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Risk assessment of population inhalation exposure to volatile organic compounds and carbonyls in urban China

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ABSTRACT

Over the past three decades, China has experienced rapid urbanization. The risks to its urban population posed by inhalation exposure to hazardous air pollutants (HAPs) have not been well characterized. Here, we summarize recent measurements of 16 highly prevalent HAPs in urban China and compile their distribution inputs. Based on activity patterns of urban Chinese working adults, we derive personal exposures. Using a probabilistic risk assessment method, we determine cancer and non-cancer risks for working females and males. We also assess the uncertainty associated with risk estimates using Monte Carlo simulation, accounting for variations in HAP concentrations, cancer potency factors (CPFs) and inhalation rates. Average total lifetime cancer risks attributable to HAPs are 2.27×10^{-4} (2.27 additional cases per 10,000 people exposed) and 2.93×10^{-4} for Chinese urban working females and males, respectively. Formaldehyde, 1,4-dichlorobenzene, benzene and 1,3-butadiene are the major risk contributors yielding the highest median cancer risk estimates, $>1 \times 10^{-5}$. About 70% of the risk is due to exposures occurring in homes. Outdoor sources contribute most to the risk of benzene, ethylbenzene and carbon tetrachloride, while indoor sources dominate for all other compounds. Chronic exposure limits are not exceeded for non-carcinogenic effects, except for formaldehyde. Risks are overestimated if variation is not accounted for. Sensitivity analyses demonstrate that the major contributors to total variance are range of inhalation rates, CPFs of formaldehyde, 1,4-dichlorobenzene, benzene and 1,3-butadiene, and indoor home concentrations of formaldehyde and benzene. Despite uncertainty, risks exceeding the acceptable benchmark of 1×10^{-6} suggest actions to reduce exposures. Future efforts should be directed toward large-scale measurements of air pollutant concentrations, refinement of CPFs and investigation of population exposure parameters. The present study is a first effort to estimate carcinogenic and non-carcinogenic risks of inhalation exposure to HAPs for the large working populations of Chinese cites.

1. Introduction

Public concern regarding organic hazardous air pollutants (HAPs) continues to grow world-wide. HAPs have a wide range of sources, effects and exposure routes. Their sources include indoor sources (Clarisse et al., 2003; Kim et al., 2001), outdoor vehicle emissions and industrial combustion (Guo et al., 2004b; Ohura et al., 2009). Their adverse health effects range from irritation of eyes, skin, mucous membranes and respiratory tract (Jones, 1999; WHO, 2010) to serious chronic illnesses such as asthma (Weisel, 2002), chronic obstructive pulmonary disease (Viegi et al., 2006), cardiovascular disease and cancer (Lewtas, 2007; WHO, 2010). Because of their relatively low boiling points and high vapor pressures, the main exposure pathway to most HAPs is through inhalation (Ramirez et al., 2012).

Due to rapid industrial and economic development over the past three decades, China has experienced large migration from rural areas to cities; urban growth and modernization; and a concomitant increase in urban air pollution (Fang et al., 2009; Zhang et al., 2013b). From 1990 to 2010, China's urban population more than doubled; net urban residential building area grew from 4 to 21 billion m^2; and the number of motor vehicles increased from 5 to 78 million (NBS, 2011). By 2007, China's formaldehyde production had reached a staggering 12,000 kt, about 4000 times what it was five decades earlier. More than 65% of formaldehyde produced is used to synthesize resins used in construction materials which are therefore a source of indoor formaldehyde pollution (Tang et al., 2009).

However, qualitative and quantitative changes in population exposure to HAPs with urbanization in China have not been characterized sufficiently, nor have the health risks of these HAP exposures. The few studies that have assessed health risks posed by HAPs in China have been limited to either indoor or outdoor exposure, or to a small number of carcinogenic HAPs, or to localized cases within small sample sizes. Zhou et al. (2011) monitored personal exposure to volatile organic compounds (VOCs) and estimated associated cancer risks (median, 4.4×10^{-5}, which means 4.4 additional cases per 100,000 people exposed) in Tianjin. However, their results were limited because formaldehyde and acetaldehyde were not considered and data was only

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collected for 12 participants. Other studies have only considered risks caused by exposure to a small suite of VOCs in indoor environments, such as residences (Guo et al., 2004a; Liu et al., 2013; Ohura et al., 2009; Weng et al., 2010), commutes (Feng et al., 2010; Li et al., 2009; Pang and Mu, 2007), public places (Weng et al., 2009), hotels (Feng et al., 2004), and hospitals (Lu et al., 2006), and hence could not give a complete picture of risk. Some studies have considered high exposure scenarios, such as recent renovation, cooking and indoor combustion, but which only evaluate a narrow suite of pollutants (Du et al., 2014; Huang et al., 2011). So far, no broad analyses of the personal exposure and associated cancer risks posed by a wide range of organic HAPs in multiple microenvironments have been done for China.

The paucity of information on HAP risk assessment has hindered risk-based regulatory decision making and regulatory actions aimed at protecting public health in China. In 2003, China's national indoor air quality standard GB/T 18883 was put into effect, which set guideline values for formaldehyde, benzene, toluene, xylenes and total volatile organic compounds (TVOCs) respectively (GB/T, 18883, 2002). However, for probable carcinogens like trichloroethene, tetrachloroethene, and for possible carcinogens like ethylbenzene, carbon tetrachloride and chloroform (IARC, 2011), the concentrations in both indoor and outdoor air is not yet regulated in China, probably due to lack of information on exposure and risk assessment. Therefore, information is required for regulatory policy.

