MODELING POLYCYCLIC AROMATIC HYDROCARBONS EMISSIONS AND AMBIENT CONCENTRATIONS IN THE UNITED STATES

A Thesis

by

JIE ZHANG

Submitted to the Office of Graduate and Professional Studies of
Texas A&M University
in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE

Chair of Committee,	Qi Ying
Committee Members,	Bill Batchelor
	Renyi Zhang
Head of Department,	Robin Autenrieth

December 2015

Major Subject: Civil Engineering

ABSTRACT

PAHs (polycyclic aromatic hydrocarbons) in the environment are of significant concern due to their high toxicity. Although PAHs are monitored in the United States (US) at the air toxics monitoring network stations, measurements alone are not sufficient to provide a complete picture of current ambient PAH levels. In this study, speciation profiles for PAHs are prepared and the Sparse Matrix Operator Kernel Emissions (SMOKE) model is used to generate the gridded national emissions of 16 priority PAHs in the US. The estimated emissions are applied in a modified Community Multi-scale Air Quality (CMAQ) model (v5.0.1) to simulate ambient concentrations of PAHs and quantify the contributions of different emission sources to the predicted concentrations. The emission modeling results show that 16-PAH emission in the US is approximately 34.8 Gg in 2011. Residential wood combustion, motor vehicles and industrial point sources are major sources of PAHs. Predicted ambient PAH concentrations by the modified CMAQ model show low biases for most species. Mean fractional bias (MFB) based on daily concentrations are generally less than 0.67, and mean fractional error (MFE) less than 1.0. Averaging the predictions over a month reduces the overall error of the prediction, as indicated by lower MFE values. Heterogeneous reactions of PAHs with O_3 on particle surface are needed to reduce the bias of the model results. Source apportionment simulations show that residential wood combustion is the most significant contributor of PAHs concentrations in winter. Motor vehicles and industrial point sources are shown to be major contributors in the US of PAHs throughout of the year.

DEDICATION

I would like to delicate this work to my dear parents. Their encouragement and support are always helping me overcome difficulties in life and study.

ACKNOWLEDGEMENTS

I would like to appreciate my advisor, Dr. Qi Ying, who is always providing patient and helpful guidance on my research and studies, which help me to achieve progress on my thesis. I also would like to express my gratitude to my committee members, Dr. Bill Batchelor and Dr. Renyi Zhang for the support of my graduate studies and research. I feel honored to have them in my thesis committee.

In addition, I would like to thank the members in our group, Dr. Jingyi Li, Dr. Hongliang Zhang, and Peng Wang for support and help during my research study.

TABLE OF CONTENTS

	Page
ABSTRACT	ii
DEDICATION	iii
ACKNOWLEDGEMENTS	iv
TABLE OF CONTENTS	v
LIST OF FIGURES	vi
LIST OF TABLES	viii
1. INTRODUCTION	1
2. GENERATION OF GRIDDED PAH EMISSIONS	7
2.1 Emission processing with the SMOKE model	7
2.2 Results	26
2.3 Conclusions	32
3. CMAQ MODEL DEVELOPMENT AND EVALUATION	34
3.1 Mechanisms description	34
3.2 Model application	41
3.3 Results	42
3.4 Discussions	56
4. SOURCE APPORTIONMENT OF PAHS	63
4.1 Methodology	63
4.2 PAHs source apportionment for three large cities	63
4.3 Conclusions	70
5. CONCLUSIONS	71
REFERENCES	72
APPENDIX	80

LIST OF FIGURES

Page

Figure 1 Residential wood combustion emission factors of 16 PAHs (profile 4642 and 91109). The unit of emission factors in gas phase is g/g TOG, and unit of emission factors in particle phase is g/g $PM_{2.5}$. 13

Figure 2 Gasoline emission factors of 16 PAHs (1101 and 91122). The unit of the emission factors in gas phase is g/g TOG, and unit of emission factors in particle phase is g/g $PM_{2.5}$. 16

Figure 3 Diesel emission factors of 16 PAHs (4674 and 91106). The unit of the emission factors in gas phase is g/g TOG, and unit of emission factors in particle phase is g/g $PM_{2.5}$. 16

Figure 4 Gridded monthly emissions of 16-PAH (column A), 7-PAH (column B), and BaP (column C) for January (row 1), April (row 2), July (row 3), and October (row 4). Units are Mg/month (10^6 g/month). 27

Figure 5 Observations sites locations in the domain.. 41

Figure 6 Observed and predicted daily PAHs concentrations for four months at all available sites. The three lines represent 10:1, 1:1, and 1:10 ratio, respectively.. 46

Figure 7 Observed and predicted monthly average PAH concentrations at all available sites. The three lines represent 10:1, 1:1, and 1:10 prediction vs observation ratio respectively.. 51

Figure 8 Monthly average surface concentrations of 16-PAH (column A), 7-PAH (column B), and BaP (column C) for January (row 1), April (row 2), July (row 3), and October (row 4). Units are μg/m^3 for 16-PAH and 7-PAH, and ng/m^3 for BaP. 54

Figure 9 Observed and predicted PAH concentrations in January 2011 with (Modified and without (Original) heterogeneous reactions of PAHs with ozone 58

Figure 10 Domain averaged PAH fractional concentration in gas and particle phases in January and July 60

Page

Figure 11 Source contribution to 16-PAH concentrations in New York, Los Angeles, and Houston in January and July 2011 65

Figure 12 Source contribution to BaP concentrations in New York, Los Angeles, and Houston in January and July 2011 69

LIST OF TABLES

Page

Table 1 US EPA 16 priority polycyclic aromatic hydrocarbons and 7 possible human carcinogens (underlined) 1

Table 2 Platform sectors for PAH species emissions generation 8

Table 3 Profiles modified in residential wood combustion........ 13

Table 4 Profiles modified in on-road sectors 14

Table 5 Profiles modified in np_oilgas and pt_oilgas sectors........ 17

Table 6 Profiles modified in c1c2rail and c3marine sectors 18

Table 7 Profiles modified in ptegu and ptegu_pk sectors 19

Table 8 Profiles modified in ptfire sector........ 20

Table 9 Other profiles modified in this study 21

Table 10 NEI 2011 Sectors Classification [35]........ 29

Table 11 Fractional contributions of major sources to total 16 PAH emissions........ 31

Table 12 PAHs reaction rate coefficients with OH radicals 34

Table 13 PAHs reaction rate coefficients with O_3 36

Table 14 KOA Regression Intercepts (A) and slopes (B) 39

Table 15 logpL0 Regression Intercepts bL and Slopes mL 40

Table 16 PAH Species Observations Sites........ 43

Table 17 Statistical analysis of model performance for daily average PAH concentrations. The unit of average observation (Ave_obs) and perdictions (Ave_Mod) is μg/m^3. 48

Table 18 Statistical analysis of model performance for monthly average PAH concentrations. The unit of the average observations (Ave_obs) and predictions (Ave_Mod) is μg/m^3. 52

Page

Table 19 Heterogeneous-reaction of particulate PAHs with O_3 56

Table 20 Statistical analysis of model performance in January when heterogeneous reactions of O_3 are included.. 59

Table 21 PAH emission factors (EF) for wood burning, coal burning and gasoline vehicles 61

Table 22 Reaction rate coefficients of PAHs used in this study and in other publications... 62

Table 23 Location of the cities used in the source apportionment analysis................. 64

1. INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are defined as a group of organic compounds containing multiple aromatic rings. PAHs in the ambient air and their photochemical oxidation products have been shown to cause human cancer [1]. Benzo[a]pyrene (BaP), the first chemical carcinogen discovered, is often applied as an indicator for PAHs exposure risk assessment [2]. US Environmental Protection Agency (US EPA) has classified 16 of the PAHs as priority pollutants based on their toxicity and potential of human exposures, among other factors. The US EPA has also classified seven of the PAHs as possible human carcinogens (see Table 1). The main national contributors to the global emission of PAHs are China, India, United States, Indonesia, Brazil, and Russia [3]. Estimated global total PAH emissions peaked in 1995 and decreased after that. The total emission is predicted to have decreased by 46-71% after 1990s [4].

Table 1 US EPA 16 priority polycyclic aromatic hydrocarbons and 7 possible human carcinogens (underlined)

#	Name	Formula	Molecular weight	Aromatic rings	Abbreviation	CAS #
1	Naphthalene	$C_{10}H_8$	128.18	2	NAPH	[91-20-3]
2	Acenaphthylene	$C_{12}H_8$	152.2	2	ACY	[208-96-8]
3	Acenaphthene	$C_{12}H_{10}$	154.21	2	ACE	[83-32-9]
4	Fluorene	$C_{13}H_{10}$	166.22	2	FLU	[86-73-7]
5	Phenanthrene	$C_{14}H_{10}$	178.24	3	PHE	[85-01-8]

Table 1 Continued

#	Name	Formula	Molecular weight	Aromatic rings	Abbreviation	CAS #
6	Anthracene	$C_{14}H_{10}$	178.24	3	ANT	[120-12-7]
7	Fluoranthene	$C_{16}H_{10}$	202.26	3	FTH	[206-44-0]
8	Pyrene	$C_{16}H_{10}$	202.26	4	PYR	[129-00-0]
9	Benzo[a]Anthracene	$C_{18}H_{12}$	228.30	4	BaA	[56-55-3]
10	Chrysene	$C_{18}H_{12}$	228.30	4	CHRY	[218-01-9]
11	Benzo[b]Fluoranthene	$C_{20}H_{12}$	252.32	4	BbF	[205-99-2]
12	Benzo[k]Fluoranthene	$C_{20}H_{12}$	252.32	4	BkF	[207-08-9]
13	Benzo[a]Pyrene	$C_{20}H_{12}$	252.32	5	BaP	[50-32-8]
14	Benzo[ghi]Perylene	$C_{22}H_{12}$	276.34	6	BghiP	[191-24-2]
15	Indeno(1,2,3-cd)Pyrene	$C_{22}H_{12}$	276.34	5	IcdP	[193-39-5]
16	Dibenz(ah)Anthracene	$C_{22}H_{14}$	278.36	5	DahA	[215-58-7]

PAHs can be directly released into the environment from natural sources, such as volcanoes and forest fires. PAH compounds also exist in crude oil, coal, and other fossil-fuel products. Most of the PAHs in the atmosphere are released from the high temperature combustion processes related to human commercial and industrial activities, such as power generation, petroleum production processes, and motor vehicle exhaust [5]. There are four major emission sources of PAHs in the United States: residential combustion, mobile, industrial processes, and open burning [3, 4].

Residential combustion consists of burning of wood, coal, oil, natural gas, or other organic substances for daily cooking, heating, and energy generation. Wood combustion from fireplaces has been proved to be a major contributor to annual emission of various air pollutants in urban areas [6, 7]. Emission factors of PAHs measured from wood combustion vary due to different wood types. For example, measured BaP emission factors are range from 0.245 mg/kg to 0.712 mg/kg wood burned for pine, oak, and eucalyptus [7], which would cause considerable uncertainty in PAHs emission estimation.

Emissions from vehicles, aircrafts, ships, locomotives, and other off-road vehicles are typically grouped into the mobile source category. Gasoline and diesel vehicles are the dominant mobile emission sources in US [8]. In order to reduce the air pollution from gasoline powered motor vehicles, vehicle designs and gasoline formulations had been changed during the 1980s and 1990s [9]. Both gasoline and diesel powered motor vehicles have higher emissions during start or idle, compared to moving exhaust. Trains, ships, and aircrafts also make significant contributions to mobile source emissions. PAH emissions from these sectors depend on the composition of the fuel. Diesel fuel consists of aliphatic hydrocarbons containing a large number of carbon atoms, which will be released in diesel exhaust [10]. In addition to fuel composition, catalyst types can also cause uncertainties. BaP emission from non-catalyst gasoline powered engines is reported to be 41.0 μg/km. However, BaP emission from catalyst gasoline powered engines is only 0.021 μg/km [10].

The dominant sources of industrial processes emissions are primary aluminum production, coke production, electric power generation, and oil gas production. The amount of emissions depends on the manufacturing processes, boiler types, and air pollution control devices [11].

To determine the risk for human beings, ambient concentrations of each PAH species need to be determined. They are influenced by transport, deposition, gas-to-particle partitioning, and chemical transformation processes [12]. Large molecular weight species are relatively easier to be adsorbed on the particle phase, while lower molecular weight species with higher vapor pressure are more abundant in the gas phase. The partitioning of PAHs between different phases also depends on the properties of each species, as well as the concentration of partitioning media, such as organic matter and element carbon content. PAH species can be partitioned into particle phase by adsorption onto particle surface (particularly black carbons (BC)) and absorption into the amorphous organic particulate organic matters (OM) [13]. Gas-to-particle partitioning theory was originally suggested by Junge [14]. Pankow [15] and Harner and Bidleman [12] developed the PAH partitioning theory by relating the PAH organic-air partitioning coefficient with octanol-air partitioning coefficient. Subsequently, it is found that BC adsorption dominates at low semi-volatile organic compounds (SVOCs) concentration because of large surface area. As concentration of organics increases, absorption of organic matter becomes progressively important [16]. Thus it is important to include BC adsorption into the gas-to-particle partitioning calculation [17].

Previous modeling studies of PAHs focused on BaP ambient concentration in Europe and

Asia. Aulinger et al. [18] introduced a PAHs partitioning mechanism into the Community Multiscale Air Quality (CMAQ) model to simulate BaP concentrations in Europe. It was found that ignoring chemical or photolytic degradation of BaP caused 4 times over-prediction of BaP. Sensitivity analysis showed that heterogeneous reaction of BaP with ozone has significant impact on BaP ambient concentration prediction [19]. It was also suggested that including the appropriate seasonal and diurnal cycles in BaP emissions is important to get a better temporal and spatial resolution of BaP concentration and deposition patterns [20]. Friedman and Selin showed that gas-to-particle partition has substantial impacts on the transport and fate of PAHs, and should be correctly accounted for in regional and global PAHs simulations [21].

In another study, Inomata et al. updated the Regional Air Quality Model (RAQM) to simulate the transport of particulate PAHs in Northeast Asia [22]. The model predicted well of 6 PAH species (PYR, CHRY, BbF, BkF, BaP, and IcdP) ambient concentration in Beijing. However, the predictions of 9 PAH species (including the above 6 PAH species and FLU, BaA, and BghiP) in the downwind Noto monitoring site in Japan differ more from observed concentrations by as much as a factor of 5. The large discrepancy of predicted and observed PAHs were studied by Thackray et al. It was concluded that regional emission estimation, model coefficients, and uncertainty of in-situ observations all contribute to the discrepancies [23].

Positive Matrix Factorization (PMF) is widely used for determination of PAHs source contributions in European and Asian cities [24-26]. Based on the previous studies, combustion of wood, coal and motor vehicles are the most significant sources of PAHs in urban atmosphere [26]. PMF, and other receptor-oriented source apportionment methods, however, only provide source contribution information at locations where measurements are available.

There are no modeling studies reported in the literatures to quantitatively determine the concentration of PAH species in the United States utilizing regional transport models. Information regarding PAHs concentrations and source contribution is helpful for determining emission control policies and analyses of PAHs impact on human health.

The objectives of this study are to (1) generate a gridded emission inventory of 16 PAH species in North America based on the most recent version of the National Emission Inventory; and (2) modify the most recent version of the CMAQ model to include gas phase decay reactions, gas-to-particle partitioning and particle phase reactions to simulation PAHs concentrations and determine contributions from major sources in the entire continental US. The emission preparation work is documented in Section 2. The CMAQ model development and application are described in Section 3. And finally, source apportionment simulations of PAHs are described in Section 4.

2. GENERATION OF GRIDDED PAH EMISSIONS

2.1 Emission processing with the SMOKE model

2.1.1 Overview of emission processing

Sparse Matrix Operator Kernel Emission (SMOKE) model is used to convert county-level annual emissions of total volatile organic compounds (VOCs) and PM in emission inventories to generate the spatial and temporal resolved PAH emissions needed for an air quality model. The detailed operation of the SMOKE model and the associated input files have been described in greater detail elsewhere [27], and thus only a short summary is provided below.

The SMOKE model reads emission inventory data files and transforms inventory species into model species matching the chemical mechanisms used in an air quality model. Subsequently, spatial distributions of emissions is determined using source specific spatial allocation surrogates or coordinates of the point source emissions. Temporal allocation of emissions are determined using source specific profiles that gradually break annual emissions to hourly emissions needed for an air quality model. During the merge step, the speciation, spatial allocation, and temporal allocation information are combined to generate gridded 1-hour resolution emissions.

Emission inventories typically include a large number of emission record for various sources. An appropriate chemical speciation profile needs to be selected based on the EPA's source classification code (SCC) associated with each emission record. A cross reference file is used to determine the appropriate chemical speciation profile for a given SCC code. Typically, due to the limitation of the available source testing data, multiple SCC codes of similar emission sources/ fuel types are mapped to use the same speciation profile. In order to generate PAH emissions, the profiles have to be modified to include the emission factors for these new species. In the speciation profiles for organic species in the gas phase, the emission factors are expressed as fraction emission of total organic gases (TOG) or VOCs. In particle phase profiles, emission factors are expressed as mass of the species emitted per unit mass emission of $PM_{2.5}$.

In this study, the 2011 National Emission Inventory (NEI) is used to estimate emissions of PAHs and other pollutants. Individual emission sectors in US used in the 2011 NEI are classified into 9 broad sectors: residential wood combustion, motor vehicles, oil and gas processes, railway and marine vessels, non-road engines, electric generation units, wildfire, other point and nonpoint sources, and Canada Mexico emission. The grouping of the individual sector is shown in Table 2.

Table 2 Platform sectors for PAH species emissions generation [28]

Broad sectors	Platform Sector	Short Name	Description
Residential wood combustion	Residential wood combustion	rwc	Nonpoint sources of residential wood combustion.

