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Estimating population exposure to ambient polycyclic aromatic hydrocarbon in the United States – Part II: Source apportionment and cancer risk assessment

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ABSTRACT

A revised Community Multiscale Air Quality (CMAQ) model was developed to simulate the emission, reactions, transport, deposition and gas-to-particle partitioning processes of 16 priority polycyclic aromatic hydrocarbons (PAHs), as described in Part I of the two-part series. The updated CMAQ model was applied in this study to quantify the contributions of different emission sources to the predicted PAH concentrations and excess cancer risk in the United States (US) in 2011. The cancer risk in the continental US due to inhalation exposure of outdoor naphthalene (NAPH) and seven larger carcinogenic PAHs (cPAHs) was predicted to be significant. The incremental lifetime cancer risk (*ILCR*) exceeds 1×10^{-5} in many urban and industrial areas. Exposure to PAHs was estimated to result in 5704 (608–10,800) excess lifetime cancer cases. Point sources not related with energy generation and the oil and gas processes account for approximately 31% of the excess cancer cases, followed by non-road engines with 18.6% contributions. Contributions of residential wood combustion (16.2%) are similar to that of transportation-related sources (mostly motor vehicles with small contributions from railway and marine vessels; 13.4%). The oil and gas industry emissions, although large contributors to high concentrations of cPAHs regionally, are only responsible of 4.3% of the excess cancer cases, which is similar to the contributions of non-US sources (6.8%) and non-point sources (7.2%). The power generation units pose the most minimal impact on excess cancer risk, with contributions of approximately 2.3%.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are organic compounds that contain multiple aromatic rings. Various forms of cancer, such as skin, lung, bladder, liver and stomach, due to PAH exposure have been reported in animal exposure studies (Boström et al., 2002). Occupational and environmental exposure to polycyclic aromatic hydrocarbons (PAH) has also been related with higher human cancer risks in epidemiological studies (Boffetta et al., 1997). The United States Environmental Protection Agency (US EPA) has classified 16 of the PAHs as priority pollutants and designated seven of the PAHs possible human carcinogens (see Table 1 of Zhang et al. (2016)). To properly evaluate the level of population exposure and cancer risk due to PAHs, the spatial and temporal distribution of ambient PAHs need to be determined. Subsequently, to design effective PAH emission control strategies, the contributions of different emission sources to the exposure and cancer risk at different locations and different time of the year need to be quantified. Part I of the two-paper series described the development of an atmospheric chemical transport model (CTM) that is capable of estimating the spatial and temporal distribution of the 16 priority PAHs. The capability of the CTM to estimate daily and monthly concentrations of the PAHs in the United States in 2011 at a spatial resolution of 36-km was demonstrated. In the second part of the study, the CTM was further applied to quantitatively determine the contributions of major sources of PAHs to the estimated population exposure and cancer risk. In the follow paragraphs in the introduction, major sources of PAHs as reported by the literature were summarized and typical source apportionment techniques for PAHs and their applications in different countries were critically reviewed.

Atmospheric PAHs are typically formed from fuel combustion sources such as motor vehicle exhaust and wood burning (Ravindra et al., 2008). Biofuel use accounted for a large fraction of annual global PAH emissions, with most of the emissions from developing countries such as India and China (Aramandla et al., 2011). In developed countries, emissions from industries and fossil fuel combustion are considered to be the major sources of PAHs (Aramandla et al., 2011). While the emission studies alone provide emissions rates of PAHs at country or regional level, they cannot be used to directly deduce the

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Table 1
Population-weighted source contributions to PAHs in the continental United States for January 2011.