In this study, we first summarize recent HAPs measurements in urban China, focusing on those HAPs with frequent occurrence in different microenvironments, and then derive a personal exposure model for working adults. Then, cancer risks due to inhalation are calculated using the method proposed by the California Office of Environmental Health and Hazard Assessment (OEHHA), with risks apportioned between indoor and outdoor sources. Because a great deal of uncertainty is often incorporated with risk assessment due to the variations in HAP concentrations, inhalation rates among individuals and toxicity of the HAPs, uncertainty in the risk estimates is also assessed by using a probabilistic risk assessment approach.

2. Methods

2.1. Urban China

In the present study, we focused on HAP exposure modeling and risk assessment in three urban areas of China, the Pearl River Delta (PRD) region, the Yangtze River Delta (YRD) region and the Bohai Rim (BR) region. All three regions have high population density and a developed economy: in 2007, the three regions accounted for more than 40% of China's entire gross domestic product (GDP). While nearly 20% of China's total population lived in these areas, they account for only about 5% of China's land area (Ness et al., 2010). Due to industrial and vehicle emissions in cities, outdoor air in these regions tends to be more polluted than in rural and suburban areas of China (Chan and Yao, 2008; Zhang et al., 2013b). The megacities Beijing, Hong Kong, Shanghai, Guangzhou, Hangzhou, Nanjing, Tianjin, and Dalian in these regions were selected as target cities for the present study.

2.2. Literature review and data collection

We searched the literature for peer-reviewed studies reporting measurements of HAPs in various microenvironments using the ISI Web of Science database for articles published prior to August 2013. The HAP measurements selected, preferably from the last 10 years (2003–2013), but at least within the last 15 years so as to reflect recent HAP emission resources and characteristics in urban China, presented useful data on HAP concentrations in one or more microenvironments.

Research studies reporting HAP measurements in different microenvironments were considered regardless of measurement duration and HAP source. Thus, studies included both short- and long-term measurements. Also, the HAPs considered included those mainly emitted from either outdoor or indoor sources, with some having both outdoor and indoor sources. Microenvironments used to determine air pollutant exposures in the present study included indoor residences (home), indoor work environment (office), commute and outdoor/other, which were defined as the representative microenvironments (Dodson et al., 2007; Weisel, 2002). The miscellaneous "other" microenvironments were assigned the same concentration distributions as the outdoors. Table 1 shows the selected references with information on study location, sampling method, sampling duration, sample size, microenvironment and the types of pollutants.

Sixteen HAPs with high abundance in different microenvironments were considered, covering the most high-risk pollutants as listed by Payne-Sturges et al. (2004) and Sax et al. (2006). These were formaldehyde, acetaldehyde, 1,3-butadiene, 1,4-dichlorobenzene, benzene, carbon tetrachloride, chloroform, ethylbenzene, styrene, tetrachloroethene, trichloroethene, toluene, m,p-xylene, o-xylene and 1,2,4-trimethylbenzene.

As raw data, namely the concentrations of individual compounds in individual samples collected during each study were not available for most studies; the reported statistics were used for analyses. To properly weight statistics from individual studies of a given city, we first compared each study's parameters, including percentiles, arithmetic mean, standard deviation, median, min, and max. We weighted equally the studies for each city/geographic region as per Loh et al. (2007). If there was more than one study for a city, the studies were weighted according to the number of unique measurements. Usually, the weighting was by the number of sampling sites; less frequently, the weighting was by the number of samples.

According to *Time use patterns in China* (NBS, 2010), the automobile is the most common mode of commuting for Chinese urban working adults. On average, working adults spend about 60% of their commuting time in an automobile. Therefore, when compiling HAP concentrations in commute microenvironments, we mainly focused on automobiles. Outdoor HAP concentrations near roadways and HAP concentrations in subways were treated as surrogates only when there was no available data. Additionally, due to lack of sufficient indoor acetaldehyde concentrations for offices, residential acetaldehyde concentrations were used as surrogates. Table 2 presents the summary statistics for the 16 HAPs in home, office, commute and outdoor/other. Table S1 (Supplementary material) shows the means and SDs for each outdoor measurement.

2.3. Exposure model and risk characterization

Fig. 1 presents the schematic diagram of risk characterization for hazardous air pollution. The current standard approach for health risk assessment was originally proposed by the U.S. National Academy of Sciences (National Academy of Sciences, 1983). The four steps of risk assessment are hazard identification, dose–response assessment, exposure assessment, and risk characterization. To estimate risk, we followed the guidance proposed by OEHHA (2003).

Risk information for the target HAPs, including cancer classification, cancer potency factor, cancer benchmark concentration and inhalation reference concentration, is presented in Table 3. Based on the weight of evidence presented by the International Agency for Research on Cancer (IARC) and Integrated Risk Information System (IRIS), all target HAPs are known to be human carcinogens, or probable and possible human carcinogens, except for toluene 1,2,4-TMB and the three xylenes. Cancer potency factors are defined as the upper-bound probability of developing cancer, assuming continuous lifetime exposure to a substance at a dose of one milligram per kilogram of body weight per day. Cancer benchmark concentrations ($\mu g/m^3$) are the exposure levels for cancer risk of one-in-one-million over a lifetime of exposure. A cancer risk of 1.0×10^{-6} is considered negligible risk (Caldwell et al., 1998; Woodruff et al., 1998). For non-cancer hazard assessment, inhalation chronic reference exposure limits (*REL*s) are used as an indicator of potential non-cancer health impacts. *REL* is defined as the concentration at