Table 2 Continued

Broad sectors	Platform Sector	Short Name	Description
Motor vehicles	Onroad for CA and TX	rateperdistance _catx	Onroad gasoline and diesel mobile sources from moving vehicles exhaust for California and Texas. Emission rate calculated by travel distance.
		ratepervehicle _catx	Onroad gasoline and diesel mobile sources from vehicles extended idle or start for California and Texas. Emission rate calculated by vehicles.
	Onroad non-refueling	rateperdistance _noRFL	The same as rateperdistance_catx, but for other states in continental US
		rateperprofile _noRFL	Onroad gasoline and diesel evaporative emissions.
		ratepervehicle _noRFL	The same as rateperdistance_catx but for other states in continental US
	Onroad refueling	rateperdistance _RFLonly	Onroad mobile refueling emissions for moving vehicles.
		rateperprofile _RFLonly	Onroad mobile refueling emissions for gasoline and diesel evaporation.
		ratepervehicle _RFLonly	Onroad mobile refueling emissions for starting or idle vehicles.
Oil and gas process	Nonpoint source oil and gas	np_oilgas	Nonpoint sources from oil and gas related processes.
	Point source oil and gas	pt_oilgas	Point sources from oil and gas production processes.
Railway and marine vessels	Class 1&2 CMV and locomotives	c1c2rail	Emission sources form locomotives and class 1 and class 2 commercial marine vessels (CMVs).
	Class3 commercial marine vessels	c3marine	Point source of class 3 commercial marine vessels.
Non-road engines	Non-road	nonroad	Nonroad equipment emissions.

Table 2 Continued

Electric generation units	EGU non-peaking units	ptegu	Point source of non-peaking electric generating units (EGUs).
	EGU peaking units	ptegu_pk	The same as ptegu sector, but only refers to EGUs that are determined to operate as peaking units.
Wildfire	Point source fires	ptfire	Point source of specific wildfires and prescribed fires
Industrial point/commercial nonpoint	Remaining nonpoint	nonpt	Other nonpoint sources not included in other platform sectors
	Remaining non-EGU point	ptnonipm	Point sources not belonged in other point source sectors, including aircraft emissions, and some rail yard emissions.
Canada and Mexico emisison	Other non-NEI nonpoint and nonroad	othar	Canada and Mexico nonpoint and nonroad mobile emissions.
	Other non-NEI onroad sources	othon	Canada and Mexico onroad mobile emissions.
	Other point sources not from the 2011 NEI	othpt	Point sources from Canada and Mexico. Also includes all non-US C3 CMV and offshore oil production processes.

2.1.2 Development of PAH speciation profiles

In this study, PAH speciation profile data for area and point source sectors are obtained from the SPECIATE database [29], or the L&E POM document [30]. The speciation profiles for mobile sources are extracted from the Motor Vehicle Emission Simulator (MOVES) [31].The SPECIATE database includes a large collection of both gas and particle phase emission profiles. Many of the profiles are used as default profiles in SMOKE to process the NEI emissions. Although PAH species are included in some of the SPECIATE profiles, they are not included in the profiles used by SMOKE model. In this study, the PAH species available for the corresponding SOMKE profiles are extracted from the SPECIATE database and added to the SMOKE profiles.

The L&E POM document, or "Locating and Estimating air emissions from sources of Polycyclic Organic Matter", is also prepared by the US EPA. It is one of a series of documents developed by the EPA to estimate emissions of air toxics. The L&E POM document was developed based on extensive literature review and database search, and includes PAH emission factors for all major combustion sources. However, the latest update of the L&E POM document was 1998, and thus some new source testing data are not included. In addition, emission factors in the L&E POM document are not split into gas and particle phase emissions, which makes it less favorable for emission estimation to support PAH air quality modeling. What is more, the emission factors in the L&E POM document are typically expressed in units of mass of PAH emitted per unit mass of fuel burned. This is not ideal either as they cannot be directly incorporated into the speciation profiles used by SMOKE. In this study, we examined data from both the SPECIATE database and L&E POM document to develop the PAH speciation profiles. To apply the L&E POM emission factors, we choose to (1) put all emissions of PAHs in the particle phase and let the partitioning code in the air quality model to redistribute the PAHs appropriately. Since the modified CMAQ model assumes instant partitioning equilibrium, putting all emissions in the particle phase would not affect the final distribution of PAHs in gas and particle phase. (2) $PM_{2.5}$ mass emission factors (mass per unit mass of fuel burned) are obtained from the "Emission Factor Listing for Criteria Pollutants" document [32], and use to convert the L&E POM emission factors into units of mass of PAH per unit mass of $PM_{2.5}$ emissions.

In the following sections, related emission profiles modified to estimate PAHs emissions are discussed in detail. The detailed emission profiles for the 16 PAH species are shown in the Appendix in Table S1.

2.1.2.1 Residential wood combustion sector

Residential wood combustion sector includes emissions from various wood burning devices, such as fireplaces, woodstoves, pellet stoves, indoor furnaces, outside wood boilers, and outdoor firepots. Although emissions vary significantly with different wood types as well as treatment technologies, only representative three profiles are used in emission processing. In this study, emission rates in both gas and particle phases of PAH species in this sector are obtained from the SPECIATE Database. Related profiles modified in this study are listed in Table 3.

Table 3 Profiles modified in residential wood combustion

Profile ID	Emission Phase	Profile Name	Ref.
4642	Gas phase	Fireplace wood combustion - oak wood	SPECIATE Database 4.0 (#4642)
91105	Particle phase	Residential Wood Combustion: HardSoft - Simplified	SPECIATE Database 4.0 (#4643-#4645)

Examples residential wood combustion emission factors are shown in Figure 1. In residential wood combustion emissions, small molecular weight species such as NAPH, ACY, and ACE are emitted as gas phase species. FLU, PHE, ANT, FTH, PYR, and BGHIP are emitted in both gas and particle phase species. The rest of the species are solely emitted in the particle phase.

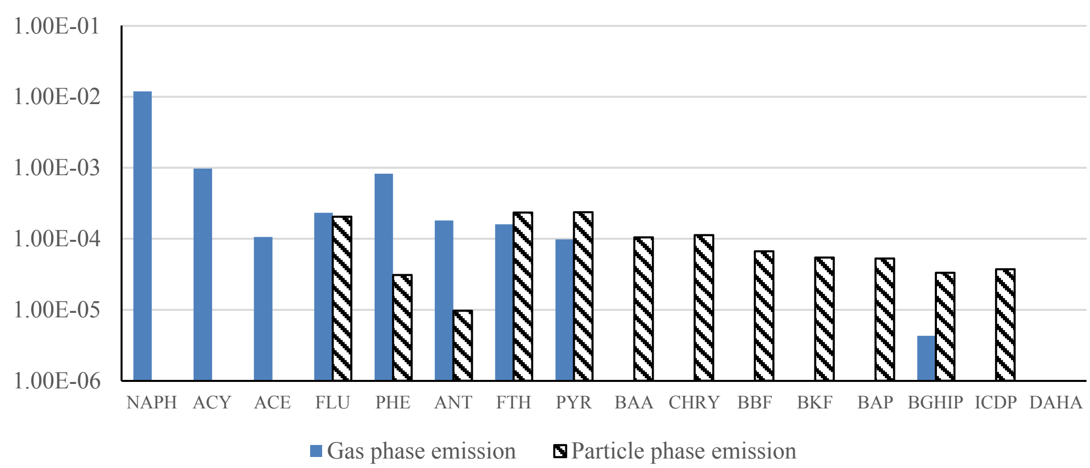


Figure 1 Residential wood combustion emission factors of 16 PAHs (profile 4642 and 91105). The unit of emission factors in gas phase is g/g TOG, and unit of emission factors in particle phase is g/g $PM_{2.5}$.

2.1.2.2 On-road vehicle sector

On-road sources are emissions from motor vehicles operated on public roadways, including passenger cars, motorcycles, minivans, sport vehicles, light duty trucks, heavy duty trucks, and buses. On-road sectors are separated into "on-road non-refueling" and "on-road refueling". On-road refueling sectors, similar to other on-road sectors, are spatially allocated to gas station locations. MOVES is integrated with SMOKE model using inputs of emission per miles traveled or emission per vehicle population data for all counties. SMOKE-MOVES model requires emission rate "lookup" tables generated from MOVES to differentiate emissions from processes, vehicle types, road types, speed, hours, etc. Sources profiles are divided into gasoline engines and diesel engines vehicles. Although the standalone version of the MOVES includes emission factor profiles for the PAHs, the profiles for the SMOKE-MOVES model do not have the necessary PAH species emission factors included and thus need to be modified. Related profiles modified in this study are listed in Table 4.

Table 4 Profiles modified in on-road sectors

Profile ID	Emission Phase	Profile Name	Ref.
1101	Gas phase	Light Duty Gasoline Vehicles	the MOVES2014 section 2.2.1
1186	Gas phase	Heavy Duty Gasoline Trucks	the MOVES2014 section 2.2.1
3150	Gas phase	Gasoline Exhaust: Non-Catalyst-Stabilized	the MOVES2014 section 2.2.1
4547	Gas phase	Gasoline Headspace Vapor - Circle K Diesel	SPECIATE Database 4.0 (#8737)

Table 4 Continued

Profile ID	Emission Phase	Profile Name	Ref.
4674	Gas phase	Diesel exhaust - medium duty trucks	the MOVES2014 section 3.2
8750	Gas phase	New extended idle mode for 2010 CDC run, 14 Dec 2012.	the MOVES2014 section 2.2.1
8762	Gas phase	Composite Profile - Non-oxygenated Gasoline Headspace Vapor	SPECIATE Database 4.0 (#8737)
87710	Gas phase	New onroad diesel exhaust profile for 2010 CDC run, 12 Nov 2012	the MOVES2014 section 3.2
877P0	Gas phase	New extended idle mode for 2010 CDC run, 14 Dec 2012.	the MOVES2014 section 3.2
8774	Gas phase	Diesel exhaust - medium duty trucks	the MOVES2014 section 3.2
91106	Particle phase	HDDV Exhaust - Simplified	the MOVES2014 section 3.2 $OC_{2.5}/PM_{2.5}=1$ in diesel exhaust
91113	Particle phase	Non-catalyst Gasoline Exhaust - Simplified	the MOVES2014 section 2.2.1 OC2.5/PM2.5 is reported in this section
91122	Particle phase	Onroad Gasoline Exhaust - Simplified	the MOVES2014 section 2.2.1
91162	Particle phase	LDDV Exhaust - Simplified	the MOVES2014 section 3.2

Gasoline emission factors of PAHs in gas phase and particle phase are shown in Figure 2. And diesel emission factors of PAHs in gas phase and particle phase are shown in Figure 3. NAPH is the most abundant species in the PAH emissions from both gasoline and diesel vehicles. Lower molecular weight species have higher emission factor in gas phase from both gasoline and diesel exhaust. In gasoline exhaust, higher molecular weight species, such as BghiP, and IcdP have higher emissions in the particle phase. However, in diesel exhaust, the larger molecular weight species have lower emission rates.

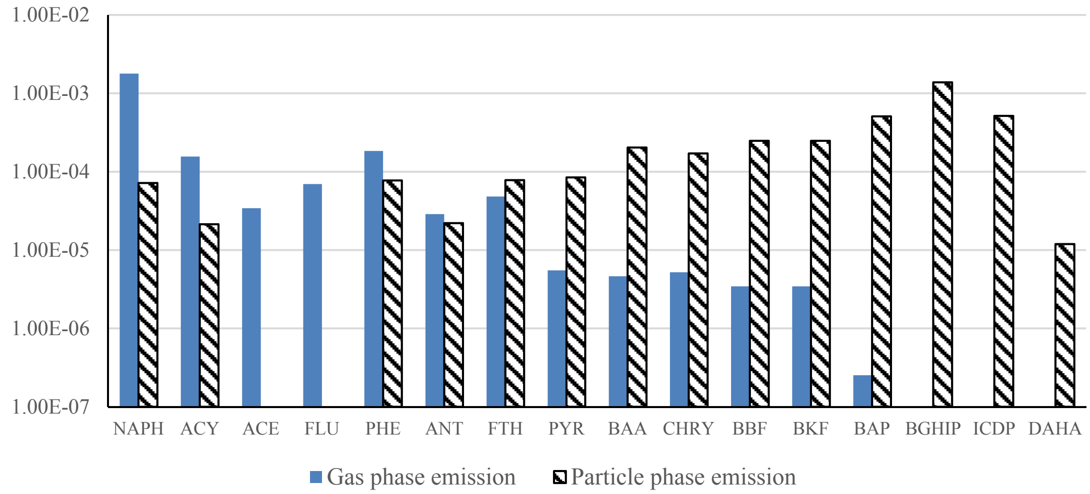


Figure 2 Gasoline emission factors of 16 PAHs (1101 and 91122). The unit of the emission factors in gas phase is g/g TOG, and unit of emission factors in particle phase is g/g $PM_{2.5}$.

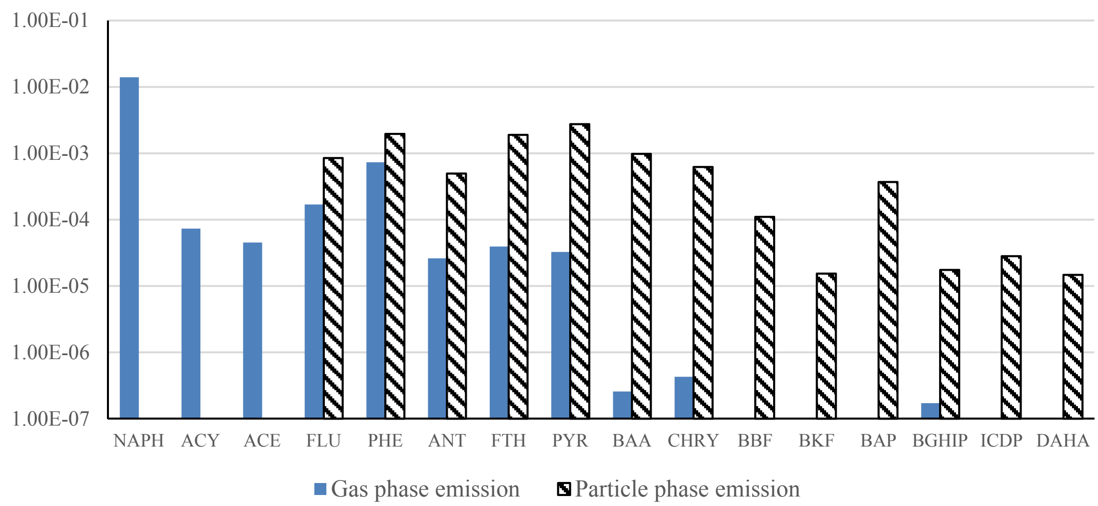


Figure 3 Diesel emission factors of 16 PAHs (4674 and 91106). The unit of the emission factors in gas phase is g/g TOG, and unit of emission factors in particle phase is g/g $PM_{2.5}$.

2.1.2.3 Oil and gas process sector

The oil and gas sectors involve both point sources and nonpoint sources sectors. Point oil gas sector emissions are submitted by states, while nonpoint oil gas sector emissions are developed by US EPA for each county in the US with oil and gas activities [28]. Related profiles modified in this study are listed in Table 5.

Table 5 Profiles modified in np_oilgas and pt_oilgas sectors

Profile ID	Emission Phase	Profile Name	Ref.
91112	Particle phase	Natural Gas Combustion - Simplified	L&E document Table 4.1.2-6 UEFL (CAP) Appendix B
91115	Particle phase	Distillate Oil Combustion - Simplified	L&E document Table 4.1.2-11 UEFL (CAP) Appendix B
91117	Particle phase	Residual Oil Combustion - Simplified	L&E document Table 4.1.2-11 UEFL (CAP) Appendix B
91136	Particle phase	Process Gas Combustion - Simplified	SPECIATE Database 4.4 (#4398 #4407 #4415)

2.1.2.4 Railway and commercial marine vessels sector

The c1c2rail sector in 2011 NEI includes emissions from locomotives and smaller commercial marine vessels considered as non-point sources, while c3marine sector includes emissions from large commercial marine vessels, which are considered as point sources. The locomotives with significant PAHs emissions are powered by diesel-fueled internal combustion engines. The marine vessels in these two sectors have either diesel-fueled internal combustion engines or residual-oil fired boilers. Related profiles modified in this study used by SMOKE to process for these two sectors are listed in Table 6.

Table 6 Profiles modified in c1c2rail and c3marine sectors

Profile ID	Emission Phase	Profile Name	Ref.
2480	Gas phase	Industrial Cluster, Ship Channel, Downwind Sample - 1993	
5674	Particle phase	Marine Vessel - Main Engine - Heavy Fuel Oil	
8774	Gas phase	Diesel exhaust - medium duty trucks	The MOVES2014 section 3.2
91106	Particle phase	HDDV Exhaust - Simplified	The MOVES2014 section 3.2

2.1.2.5 Non-road engines sector

Non-road engines emission sector contains exhaust, evaporation, and refueling emissions from non-road engines (except emissions from commercial marine vessels, locomotives, and aircrafts). All non-road emissions are collected at county level. The same profiles used in on-road sectors are used to speciate VOC emission into PAHs emissions for non-road sources.

2.1.2.6 Electric generation units sector

The EGU sectors include the ptegu and ptegu_pk sectors in the NEI 2011 point inventory.

US EPA split the electric power generation emissions into two sectors to facilitate analysis of the impact of emissions from units that are running only during peak electricity generator hours. Related profiles modified in this study are listed in Table 7.

