



Levels and neurodevelopmental effects of polycyclic aromatic hydrocarbons in settled house dust of urban dwellings on preschool-aged children in Nanjing, China

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ABSTRACT

This study investigated levels and possible determinants of polycyclic aromatic hydrocarbons (PAHs) in settled house dust of urban dwellings with preschool-aged children in Nanjing, China. The possible neurodevelopmental effects of house-dust PAHs on the children were also investigated. Settled house dust was collected from 203 urban houses. Fifteen PAHs were measured using an HPLC with a fluorescence detector and wavelength programming. The Child Behavior Checklist and the Gesell Development Inventory were used to evaluate the overall development of the children. PAHs were detected in settled house dust of >90% of houses, where high molecular weight PAHs predominated. Most PAHs with two or three rings were found at significantly higher levels in indoor compared with outdoor dust. PAH isomeric ratios showed that PAHs with four or more rings might be derived from outdoor coal burning sources. Naphthalene was found at lower concentrations in houses with more hours of natural ventilation, while pyrene, chrysene (Chr) and benzo[k]fluoranthene (BkF) were present at higher concentrations in these houses. A higher floor level correlated with lower levels of PAHs, especially those with four or more rings, while older houses had higher PAH levels. Benzo[a]pyrene (BaP) and indeno[1,2,3-cd]pyrene levels were positively associated with most behavioral problem scores. Higher BaP and benzo[g,h,i]perylene levels showed lower Gesell language development quotient (DQs), and higher Chr and BkF were associated with lower Gesell social skill DQs. In conclusion, 15 PAHs are ubiquitous in urban settled house dust, where outdoor coal burning and indoor cooking oil fumes are the two main sources of PAHs in Nanjing, China. Natural ventilation, floor level and residence age potentially influence house dust PAH levels. The potential adverse effect of postnatal exposure to PAHs on the behavior and neurodevelopment of preschool-aged children requires follow up in larger studies.

Keywords: Urban house dust, polycyclic aromatic hydrocarbons, child exposure, neurodevelopment

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1. Introduction

Concentrations of chemical contaminants, such as semi-volatile organic compounds like polycyclic aromatic hydrocarbons (PAHs), flame retardants and pesticides, are often higher in indoor air than in outdoor air (Rudel and Perovich, 2009). Many indoor contaminants absorb onto particulate matter, which is initially suspended in air and later settles as dust. This has led to settled house dust being considered as an exposure medium and a global indicator of residential contamination (Butte and Heinzow, 2002; Liou et al., 2002), particularly for infants and toddlers, who are at highest risk because of their hand-to-mouth activities.

Prenatal PAH exposure has been linked to neurodevelopmental toxicity. For example, maternal PAH exposure during pregnancy has been associated with IQ deficits (Perera et al., 2009; Edwards et al., 2010) and cognitive developmental delay (Perera et al., 2006; Edwards et al., 2010). Given that neurodevelopmental processes such as myelination are not completed until adolescence (Rice and Barone, 2000), and that inadvertent dust ingestion could be responsible for as much as 42% of non-dietary PAH exposure in young children (Gevao et al., 2007), settled house dust may be an important source of PAH uptake in children.

Studies have reported PAH levels in settled house dust in different localities and countries (Rudel et al., 2003; Maertens et al., 2004; Gevao et al., 2007; Maertens et al., 2008; Roberts et al., 2009; Langer et al., 2010). However, to the best of our knowledge, only one study reported the PAH floor loadings in China, with only 14 settled house dust samples for small rural villages (Naspinski et al., 2008). In this study, PAH concentrations in urban settled house dust were measured in Nanjing, China, to evaluate urban indoor PAH pollution, their possible determinants and sources. Simultaneously, the behavior and neurodevelopment of preschool-aged children in these houses were tested to find any association with the corresponding PAH levels.

2. Methods

2.1. Subject recruitment

Recruitment questionnaires ($n=500$) were sent to families with preschool-aged children (4–5 years) randomly selected from the four largest public kindergartens in the Gulou district of Nanjing, China, of which 464 answered and returned the questionnaire. The exclusion criteria we used included (1) living in the current house for less than 1 year; (2) birthing problems, such as delivery injuries or a low birth weight; (3) neonatal problems, such as asphyxia,

intracranial hemorrhage, hypoxic–ischemic encephalopathy and severe jaundice; (4) acquired disabilities, including poisoning, encephalitis, meningitis, encephalopathy, cerebral trauma and cerebral injuries following convulsions; and (5) developmental dysplasia or other developmental defects influenced by malnutrition or inherited metabolic diseases. Subject recruitment is also described elsewhere (Wang et al., 2012). Based on these criteria, 400 preschool-aged children were selected for the house investigation and the measures of behavior and neurodevelopment. During the field investigation, 146 families opted out of the survey because of the in-house sampling date conflicted or because they thought there were too many questions in the 113-item Child Behavior Checklist. Of the remaining 254 families, because of the invasive blood sampling, only 98 children provided finger-prick blood and performed the Gesell Development Inventory test (Table 1). Informed consent was obtained from all participants and the Institutional Ethics Committee of Nanjing Medical University and Institutional Review Board approved the study.

