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Association of PAHs and BTEX exposure with lung function and respiratory symptoms among a nonoccupational population near the coal chemical industry in Northern China

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ABSTRACT

Emissions (particularly aromatic compounds) from coal industries and biomass fuels combustion lead to high health risks for neighboring residents. To investigate the association of polycyclic aromatic hydrocarbons (PAHs) and benzene, toluene, ethylbenzene and 1,2-dimethylbenzene (BTEX) exposure with lung function and respiratory symptoms among adults and children near the coal-chemical industry in Northern China, adults and children from a county dotted with coal chemical industry were chosen as subjects for investigation (investigated area, IR). The control group consisted of adults and children from an agricultural county (control area, CR). The environmental and urinary PAH and BTEX levels of adults and children were determined by isotope dilution liquid chromatography coupled with tandem mass spectrometry. The Mann-Whitney *U* test and multivariate linear regression models were used to analyze the relationship between pollutant exposure and the respiratory system. The results showed that in an ambient environment, levels of PAHs and BTEX in the IR were significantly higher than those in the CR. Particularly, the concentration profiles for air samples were IR > CR and indoor > outdoor. Both for adults and children, the geometric (GM) concentrations of urinary PAHs and BTEX from the IR were significantly higher than those measured in the CR. Additionally, the urinary PAH exposure level profiles of smokers were higher than those of nonsmokers, indicating that indoor air and smoking were both important nonoccupational exposure sources. The decline of the forced expiratory in the first second (FEV_1, %) and the forced expiratory middle flow rate ($FEF_{25\%}$) in children were associated with increasing urinary PAH metabolite levels ($p < 0.05$). The increase in urinary 1-OHN, 3-OHPhe, 4-OHPhe and 1-OHP levels could be linked to a decrease in FEV1 ($r = -0.179$, $p < 0.05$) and $FEF_{25\%}$ with the coefficient of -0.166, -0.201 and -0.175 ($p < 0.05$), respectively. Medical examinations and lung function tests indicated that residents in the IR had higher occurrences of chest inflammation or declining lung function than residents in the CR. Moreover, exposure to PAHs and BTEX could decrease child lung function, though decreased lung function was not observed in adults. Both urinary monitoring and lung function data showed that children were more sensitive to PAH and BTEX exposure than adults.

1. Introduction

Northern China is the main coal mining and consumption area. The coal chemical industry has emitted large quantities of toxic wastes, particularly polycyclic aromatic hydrocarbons (PAHs), benzene, toluene, ethylbenzene and 1,2-dimethylbenzene (BTEX), into the environment in Northern China. PAHs are formed during the incomplete combustion of fossil fuels. BTEX is a widely used chemical solvent and organic intermediate with high volatility. Studies show high levels of PAHs and BTEX in the most serious smog area in Northern China (Wang et al., 2006; Wang et al., 2011a, 2011b), and residents of Northern China have a higher body burden of PAHs and BTEX than people in other cities of China (Wang et al., 2011a, 2011b; Duan et al., 2003; Xia et al., 2010; Chen et al., 2016).

Long-term exposure to PAHs and BTEX irritates the respiratory system, causing symptoms such as asthma, inflammation and cough

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(Alshaarawy et al., 2013; Wichmann et al., 2009; Wickramasinghe et al., 2012; Rosa et al., 2011), particularly in children (Barraza-Villarreal et al., 2014; Jung et al., 2014). Toxicological and epidemiological studies have confirmed that lung is the target organ for PAHs (Gentner and Weber, 2011; Marie-Desvergne et al., 2010; Cioroiu et al., 2013). After oral exposure, BaP-diol epoxide (BPDE)-DNA adducts (a biomarker of genetic damage and exposure for PAHs) were significantly increased in the lung, liver, and mononucleated blood cells of the BaP-treated rats, with the highest levels found in the lungs (Marie-Desvergne et al., 2010). Clinical studies also confirmed that the lung tissue level of benzo(a)anthracene (a carcinogenic PAH) was significantly higher ($p < 0.05$) in patients from urban areas than from rural areas (Cioroiu et al., 2013). Chronical exposure to high levels of PAHs and BTEX in the air may influence lung function and even lead to a higher occurrence of lung cancer (Kim et al., 2014).

Mechanism and population research indicate that chronic respiratory symptoms are associated with damage induced by oxidative stress (Noh et al., 2015; Li et al., 2015). Lung function is an early and important indicator used to assess the influence of air pollution on respiratory systems. Unlike epidemic investigation through questionnaires, lung function testing is an objective outcome not affected by life habits and bias.

Studies of the relationship between PAH and BTEX exposure and lung function were established with monitoring data on gaseous levels (Mölter et al., 2013; Cakmak et al., 2014; Jedrychowski et al., 2015a, 2015b; Gentner and Weber, 2011), and models were used to predict the real human exposure and assess the health risks. However, due to the individual difference and the uncertainty of exposure time and sites, the estimates often deviate from real exposure. Biomonitoring metabolites of PAHs and BTEX in human specimen is a practical approach to assess exposure levels through inhalation, dietary and dermal contact overcoming the drawbacks of environmental monitoring alone. Though only short-term exposure periods are indicated (no longer than 48 h according to the excretions of different targets) (Li et al., 2012), a large sample size could represent the real exposure of the cohort population. Moreover, compared with blood samples or other tissue sampling, urine is easily collected and handled.