Table 7 Profiles modified in ptegu and ptegu_pk sectors

Profile ID	Emission Phase	Profile Name	Ref.
0008	Gas phase	Reciprocating Diesel Engine	the MOVES2014 section 3.2
0009	Gas phase	Reciprocating Distillate Oil Engine	the MOVES2014 section 3.2
1084	Gas phase	Residential Wood Combustion (C-1 - C-6)	SPECIATE Database 3.2 (#1167) Schauer et al. [7]
2420	Gas phase	Degreasing, All Processes/All Industries	SPECIATE Database 4.1 (#7196)
2485	Gas phase	Composite of 21 Fugitive Emission Profiles from Petroleum Industry Facilities - 1993	SPECIATE Database 3.2 (#1192 #1195)
5565B	Gas phase	Aircraft Landing/Takeoff (LTO) - Commercial	SPECIATE Database 4.3 (#8862)
8745	Gas phase	Composite Profile - Degreasing: Cold Cleaning	SPECIATE Database 4.0 (#8745)
91106	Particle phase	HDDV Exhaust - Simplified	the MOVES2014 section 3.2
91110	Particle phase	Sub-Bituminous Combustion - Simplified	L&E document Table 4.1.2-9 UEFL (CAP) Appendix B
91112	Particle phase	Natural Gas Combustion - Simplified	L&E document Table 4.1.2-6 UEFL (CAP) Appendix B
91114	Particle phase	Wood Fired Boiler - Simplified	SPECIATE Database 4.4 (#8898-#8903)
91115	Particle phase	Distillate Oil Combustion - Simplified	L&E document Table 4.1.2-11 UEFL (CAP) Appendix B

Table 7 Continued

Profile ID	Emission Phase	Profile Name	Ref.
91117	Particle phase	Residual Oil Combustion - Simplified	L&E document Table 4.1.2-11 UEFL (CAP) Appendix B
91119	Particle phase	Kraft Recovery Furnace - Simplified	L&E document Table 4.9.1-1 UEFL (CAP) Appendix B
91125	Particle phase	PM_{SO2} Controlled Lignite Combustion - Simplified	SPECIATE Database 4.4 (#4367 #4368 #4369 #4370)
91126	Particle phase	Solid Waste Combustion - Simplified	L&E document Table 4.3.2-1 UEFL (CAP) Appendix B
91136	Particle phase	Process Gas Combustion - Simplified	SPECIATE Database 4.4 (#4398 #4407 #4415)
91145	Particle phase	Petroleum Ind - Avg - Simplified	SPECIATE Database 4.4 (#4394 - #4401 #4403)
91147	Particle phase	Overall Average/Default - Simplified	

2.1.2.7 Fires sector

The ptfire sector includes point source emissions from wildfire and prescribed burning processes with daily data. However, open burning and agricultural burning emissions without daily data are assigned to the nonpoint sector. Related profiles modified in this study are listed in Table 8.

Table 8 Profiles modified in ptfire sector

Profile ID	Emission Phase	Profile Name	Ref.
5560	Gas phase	Miscellaneous burning-Forest Fire	SPECIATE Database 4.4 (#8743)

Table 8 Continued

Profile ID	Emission Phase	Profile Name	Ref.
91102	Particle phase	Wildfires - Simplified	SPECIATE Database 4.4 (#4463 #4464 #4465 #4466 #4468 #423212.5)
91109	Particle phase	Prescribed Burning - Simplified	SPECIATE Database 4.4 (#4463 #4464 #4465 #4466 #4468 #423212.5)

2.1.2.8 Other nonpoint and point source sectors

The sectors in this group includes the following emission sources: the industrial, commercial, and residential combustion stationary emissions; chemical manufacture; solvent utilization; chemicals storage and transport; waste disposal and treatment; industrial metal production; and agricultural burning. Other point source emissions in the 2011 NEI are also grouped into this category. Other profiles modified in this study are listed in Table 9.

Table 9 Other profiles modified in this study

Profile ID	Emission phase	Profile name	Ref.
0000	Gas phase	Added for 2002ac othar - zero emissions	SPECIATE Database 3.2 (#0000)
0079	Gas phase	Chemical Manufacturing - Flares	L&E document Table 4.3.8-1 UEFL (CAP) Appendix B

Table 9 Continued

Profile ID	Emission phase	Profile name	Ref.
1007	Gas phase	Mineral Products - Asphaltic Concrete	SPECIATE Database 3.2 (#1007)
1064	Gas phase	Olefins Production - Ethylene - Compressor Lube Oil Vent	SPECIATE Database 4.3 (#8861)
1095	Gas phase	Textile Products - General Fabric Operations - Dyeing and Curing	SPECIATE Database 3.2 (#1095)
1096	Gas phase	Textile Products - General Fabric Operations - Tenter Frame	SPECIATE Database 3.2 (#1096)
1189	Gas phase	Pulp and Paper Industry - Plywood Veneer Dryer	SPECIATE Database 4.4 (#8787 #8805 - #8810 #8814 - #8820)
1192	Gas phase	Degreasing	SPECIATE Database 3.2 (#1192)
1193	Gas phase	Dry cleaning	SPECIATE Database 3.2 (#1193)
1194	Gas phase	Autobody Repair	SPECIATE Database 3.2 (#1194)
1195	Gas phase	Degreasing - Composite	SPECIATE Database 3.2 (#1195)
1196	Gas phase	Drycleaning - Composite	SPECIATE Database 3.2 (#1196)
1202	Gas phase	Primary Aluminum Production	SPECIATE Database 3.2 (#1202)
2508	Gas phase	Vehicle Exhaust - Juarez rush hour traffic - 1996	the MOVES2014 section 3.2
3002	Gas phase	Landfills	SPECIATE Database 3.2 (#5652-#5654)
3066	Gas phase	Consumer Products: General Purpose Cleaners - Aerosols	SPECIATE Database 4.0 (#3123)
3127	Gas phase	Aerosol Coatings: Metallic Pigmented Coatings	SPECIATE Database 4.0 (#3128)

Table 9 Continued

Profile ID	Emission phase	Profile name	Ref.
3131	Gas phase	Aerosol Coatings: Auto Body Primers	SPECIATE Database 4.0 (#3128)
3134	Gas phase	Aerosol Coatings: Exact Match Automotive Coatings	SPECIATE Database 4.0 (#3128)
3135	Gas phase	Aerosol Coatings: Ground/Traffic/Marking Coatings	SPECIATE Database 4.0 (#3128)
3137	Gas phase	Aerosol Coatings: Vinyl/Fabric/Leather/Polycarb Coatings	SPECIATE Database 4.0 (#3128)
3138	Gas phase	Aerosol Coatings: Coatings (Unspecified)	SPECIATE Database 4.0 (#3128)
3139	Gas phase	Architectural Coatings: Solvent Borne	SPECIATE Database 4.0 (#3149)
3140	Gas phase	Architectural Coatings: Water Borne	SPECIATE Database 4.0 (#3149)
3141	Gas phase	Thinning Solvent/Mineral Spirits	SPECIATE Database 4.0 (#3141)
3142	Gas phase	Consumer Products Composite: Adhesives And Sealants	SPECIATE Database 3.2 (#8523 #8525)
3144	Gas phase	Consumer Products Composite: Solvents And Coating Related Products	SPECIATE Database 4.0 (#3123)
3146	Gas phase	Consumer Products Composite: Personal Care Products	SPECIATE Database 4.0 (#3123)
3147	Gas phase	Consumer Products Composite: Personal Care Products	SPECIATE Database 4.0 (#3123)
3149	Gas phase	Aerosol Coatings: Overall Composite	SPECIATE Database 4.4 (#3128)
3161	Gas phase	Diesel Exhaust - Farm equipment	the MOVES2014 section 3.2
4458	Gas phase	Paraffinic Petroleum Distillate	SPECIATE Database 4.4 (#4435)

Table 9 Continued

Profile ID	Emission phase	Profile name	Ref.
4553	Gas phase	Meat charbroiling	SPECIATE Database 4.4 (#4553)
4651	Gas phase	Cooking vegetables - Stir frying in canola oil	SPECIATE Database 4.4 (#4651)
4652	Gas phase	Cooking potatoes - Deep frying in hydrogenated oil	SPECIATE Database 4.4 (#4652)
4659	Gas phase	Cigarette smoke	SPECIATE Database 4.4 (#4659)
4730	Gas phase	External Combustion - Pulp and Paper Mills Kraft Process Recovery Boiler	SPECIATE Database 4.4 (#4730-#4732)
5565B	Gas phase	Aircraft Landing/Takeoff (LTO) - Commercial	SPECIATE Database 4.4 (#1097-#1099 #5565 #8876 #8877)
8520	Gas phase	Consumer and Commercial Products: Automotive Aftermarket Products: All Automotive Aftermarket Products	SPECIATE Database 4.4 (#8500 #8523 #8525-#8527 #8531)
8744	Gas phase	Composite Profile - Architectural Coatings: Solvent Borne and water borne	SPECIATE Database 4.4 (#3149)
91103	Particle phase	Agricultural Burning - Simplified	SPECIATE Database 4.4 (#8943)
91105	Particle phase	Residential Wood Combustion: HardSoft – Simplified; Assignment basis: Mexico SCC	SPECIATE Database 3.2 (#1167) Schauer et al. [7]
91108	Particle phase	Paved Road Dust - Simplified	SPECIATE Database 4.4 (#4656 #4657 #4658)
91116	Particle phase	Charbroiling - Simplified	L&E document Table 4.12.9-1 UEFL (CAP) Appendix B
91127	Particle phase	Cement Production - Simplified	L&E document Table 4.8 UEFL (CAP) Appendix B

Table 9 Continued

Profile ID	Emission phase	Profile name	Ref.
91132	Particle phase	Secondary Aluminum - Simplified	L&E document Table 4.4.1-1 UEFL (CAP) Appendix B
91135	Particle phase	Meat Frying - Simplified	SPECIATE Database 4.4 (#4653 #4654)
91137	Particle phase	Aluminum Production - Simplified	L&E document Table 4.4.1-1 UEFL (CAP) Appendix B
91138	Particle phase	Lime Kiln - Simplified	L&E document Table 4.9.2-1 UEFL (CAP) Appendix B
91139	Particle phase	Sintering Furnace - Simplified	L&E document Table 4.4.2-1 UEFL (CAP) Appendix B
91140	Particle phase	Charcoal Manufacturing - Simplified	
91146	Particle phase	Slash Burning - Simplified	SPECIATE Database 4.4 (#4467)
91148	Particle phase	Asphalt Roofing - Simplified	L&E document Table 4.6-2 UEFL (CAP) Appendix B
91155	Particle phase	Residential Coal Combustion - Simplified	L&E document Table 4.1.2-9 UEFL (CAP) Appendix B
91156	Particle phase	Residential Natural Gas Combustion - Simplified	L&E document Table 4.1-9 UEFL (CAP) Appendix B
91157	Particle phase	Cast Iron Cupola - Simplified	L&E document Table 4.4.4-1 UEFL (CAP) Appendix B
91158	Particle phase	Secondary Copper - Simplified	L&E document Table 4.4.1-1 UEFL (CAP) Appendix B
91159	Particle phase	Asphalt Manufacturing - Simplified	
91168	Particle phase	Secondary Lead - Simplified	L&E document Table 4.4.1-1 UEFL (CAP) Appendix B
91175	Particle phase	Potato Deep-Frying - Simplified	SPECIATE 4.0 (#3915-#3919)
91177	Particle phase	Sludge Combustion - Simplified	L&E document Table 4.3.3-1 UEFL (CAP) Appendix B

Table 9 Continued

Profile ID	Emission phase	Profile name	Ref.
91178	Particle phase	Lead Production - Simplified	L&E document Table 4.4.1-1 UEFL (CAP) Appendix B
92018	Particle phase	Cigarette Smoke - Simplified	L&E document Table 4.12.4-1 UEFL (CAP) Appendix B

2.1.2.9 Canada and Mexico emissions

This sector includes the on-road, point, and nonpoint emission sources located within the domain but out of US. Because of the long range transport, they would have impacts on the predicted ambient concentrations of PAHs in the US. Profiles used in this sector are included in the other US emission sectors.

2.2 Results

2.2.1 Regional distribution of PAH emissions

In this study, emissions of 16 priority PAH species in January, April, July, and October of 2011 are generated, which represent emissions in winter, spring, summer, and fall respectively.

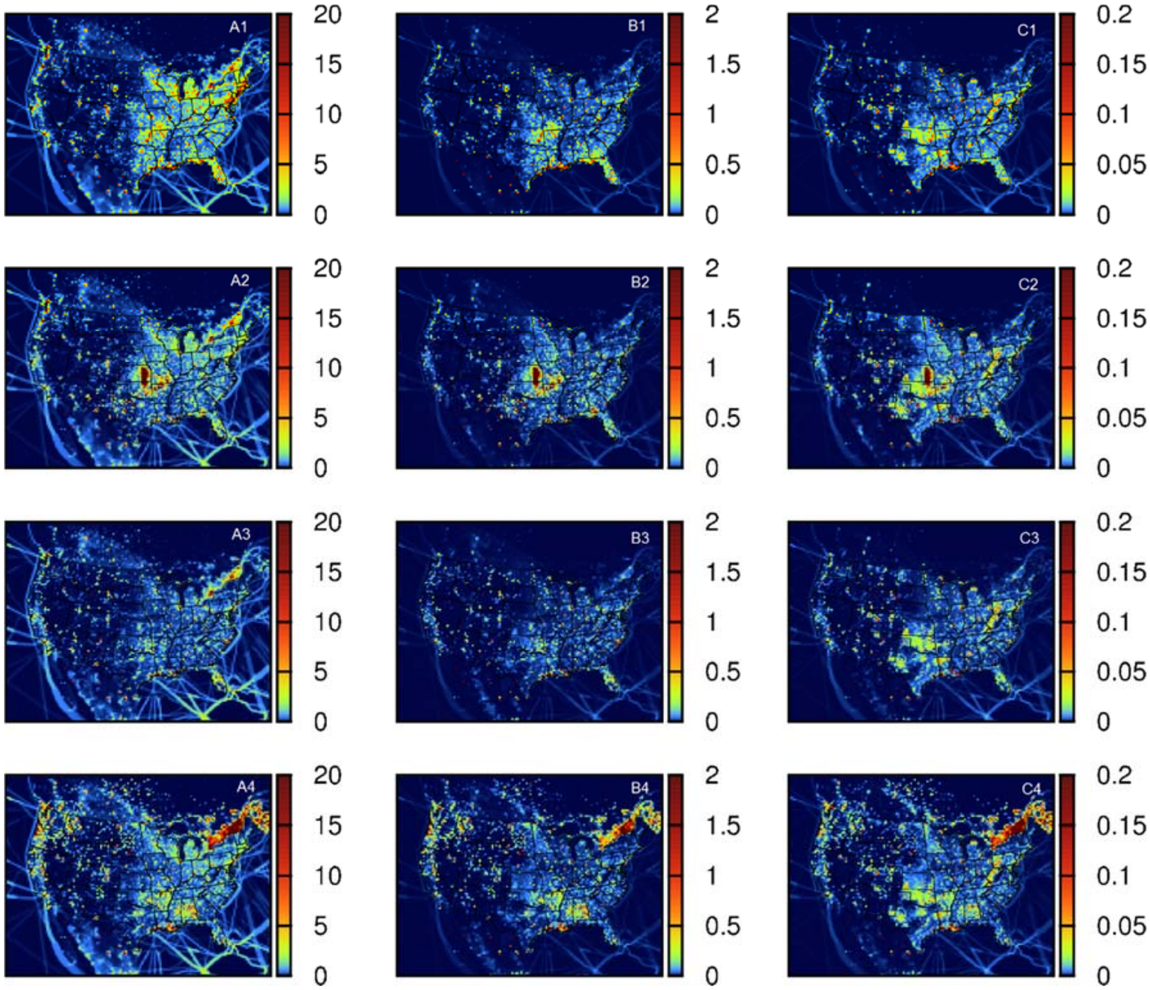


Figure 4 Gridded monthly emissions of 16-PAH (column A), 7-PAH (column B), and BaP (column C) for January (row 1), April (row 2), July (row 3), and October (row 4). Units are Mg/month (10^6 g/month).

In the reginal plots of PAHs emission (see Figure 4), 16-PAH is the sum of the emissions of the 16 PAHs listed in Table 1. 7-PAH is the sum of the emissions of the 7 PAH species known to cause cancer [33]. Monthly emission is generated for January, April, July, and October to represent winter, spring, summer, and fall season separately. The emissions in the upper layers are combined into the surface layer, when calculating the total emissions. In January, a lot of PAHs emissions are from area sources in urbans, such as residential wood combustion. Further examination of the diurnal variation of the PAHs shown that there are higher PAHs emissions at night. That is because most PAHs are released from heat generation, which is related with temperature and human activities. There is a high emission area located in Kansas and northern Alabama in April, which is caused by wildfire in Flint Hill. It is the largest contiguous area of tallgrass prairie remaining today, where burn regimes are implemented during spring and fall [34]. Wildfire emits a large amount of PAHs into the air. Most emissions in July are from point sources, such as power plants and industrial processes. In October, there are a lot emissions near the border of United States and Canada. As large cities of Canada are located at its Southeast boundary, and average temperature in the region is already less than 10 °C in fall, this large emission is likely caused by residential heating and urban motors in that area. However, in this study, the latest inventories in Canada area is developed in 2005, which may lead to over-estimation of emissions for 2011, because current study indicates the global total emission of PAHs is reducing in recent years [4].

2.2.2 Annual emissions

Then annual emissions are estimated as three times the sum of the four month emission estimated in this study. Annual emissions of PAHs from different sources and total PAH emissions are compared with EPA estimations of the PAHs [35], which are also based on the 2011 NEI data. The EPA reported emissions are grouped into 7 major sectors (without wildfire, and Canada and Mexico emissions) but the individual sectors used in EPA's analysis are different from the 24-sector used by the 2011 NEI for MOKE processing. The EPA classification is shown in Table 10. The annual 16-PAH, 7-PAH and BaP emissions based on this study is 34.8 Gg/year, 4.5 Gg/year, and 0.55 Gg/year. In comparison of the EPA estimation is 15.4 Gg/year, 0.4 Gg/year, and 0.09 Gg/year, respectively. The difference in the predictions from this study and the EPA's estimations are most likely due to different PAH speciation profiles used to process the NEI emission data. The speciation profiles used by the EPA appear to have missed quite a number of PAHs in various sectors. Using four month emissions to estimate annual emission might also have caused the different in annual emission estimations.

