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Time trends of polycyclic aromatic hydrocarbon exposure in New York city from 2001 to 2012: Assessed by repeat air and urine samples

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ABSTRACT

Background: Exposure to air pollutants including polycyclic aromatic hydrocarbons (PAH), and specifically pyrene from combustion of fuel oil, coal, traffic and indoor sources, has been associated with adverse respiratory health outcomes. However, time trends of airborne PAH and metabolite levels detected via repeat measures over time have not yet been characterized. We hypothesized that PAH levels, measured repeatedly from residential indoor and outdoor monitors, and children's urinary concentrations of PAH metabolites, would decrease following policy interventions to reduce traffic-related air pollution.

Methods: Indoor PAH (particle- and gas-phase) were collected for two weeks prenatally ($n=98$), at age 5/6 years ($n=397$) and age 9/10 years ($n=198$) since 2001 and at all three age-points ($n=27$). Other traffic-related air pollutants (black carbon and $PM_{2.5}$) were monitored indoors simultaneous with PAH monitoring at ages 5/6 ($n=403$) and 9/10 ($n=257$) between 2005 and 2012. One third of the homes were selected across seasons for outdoor PAH, BC and $PM_{2.5}$ sampling. Using the same sampling method, ambient PAH, BC and $PM_{2.5}$ also were monitored every two weeks at a central site between 2007 and 2012. PAH were analyzed as semivolatile PAH (e.g., pyrene; MW 178–206) ($\sum_8 PAH_{semivolatile}$: Including pyrene (PYR), phenanthrene (PHEN), 1-methylphenanthrene (1-MEPH), 2-methylphenanthrene (2-MEPH), 3-methylphenanthrene (3-MEPH), 9-methylphenanthrene (9-MEPH), 1,7-dimethylphenanthrene (1,7-DMEPH), and 3,6-dimethylphenanthrene (3,6-DMEPH)) and the sum of eight nonvolatile PAH ($\sum_8 PAH_{nonvolatile}$: Including benzo[a]-anthracene (BaA), chrysene/iso-chrysene (Chry), benzo[b]fluoranthene (BbFA), benzo[k]fluoranthene (BkFA), benzo[a]pyrene (BaP), indeno[1,2,3-c,d]pyrene (IP), dibenzo[a,h]anthracene (DahA), and benzo[g,h,i]perylene (BghiP); MW 228–278). A spot urine sample was collected from children at child ages 3, 5, 7 and 9 between 2001 and 2012 and analyzed for 10 PAH metabolites.

Results: Modest declines were detected in indoor BC and $PM_{2.5}$ levels between 2005 and 2012 (Annual percent change [APC] $= -2.08\%$ [$p=0.010$] and -2.18% [$p=0.059$] for BC and $PM_{2.5}$, respectively), while a trend of increasing pyrene levels was observed in indoor and outdoor samples, and at the central site during the comparable time periods (APC $=4.81\%$, 3.77% and 7.90%, respectively; $p < 0.05$ for all). No significant time

Abbreviations: APC, annual percent change; BC, black carbon; CCCEH, Columbia Center for Children's Environmental Health; DEP, diesel exhaust particle; EC, elemental carbon; ETS, environmental tobacco smoke; I/O ratio, indoor-to-outdoor ratio; NYC, New York city; PAH, polycyclic aromatic hydrocarbons; $\sum_8 PAH_{semivolatile}$, sum of 8 low molecular-weight-PAH ≤ 206; $\sum_8 PAH_{nonvolatile}$, sum of 8 high molecular-weight-PAH ≥ 228; 1-OH-NAP, 1-Naphthol; 2-OH-NAP, 2-Naphthol; 2-OH-FLUO, 2-hydroxyfluorene; 3-OH-FLUO, 3-hydroxyfluorene; 9-OH-FLUO, 9-hydroxyfluorene; 1-OH-PHEN, 1-hydroxyphenanthrene; 2-OH-PHEN, 2-hydroxyphenanthrene; 3-OH-PHEN, 3-hydroxyphenanthrene; 4-OH-PHEN, 4-hydroxyphenanthrene; 1-OH-PYR, 1-hydroxypyrene; PM, particulate matter; PUF, polyurethane foam

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trend was observed in indoor $\sum_8 PAH_{nonvolatile}$ levels between 2005 and 2012; however, significant opposite trends were detected when analyzed seasonally (APC= −8.06% [$p<0.01$], 3.87% [$p<0.05$] for nonheating and heating season, respectively). Similarly, heating season also affected the annual trends (2005–2012) of other air pollutants: the decreasing BC trend (in indoor/outdoor air) was observed only in the nonheating season, consistent with dominating traffic sources that decreased with time; the increasing pyrene trend was more apparent in the heating season. Outdoor $PM_{2.5}$ levels persistently decreased over time across the seasons. With the analyses of data collected over a longer period of time (2001–2012), a decreasing trend was observed in pyrene (APC= −2.76%; $p<0.01$), mostly driven by measures from the nonheating season (APC= −3.54%; $p<0.01$). In contrast, levels of pyrene and naphthalene metabolites, 1-hydroxypyrene and 2-naphthol, increased from 2001 to 2012 (APC=6.29% and 7.90% for 1-hydroxypyrene and 2-naphthol, respectively; $p<0.01$ for both).

