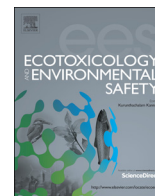




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# Ecotoxicology and Environmental Safety

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## Benzene homologues in environmental matrixes from a pesticide chemical region in China: Occurrence, health risk and management

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### ARTICLE INFO

#### Article history:

Received 21 October 2013

Received in revised form

28 January 2014

Accepted 28 January 2014

Available online 15 April 2014

#### Keywords:

BTEX

Risk assessment

Multi-media

Pesticide plant

### ABSTRACT

The contamination status and health risks of benzene, toluene, ethylbenzene and xylene (BTEX) in air, soil, dust and groundwater were evaluated in a pesticide chemical region located in Hebei province, China. The concentrations of BTEX in air ranged from 7.80 to 238  $\mu\text{g}/\text{m}^3$  and those in soil and dust ranged from lower than limit of detection (LOD) to 32,360  $\text{ng}/\text{g}$  dw, and those in groundwater varied from 2.68 to 98.6  $\mu\text{g}/\text{L}$ . Generally, the levels of BTEX in multimedia matrixes were all below the standards established in China. Health risk assessment was performed based on the monitoring data via inhalation, dermal contact and ingestion pathways and hazard quotient (HQ) was calculated to be on the order of  $10^{-7}$ , below  $10^{-6}$ , and Hazard index (HI) levels of BTEX were lower than 1.0. However, both HQ and HI ascended with an increase in work experience/exposure. Integrated risk management was proposed to eliminate BTEX pollution and to protect occupational health of workers in those industries.

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### 1. Introduction

Benzene, toluene, ethylbenzene and xylenes (BTEX) have been extensively used as the raw materials, intermediates and organic solvents in industrial and pesticide production process (Hu et al., 2010). Due to their volatility, relatively high water solubility and low  $K_{ow}$  values (Dou et al., 2008), BTEX may be dispersed in the environment related to meteorological conditions, tend to redistribute pollutants through advective and convective transports on both regional and large scales (Chen and Brune, 2012; Kumar and Viden, 2007). BTEX are released from a variety of sources, including combustion products of wood and fuels, traffic, industrial paints, adhesives, degreasing agents, and aerosols (von Eckstaedt et al., 2012; Na et al., 2004). Especially, pollution accidents of BTEX have significant implications on the surrounding environment. BTEX contaminants have been widely detected at numerous chemical industrial facilities and oil basin, especially the areas nearby the petroleum and related product manufactures. As neurotoxicants, BTEX are capable of causing abnormality, sudden change and a various cancers such as leukemia (Smith et al., 2007). The presences of BTEX in the environment are hazardous to public health (Weichenthal et al., 2012; Kuo et al., 1998) and ecological concerns

due to their toxicity and potential to be bio-accumulated throughout food chain. They are regarded as environmental priority pollutants in many countries (Dutta et al., 2009). Benzene has been classified as Group1 human health hazards by the International Agency for Research on Cancer (IARC, 2011). Chronic exposure to toluene, ethylbenzene and xylenes has been associated with adverse effects on the nervous system, respiratory system, liver and kidneys (Durmusoglu et al., 2010). Therefore, it is necessary to assess the contaminations and potential risks of these compounds in environment, particularly, the health risk impact to people who live and/or stay in the plants using BTEX as materials.