Table 10 NEI 2011 Sectors Classification [35]

Main Sectors	EPA NEI 2011 Sectors
Residential wood combustion	Fuel Comb – Residential – Wood
Motor vehicles	Mobile – On-road Diesel Heavy Duty Vehicles
	Mobile – On-road Diesel Light Duty Vehicles
	Mobile – On-road non-Diesel Heavy Duty Vehicles
	Mobile – On-road non-Diesel Light Duty Vehicles
Oil gas process	Industrial Processes - Oil & Gas Production
Railway and vessels	Mobile – Locomotives Mobile - Commercial Marine Vessels
Non-road engines	Mobile - Non-Road Equipment - Diesel Mobile - Non-Road Equipment - Gasoline Mobile - Non-Road Equipment - Other

Table 10 Continued

Main Sectors	EPA NEI 2011 Sectors
Other point and nonpoint	Bulk Gasoline Terminals
	Commercial Cooking
	Fuel Comb - Comm/Institutional - Biomass
	Fuel Comb - Comm/Institutional - Coal
	Fuel Comb - Comm/Institutional - Natural Gas
	Fuel Comb - Comm/Institutional - Oil
	Fuel Comb - Comm/Institutional - Other
	Fuel Comb - Residential - Natural Gas
	Fuel Comb - Residential - Oil
	Fuel Comb - Residential - Other
	Gas Stations
	Miscellaneous Non-Industrial NEC
	Solvent - Consumer & Commercial Solvent Use
	Solvent - Degreasing
	Solvent - Dry Cleaning
	Solvent - Graphic Arts
	Solvent - Industrial Surface Coating & Solvent Use
	Solvent - Non-Industrial Surface Coating
	Industrial Processes - Cement Manuf
	Industrial Processes - Chemical Manuf
	Industrial Processes - Ferrous Metals
	Industrial Processes - Mining
	Industrial Processes - NEC
	Industrial Processes - Non-ferrous Metals
	Industrial Processes - Petroleum Refineries
	Industrial Processes - Pulp & Paper
	Industrial Processes - Storage and Transfer
	Mobile - Aircraft
	Waste Disposal

Table 10 Continued

Main Sectors	EPA NEI 2011 Sectors
Electric generation units	Fuel Comb - Electric Generation - Biomass
	Fuel Comb - Electric Generation - Coal
	Fuel Comb - Electric Generation - Natural Gas
	Fuel Comb - Electric Generation - Oil
	Fuel Comb - Electric Generation - Other
	Fuel Comb - Industrial Boilers, ICEs - Biomass
	Fuel Comb - Industrial Boilers, ICEs - Coal
	Fuel Comb - Industrial Boilers, ICEs - Natural Gas
	Fuel Comb - Industrial Boilers, ICEs - Oil
	Fuel Comb - Industrial Boilers, ICEs - Other

Fractional contributions of each major sector to PAH emissions have been calculated and compared with the EPA estimation as well as estimations from other studies, as shown in Table 11. The detailed annual emission estimation is shown in the Appendix in the Table S2. Wildfire and open burning emissions are not used in the analysis as these emissions are more likely to be very different from month-to-month, and the four-month emission estimated in this study is not accurate to estimate their annual emissions.

Table 11 Fractional contributions of major sources to total 16 PAH emissions

Sources	This study	EPA estimation	Other studies [3, 4, 36, 37]
Residential wood combustion	38.57%	21.57%	16.0 – 57.7%
Motor vehicles	13.80%	53.27%	17.6 – 24.8%
Electric generation units	1.02%	2.77%	1.0 – 5.1%
Non-road engines	8.46%	6.03%	–
Oil gas process	2.78%	0.21%	8.7%
Railway and marine vessels	4.73%	0.74%	–

Table 11 Continued

Sources	This study	EPA estimation	Other studies [3, 4, 36, 37]
Other point and nonpoint	30.64%	15.40%	7.0 – 41.2%

The major differences in the estimated emission are the larger contributions due to other point and nonpoint sources and lower contributions due to motor vehicles. PAHs emissions from other sector are mainly based on industrial point source with large emission factors. For diesel engines manufactured in 2007 and later, changed composition of oil compounds and advanced emission controls reduced the total emission amount both of VOCs and PAHs. Gasoline containing ethanol also has less PAHs released from engines exhaust [31]. So PAHs emissions from motor vehicles have been reduced in recent year. Thus, the lower contributions of motor vehicles to total PAH emission at national level are not unexpected. Even their overall contributions are low, they can still be one of the major sources of PAHs in urban areas. Due to the large emission factors of PAHs from wood combustion, residential wood burning is a dominant source of PAHs emissions in urban areas. Wildfire also has great amount of PAHs emission, however those emission points are located around forested areas and lasted short period of time, which have low influence on urban concentrations.

2.3 Conclusions

In conclusion, there are higher area source PAHs emissions in winter from residential heating due to low temperature. In summer, point sources from power plants are dominant in PAHs emission. Emissions from motor vehicles are not influenced by seasons. Most PAHs are emitted in the eastern part and west coastal cities in United States. Big cities located in the Northeast of US have the highest emission rate of total 16 PAHs. Emissions of large molecular weight species (7-PAH, and BaP) are higher in the Southeast of US than other parts of the country.

3. CMAQ MODEL DEVELOPMENT AND EVALUATION

3.1 Mechanisms description

3.1.1 Gas phase photochemical mechanism

Reactions with oxidants in the troposphere are significant loss pathways of PAHs. Those oxidants include OH, NO_3 and O_3 [38].

PAH-OH reaction rates have been reported in various publications. The second order reaction rate coefficients ($k_{2,OH}$) used in this study are listed in Table 12. Using the global 12-hour average concentration of OH of 2×10^6 molecules cm^{-3} [39], the half-life of PAHs are estimated and also shown in Table 12.

Table 12 PAHs reaction rate coefficients with OH radicals

Species	$k_{2,OH}$ ($cm^3 molecules^{-1} s^{-1}$)	Half-life (hr)	Ref.
NAPH	2.16E-11*	4.5	Atkinson [40]
ACY	1.10E-10	0.9	Atkinson and Aschmann [41]
ACE	7.33E-11	1.3	Reisen and Arey [42]
			Brubaker and Hites [43]
			Atkinson and Aschmann [41]
			Klopffer et al. [44]
			Banceu et al. [45]
			Klamt [46]
FLU	1.40E-11	6.9	Kwok et al. [47]
			Brubaker and Hites [43]
			Klopffer et al. [44]

Table 12 Continued

Species	$k_{2,OH}$ ($cm^3 molecules^{-1} s^{-1}$)	Half-life (hr)	Ref.
PHE	2.74E-11	3.5	Biermann et al. [48]
			Atkinson [49]
			Kwok et al [47]
			Brubaker and Hites [43]
			Lee et al. [50]
ANT	1.30E-10	0.7	Atkinson [49]
			Biermann et al. [48]
FTH	1.10E-11	8.8	Brubaker and Hites [43]
PYR	5.00E-11	1.9	Atkinson et al. [51]
BaA	5.00E-11	1.9	
CHRY	5.00E-11	1.9	
BbF	1.86E-11	5.2	
BkF	5.36E-11	1.8	
BaP	3.85E-10	0.3	
DahA	5.00E-11	1.9	
BghiP	5.00E-11	1.9	
IcdP	4.47E-10	0.2	

* is the reaction rate under 298K. NAPH reaction rate is time dependent as $k_{OH} = 1.07\times10^{-12} \exp(895/T)$

Reaction rate coefficients of PAHs with O_3 are several orders of magnitude smaller than PAH-OH reactions, however, they cannot be neglected due to high concentration of ozone in troposphere (6.9×10^{11} molecules cm^{-3}, as 2011 annual average [38]). The second order reaction rate coefficients ($k_{2,O3}$) and the half-life based on the 2011 annual average ozone concentration are listed in Table 13. For ACE, the reaction rate coefficient is large enough that the O_3 reaction is as important as the OH reaction.

Table 13 PAHs reaction rate coefficients with O_3

Species	$k_{1,O3}$ ($cm^3 molecules^{-1} s^{-1}$)	Half-life (hr)	Ref.
NAPH	2.5E-19	1113.0	Atkinson et al. [52, 53]
ACY	5.5E-16	0.5	Atkinson and Aschmann [54]
ACE	5.0E-19	558.1	Atkinson and Aschmann [54]
PHE	4.0E-19	697.6	Kwok et al. [47]

NO_3 can react with PAH molecules by breaking C = C like OH radicals. However, NO_3 reaction rates with PAHs are generally slow. For example, the second reaction rate coefficient of naphthalene is 3.3×10^{-28} cm^3 $molecules^{-1}$ s^{-1} [54]. According to global 12-hour average concentration of NO_3, which is 5×10^8 molecules cm^{-3} [39], the pseudo-first order reaction rate coefficient is only 1.65×10^{-19} s^{-1}, which is 10^{12} times less than that of ozone, and 10^{13} times less than that of OH. Thus, considering the slow reactions of PAHs with NO_3, these reactions are not included in the current study.

In this study, the gas phase SAPRC99 photochemical mechanism [55] is modified to include gas phase reactions of PAH species with OH and ozone, using the reaction rate coefficients listed in Table 12 and Table 13. Only the NAPH + OH reaction is treated in detail, following that of Zhang et al. [56]. For other species, they are treated as decay reactions without reactive reaction products. As the concentrations of the PAHs are low, this simplified treatment does not expect to significantly change the OH budget and the atmospheric oxidation capacity in general. The gas phase PAH species reactions with OH and ozone are listed in the Appendix Table S3.

3.1.2 Gas-to-particle partitioning of PAHs

Considering both adsorption of PAHs on BC and absorption into OM and assuming that gas-organic partitioning coefficients of PAHs are empirically related to the partitioning coefficient between octanol and gas phases. Lohmann and Lammel [17] derived an equation to calculate the gas-particle phase partitioning coefficient, K_p, as shown in equation (1):

$$K_p(m^3/\mu g) = 10^{-12}\left(f_{OM}\frac{MW_{oct}\,\gamma_{oct}}{MW_{OM}\,\gamma_{OM}\,\rho_{oct}}K_{oa} + f_{BC}\frac{a_{atm-BC}}{a_{soot}\ \rho_{BC}}K_{soot-air}\right) \qquad (1)$$

where f_{OM} and f_{BC} are the fractions of OM and BC in the fine particles, respectively. K_{oa} is the partitioning coefficient between octanol and gas phase. $K_{soot-air}$ is the partitioning coefficient between air and soot. MW_{oct} is the molecular weight of octanol, g/mol; and MW_{OM} is organic matter molecular weight, g/mol. γ_{oct} and γ_{OM} are activities of the PAH in octanol and OM, respectively. ρ_{oct} and ρ_{BC} are the density of octanol and BC, respectively, g/cm^3. The octanol density is taken as 0.824 g/cm^3 and ρ_{BC} is assumed to be 2.2 g/cm^3, which is consistent with the value used in the CMAQ model. a_{atm-BC} and a_{soot} are specific surface area of atmospheric black carbons and diesel soot, respectively, m^2/m^3.

In this study, the PAHs are assumed to exist solely in the fine mode aerosol in the CMAQ model (i.e. the J mode), thus $f_{OM} = m_{OM}/PMJ$, and $f_{BC} = m_{BC}/PMJ$, where m_{OM} and m_{BC} are mass concentration of OM and BC in the fine mode, respectively. PMJ is the mass concentration of the fine mode aerosol. A more detailed study that allows PAHs to partitioning into all three modes in the CMAQ model (ultrafine, fine and coarse) shows that the amount of PAH in the ultrafine and coarse modes are several orders of magnitude lower, thus the current treatment is not expected to introduce significant errors in the PAH partitioning predictions. With assumption $\gamma_{oct}/\gamma_{OM} = 1$, $MW_{oct}/MW_{OM} = 1$ [12], and $a_{atm-BC}/a_{soot} = 1$, equation (1) can be simplified to equation (2).

$$K_p = 10^{-12}\left(\frac{m_{OM}K_{OA}}{\rho_{oct}} + \frac{m_{BC}K_{soot-air}}{\rho_{BC}}\right)PMJ^{-1} \tag{2}$$

Octanol-air partitioning coefficient varies with different atmospheric temperature. According to Odabasi et al [57], $\log K_{OA}$ can be estimated with temperature dependence linear regression, as shown in equation (3).

$$log\,K_{OA} = A + B/T \tag{3}$$

where the intercepts (A) and slopes (B) are derived from regression of measured K_{OA} under different temperatures. A and B values for 14 of the 16 PAH species (except for naphthalene and pyrene) are following Odabasi et al [57] recommendation. For naphthalene and pyrene, K_{OA} under different temperatures are obtained from a fragment constant method described by literature [58] to determine the A and B values. The A and

B values for the 16 PAH species are shown in Table 14.

Table 14 K_{OA} Regression Intercepts (A) and slopes (B)

Name	B	A
NAPH	3617	-7.05
ACY	2476	-1.97
ACE	2597	-2.20
FLU	2833	-2.61
PHE	3293	-3.37
ANT	3316	-3.41
FTH	3904	-4.34
PYR	5010	-7.87
BaA	4746	-5.64
CHRY	4754	-5.65
BbF	5285	-6.40
BkF	5301	-6.42
BaP	5382	-6.50
DahA	5887	-7.17
BghiP	5834	-7.03
IcdP	5791	-7.00

As suggested by Dachs et al. [59] and van Noort [60], $K_{soot-air}$ values for each species can be estimated from the subcooled vapor pressure (p_L^0, Pa), and BC specific area (a_{BC}, m^2/g), as shown in equation (4).

$$log\,K_{soot-air} = -0.85 log p_L^0 + 8.94 - log(\frac{998}{a_{BC}}) \quad (4)$$

In this study, diesel soot specific BET surface area based on NIST standard reference material SRM 1650, which is approximately 90 m^2/g, is used as a_{BC}. The temperature dependence of p_L^0 can be calculated by the temperature regression with intercept b_L and slope m, as shown in equation (5):

$$log\, p_L^0(Pa) = m_L/T + b_L \quad (5)$$

p_L^0 values under selected temperatures are obtained from EPI Suit software, which is developed by the US EPA (http://www.epa.gov/oppt/exposure/pubs/episuitedl.htm). The intercept b_L and the slope m_L for 16 PAHs are shown in Table 15.

Table 15 logp_L^0 Regression Intercepts b_L and Slopes m_L

Name	m_L	b_L
NAPH	-4156.5	14.669
ACY	-4748.2	15.144
ACE	-4742.5	15.138
FLU	-4979.8	15.343
PHE	-5300.2	15.534
ANT	-5834.6	16.027
FTH	-5723.1	15.812
PYR	-6098.9	16.114
BaA	-6086.6	15.970
CHRY	-6990.5	16.762
BbF	-6521.9	16.392
BkF	-7083.4	16.775
BaP	-7025.1	16.676
DahA	-7723.8	17.170
BghiP	-7426.1	17.020
IcdP	-7336.0	16.824

The gas-particle partitioning scheme described by equations (2) to (5) are implemented in the CMAQ model.

3.2 Model application

The revised model is used to predict ambient PAH concentrations in the entire continental United States in January, April, July, and October, 2011. Figure 5 shows the model domain and the locations of observation sites.

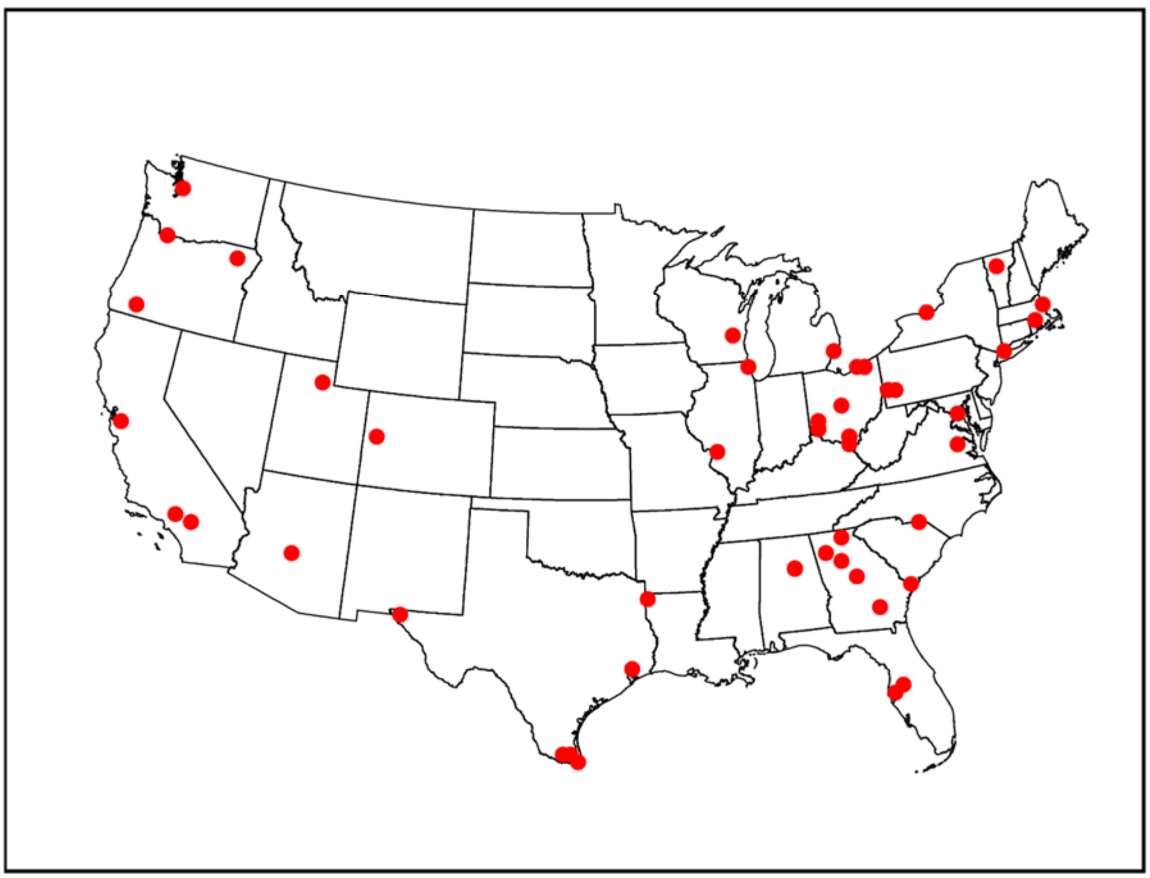


Figure 5 Observations sites locations in the domain

The meteorological inputs are generated using the Weather Research and Forecasting (WRF) model version 3.6. The simulations are initialized using the North American Regional Reanalysis (NARR) data (from National Oceanic and Atmospheric Administration (NOAA), www.esrl.noaa.gov/psd/data/gridded/data.narr.html) with 32-km horizontal resolution and 3-h time resolution, for all variables except soil moisture, which was initialized using predictions from the North American Land Data Assimilation System (NLDAS). Emissions are generated based on the NEI 2011, using SMOKE and the profiles updated with PAH species, as described in Section 2. Biogenic emissions are generated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) [61].

3.3 Results

3.3.1 Observation data

Observations of PAHs in the entire continental United States are downloaded from EPA's Air Toxic Website http://www.epa.gov/ttnamtil/toxdat.html#data. The 24-hr daily average concentrations are monitored every six days. Only data above the Method Detection Limite (MDL) are retained in the analysis. Overall there are 61 sites observing PAH species ambient concentrations. The details of the monitoring sites are listed in Table 16, and their locations are shown in Figure 5.