Table 1. Characteristics of the subjects and their houses (n=203)

	Contents
Children's characteristics	
Sex (n (%)) males)	104 (51)
Age (n (%))	
4	100 (49)
5	103 (51)
CBCL scores (n)	203
DQs (n)	98
Blood Pb levels (µg/L, n=98, [mean±SD (Min, Max)])	45±15 (30, 75)
House characteristics	
Residence age [mean±SD (Min, Max)]	16±8 (1, 60)
Floor level [mean±SD (Min, Max)]	5±5 (1, 28)
Frequency of natural ventilation [n (%)]	
<1 hr/d	127 (63)
1–4 hr/d	63 (31)
>4 hr/d	13 (6)
Smoking status [n (%)]	
No	118 (58)
Yes	85 (42)
Near to a main road (<500 meters)	
No	99 (49)
Yes	104 (51)
Vacuum or wipe cleaning frequency [n (%)]	
1–3 d	118 (58)
4–7 d	52 (26)
7–14 d	21 (10)
>14 d	12 (6)
Kitchen type [n (%)]	
Open	61 (30)
Separate	142 (70)

2.2. Building investigations and dust sampling

Settled house dust was sampled from the houses of all subjects recruited during the non-heating season from March to June 2011. One sample of dust from each house was collected using commercial vacuum cleaners and paper bags on the floor surface in each dwelling where preschool-aged children commonly played. Simultaneously, settled dust on an outdoor ground surface

near the house was sampled with a hand-held brush. For further details of the sampling process, please see the Supporting Material (SM). In total, 230 samples of indoor settled house dust and 150 outdoor settled dust samples were collected. After excluding mislabeled, mis-stored and insufficient dust samples (26 samples and one single-family home dust sample were excluded), a total of 203 samples of indoor dust and 110 paired outdoor dust samples from low or high-rise apartment buildings were available for the final analysis. During dust sampling, the caregivers were asked to provide information about residence age, floor level, household smoking, kitchen type (open or separate) and cooking habits (once, twice or three times per day), frequency of natural ventilation through open windows (<1 hr/d, 1–4 hr/d or >4 hr/d), vacuum or wipe cleaning frequency (every 1–3 d, 4–7 d, ≤14 d or >14 d) and whether the residence was near a main road (<500 meters from the child's home to a two- or more-lane motorway) (Table 1).

2.3. PAH laboratory analysis

Pretreatment and laboratory measurements of PAHs in dust were carried out following the method described by Lu et al., (2011). Particles were separated from fibers by sieving through a mesh that was <150 µm in size. Acetone was added to each sieved dust sample and extraction was performed in an ultrasonic bath. The supernatant was concentrated and then spiked with methyl cyanide to 0.1 mL. Using methyl cyanide and water as the mobile phase, PAHs were separated by gradient elution and detected with a fluorescence detector and wavelength programming. Targeted PAHs included: naphthalene (Nap), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fla), pyrene (Pyr), benz[a]anthracene (BaA), chrysene (Chr), benzo[b]-fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), dibenz[a,h]anthracene (DBA), benzo[g,h,i]perylene (BP) and indeno[1,2,3-cd]pyrene (IP). The fifteen PAHs were efficiently separated within 28 min and showed good linearity within the 0.5–50 ng/mL range. The absolute recovery efficiencies were 62–110% and the relative standard deviation (RSD) ranged between 1.4 and 5.9% (Lu et al., 2011). The instrumental detection limit (IDL) was determined on the basis of response at a signal-to-noise ratio (S/N) of 3. The method detection limits (MDL) under the present chromatographic conditions were calculated using the IDLs, the volume of extracts, and the sample weights.

2.4. Measures of child behavior and neurodevelopment

All 203 children in the houses where dust samples were collected were asked to check their behavioral and neurodevelopment problems. Behavioral problems were measured by caregiver report on the 113-item Child Behavior Checklist (CBCL) for children 4–16 years of age (Achenbach, 1991), which collects information on child behaviors occurring in the past 6 months and has been proved workable in China (Xin and Zhang, 1992). The caregivers completed the CBCL with guidance as needed from the research workers. The syndrome scores were computed for all eight syndrome domains (anxious/depressed, withdrawn/depressed, somatic complaints, social problems, thought problems, attention problems, compulsive and aggressive behavior) by summing the scores on the specific items, yielding a continuous raw score. Based on responses to the syndrome scales, internalizing, externalizing and total problem scores were obtained.

The Gesell Development Inventory has been translated and standardized by the Beijing Children's Health Care Institute, which consists of five behavioral domains: adaptive, gross motor, fine motor, language and personal social behaviors (Zhu et al., 1983). The items used for the five behavioral domains included: (1) adaptive behavior: eye-hand coordination, imitation, object recovery, comprehension, discriminative performance, perception, completion and number conception; (2) gross motor behavior: postural responses, balance of the head, and posture at standing,

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