The typical pesticide chemical region is located in Hebei province, northern China, and pesticide production are the leading industries, which accounted for an important proportion of the GDP of the region and contributed 14 percent to the county's gross industrial output value in 2011. The regional annual dominant wind is NNE with a frequency of 13 percent and the dominant wind in spring is NNW. The climate in this region is East Asian continental monsoon climate, and is drought in temperate Asia and Mongolia with an average annual precipitation of 414.4 mm. There are three pesticide-producing factories (namely plants A, B and C, respectively) in the region, whose products are exported to the international market, columned the second national pesticide export industry, have important impact on local economic development. All of them used and stored a large number of organic chemicals, which may generate BTEX-containing pollutants during the production process. Therefore,

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it is a typical chemical enterprise area for investigating spatial distribution, composition characteristics and pollution status of BTEX in multi-medium environment. Meanwhile, a health risk assessment of BTEX was conducted based on the monitoring data. All investigated pesticide-producing factories in present study belong to the medium-sized enterprises (SMEs). The SMEs have been a significant engine of China's economic transition. While owing to the lack of specific management countermeasures, environmental pollution and accidents of the SMEs have plagued China's enterprise development and environmental management (He et al., 2014). The present study was conducted as a systematic investigation to trace sources, fates and risk of toxic substances in various environmental media from inside and/or outside of the SMEs in China. The objectives of the present study were to improve understanding of the contamination and potential sources of BTEX in the pesticide-producing area, to assess the health risk and negative impacts on occupational people, and then to provide scientific information for effective control and management of BTEX in the related chemical industrial region.

## 2. Materials and methods

### 2.1. Sample collection

In May 2012, a total of twenty samples were collected from air, surface soil, dusts and groundwater in an industrial district located in Hebei province in the north part of China. Detailed information of sample size in different environmental matrixes from each factory was presented in Fig. 1. The sampling process and the analysis procedure of atmospheric BTEX were mainly conducted based on the thermal desorption–gas chromatography method. Air samples were drawn through an absorption tube filled with Tenax (200 mg, Markes) at a flow rate of 20 ml/min for 20 min. Groundwater samples were obtained from the wells in the factories used for production and drinking. Samples were collected after 3 min flushing in 40 ml headspace vials (Agilent). The flow rate was reduced to avoid introducing bubbles during sampling. Hydrochloric acid (Merck, 37 percent) was added to acidify the water samples (pH < 2). The vials were immediately sealed with aluminum crimp caps (Agilent) with Teflon-faced septa (Agilent) and shaken to mix the content. All samples were transported in cooled containers and stored in the dark at 4 °C for a maximum of 5 days. Surface soil samples (0–20 cm) were taken from uncovered ground in both plants A and B but not in factory C where no ground was exposed, and obtained by mixing at least five adjacent topsoil samples (approximately 10 × 10 m). Approximately 200 g of dust composite samples were collected at

upwind and downwind in production and living area in the factories using vacuum cleaner from four corners at the area of 100 m<sup>2</sup>. Any soil or dusts off the vial threads were quickly brushed and the vials were immediately sealed with the septum and screw-cap. Store samples on icebox at 4 °C.

### 2.2. Instrument analysis

**Air:** the air samples were analyzed by a gas chromatography (GC) (GC-2010 SHIMADZU, Japan) equipped with a Photo Ionization Detector after thermal desorption (Pekin Elmer Turbo Matrix 650). GC parameters: helium was used as carrier gas at 30 ml/min, initial oven temperature 40 °C and were hold for 4 min, and to 40–120 °C at 10 °C/min and were hold for 13 min. Temperature of FID detector was 300 °C, H<sub>2</sub> was used as the carrier gas at rate of 40 ml/min.

**Water:** the sampling process and the analysis procedure of aqueous BTEX were conducted mainly based on the US Environmental Protection Agent (USEPA) method 5030c using a GC/MS (Agilent 7890 N/5975C) equipped with an automated headspace sampler (Teledyne Tekmar Aquatek 70), a purge and trap instrument (Tekmar Atomx). The chromatographic column was DB-VRX (60 m, 0.25 mm, 1.4 μm) and helium was used as the carrier gas at 0.9 ml/min. The inlet temperature was 200 °C and the split ratio was 10:1. The temperature program was set as: initial temperature was 45 °C and was hold for 10 min, then was increased up to 190 °C at 12 °C/min and was hold for 2 min, and was finally increased up to 225 °C at 6 °C/min and was hold for 1 min.