Conclusions: Multiple NYC legislative regulations targeting traffic-related air pollution may have led to decreases in $\sum_8 PAH_{nonvolatile}$ and BC, especially in the nonheating season. Despite the overall decrease in pyrene over the 2001–2012 periods, a rise in pyrene levels in recent years (2005–2012), that was particularly evident for measures collected during the heating season, and 2-naphthol, indicates the contribution of heating oil combustion and other indoor sources to airborne pyrene and urinary 2-naphthol.

1. Introduction

Exposure to traffic-related air pollutants such as polycyclic aromatic hydrocarbons (PAH), black carbon (BC), and particulate matter less than 2.5 μm in diameter ($PM_{2.5}$) may induce the development of asthma or trigger asthma symptoms (Jung et al., 2012a, 2012b; Miller et al., 2004; Perzanowski et al., 2013; Rosa et al., 2011; Spira-Cohen et al., 2011). For example, our group at the Columbia Center for Children's Environmental Health (CCCEH) reported that young children from Northern Manhattan who are exposed repeatedly to high PAH (i.e., semivolatile pyrene) during prenatal and early childhood periods may be at greater risk of having asthma (Jung et al., 2012b). Several of the PAH also have been associated with the development of indoor allergen sensitization in early childhood (Miller et al., 2010; Perzanowski et al., 2013).

Major outdoor sources of PAH, BC and $PM_{2.5}$ include traffic emissions derived from diesel/gasoline vehicles and heating and power sources such as oil, coal, and biomass, and indoor sources include cooking, residential heating, environmental tobacco smoke (ETS), and candles/incense (Lewtas, 2007). While nonvolatile PAH (i.e., high-molecular weight PAH ≥ 228; e.g., benzo(a)pyrene) are generated predominantly by incomplete combustion, semivolatile PAH (i.e., low-molecular weight PAH ≤ 206; e.g. pyrene) are emitted from both incomplete combustion and petrogenic (i.e., produced by petroleum) sources such as direct evaporation from petroleum products, oil leaks, and the ground (Ma et al., 2010; Zhang and Tao, 2009). Major sources of New York City (NYC) ambient $PM_{2.5}$ concentrations include long-range transported sulfate, upwind photochemical production of sulfate and other secondary organic carbon, traffic emissions, and steel dust (Cyrys et al., 2003; Lall et al., 2011). Residential and commercial heating oil (residual oil; types #4 and #6) combustion also is considered a major source of NYC BC and $PM_{2.5}$ (Cornell et al., 2012). The observation that indoor/outdoor (I/O) concentration ratios of semivolatile PAH are greater than 1 in NYC homes indicates that indoor sources of semivolatile PAH can be important. The I/O ratios of nonvolatile PAH and BC less than 1 or close to 1 suggest that the indoor concentrations of nonvolatile PAH and BC arise predominantly from the transport of outdoor air into the indoor environment (Jung et al., 2010a; Kinney et al., 2002).

In an effort to diminish levels of air pollutants, NYC implemented multiple legislative regulations since 2000. These included the conversion to ultra-low sulfur diesel fuel (ULSD)[1] and use of the best available retrofit technology (BART)[1] on city-owned or operated diesel fuel-powered vehicles such as garbage trucks, school buses and fleet trucks, and non-road vehicles (e.g., construction vehicles, generators) in 2007 as well as the conversion and replacement of Metropolitan Transportation Authority diesel vehicles to low emission diesel and alternative fuel vehicles in 2000 (MTA, 2013). In 2009, the existing idling law was strengthened further near school districts; idling in excess of 1 min was prohibited near any public or non-public school.[2] More recently, in 2011, the Clean Heat Rule was initiated to phase out the use of dirty fuel oils (low grade, types #4 and #6) for heating by 2012 with a conversion to a ultra-low sulfur version of #2 oil, or natural gas, as well as prohibiting any new boilers issued for #4 or #6 fuel oil and eliminating renewals issued for #6 fuel oil.

Coincident with Clean Air Act provisions since 1970s and the earlier NYC bus fleet-wide plan to reduce diesel emissions, and increase the use of cleaner fuels since 2000, our previous study found that the average levels of nonvolatile PAH in NYC, measured using 48-h personal air monitoring in our cohort, have declined between 1998 and 2006 (Narvaez et al., 2008). However, time trends of airborne PAH and PAH metabolite levels via repeat measures in the same homes and individuals over time have not been previously characterized. The objectives of this study were to (1) determine annual trends in PAH and other traffic-related pollutants (i.e., BC and $PM_{2.5}$) using a more comprehensive and longer monitoring data set that included repeated measures from children's indoor and outdoor environments prenatally through age 9/10 as well as from the central monitoring site; and (2) examine annual trends of PAH metabolites in urine after adjusting for potential covariates such as age, race/ethnicity and indoor sources. Our approach was to utilize data on airborne traffic-related air pollutants from children's residences and PAH urinary metabolites collected as part of the CCCEH longitudinal birth cohort study in Northern Manhattan and the South Bronx, New York. We hypothesized that (1) airborne PAH, BC and $PM_{2.5}$ levels, measured repeatedly from indoor and outdoor residential monitors and at the central site, and PAH metabolite levels would decrease from 2001 to 2012, in parallel with policy interventions to reduce traffic-related air pollution and (2) decreasing trends of traffic-related air pollutants would be more apparent during the nonheating season when residential heating oil is not used.

2. Materials and methods

2.1. Study design and population

Nonsmoking African American or Dominican women ages 18–35 living in Northern Manhattan and the South Bronx were enrolled during pregnancy

[1] Local Law 39, 40, 41, 42 and 77.

[2] Local Law 4, 5, and 25.