Table 16 PAH Species Observations Sites

NO.	State	Site Code	Longitude	Latitude	Setting	City or County
s1	AL	010735502	-86.8239	33.5448	Urban And Center City	Birmingham
s2	AL	010735503	-86.8029	33.5684	Urban And Center City	Birmingham
s3	AL	010735505	-86.8055	33.55663	Urban And Center City	Birmingham
s4	AL	010736004	-86.7964	33.56528	Urban And Center City	Birmingham
s5	AZ	040139997	-112.096	33.50373	Urban And Center City	Phoenix
s6	CA	060371103	-118.227	34.06659	Urban And Center City	Los Angeles
s7	CA	060658001	-117.416	33.99958	Suburban	Rubidoux
s8	CA	060850005	-121.895	37.3485	Urban And Center City	San Jose
s9	CO	080770018	-108.562	39.06425	Urban And Center City	Grand Junction
s10	DC	110010043	-77.0132	38.92185	Urban And Center City	Washington
s11	FL	120573002	-82.2304	27.96565	Rural	Valrico
s12	FL	121030026	-82.7146	27.85004	Suburban	Pinellas Park
s13	GA	130210012	-83.5435	32.80541	Rural	Macon
s14	GA	130510021	-81.0488	32.06923	Suburban	Savannah
s15	GA	130690002	-82.7501	31.51329	Rural	Coffee
s16	GA	130850001	-84.0598	34.37632	Rural	Dawson
s17	GA	130890002	-84.2903	33.68801	Suburban	DeKalb
s18	GA	132230003	-85.0453	33.9285	Rural	Paulding
s19	IL	170314201	-87.7992	42.14	Suburban	Northbrook
s20	KY	210430500	-82.9883	38.23833	Rural	Carter
s21	MA	250250042	-71.0825	42.32944	Urban And Center City	Suffolk
s22	MI	261630033	-83.1496	42.30754	Suburban	Dearborn
s23	MO	295100085	-90.1987	38.65644	Urban And Center City	St. Louis
s24	NY	360050080	-73.9201	40.83606	Urban And Center City	New York
s25	NY	360551007	-77.5481	43.1462	Urban And Center City	Rochester
s26	OH	390170003	-84.3543	39.4938	Urban And Center City	Middletown
s27	OH	390350038	-81.6824	41.47701	Urban And Center City	Cleveland
s28	OH	390350069	-81.6378	41.519	Urban And Center City	Cleveland
s29	OH	390351002	-81.8187	41.39629	Suburban	Brook Park

Table 16 Continued

NO.	State	Site Code	Longitude	Latitude	Setting	City or County
s30	OH	390490034	-82.9944	40.00274	Urban And Center City	Columbus
s31	OH	390610014	-84.479	39.19433	Suburban	Cincinnati
s32	OH	390610042	-84.5512	39.10492	Urban And Center City	Cincinnati
s33	OH	390610044	-84.7116	39.13837	Suburban	Addyston
s34	OH	390610045	-84.5187	39.17093	Urban And Center City	Cincinnati
s35	OH	390810017	-80.6156	40.36644	Urban And Center City	Steubenville
s36	OH	391450020	-82.8225	38.60934	Rural	Scioto
s37	OH	391450021	-82.8296	38.60066	Rural	Franklin Furnace
s38	OH	391450022	-82.8348	38.58808	Rural	Franklin Furnace
s39	OH	391555504	-80.8127	41.2351	Urban And Center City	Warren
s40	OR	410290133	-122.8792	42.3141	Urban And Center City	Medford
s41	OR	410292129	-122.88	42.33155	Rural	Franklin Furnace
s42	OR	410350004	-121.7314	42.1903	Suburban	Altamont
s43	OR	410390060	-123.0837	44.0263	Urban And Center City	Eugene
s44	OR	410390062	-123.1615	44.0729	Urban And Center City	Eugene
s45	OR	410510246	-122.679	45.5613	Urban And Center City	Portland
s46	PA	420030064	-79.8681	40.32377	Urban And Center City	Liberty
s47	RI	440070022	-71.415	41.80795	Urban And Center City	Providence
s48	SC	450250001	-80.1988	34.61537	Rural	Chesterfield
s49	TX	480610006	-97.4938	25.8925	Urban And Center City	Brownsville
s50	TX	481410053	-106.501	31.75853	Urban And Center City	El Paso
s51	TX	482011039	-95.12849	29.67005	Urban And Center City	Deer Park
s52	TX	482030002	-94.16744	32.669	Rural	Harrison
s53	TX	482150043	-98.29107	26.22623	Suburban	Mission
s54	TX	482151048	-97.93726	26.13108	Urban And Center City	Mercedes
s55	TX	484790016	-99.5203	27.5113	Suburban	Laredo
s56	TX	490110004	-111.8845	40.90297	Suburban	Bountiful
s57	TX	500070007	-72.86884	44.52839	Rural	Underhill

Table 16 Continued

NO.	State	Site Code	Longitude	Latitude	Setting	City or County
s58	UT	510870014	-77.40041	37.55655	Suburban	East Highland Park
s59	VT	530330080	-122.3081	47.56833	Urban And Center City	Seattle
s60	VA	540095501	-80.59532	40.33564	Urban And Center City	Follansbee
s61	WA	550270001	-88.62111	43.46611	Rural	Horicon

As the measured concentrations of each species are the sum of the gas and particle phases concentrations, the CMAQ model predictions of gas and particle phase concentrations are also combined using equation (6):

$$C_T = \frac{MW \cdot C_G \cdot P}{RT} + C_P \tag{6}$$

where C_T is total concentration of PAH species, in μg/m^3; C_P is particle phase concentration of PAH species, in μg/m^3; C_G is gas phase concentration of PAH species, in ppmv. MW is the molecular weight of the PAH species, in g/mol; P is the atmospheric pressure, in Pa; R is the ideal gas law constant, which is 8.314 m^3 Pa K^{-1}mol^{-1}; T is atmospheric temperature, in K. Atmospheric pressure and temperature are obtained from the meteorological fields generated by WRF. Predicted hourly concentrations are averaged to compare with daily observations.

3.3.2 Model Performance Evaluation

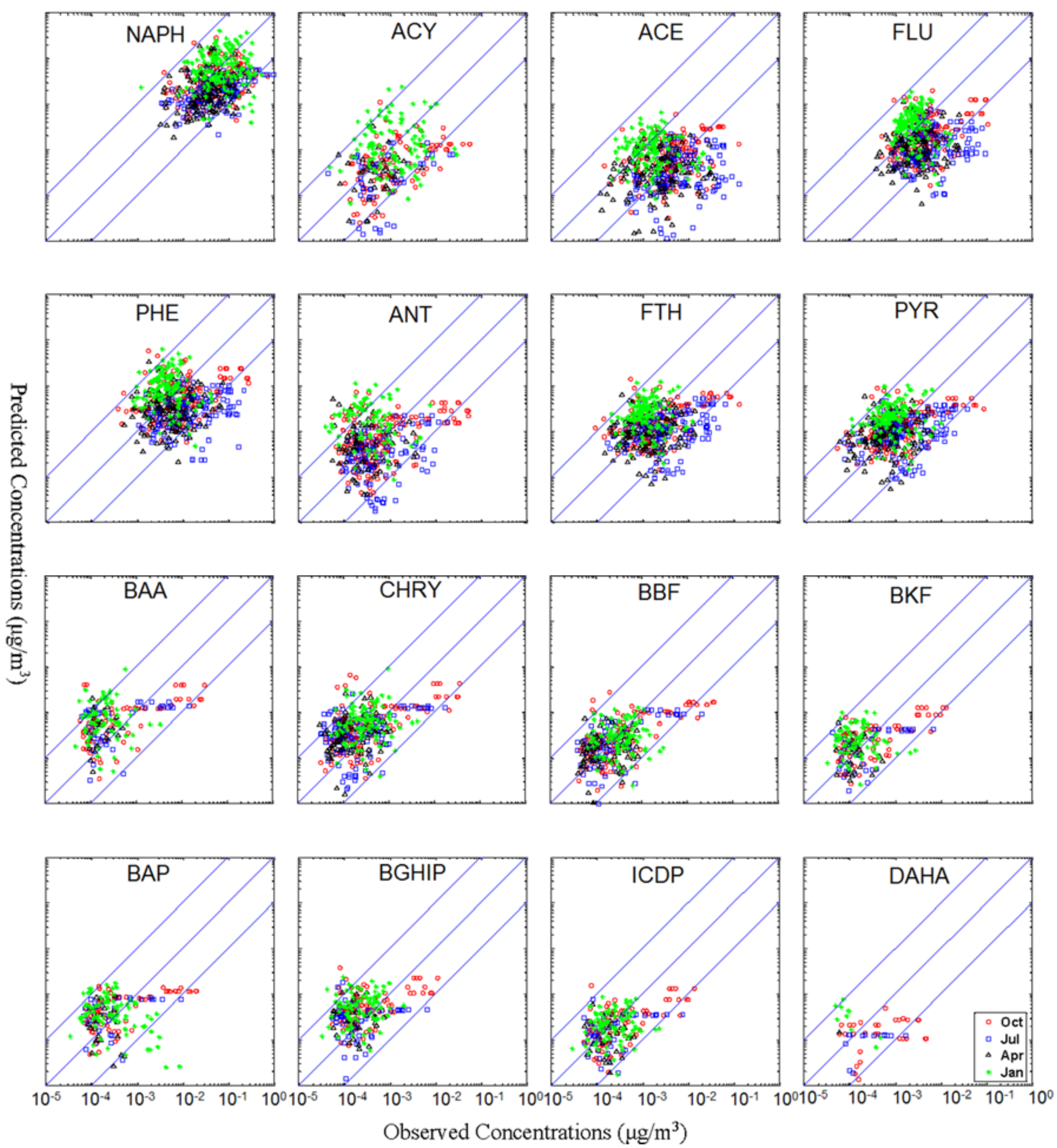


Figure 6 Observed and predicted daily PAHs concentrations for four months at all available sites. The three lines represent 10:1, 1:1, and 1:10 ratio, respectively

To evaluate model performance, predicted concentrations are compared to all available daily average observations first, as shown in Figure 6.

In the plots, each point represents the comparison of daily value at a sampling site. The overall bias for most species appears to be small, and most predictions are generally within the same order of magnitude of the observed concentrations. Predictions of some small molecular weight PAH species, such as ACE, FLU, PHE, ANT and FTH, are lower than observations in summer. Some species, such as PHE, ANT, PYR, BaA, and CHRY, are over-predicted in winter. For most species, the highest concentrations are under-predicted. For sites that are influenced by local emissions, this under-prediction might be caused by the relatively coarse grid resolution used in this study.

The mean fractional bias (MFB) and mean fractional error (MFE) [62] are calculated using equation (7) and (8):

$$MFB = \frac{2}{N}\sum_{i=1}^{N}\frac{(C_{m,i} - C_{o,i})}{(C_{m,i} + C_{o,i})} \quad (7)$$

$$MFE = \frac{2}{N}\sum_{i=1}^{N}\frac{|C_{m,i} - C_{o,i}|}{(C_{m,i} + C_{o,i})} \quad (8)$$

In the above equations, N is the number of daily average concentrations for each PAH. C_m and C_o are predicted and observed daily average concentrations respectively. MFB ranges from -2 to 2, while MFE ranges from 0 to 2. There is no established performance criteria for predicted ambient PAH concentrations so far. Because of the low concentrations, wide concentration ranges, and large uncertainty in emission estimations, we propose that acceptable criteria for MFB is from -1.3 to 1.3, and acceptable criteria for MFE is from 0 to 1.3, which indicate that predicted concentrations are from 0.2 to 5 times of the observed concentrations. The statistical analysis of model performance in four months is shown in Table 17.

Table 17 Statistical analysis of model performance for daily average PAH concentrations. The unit of average observation (Ave_obs) and perdictions (Ave_Mod) is µg/m^3.

Species	January				April			
	Ave_Obs	Ave_Mod	MFB	MFE	Ave_Obs	Ave_Mod	MFB	MFE
NAPH	1.85E-01	8.64E-02	-0.24	0.88	1.46E-01	3.02E-02	-0.63	0.96
ACY	2.49E-03	2.64E-03	0.07	1.07	7.41E-04	4.53E-04	-0.47	0.92
ACE	2.89E-03	1.32E-03	-0.31	0.80	4.52E-03	4.89E-04	-1.20	1.27
FLU	3.34E-03	4.47E-03	0.25	0.82	4.25E-03	1.66E-03	-0.68	0.88
PHE	6.27E-03	1.25E-02	0.58	1.00	9.01E-03	4.07E-03	-0.50	0.92
ANT	6.41E-04	2.34E-03	0.93	1.24	5.37E-04	7.21E-04	0.27	1.00
FTH	1.60E-03	2.97E-03	0.57	0.92	2.25E-03	1.17E-03	-0.31	0.85
PYR	1.23E-03	2.57E-03	0.68	1.01	1.12E-03	1.05E-03	0.11	0.82
BaA	3.15E-04	1.08E-03	0.91	1.18	1.91E-04	5.51E-04	0.70	0.80
CHRY	4.62E-04	8.38E-04	0.50	0.86	2.32E-04	4.30E-04	0.52	0.89
BbF	4.74E-04	3.51E-04	-0.16	0.70	2.37E-04	1.90E-04	-0.07	0.76
BkF	2.35E-04	3.09E-04	0.46	0.89	1.50E-04	1.60E-04	-0.06	0.68
BaP	6.11E-04	4.92E-04	0.27	1.01	2.09E-04	3.02E-04	0.16	0.86
BghiP	3.26E-04	6.41E-04	0.62	0.90	2.20E-04	3.64E-04	0.38	0.84
IcdP	3.15E-04	2.64E-04	-0.04	0.70	2.37E-04	1.57E-04	-0.41	0.83
DahA	1.45E-04	3.68E-04	0.79	1.31	5.86E-05	1.46E-04	0.86	0.86

Table 17 Continued

Species	July				October			
	Ave_Obs	Ave_Mod	MFB	MFE	Ave_Obs	Ave_Mod	MFB	MFE
NAPH	1.93E-01	2.31E-02	-0.92	1.04	3.52E-01	4.35E-02	-0.74	1.04
ACY	2.27E-03	4.04E-04	-0.81	1.16	4.27E-03	8.14E-04	-0.73	1.03
ACE	1.38E-02	5.01E-04	-1.66	1.66	8.86E-03	9.76E-04	-1.26	1.33
FLU	1.25E-02	1.70E-03	-1.16	1.23	9.86E-03	2.89E-03	-0.76	0.98
PHE	2.87E-02	3.77E-03	-1.09	1.19	2.09E-02	7.25E-03	-0.47	0.93
ANT	1.63E-03	4.87E-04	-0.54	0.94	3.63E-03	1.11E-03	-0.03	0.93
FTH	7.48E-03	1.44E-03	-0.81	1.03	6.71E-03	2.13E-03	-0.30	0.85
PYR	3.67E-03	1.31E-03	-0.34	0.90	4.35E-03	1.65E-03	-0.05	0.78
BaA	1.36E-03	8.01E-04	0.36	0.99	3.20E-03	1.05E-03	0.24	1.01
CHRY	8.35E-04	5.04E-04	0.27	0.98	2.20E-03	8.44E-04	0.31	0.90
BbF	1.17E-03	3.34E-04	-0.18	0.80	2.34E-03	4.65E-04	-0.24	0.79
BkF	6.92E-04	2.23E-04	-0.40	0.91	1.24E-03	3.57E-04	-0.17	0.86
BaP	9.17E-04	4.75E-04	0.07	0.86	2.02E-03	6.12E-04	0.08	0.96
BghiP	5.01E-04	3.66E-04	0.19	0.94	9.72E-04	7.10E-04	0.42	0.89
IcdP	6.07E-04	1.98E-04	-0.34	0.82	1.20E-03	3.28E-04	-0.30	0.81
DahA	4.37E-04	1.07E-04	-0.89	0.97	1.03E-03	1.71E-04	-0.84	1.12

In January, concentrations of some PAH species are slightly over-predicted, but all MFB and MFE values are within the suggested performance criteria. A lot of species are predicted well with MFB between -0.67 and 0.67 (12 out of 16). For over half of the 16 species, MFE is generally less than 1.0. The model performance of MFB in April is good as January with 12 species MFB within -0.67 and 0.67. Under-prediction of some species is quite obvious in July, such as NAPH, ACY, ACE, FLU, and PHE, especially for ACE. These species with large errors are predominantly partitioned into the gas phase, and higher temperature in summer time leads to higher fractions of these species in the gas phase. The negative bias in predicted concentrations could be caused by over-estimating their gas phase reaction rate coefficients, over-estimation of the gas phase fraction due to uncertainty in the partitioning coefficient, or under-estimating the emission of these species in July. In October simulation, most species are predicted well in terms of MFB (11 out of 16 within -0.67 to 0.67). MFE values are approximately 1.0.

As the health effects due to exposure to ambient PAHs are generally chronic, the ability of the model in predicting long-term averages needs to be evaluated as well. The predicted monthly average concentrations are calculated using all the hourly predictions within each month. The observed concentrations during each month are averaged to present average concentration for the month. Only stations with at least four valid observations are used in the model performance evaluation, as it is generally requested that 75% of all the available observations should be available to calculate the monthly average. Comparison of predicted vs. observed monthly average concentrations is shown in Figure 7.