**Soil and dust:** Sampling process and the analysis procedure were conducted with the USEPA method 5035 A and 8260c using a GC/MS (Agilent 7890N/5975C) equipped with an automated headspace sampler (Teledyne Tekmar Aquatek 70), a purge and trap instrument (Tekmar Atomx). The chromatographic column was DB-624 (30 m, 0.25 mm, 1.4 μm) and helium was used as the carrier gas at 0.9 ml/min. The inlet temperature was 200 °C and the split ratio was 10:1. The temperature program was set as: initial temperature was 38 °C and hold for 1.8 min, and was increased up to 120 °C at 10 °C/min, and finally increased up to 210 °C at 15 °C/min.

### 2.3. Quality assurance and quality control

Recovery studies of BTEX were conducted to demonstrate the efficiency of the methods. The recovery rates ranged from 99 percent to 103 percent for air samples, 96 percent to 119 percent for soil/dust samples and 87 percent to 110 percent for groundwater samples, respectively. The detected limits for the air, soil/dust and groundwater samples were 0.50 μg/cm<sup>3</sup>, 0.50 ng/g dw and 0.50 μg/L, respectively.

### 2.4. Health risk assessment

The health risk assessment models recommended by USEPA were used to quantify the potential risk indexes of BTEX according to the monitored data.

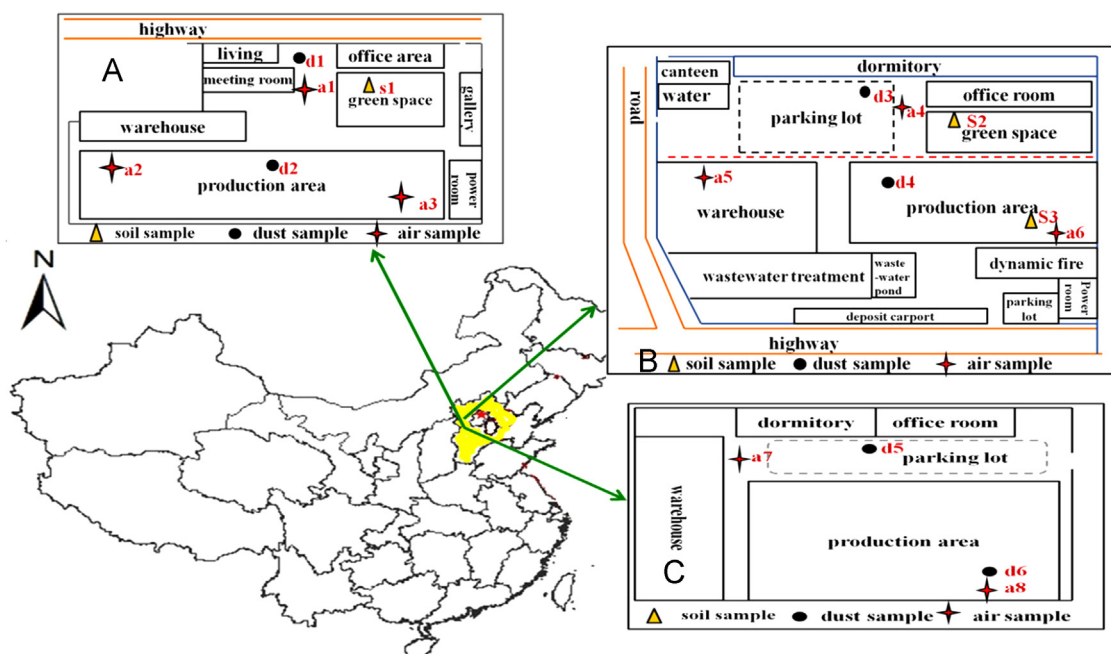


Fig. 1. Sampling area and sites in typical pesticide chemical region.

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