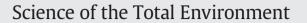
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Ambient air benzene at background sites in China's most developed coastal regions: Exposure levels, source implications and health risks

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HIGHLIGHTS

• Background benzene had higher cancer risk in China's developed regions.

· Coal/biofuel burning for heating increased benzene levels in winter in north China.

• Industrial emission contributed substantially to benzene in the Yangtze River Delta.

• Vehicle exhaust was the main source of benzene in the Pearl River Delta.

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ABSTRACT

Benzene is a known human carcinogen causing leukemia, yet ambient air quality objectives for benzene are not available in China. The ambient benzene levels at four background sites in China's most developed coastal regions were measured from March 2012 to February 2013. The sites are: SY_{NECP} , in the Northeast China Plain (NECP); YC_{NCP} , in the North China Plain (NCP); TH_{YRD} , in the Yangtze River Delta (YRD) and DH_{PRD}, in the Pearl River Delta (PRD). It was found that the mean annual benzene levels (578-1297 ppt) at the background sites were alarmingly higher, especially when compared to those of 60–480 pptv monitored in 28 cities in the United States. Wintertime benzene levels were significantly elevated at both sites (SY_{NECP} and YC_{NCP}) in northern China due to heating with coal/biofuels. Even at these background sites, the lifetime cancer risks of benzene (1.7-3.7E-05) all exceeded 1E-06 set by USEPA as acceptable for adults. At both sites in northern China, good correlations between benzene and CO or chloromethane, together with much lower toluene/benzene (T/B) ratios, suggested that benzene was largely related to coal combustion and biomass/biofuel burning. At the DH_{PRD} site in the PRD, benzene revealed a highly significant correlation with methyl tert-butyl ether (MTBE), indicating that its source was predominantly from vehicle emissions. At the TH_{YRD} site in the YRD, higher T/B ratios and correlations between benzene and tetrachloroethylene, or MTBE, implied that benzene levels were probably affected by both traffic-related and industrial emissions.

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1. Introduction

Benzene has been classified as a known Group I human carcinogen by the International Agency for Research on Cancer (IARC) (WHO, 1999). It has been estimated that a lifetime exposure of $1 \,\mu g/m^3$ of benzene leads to about 6 cases of leukemia per 1,000,000 inhabitants (WHO, 2000). Benzene may also cause central nervous system depression, cardiac arrhythmias, respiratory irritation and aplastic anemia (USEPA, 2002). As one of the most important hazardous air pollutants with adverse health effects, benzene in ambient air is widely regulated in many countries, or regions, to protect public health. For example, an annual limit of 5 μ g/m³ was recommended for benzene in ambient air by the European Union (EU, 2008), and the Inhalation Minimal Risk Level (MRL, at a cancer risk of 1 in 10,000) of 4.0 ppbv was established by the United States Environmental Protection Agency for benzene (USEPA, 2009a). These guidelines, or limits, for ambient benzene levels, however, are not available in China.

Since benzene has been forbidden to be used in solvents, the main emission sources are from combustion processes, including biomass/ biofuel burning and fossil fuel combustion (Andreae and Merlet, 2001; Moreira dos Santos et al., 2004; Liu et al., 2008). The rapid economic growth in recent decades has resulted in coal consumption in China

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reaching 2.41 billion tons Standard Coal Equivalent (SCE) by 2012, the number of vehicles surging to over 100 million with 37.5 million tons of gasoline, and 107 million tons of diesel being consumed by the transportation sector (NBSC, 2013a). Besides fossil fuels, it was estimated that 110–158 million tons per year of agricultural residues were burned in situ (Streets et al., 2003; Yan et al., 2006), and 522–568 million tons of stalks and firewood were used for cooking and heating in rural areas in China (NBSC, 2008). This huge fuel consumption induces a large amount of primary air pollutants, including benzene, being emitted into the atmosphere (Klimont et al., 2002; Wei et al., 2008, 2014). As estimated by Wei et al. (2014), total emissions of benzene in China were up to 1.06 million tons in 2005. With these massive benzene emissions, there are serious concerns for public health, and the regulation of ambient benzene level is of high importance in China.