	C123	EGU	MVH	NPT	NRD	OGS	OPT	RWC	OTH
NAPH	1.1%	0.3%	12.9%	7.0%	4.1%	0.1%	5.3%	60.1%	9.1%
ACE	1.3%	0.8%	17.2%	2.0%	8.4%	1.7%	27.3%	34.6%	6.8%
ACY	0.4%	0.0%	10.9%	0.2%	4.2%	0.3%	5.7%	75.8%	2.4%
FLU	1.5%	0.1%	19.8%	0.3%	9.9%	0.2%	5.4%	51.1%	11.7%
PHE	1.3%	0.1%	15.1%	0.6%	5.9%	1.1%	11.4%	55.2%	9.4%
ANT	1.6%	0.1%	14.3%	0.8%	6.9%	0.2%	8.7%	64.2%	3.2%
FTH	2.2%	0.2%	15.7%	2.0%	8.1%	0.2%	12.9%	53.5%	5.2%
PYR	3.0%	0.1%	20.9%	1.3%	10.7%	2.0%	12.2%	44.9%	4.8%
CHRY	2.0%	0.2%	15.6%	3.0%	8.8%	0.5%	14.1%	52.0%	3.8%
BaA	2.5%	0.1%	16.0%	1.5%	9.2%	2.4%	30.5%	35.1%	2.6%
BbF	0.6%	0.4%	16.5%	3.4%	9.5%	0.0%	14.2%	52.0%	3.5%
BkF	0.1%	0.3%	17.2%	1.7%	9.5%	0.0%	17.9%	49.0%	4.3%
BghiP	0.0%	0.1%	27.1%	0.5%	16.6%	13.0%	26.4%	14.3%	2.0%
BaP	1.2%	0.1%	19.4%	1.4%	11.3%	7.7%	31.0%	25.9%	2.0%
IcdP	0.2%	0.2%	30.6%	1.1%	18.1%	0.4%	9.1%	37.4%	3.0%
DahA	1.4%	1.3%	14.6%	25.9%	9.6%	0.0%	39.6%	0.0%	7.5%
15PAH	1.5%	0.1%	16.7%	0.9%	7.9%	1.3%	12.3%	52.1%	7.2%
∑cPAH	1.4%	0.2%	18.1%	2.3%	10.4%	2.0%	21.2%	41.3%	3.2%
TEC_{cPAH}(low)	1.3%	0.2%	19.1%	2.5%	11.1%	5.8%	29.0%	28.5%	2.5%
TEC_{cPAH}(high)	1.3%	0.4%	17.6%	5.3%	10.3%	3.6%	27.1%	31.0%	3.3%

C123: (locomotives and class 1–3 marine vessels), EGU (point sources of electric generation units), MVH (motor vehicles), NPT (non-point sources), NRD (non-road engines), OGS (oil and gas processes), OPT (other industrial point sources), RWC (residential wood combustion) and OTH (non-US sources; also includes all non-US C3 CMV and offshore oil production processes). 15PAH is the sum of the concentrations of all the PAHs except NAPH. ∑cPAH is the sum of the concentrations of the seven carcinogenic PAHs (cPAHs).

contributions of different sources to ambient PAH concentrations due to spatiotemporal variation of the emissions and regional transport and transformation of emitted PAHs in the atmosphere (Inomata et al., 2012).

Traditionally, receptor-oriented source apportionment techniques have been applied to determine contributions of different sources to the observed PAH concentrations. The most commonly applied techniques including Chemical Mass Balance (CMB) (Yang and Chen, 2004), Principle Component Analysis (PCA) (Dong and Lee, 2009; Li et al., 2006; Ma et al., 2011; Mai et al., 2003; McDonough et al., 2014; Mishra et al., 2016), Positive Matrix Factorization (PMF) (Callen et al., 2014; Xie et al., 2014), and multilinear regression (Harrison et al., 1996; Masiol et al., 2012). Comparison studies showed that these receptor-oriented methods generally give comparable PAH source apportionment results (Larsen and Baker, 2003; Teixeira et al., 2015). In addition, some studies used carbon isotope dating to differentiate contributions from biomass burning to fossil fuel combustion (Mandalakis et al., 2005; Sheesley et al., 2009; Zencak et al., 2007). While the molecular diagnostic ratio (MDR) technique has been applied in several studies (Fu et al., 2010; Schifman and Boving, 2015; Zhang et al., 2005), many PAH MDRs vary significantly in space and time due to different reaction rates in the atmosphere and the estimates were not valid source markers (Amador-Munoz et al., 2013; Katsoyiannis et al., 2011; Tobiszewski and Namiesnik, 2012). Major sources identified as contributing to ambient PAH concentrations include open burning of agriculture residuals (Chen et al., 2008), gasoline and diesel vehicles (Larsen and Baker, 2003), coal combustion (Chen et al., 2011), wood combustion (Piazzalunga et al., 2013) and industrial sources such as refineries (Motelay-Massei et al., 2007).

The receptor-oriented techniques are useful in that they are based on observations and do not need information on emissions and detailed knowledge of physical and chemical processes of PAHs in the atmosphere. However, as the PAH species are not strictly inert, concentrations of the measured PAH concentrations need to be adjusted to compensate for the differential reactive decay of the PAH species before they can be applied in some of the techniques such as CMB and MDR. Such adjustments are difficult to perform without detailed information on the spatial and temporal history of the PAH species. Another limitation of the receptor-oriented methods is that they can only provide source apportionment information in the location where observations are made. To overcome the data gap, regional chemical transport models (CTMs) can be used. Currently, only a few studies used a regional model to determine source contributions of PAHs (San Jose et al., 2013; Zhang and Ying, 2012). In a recent study, Shen et al. (2014) estimated source contributions to lung cancer risk from Benzo[α]pyrene (BaP) exposure using a global chemical transport model. However, they didn't include other possible carcinogenic PAHs in their calculations.