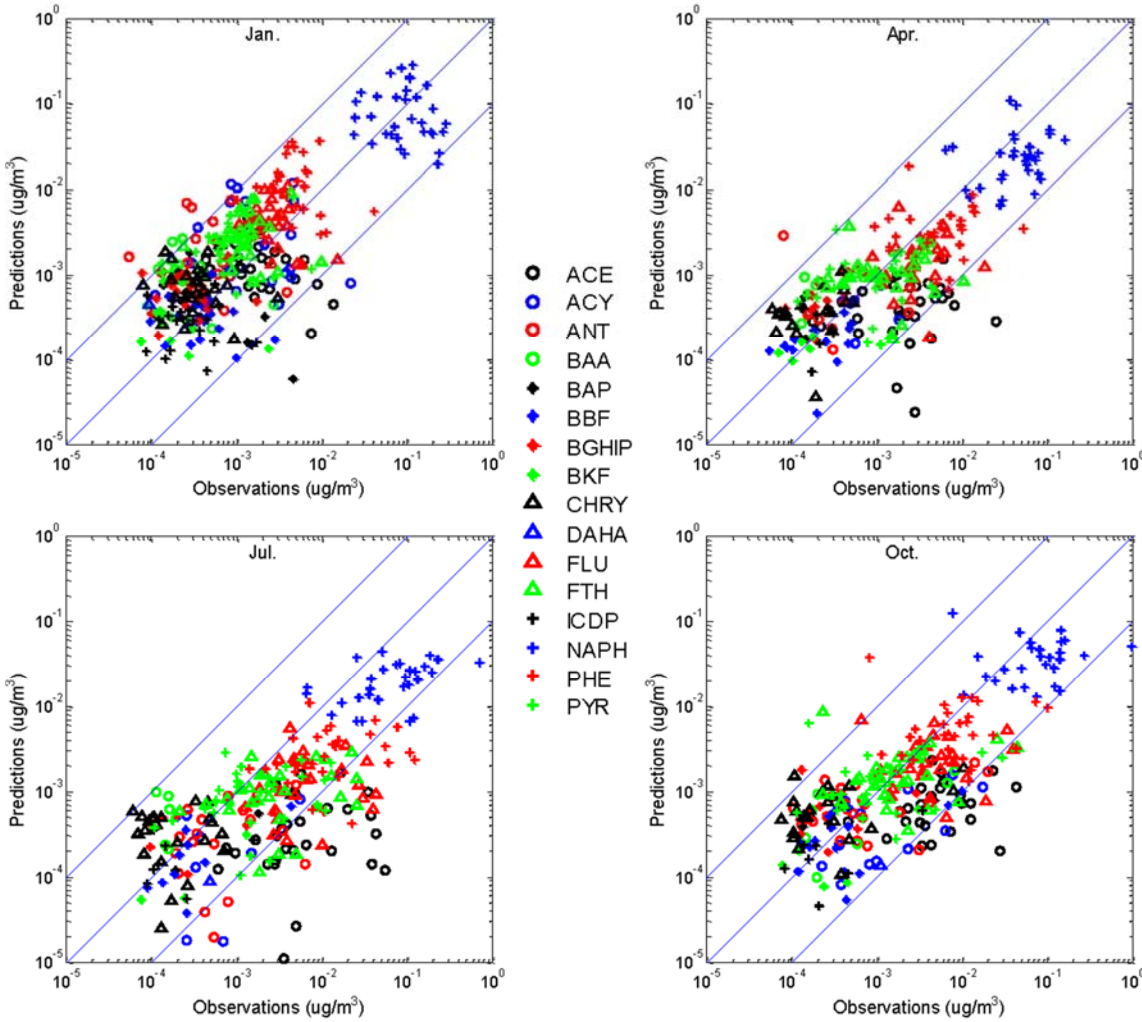


Figure 7 Observed and predicted monthly average PAH concentrations at all available sites. The three lines represent 10:1, 1:1, and 1:10 prediction vs observation ratio respectively

It can be seen clearly that concentrations of the 16 PAH species span almost three orders of magnitude (10^{-4} – 10^{-1} μg/m^3). The predicted concentrations also span the same orders of magnitude, and are in general agreement with the observations. As with the daily average concentrations, the largest under-prediction occurs in July, particularly for ACE.

Slight over-prediction occurs in January for some species. The simulations agree with observations better in April and October.

Table 18 Statistical analysis of model performance for monthly average PAH concentrations. The unit of the average observations (Ave_obs) and predictions (Ave_Mod) is μg/m^3.

Species	January				April			
	Ave_Obs	Ave_Mod	MFB	MFE	Ave_Obs	Ave_Mod	MFB	MFE
NAPH	1.41E-01	9.63E-02	-0.19	0.83	5.22E-02	2.83E-02	-0.55	0.82
ACY	3.17E-03	3.84E-03	0.22	1.10	8.98E-04	4.30E-04	-0.62	0.62
ACE	2.79E-03	1.49E-03	-0.25	0.79	3.66E-03	4.70E-04	-1.20	1.25
FLU	3.29E-03	4.87E-03	0.36	0.82	3.67E-03	1.63E-03	-0.62	0.81
PHE	6.55E-03	1.43E-02	0.66	0.99	8.08E-03	3.99E-03	-0.43	0.84
ANT	7.39E-04	2.60E-03	0.92	1.18	4.85E-04	7.49E-04	0.38	0.84
FTH	1.71E-03	3.31E-03	0.64	0.91	2.03E-03	1.14E-03	-0.25	0.80
PYR	1.33E-03	2.79E-03	0.75	1.00	1.02E-03	1.02E-03	0.16	0.75
BAA	4.18E-04	1.12E-03	0.95	1.24	1.53E-04	6.28E-04	1.13	1.13
CHRY	4.81E-04	8.54E-04	0.61	0.86	2.22E-04	4.21E-04	0.63	0.82
BBF	5.20E-04	3.91E-04	-0.11	0.58	2.47E-04	1.90E-04	-0.14	0.61
BKF	3.14E-04	3.45E-04	0.46	0.84	1.25E-04	1.57E-04	0.22	0.30
BAP	7.52E-04	5.46E-04	0.17	1.00	1.70E-04	3.86E-04	0.71	0.71
BGHIP	3.57E-04	6.83E-04	0.70	0.84	2.13E-04	4.22E-04	0.65	0.68
ICDP	3.38E-04	3.08E-04	0.05	0.63	2.36E-04	1.99E-04	-0.26	0.47
DAHA	9.28E-05	4.46E-04	1.31	1.31	-	-	-	-

Table 18 Continued

Species	July				October			
	Ave_Obs	Ave_Mod	MFB	MFE	Ave_Obs	Ave_Mod	MFB	MFE
NAPH	1.02E-01	2.13E-02	-0.94	1.07	2.78E-01	4.01E-02	-0.72	0.95
ACY	1.27E-03	2.85E-04	-1.01	1.20	3.34E-03	5.32E-04	-0.99	1.09
ACE	1.20E-02	4.45E-04	-1.67	1.67	8.11E-03	7.84E-04	-1.36	1.36
FLU	1.07E-02	1.49E-03	-1.20	1.22	7.33E-03	2.49E-03	-0.65	0.84
PHE	2.48E-02	3.22E-03	-1.13	1.18	1.49E-02	6.96E-03	-0.36	0.75
ANT	1.33E-03	3.69E-04	-0.70	0.87	2.07E-03	7.72E-04	0.09	0.82
FTH	6.19E-03	1.14E-03	-0.87	1.01	4.41E-03	1.93E-03	-0.14	0.69
PYR	2.91E-03	1.06E-03	-0.40	0.85	2.75E-03	1.49E-03	0.11	0.70
BAA	6.50E-04	6.56E-04	0.65	1.00	2.31E-03	7.40E-04	-0.16	1.03
CHRY	4.27E-04	3.91E-04	0.30	0.94	1.06E-03	7.03E-04	0.58	1.07
BBF	6.32E-04	2.40E-04	-0.41	0.66	1.24E-03	3.65E-04	-0.21	0.72
BKF	5.40E-04	1.42E-04	-0.92	0.92	7.83E-04	2.25E-04	-0.52	0.95
BAP	7.03E-04	3.43E-04	-0.14	0.87	1.68E-03	5.73E-04	0.20	1.15
BGHIP	3.50E-04	3.61E-04	0.36	1.00	5.04E-04	6.83E-04	0.61	0.96
ICDP	3.81E-04	1.51E-04	-0.33	0.60	5.60E-04	2.48E-04	-0.15	0.72
DAHA	4.86E-04	8.78E-05	-1.39	1.39	1.12E-03	1.36E-04	-1.56	1.56

The MFB and MFE for the monthly average concentrations are also calculated and shown in Table 18.

3.3.3 Regional distribution of PAHs

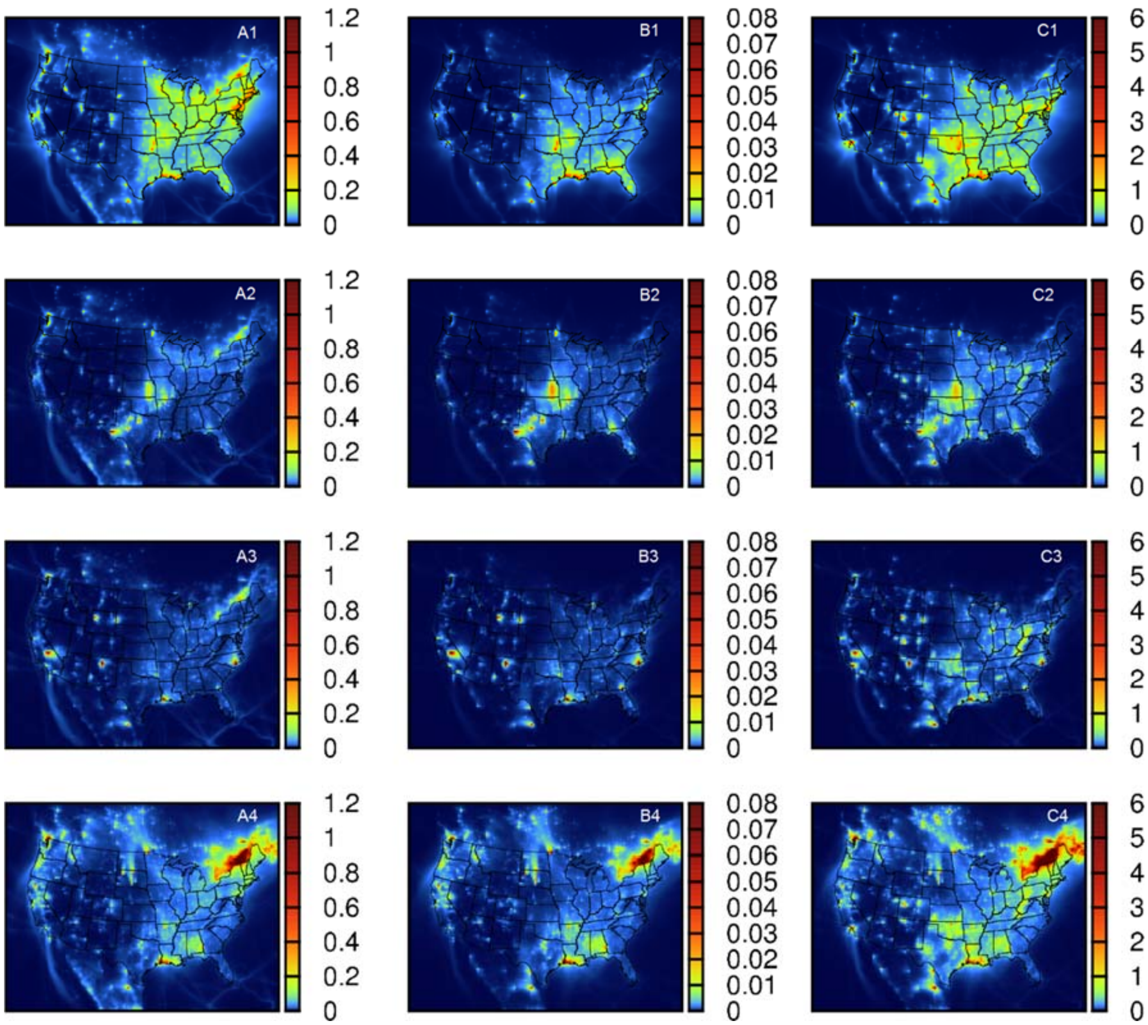


Figure 8 Monthly average surface concentrations of 16-PAH (column A), 7-PAH (column B), and BaP (column C) for January (row 1), April (row 2), July (row 3), and October (row 4). Units are μg/m^3 for 16-PAH and 7-PAH, and ng/m^3 for BaP.

Figure 8 shows the regional distribution of predicted monthly average concentrations of 16-PAH, 7-PAH, and BaP. In winter, 16-PAH concentrations in eastern US are approximately 0.2 μg/m^3, with higher concentrations exceeding 0.4 μg/m^3 at some large urban centers, such as New York, Boston, and Washington. The 7-PAH concentrations in southeastern US are higher than the northeastern part, with concentrations reaching 0.02 μg/m^3 near the coast of Louisiana. Comparatively, concentrations of 7-PAH in the northeastern are approximately 0.005 μg/m^3. Large areas in the eastern US and several west coastal regions have high BaP concentrations in January, which exceed the European Union target value of 1.0 ng/m^3 for annual average ambient BaP concentrations [63].

PAH concentrations are lower in spring (April) and summer (July). 16-PAH concentrations in most part of the eastern US are less than 0.1 μg/m^3 and 7-PAH concentrations close to 0.005 μg/m^3. For BaP concentrations, most areas in the US have concentrations lower than 1.0 ng/m^3, except for hot spots in big cities or wildfire locations. The April high concentrations in less populated Kansas is due to open burning, leading to elevated concentrations of 16-PAH (~ 0.2 μg/m^3), 7-PAH (~ 0.02 μg/m^3), and BaP(>1.0 ng/m^3). In fall (October), there are extremely high concentrations of PAHs near the US-Canada border in the northeastern US. 16-PAH concentration reaches 0.2 μg/m^3, and BaP concentrations are higher 1.0 ng/m^3 in large cities in the northeast. Concentrations in the coastal areas in Louisiana and part of Texas along the Gulf of Mexico also show high PAH emissions. 16-PAH, 7-PAH and BaP concentrations exceed 0.6 μg/m^3, 0.05 μg/m^3, and 3.0 ng/m^3, respectively. As these areas have extensive oil-gas related activities, it is expected that contributions from industrial processes are significant.

3.4 Discussions

3.4.1 Heterogeneous oxidation of particle-bond PAHs by ozone

It has been widely reported that heterogeneous oxidation of PAHs on particle surface by oxidants, such as OH [64-67], ozone [68-74], and NO_3 [75], can be important pathways that affect the life time, and thus, the ambient concentration of PAHs in atmosphere. The results shown in the previous section do not consider particle phase oxidations. Although a number of experimental studies have been carried out to understand the oxidation mechanism and quantify the reaction rate coefficients, very different results have been reported in the literature and the rates apparently vary with the substrates, making it difficult to implement them into a regional chemical transport model. In this section, sensitivity simulations are conducted to evaluate the importance of the heterogeneous oxidation due to ozone in the prediction of PAH concentrations. The heterogeneous oxidation of PAH in this study is treated as second order reactions. The reaction rate coefficients of O_3 with PAHs have been reviewed by Perraudin et al [74]. Table 19 shows the reaction rate coefficients ($k_{2(het),O3}$) used in this study. The half-lives of the PAHs due to an average concentration of ozone of 6.9×10^{11} molecules/cm^3 are also shown in Table 19.

Table 19 Heterogeneous-reaction of particulate PAHs with O_3

Species	$k_{2\,(het),O3}$ (cm^3 molecules^{-1} s^{-1})	Half-life (hr)
NAPH	9.0E-19	310.1
PHE	2.35E-17	11.9

Table 19 Continued

Species	$k_{2\ (het),O3}$ (cm^3 molecules^{-1} s^{-1})	Half-life (hr)
ANT	1.2E-16	2.3
FTH	1.7E-17	16.5
PYR	5.9E-17	4.7
BaA	5.75E-17	4.9
CHRY	2.3E-17	12.1
BkF	2.75E-17	10.1
BaP	9.7E-17	2.9
IcdP	2.8E-17	10.0

After modifications that considering heterogeneous reaction with O_3, there is no obvious change in NAPH, because the concentration in particle phase is low and the extremely long half-life of NAPH in the particle phase. As shown in Figure 9, most of the other 9 species have shown decreased concentrations. As expected, the species with fastest O_3 reaction rates (ANT, BaP, PYR, and BaA) show the most significant decrease. As these species are over-predicted in the original simulation, including surface heterogeneous reactions improves the model performance in both MFB and MFE as shown in Table 20. MFB decreases by approximately 18% on average, and MFE decreases by 7%. MFB of NAPH and PHE has changed little. After modification, BaP concentration in January is predicted well with an MFB of -15% for daily average concentrations.