This study aims to investigate the influence of coal combustion emissions on respiratory systems of the neighboring population. We measured PAHs and BTEX in related environmental media (including soil, air and ground water) and in the urine levels of PAH and BTEX metabolites in a cohort population. Epidemiology investigations, medical examinations, environmental and urinary exposure levels and lung function were combined to comprehensively assess the effects of PAH and BTEX exposure on the lung function and respiratory systems of adults and children.

2. Materials and method

2.1. Study site selection

Two counties in Northern China were selected as the investigated area (IR) for their 5 coke oven plants and other pollution sources, including 18 controlled-exhaust emission sites, 30 fugitive exhaust-emission sites and 8 directly discharged waste water sites. The IR areas were constrained in residential areas < 10 km from the coke oven plants. Soil, air, and water were collected. Four typical sites were selected as the control area (CR), where all environmental samples were collected in a similar way. Air, water and soil sample were collected from July 2012 to August 2012. All sample information is listed in Table 1.

2.2. Sampling of air, water and soil

2.2.1. Air

Both indoor and outdoor air samples were collected. Three pairs of PAH and BTEX samples (one sample every day) from outdoor and indoor air were collected by a medium-volume sampler (SIBATA, Japan) and a low-volume sampler (Tianhong, China), respectively. For analysis of PAHs, air were pumped through glass fiber filter (Whatman, England) at 100–300 L/min for 24 h and 0.5 L/min for 8 h and subsequently through polyurethane foam/XAD plugs for outdoor and indoor air, respectively. BTEX was collected through TENAX adsorption tubes. Daily meteorological data (i.e., wind speed, wind direction, temperature, relative humidity and pressure) were collected simultaneously.

2.2.2. Water

The water samples were collected from river, well water and irrigation water (i.e., river and well). After collection into the clean glass bottles, the water samples were protected from light, shipped to laboratories and stored at 4 °C until chemical analysis.

2.2.3. Soil

At each site, soil samples were collected by a surface soil collector (DIK-1621) and combined to form a composite sample for the site. Samples were sealed in a clean glass container, transported to the laboratory and stored at −20 °C until chemical extraction.

2.3. Sample pretreatment and instrumental analysis methods

Air, water and soil were pretreated and analyzed according to the method of Ministry of Environmental Protection (MEP) of China (MEP, 1989, 2003 and 2010) and the U.S. EPA (Ministry of Environmental Protection, 1986 and 1996, 1999, 2006). We measured 16 priority PAHs (i.e., naphthalene (Nap), acenaphthylene (Acey), acenaphthene (Acet), fluorene (Fl), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), chrysene (Chr), benz(a)anthracene (BaA), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), indeno(1,2,3-c,d)pyrene (IcdP), dibenz(a,h)anthracene (DahA), benzo(g,h,i)pyrene (BghiP), benzo(a)fluoranthene (BaF) and benzo(e)pyrene (BeP)), as well as four BTEX compounds (i.e., benzene, toluene, ethylbenzene (EB) and 1,2-dimethylbenzene (1,2-DB)) in air, water and soil samples. The MDLs of PAHs in the air, water and soil samples were 0.00062–0.524 ng/m^3, 0.0009–1.2 ng/L and 0.002–20.8 ng/g, respectively. The MDLs of the BTEX in the air, water and soil samples were 0.0423–0.0800 μg/m^3, 0.15–0.53 ng/L and 0.5–3.0 μg/kg, respectively.

2.4. Participant recruitment

The study participants were recruited within the environmental sample locations. Four thousand and ninety-one residents from the IR and CR, including adults aged 20–75 and children aged 6–12, were randomly questioned in the epidemiological investigation. Then, 480 residents from the IR and 299 residents from the CR who were considered sensitive to environmental exposure were chosen for physical examination of respiratory symptoms such as chronic bronchitis, asthma, emphysema for adults, upper respiratory tract infection, other chronic lower respiratory symptoms and allergic rhinitis. Consequently, 583 people were surveyed by urinary monitoring of PAHs and BTEX, and 406 people were tested for lung function. All the adults had lived in the area for > 2 years, and the average time was 38 ± 18 years. All the children had lived in the area for > 1 year.

Every family was required to answer a questionnaire, including identification of the family member, their primary diet, the house structure, energy resources and structure, as well as the family financial condition, etc. Every adult participant or the parents of every child participants were required to fill out a consent form and answer a personal questionnaire including name, family address, gender, age, lifestyle habits, health status and smoking information (i.e., smoker and nonsmoker). Their personal information is listed in Table 1. The epidemiological investigation, lung function tests and medical