Few studies have been reported in the peer-reviewed literature on PAH concentrations, sources and population health risk in the US, and most published studies (McDonough et al., 2014; Naumova et al., 2002; Schifman and Boving, 2015; Simcik et al., 1999) focused on PAH concentrations and sources using local scale models but did not evaluate the regional population health risk. As reported in Part I of this two paper series, concentrations of PAHs are high in many places in the US. This makes it necessary to quantify the contributions of different emission sources to the predicted ambient PAH concentrations so that effective emission control strategies can be formulated.

Thus, the objectives of this study are to provide a regional source apportionment of PAHs using a regional chemical transport model and to evaluate the contributions of different sources to excess cancer risk. To our knowledge, this is the first study to provide a national level prediction of PAH source apportionment information and cancer risk assessment in the continental US.

2. Methods

2.1. Modeling PAHs using the CMAQ model

The Community Multiscale Air Quality (CMAQ) model version 5.0.1 developed by the US EPA was expanded to include treatment of emission, reactions, transport, deposition and gas-to-particle partitioning processes to predict the gas and particle phase concentrations of 16 priority PAH concentrations, including the 7 carcinogenic PAH (cPAH) species, Benzo[*a*]anthracene (BaA), Chrysene (CHRY), Benzo[*b*]fluoranthene (BbF), Benzo[*k*]fluoranthene (BkF), Benzo[α]pyrene (BaP), Indeno[1,2,3-

Table 2
Population-weighted source contributions to PAHs in the continental United States for July 2011.

	C123	EGU	MVH	ARE	NRD	OGS	OPT	RWC	OTH
NAPH	3.0%	0.9%	23.9%	15.1%	13.9%	0.2%	13.5%	3.7%	25.8%
ACE	1.9%	1.0%	14.2%	1.9%	15.5%	1.6%	43.1%	1.2%	19.5%
ACY	2.0%	0.1%	24.1%	0.5%	19.8%	1.2%	25.1%	5.9%	21.2%
FLU	2.7%	0.2%	20.3%	0.3%	23.3%	0.2%	9.2%	2.6%	41.2%
PHE	2.8%	0.1%	20.1%	0.7%	15.5%	1.4%	22.5%	2.7%	34.2%
ANT	4.7%	0.2%	25.7%	1.4%	26.0%	0.4%	22.8%	4.6%	14.1%
FTH	4.6%	0.3%	22.2%	1.9%	24.6%	0.3%	24.9%	3.3%	17.8%
PYR	5.2%	0.2%	23.4%	1.3%	26.0%	2.8%	23.0%	2.4%	15.6%
CHRY	4.7%	0.4%	19.4%	3.2%	25.9%	0.7%	28.6%	4.0%	13.1%
BaA	4.1%	0.2%	15.2%	1.3%	20.1%	3.0%	46.3%	2.1%	7.7%
BbF	1.5%	0.8%	14.8%	3.4%	26.7%	0.0%	31.4%	4.6%	16.8%
BkF	0.2%	0.7%	11.5%	1.7%	22.0%	0.0%	36.6%	3.8%	23.4%
BghiP	0.1%	0.1%	9.7%	0.4%	24.9%	10.5%	49.7%	0.6%	4.0%
BaP	1.7%	0.2%	10.3%	1.0%	18.0%	7.6%	53.9%	1.2%	6.1%
IcdP	0.4%	0.3%	18.6%	1.4%	43.5%	0.6%	19.1%	3.0%	13.2%
DahA	1.8%	1.3%	8.0%	8.8%	14.5%	0.0%	45.5%	0.0%	20.2%
15PAH	3.1%	0.3%	19.5%	1.1%	20.7%	1.8%	25.4%	2.7%	25.5%
∑cPAH	2.8%	0.4%	14.9%	2.0%	23.4%	2.6%	39.8%	2.8%	11.4%
TEQ_{cPAH}(low)	2.0%	0.3%	11.3%	1.6%	19.3%	5.9%	50.2%	1.5%	7.8%
TEQ_{cPAH}(high)	2.2%	0.5%	11.8%	3.0%	19.6%	3.8%	46.2%	1.8%	11.1%

C123: (locomotives and class 1–3 marine vessels), EGU (point sources of electric generation units), MVH (motor vehicles), NPT (non-point sources), NRD (non-road engines), OGS (oil and gas processes), OPT (other industrial point sources), RWC (residential wood combustion) and OTH (non-US sources; also includes all non-US C3 CMV and offshore oil production processes). 15PAH is the sum of the concentrations of all the PAHs except NAPH. ∑cPAH is the sum of the concentrations of the seven carcinogenic PAHs (cPAHs).

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