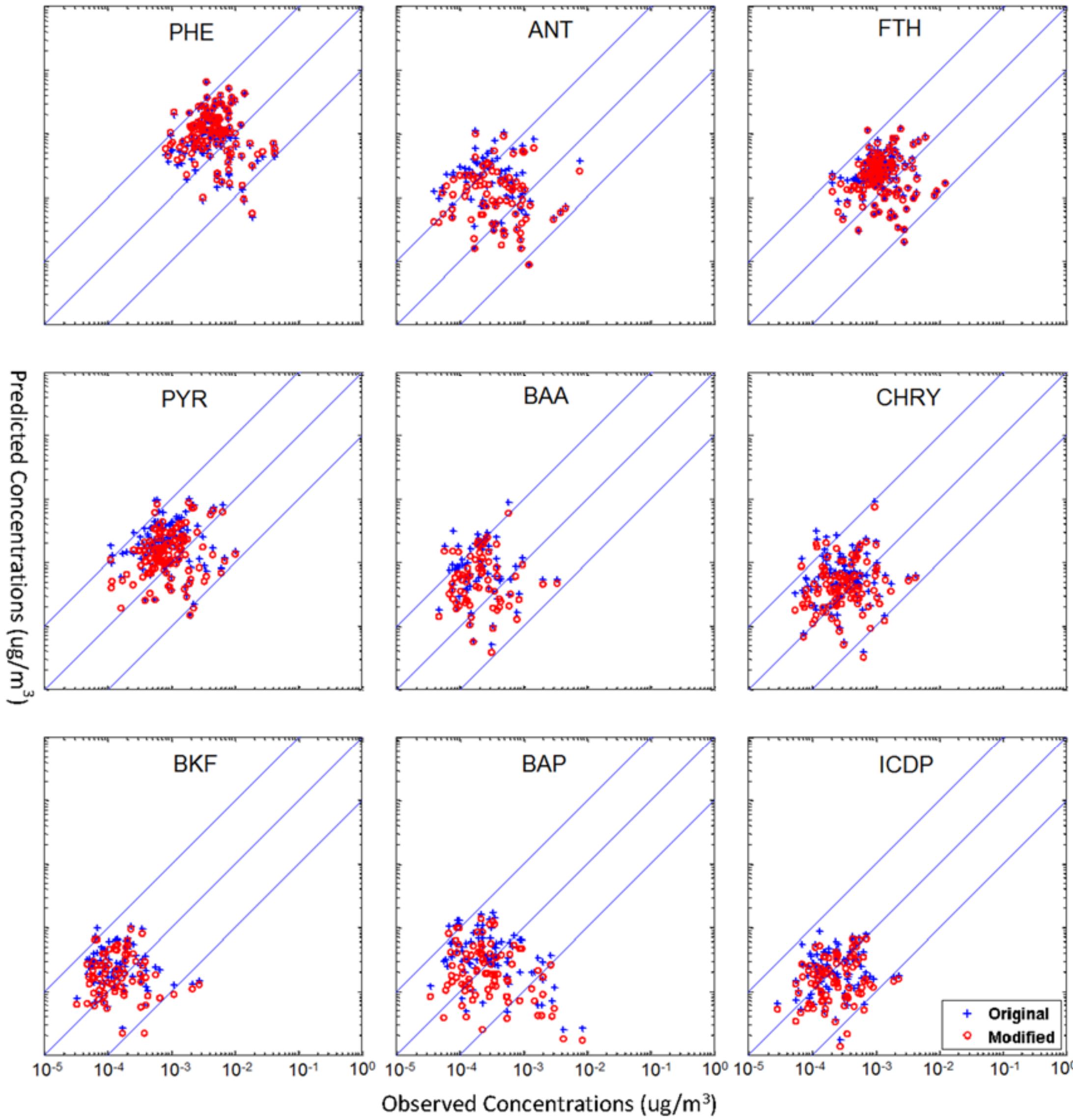
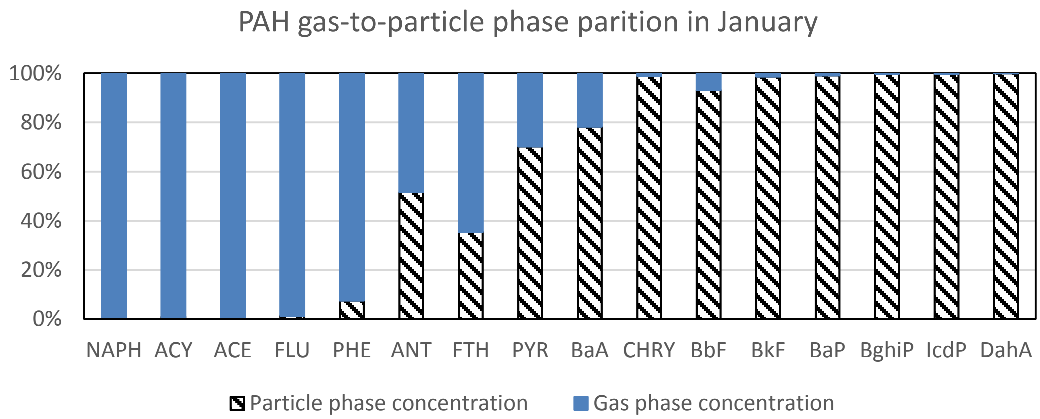


Figure 9 Observed and predicted PAH concentrations in January 2011 with (Modified) and without (Original) heterogeneous reactions of PAHs with ozone

Table 20 Statistical analysis of model performance in January when heterogeneous reactions of O_3 are included.

Species	Original		Modified	
	MFB	MFE	MFB	MFE
NAPH	-0.24	0.88	-0.23	0.88
PHE	0.58	1.00	0.65	1.04
ANT	0.93	1.24	0.79	1.17
FTH	0.57	0.92	0.49	0.85
PYR	0.68	1.01	0.42	0.80
BaA	0.91	1.18	0.69	1.08
CHRY	0.50	0.86	0.25	0.75
BkF	0.46	0.89	0.22	0.78
BaP	0.27	1.01	-0.15	0.94
IcdP	-0.04	0.70	-0.30	0.70

The heterogeneous reactions of PAH with ozone also influenced on predicted concentrations in July. There is no obvious change of species including NAPH, ANT, FTH, PYR, and BaA (less than 3%). As shown in Figure 10, compared to PAH partition in January, ANT, FTH, PYR, and BaA in particle phase are reduced significantly over the entire domain. With lower fraction of the species in the particles, the impact of including the heterogeneous reactions become less significant. The predicted concentrations of four other species have been decreased slightly, but MFB is still within the range of ±0.67.



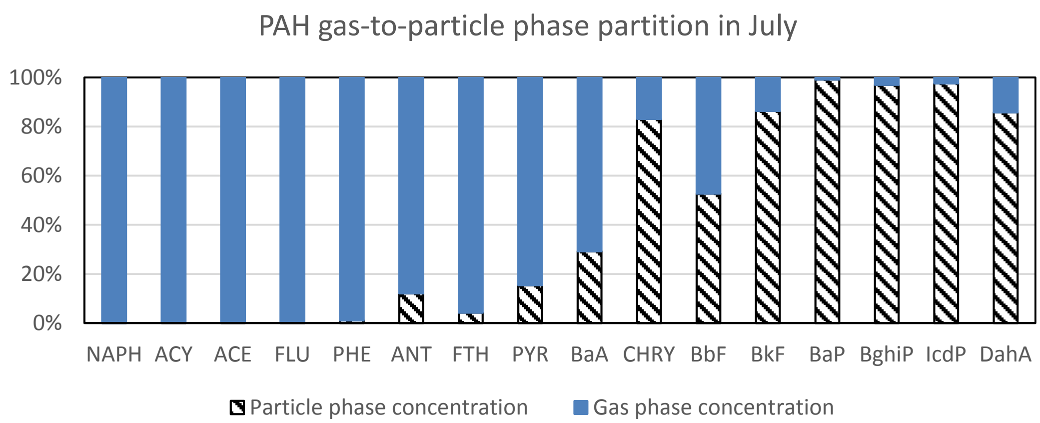


Figure 10 Domain averaged PAH fractional concentration in gas and particle phases in January and July

3.4.2 Uncertainty in emission factors and reaction rate coefficients

The uncertainties of PAHs ambient concentrations are expected to be significant due to uncertainties in emissions estimation, reaction mechanisms, and other model input data, such as meteorological fields. The large diversity in fuels and combustion conditions leads to large uncertainties in emission factors even for the same source. Table 21 shows the ranges of emission factors of 16 PAHs in residential wood combustion, coal burning, and gasoline vehicles based on a literature review.

Table 21 PAH emission factors (EF) for wood burning, coal burning and gasoline vehicles

Species	Wood combustion EF (mg/kg wood) [7]		Coal burning EF (mg/kg coal) [76]	Gasoline EF (μg/km) [9]	
Phase	Gas	Particle	Total	Gas	Particle
NAPH	227		0-142	1000-50000	
ACY	9.99-18.6			37.0-2180	
ACE	0.893-2.02			6.55-177	
FLU	2.61-4.44			9.72-358	20.1
PHE	8.14-15.7	0.07-0.67	31-239	21.7-622	434
ANT	1.76-3.44	0.0061-0.23	0-105	3.69-148	106
FTH	3.05-3.75	0.51-3.95	40-614	4.25-160	0.069-152
PYR	1.87-2.70	0.58-3.78	7-561	4.28-160	0.077-217
BaA	0-0.032	0.53-1.22	0-70	0.181-4.80	0.097-51.9
CHRY	0-0.027	0.59-1.14		0.451-5.07	0.206-52.1
BbF		0.33-0.79	0-482		0-37.3
BkF		0.29-0.67	0-273		0.083-32.7
BaP		0.24-0.71	0-194		0.021-41.0
BghiP	0.007-0.082	0.35-0.84	0-291		
IcdP		0.168-0.518	0-158		0.436-92.0

For example, emission factors of BaP in the gasoline vehicle exhaust particles can vary by thousands of times, as is true for many other PAH species. This large uncertainty in emission estimation can lead to significant errors in the simulated concentration and source contribution. In addition to emissions, parameters in the reaction kinetics and partitioning of PAHs could also lead to uncertainties in the predicted concentrations. Table 22 shows the reaction rate coefficients of PAHs with OH and O_3 used in this study and summarized experimental data in the literature [38]. (K_{OA} and p_L^0 have minor uncertainties of 3.5-5 % and 1.6-11.8 % respectively, as reported in [57])

Table 22 Reaction rate coefficients of PAHs used in this study and in other publications

Species	k_{OH} (cm^{-3} $molecules^{-1}$ s^{-1})		k_{O3} (cm^{-3} $molecules^{-1}$ s^{-1})	
	This Study	Other Publications	This Study	Other Publications
NAPH	1.07E-12	1.9E-11 – 2.7E-11	2.5E-19	2.0E-19 – 3.0E-19
ACY	1.10E-11	1.1E-10 – 1.3E-10	5.5E-16	1.6E-16 – 5.5E-16
ACE	1.03E-11	5.8E-11 – 1.0E-10	5.0E-19	<5.0E-19
FLU	1.40E-11	9.9E-12 – 1.6E-11		
PHE	1.27E-11	1.3E-11 – 3.4E-11	4.0E-19	4.0E-19
ANT	1.30E-10	1.3E-11 – 2.0E-10		
FTH	1.10E-11	1.1E-11		
PYR	5.00E-11	5.0E-11		

4. SOURCE APPORTIONMENT OF PAHS

4.1 Methodology

As shown in the previous section, concentrations of PAHs are elevated 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 an effective emission control strategy can be formulated. In this study, contributions from nine major sectors are considered: electric generation units (egu), motor vehicles (mobile), non-road engines (nonroad), oil gas process (oilgas), residential wood combustion (rwc), railway and vessel emissions (c1c2c3), commercial nonpoint sources (nonpt), industrial point source (ptnonipm), and Canada Mexico emissions (canmex). The grouping of the original 24 NEI sectors to the nine groups is discussed in section 2.2. Source apportionment simulations are conducted for January and July, representing winter and summer conditions, respectively. As contribution of each source to PAH concentrations is generally additive, in each of the source apportionment run, PAH emission from a single emission sector is included. Emissions of other species are kept the same as the basic case simulations discussed in the previous section.

4.2 PAHs source apportionment for three large cities

Three of the top 5 of most populous cities (New York #1, Los Angeles #2, and Houston

#4) are chosen in this analysis. The three cities are chosen because they are expected to have different dominating sources. All three cities have a large vehicle population. New York, located further north than the remaining two cities, have colder winters and heavy emission from residential wood combustion. Houston, the biggest city in Texas, has large petrochemical related industries. The coordinates of the three cities are shown in Table 23.

Table 23 Location of the cities used in the source apportionment analysis

City	State	Longitude	Latitude
New York	NY	-73.9797	40.7033
Los Angeles	CA	-118.4117	34.0205
Houston	TX	-95.4013	29.8172

In the following, the monthly average concentrations of 16-PAH and BaP and sources that contribute to their ambient concentrations are discussed.

Among these three cities, New York is the one with highest ambient PAH concentrations. The monthly average 16-PAH is approximately 0.24 μg/m^3 in winter, and 0.1 μg/m^3 in summer. 16-PAH concentration in Los Angeles does not show significant seasonal variations, and is approximately 0.04 μg/m^3 in both seasons. However, 16-PAH concentration in Houston changes significantly from 0.03 μg/m^3 in summer to 0.12 μg/m^3 in winter.

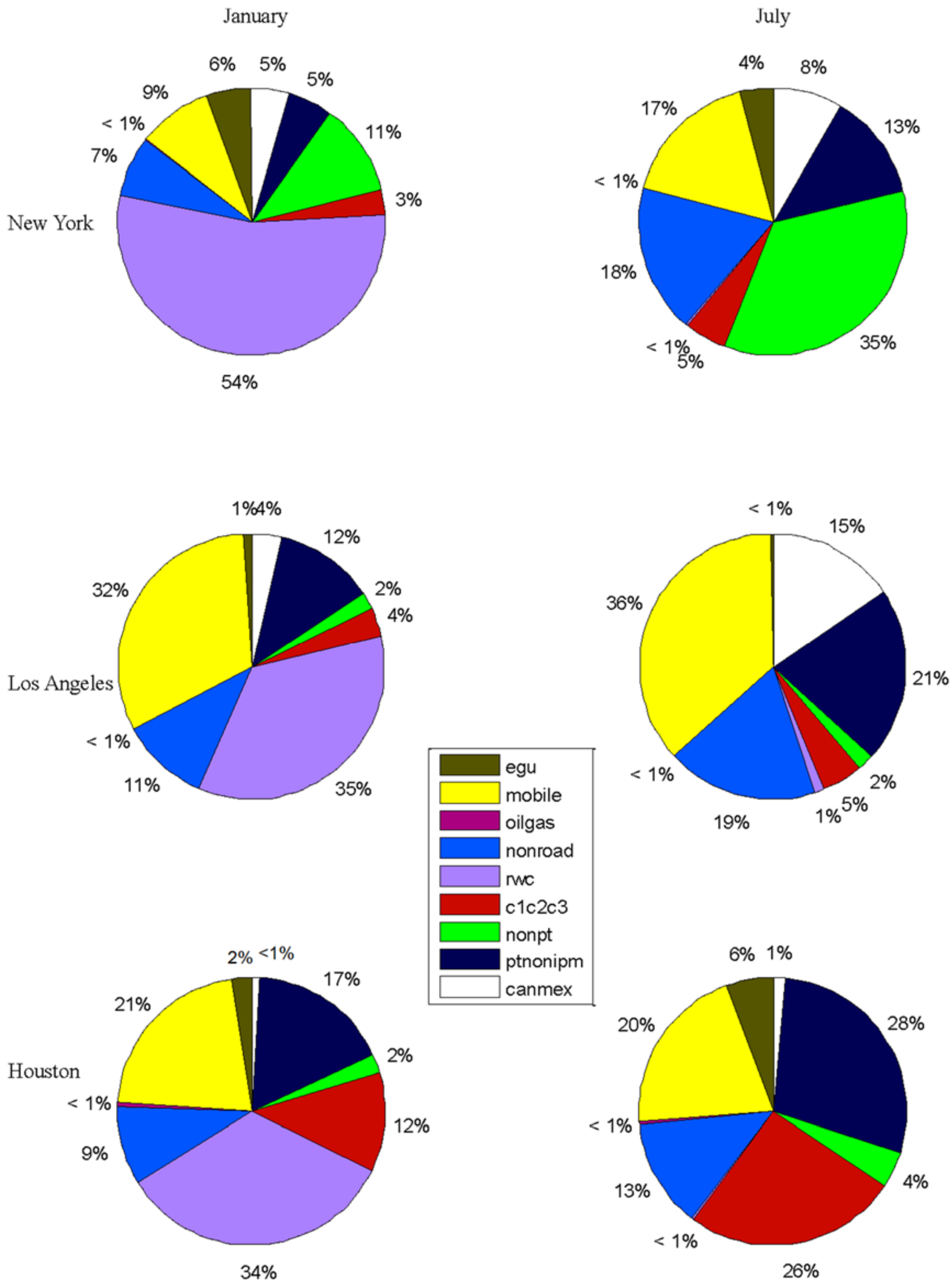


Figure 11 Source contribution to 16-PAH concentrations in New York, Los Angeles, and Houston in January and July 2011

Figure 11 shows the contribution of each source to 16-PAH ambient concentrations in the three cities. Residential wood combustion is the largest contributor in all three cities, accounting for 54% of the 16-PAH in New York, and 34%-35% in Los Angeles and Houston in the winter. Higher contributions in New York are expected as the winter there is much colder. However, in summer time residential wood combustion is less significant. The absolute contributions of residential wood combustion to 16-PAH in January are 0.148 $\mu g/m^3$, 0.0183 $\mu g/m^3$, and 0.0352 $\mu g/m^3$for New York, Los Angeles, and Houston, respectively.

Motor vehicles are always an important source of 16-PAH in the three cities in both January and July. In January, they contribute to 9%, 32%, and 21% of 16-PAH in New York, Los Angeles, and Houston, respectively, and their contributions are generally slightly higher in July. The absolute contributions of motor vehicles to 16-PAH are approximately 0.02 $\mu g/m^3$ in all three cities, which does not change much between different seasons.

The industrial point source sector is another important contributor to 16-PAH concentration in both winter and summer, ranging from 5% in January New York to 26% in July Houston. It is not surprising that its contribution in Houston is the highest as there are a lot of point emissions from industrial sources. The absolute contribution to 16-PAH due to industrial point source sector is approximately 0.0063 $\mu g/m^3$ in Los Angeles and 0.0177 $\mu g/m^3$ in Houston during winter time. The non-point sector is a large sector in New

York, and the concentration from this sector is approximately 0.03 μg/m^3. It is a significant source of 16-PAH in New York, with a relative contribution of 5% (0.015 μg/m^3) in January and 13% (0.013 μg/m^3) in July. However, it is less important in other two cities.

New York has the highest 16-PAH concentrations of 0.02 μg/m^3 from the non-road sector in both winter and summer. However, its relative contribution is higher in summer (7% in winter vs. 18% in summer) due to lower total concentration. Relative contribution of non-road sector to 16-PAH is also high in Los Angeles, accounting for 11% (0.0056 μg/m^3) in winter and 19% (0.0069 μg/m^3) in summer. Relative contribution of non-road source to 16-PAH is lower in Houston, with 9% (0.0097 μg/m^3) in winter and 13% (0.0038 μg/m^3) in summer.

All three cities are large port cities with heavy commercial marine vessels activities. Port of Houston, Port of New York, and Port of Los Angeles rank the 2nd, 3rd, and 8th in terms of the total tonnage of goods handled, respectively. The concentration of 16-PAH in Houston from that sector is more than 0.01 μg/m^3 in winter and 0.0074 μg/m^3 in summer, accounting for 12% and 26% of total 16-PAH concentrations, respectively. The contributions from commercial marine vessel activities to 16-PAH in two other cities are approximately 5%.

New York is close to the US-Canada border and Los Angeles and Houston are close to the US-Mexico border, so they might be influenced by the emission source from these two countries. Concentration of 16-PAH caused by Canadian emissions reaching in New York is 0.013 μg/m^3 (5%) in winter and 0.009 μg/m^3 (8%) in summer. Los Angeles has a significant concentration fraction impacted by Mexican emissions (4% in winter, and 15% in summer). However, Houston experiences least influence from Mexico (less than 1% in both winter and summer).

In addition to the contributions to 16-PAH concentrations, source contributions to individual PAHs are also determined. In the following, the source contributions to BaP are discussed in greater detail. BaP concentration in winter is close to 1 ng/m^3 in New York and Houston (~1.0 ng/m^3), but in summer it falls below 1 ng/m^3 in both cities (~0.81 ng/m^3 and ~0.44 ng/m^3). BaP concentration in Los Angeles is within 0.54 - 0.74 ng/m^3 in both seasons. Figure 12 shows the source apportionment of BaP for the three cities in both seasons. Contrarily to 16-PAH, contributions of residential wood combustion to BaP in winter is small, ranging from 2-3% in Los Angeles and Houston, and 8% in New York.

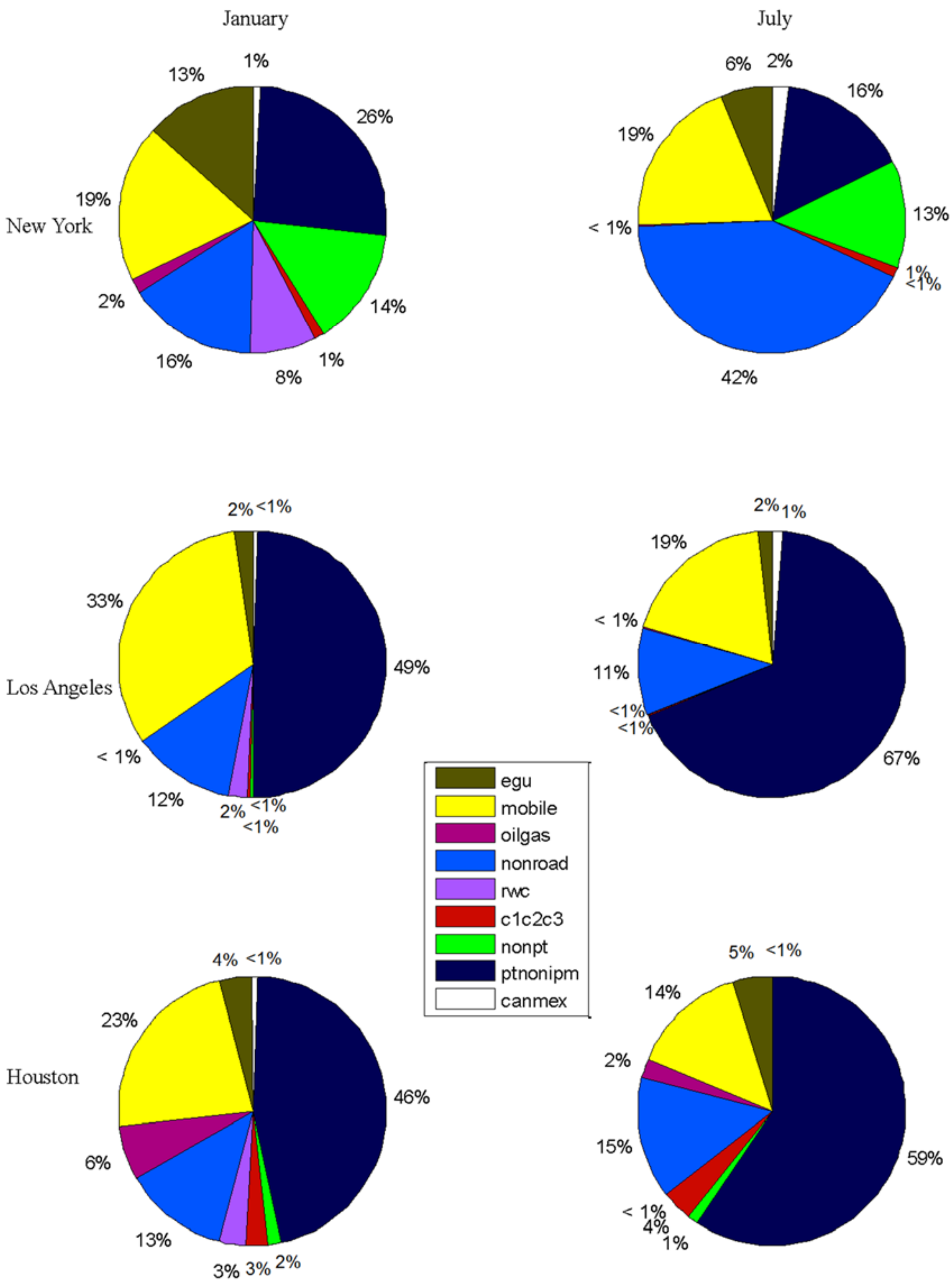


Figure 12 Source contribution to BaP concentrations in New York, Los Angeles, and Houston in January and July 2011

Industrial point source sector is the most significant contributor of BaP for Los Angeles and Houston, accounting for approximately 50% in winter, and 60%-70% in summer. Its contributions are lower in New York, accounting for 26% in winter and 16% in summer (0.31 ng/m^3 and 0.13 ng/m^3, respectively).

The non-road and mobile sectors are both important sources of BaP for the three cities. The mobile source sector accounts for 19-33% of BaP in January, and 15-20% in July. Contributions of non-road emissions are similar to those of on-road mobile sources, accounting for 12-16% in January and 11-42% in July. Contrarily to 16-PAH, the transport distance of BaP is shorter, so all three cities are less impacted by the emissions from neighboring countries. In Houston, oil and gas emissions accounts a non-negligible fraction (6% in January, and 2% in July) of BaP, although their contributions to 16-PAH are much lower in all three cities.

4.3 Conclusions

For both 16-PAH and BaP, concentrations in Los Angeles are lowest among the three cities in two seasons, while concentrations in New York are highest in winter time. Residential wood combustion makes a significant contribution to 16-PAH in winter, but little contribution to BaP. Motor vehicles and non-road engines are both important sources of 16-PAH and BaP concentrations. The industrial point source sector contributes large fractional concentrations to both 16-PAH and BaP, especially in Houston.

5. CONCLUSIONS

In this study, gridded emissions of 16 priority PAH species in the continental US for 2011 were generated using the 2011 NEI and the updated speciation profiles based on the PAH emission factors in the SPECIATE database, the L&E POM document and the MOVES database. The total 16-PAH emissions in the US are estimated to be approximately 34.8 Gg in 2011. It is 2 times higher than the estimated emissions by the US EPA, which is based on the same 2011 NEI. Residential wood combustion, industrial point/commercial nonpoint sources and mobile sources account for 39%, 31% and 14% of the total 16-PAH emissions. A modified gas phase photochemical mechanism based on SAPRC99 and the AERO5 aerosol module are implemented in the CMAQ model (v5.0.1) to simulate the emission, transport, reactions and gas-to-particle partitioning of PAHs in the entire continental US in January, April, July and October 2011. The predicted concentrations of PAHs generally agree with the observed daily average PAH concentrations at 61 air toxics monitoring sites. Concentrations of observed PAH concentrations of different species spans three orders of magnitude, which is well reproduced by the simulation. The MFB and MFE for the 16 PAHs based on daily concentrations are generally less than 0.67 and MFE less than 1.0. The MFE based on monthly average concentrations are lower, suggesting that the model can better predict the monthly PAH concentrations. Heterogeneous reactions of PAH with ozone improves the simulations of several PAH species, including BaP and ANT. Residential wood combustion, motor vehicles, industrial point sources are major contributors to PAHs concentrations.

REFERENCES

[1] Lewtas, J., *Complex-Mixtures of Air-Pollutants - Characterizing the Cancer Risk of Polycyclic Organic-Matter*. Environmental Health Perspectives, 1993. **100**: p. 211-218.

[2] IARC, *IARC Monographs on the Evaluation of Carcinogenic Risks to Humans*, in *Some Non-heterocyclic Polycyclic Aromatic Hydrocarbons and Some Related Exposures*. 2010, World Health Organization International Agency for Research on Cancer: Lyon, France.

[3] Zhang, Y.X. and S. Tao, *Global Atmospheric Emission Inventory of Polycyclic Aromatic Hydrocarbons (PAHs) for 2004.* Atmospheric Environment, 2009. **43**(4): p. 812-819.

[4] Shen, H.Z., et al., *Global Atmospheric Emissions of Polycyclic Aromatic Hydrocarbons from 1960 to 2008 and Future Predictions.* Environmental Science & Technology, 2013. **47**(12): p. 6415-6424.

[5] Kavouras, I.G., et al., *Source Apportionment of Urban Particulate Aliphatic and Polynuclear Aromatic Hydrocarbons (PAHs) Using Multivariate Methods.* Environmental Science & Technology, 2001. **35**(11): p. 2288-2294.

[6] McDonald, J.D., et al., *Fine Particle and Gaseous Emission Rates from Residential Wood Combustion.* Environmental Science & Technology, 2000. **34**(11): p. 2080-2091.

[7] Schauer, J.J., et al., *Measurement of Emissions from Air Pollution Sources. 3. C-1-C-29 Organic Compounds from Fireplace Combustion of Wood.* Environmental Science & Technology, 2001. **35**(9): p. 1716-1728.

[8] Gertler, A.W., *Diesel vs. Gasoline Emissions: Does PM from Diesel or Gasoline Vehicles Dominate in the US?* Atmospheric Environment, 2005. **39**(13): p. 2349-2355.

[9] Schauer, J.J., et al., *Measurement of Emissions from Air Pollution Sources. 5. C-1-C-32 Organic Compounds from Gasoline-powered Motor Vehicles.* Environmental Science & Technology, 2002. **36**(6): p. 1169-1180.

[10] Schauer, J.J., et al., *Measurement of Emissions from Air Pollution Sources. 2. C-1 through C-30 Organic Compounds from Medium Duty Diesel Trucks.* Environmental Science & Technology, 1999. **33**(10): p. 1578-1587.

[11] Yang, H.H., et al., *PAH Emission from Various Industrial Stacks.* Journal of Hazardous Materials, 1998. **60**(2): p. 159-174.

[12] Harner, T. and T.F. Bidleman, *Octanol-air Partition Coefficient for Describing Particle/Gas Partitioning of Aromatic Compounds in Urban Air.* Environmental Science & Technology, 1998. **32**(10): p. 1494-1502.

[13] Dvorska, A., et al., *Polycyclic Aromatic Hydrocarbons in Background Air in Central Europe - Seasonal Levels and Limitations for Source Apportionment.* Atmospheric Environment, 2012. **46**: p. 147-154.

[14] Junge, C.E., *In Fate of Pollutants in the Air and Water Environment*, Wiley, Editor. 1977: New York. p. 7-25.

[15] Pankow, J.F., *An Absorption-Model of Gas-Particle Partitioning of Organic-Compounds in the Atmosphere.* Atmospheric Environment, 1994. **28**(2): p. 185-188.

[16] Lohmann, R., *The Emergence of Black Carbon as A Super-sorbent in Environmental Chemistry: The End of Octanol?* Environmental Forensics, 2003. **4**(3): p. 161-165.

[17] Lohmann, R. and G. Lammel, *Adsorptive and Absorptive Contributions to the Gas-particle Partitioning of Polycyclic Aromatic Hydrocarbons: State of Knowledge and Recommended Parametrization for Modeling.* Environmental Science & Technology, 2004. **38**(14): p. 3793-3803.

[18] Aulinger, A., V. Matthias, and M. Quante, *Introducing A Partitioning Mechanism for PAHs into the Community Multiscale Air Quality Modeling System and Its Application to Simulating the Transport of Benzo(a) pyrene over Europe.* Journal of Applied Meteorology and Climatology, 2007. **46**(11): p. 1718-1730.

[19] Bieser, J., et al., *Impact of Emission Reductions between 1980 and 2020 on Atmospheric Benzo[a]pyrene Concentrations over Europe.* Water Air and Soil Pollution, 2012. **223**(3): p. 1393-1414.

[20] Bewersdorff, I., et al., *The Effect of Temporal Resolution of PAH Emission Data on Transport and Deposition Patterns Simulated with the Community Multiscale Air Quality Modelling System (CMAQ).* Meteorologische Zeitschrift, 2009. **18**(1): p. 41-53.

[21] Friedman, C.L. and N.E. Selin, *Long-Range Atmospheric Transport of Polycyclic Aromatic Hydrocarbons: A Global 3-D Model Analysis Including Evaluation of Arctic Sources.* Environmental Science & Technology, 2012. **46**(17): p. 9501-9510.

[22] Inomata, Y., et al., *Emission and Atmospheric Transport of Particulate PAHs in Northeast Asia.* Environmental Science & Technology, 2012. **46**(9): p. 4941-4949.

[23] Thackray, C.P., et al., *Quantitative Assessment of Parametric Uncertainty in Northern Hemisphere PAH Concentrations.* Environmental Science & Technology, 2015. **49**(15): p. 9185-9193.

[24] Gao, B., et al., *Tracer-based Source Apportionment of Polycyclic Aromatic Hydrocarbons in PM2.5 in Guangzhou, Southern China, Using Positive Matrix Factorization (PMF).* Environmental Science and Pollution Research, 2013. **20**(4): p. 2398-2409.

[25] Jang, E., M.S. Alam, and R.M. Harrison, *Source Apportionment of Polycyclic Aromatic Hydrocarbons in Urban Air Using Positive Matrix Factorization and Spatial Distribution Analysis.* Atmospheric Environment, 2013. **79**: p. 271-285.

[26] Larsen, R.K. and J.E. Baker, *Source Apportionment of Polycyclic Aromatic Hydrocarbons in the Urban Atmosphere: A Comparison of Three Methods.* Environmental Science & Technology, 2003. **37**(9): p. 1873-1881.

[27] US EPA, *SMOKE v3.5.1 User's Manual*. 2013.

[28] Eyth, A., Zubrow, A., Mason, R., *Technical Support Document (TSD): Preparation of Emissions Inventories for the Version 6.1, 2011 Emissions Modeling Platform*. 2014, U.S. EPA.

[29] Mobley, J.D., et al., *SPECIATE - EPA's Database of Speciated Emission Profiles.* Air Pollution Modeling and Its Application Xix, 2008: p. 665-666.

[30] US EPA, *Locating and Estimating Air Emissions from Sources of Polycyclic Organic Matter*. 1998.

[31] US EPA, *Air Toxic Emissions from On-road Vehicles in MOVES2014*. 2014.

[32] US EPA, *Uncontrolled Emission Factor Listing for Criteria Air Pollutants*. 2001.

[33] Brenner, R., *National Emission Standards for Hazardous Air Pollutans: Revision of Source Category List for Standards Under Section 112(c)(6) and 112(k) of the Clean Air Act.* Federal Register notices, 2002. **67**(217): p. 5.

[34] Wilgers, D.J. and E.A. Horne, *Effects of Different Burn Regimes on Tallgrass Prairie Herpetofaunal Species Diversity and Community Composition in the Flint Hills, Kansas.* Journal of Herpetology, 2006. **40**(1): p. 73-84.

[35] US EPA, *The 2011 National Emission Inventory*. 2015.

[36] Cooke, M. and A.J. Dennis, *Chemical Analysis and Biological Fate: Polynuclear Aromatic Hydrocarbons*. 1981, Battelle Press, Columbus, OH.

[37] Ramdahl, T., I. Alfheim, and A. Bjørseth, *PAH Emission from Various Sources and Their Evolution over the Last Decades*, in *Mobile Source Emissions Including Policyclic Organic Species*. 1983, Springer. p. 277-297.

[38] Keyte, I.J., R.M. Harrison, and G. Lammel, *Chemical Reactivity and Long-range Transport Potential of Polycyclic Aromatic Hydrocarbons - A Review.* Chemical Society Reviews, 2013. **42**(24): p. 9333-9391.

[39] Atkinson, R. and J. Arey, *Mechanisms of the Gas-phase Reactions of Aromatic Hydrocarbons and PAHs with OH and NO3 Radicals.* Polycyclic Aromatic Compounds, 2007. **27**(1): p. 15-40.

[40] Atkinson, R., *Kinetics and Mechanisms of the Gas-Phase Reactions of the Hydroxyl Radical with Organic-Compounds under Atmospheric Conditions.* Chemical Reviews, 1986. **86**(1): p. 69-201.

[41] Atkinson, R. and S.M. Aschmann, *Kinetics of the Gas-Phase Reactions of Alkylnaphthalenes with O-3, N2o5 and Oh Radicals at 298+/-2-K.* Atmospheric Environment, 1987. **21**(11): p. 2323-2326.

[42] Reisen, F. and J. Arey, *Reactions of Hydroxyl Radicals and Ozone with Acenaphthene and Acenaphthylene.* Environmental Science & Technology, 2002. **36**(20): p. 4302-4311.

[43] Brubaker, W.W. and R.A. Hites, *OH Reaction Kinetics of Polycyclic Aromatic Hydrocarbons and Polychlorinated Dibenzo-p-dioxins and Dibenzofurans.* Journal of Physical Chemistry A, 1998. **102**(6): p. 915-921.

[44] Klopffer, W., et al., *Quantitative Presentation of Photochemical Transformation Processes in the Troposphere.* Chemiker-Zeitung, 1986. **110**(2): p. 57-61.

[45] Banceu, C.E., et al., *Reactions of Methylated Naphthalenes with Hydroxyl Radicals under Simulated Atmospheric Conditions.* Polycyclic Aromatic Compounds, 2001. **18**(4): p. 415-425.

[46] Klamt, A., *Estimation of Gas-Phase Hydroxyl Radical Rate Constants of Organic-Compounds from Molecular-Orbital Calculations.* Chemosphere, 1993. **26**(7): p. 1273-1289.

[47] Kwok, E.S.C., et al., *Reactions of Gas-Phase Phenanthrene under Simulated Atmospheric Conditions.* Environmental Science & Technology, 1994. **28**(3): p. 521-527.

[48] Biermann, H.W., et al., *Kinetics of the Gas-Phase Reactions of the Hydroxyl Radical with Naphthalene, Phenanthrene, and Anthracene.* Environmental Science & Technology, 1985. **19**(3): p. 244-248.

[49] Atkinson, R., et al., *Evaluated Kinetic and Photochemical Data for Atmospheric Chemistry .3. Iupac Subcommittee on Gas Kinetic Data Evaluation for Atmospheric Chemistry.* Journal of Physical and Chemical Reference Data, 1989. **18**(2): p. 881-1097.

[50] Lee, W., P.S. Stevens, and R.A. Hites, *Rate constants for the Gas-phase Reactions of Methylphenanthrenes with OH as A Function of Temperature.* