by exposed individuals and which can therefore reduce the uncertainty of risk assessment using exposure parameters. The next meaningful step should be to compare the differences between tranditional air sampling and nasal filters in the same dimension under air pollution, and further refine calibrated parameters of the exposure

model.

5. Conclusions

Our results provide evidence that the health risk analysis for exposed populations based on traditional air particulate sampling methods ignores individual differences in exposure pathways and behaviors, and significantly underestimates inhalable PM, other airborne pollutants and health risks of exposed individuals under a severe haze event. We recommend more accurate sampling methods be adopted for different exposed individuals to effectively reduce evaluation model uncertainty of the associated health hazards and risk predictions. We also recommend implementing a more effective protection strategies during haze episodes to reduce PM pollution and to protect exposed populations. In addition, the comparison between active sampling and traditional air pumping passive sampling methods should be carried out, and the correction coefficient of the exposure evaluation model should be established to quickly and accurately obtain individual exposure risk under air pollution conditions. Although the method of nasal filters in this study can directly obtain the amount of PM and PM-loaded pollutants actually inhaled by the exposed population, this method has its limitations in practical application. For example, a large number of volunteers are needed, the wearing process affects breathing, the consumption cost of nasal filters needs to be considered, and it is not suitable for air pollution monitoring and risk assessment in larger regions and large populations. Therefore, it is also recommended to divide the sensitive groups and hot spot of PM pollution through fixed site and mobile site monitoring, and then select key groups in representative areas to wear nasal filters for PM monitoring and risk analysis.

CRediT authorship contribution statement

Wei Guo: Writing – review & editing, Writing – original draft, Project administration, Funding acquisition. Xinyou Zhang: Methodology, Data curation. Junhui Yue: Writing – review & editing, Investigation. Yue Gao: Writing – review & editing. Martin R. Tillotson: Writing – review & editing. Xu Zhao: Writing – review & editing, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary material

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