More importantly, China's east-west imbalance in economic status and population density makes the health risks of ambient benzene a more alarming problem. The ten developed coastal provinces or municipalities, including Liaoning, Hebei, Beijing, Tianjin, Shandong, Jiangsu, Shanghai, Zhejiang, Fujian and Guangdong, covered 10.9% of China's land area. However, together they contained 40.6% of China's population in 2012, and they accounted for 61.3% of the Gross Domestic Product (GDP); 68.6% of the Gross Industrial Output; 27.9% of grain production; 54.8% of vehicle numbers and 50.5% of fossil fuel consumption (NBSC, 2013a). This implies that a larger portion of China's benzene emissions are concentrated in the densely populated coastal regions and, therefore, a large proportion of the population is exposed to elevated benzene levels in the ambient air. Previous studies in urban areas of this region, such as Beijing (Liu et al., 2009; Zhang et al., 2012a,b; Li et al., 2014) and Guangzhou (Wang et al., 2002; Barletta et al., 2008; Zhang et al., 2013), showed much higher benzene levels when compared to those reported in Europe, or in the United States. Even early in January–February 2001, benzene was found to pose potential health risks from a survey of volatile organic compounds (VOCs) in 43 Chinese cities (Barletta et al., 2005), mostly in the eastern areas. In China, the leukemia incidence rate was 4.3 per 100,000 in 2012 (Ferlay et al., 2013), with urban areas showing a higher occurrence than rural areas (Chen et al., 2012; Liu et al., 2013). In 2012, leukemia cases reported in China reached 65,778, accounting for 18.7% of the world's total leukemia occurrences (Ferlay et al., 2013). It is of high importance to investigate whether benzene exposure levels in China is a factor contributing to leukemia incidence.

To date, a systematic health risk assessment for benzene exposure has not been undertaken in China, and very few health risk studies are available with a focus on urban environments (Zhou et al., 2011; Zhang et al., 2012a; Li et al., 2014). The results of previous studies in urban areas cannot be regarded as representative of those in the whole developed coastal regions because field measurements were conducted at sites close to emission sources, and conducted over very short time spans. Monitoring of benzene levels at more background sites would, therefore, not only reveal background exposure levels, but also reflect the impact of regional social–economic changes. Unfortunately, monitoring at background or remote locations is scarce and sporadic in China (Guo et al., 2004; Tang et al., 2009; Suthawaree et al., 2010; Xue et al., 2013; Zhang et al., 2014). To fully understand the exposure levels and the potential health risks, long-term measurements are needed. Furthermore, identifying major sources of benzene is crucial to reducing emissions.

In this study, we monitored benzene levels at four background sites in China's most developed coastal regions concurrently for a one-year period. The sites were located in the Northeast China Plain (NECP), the North China Plain (NCP), the Yangtze River Delta (YRD) region, and in the Pearl River Delta (PRD) region. The purposes of this study are: (1) to investigate the background exposure levels of benzene in the most developed coastal regions; (2) to explore the main emission sources of benzene and their spatiotemporal patterns; and (3) to estimate the potential health risks of benzene in these developed regions.

2. Methodology

2.1. Description of sampling sites

The geographical locations of the sampling sites are shown in Fig. 1. The sites are all research stations included in the Chinese Ecosystem Research Networks (CERN, available at: http://www.cern.ac.cn/0index/index.asp/; last access: 19 June 2014). They are all located at rural or remote areas without factories and workshops in the neighborhood, and they are suitable sites to monitor regional background atmospheric compositions.

The SY_{NECP} site (Shenyang; 41.50°N, 123.40°E) is located in the Northeast China Plain (NECP), which has long been a famous base of China's heavy industry and a center of mining industry since 1953. Located in the center of the Liaoning Province, the SY_{NECP} site can receive air pollutants from the surrounding industrial cities. The site is 35 km south of the provincial capital Shenyang, 38 km northwest of Benxi, 32 km to the northeast of Liaoyang and 55 km to the northeast of Anshan, making it a good regional monitoring station.

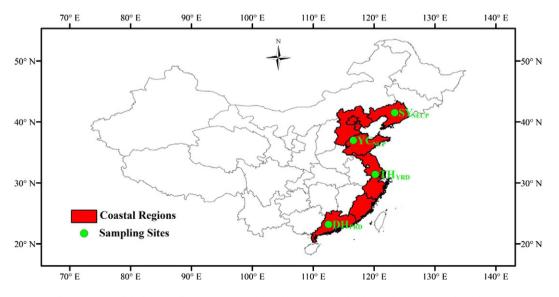


Fig. 1. Locations of the four sampling sites (green cycle) in China's most developed coastal regions (red colored areas).

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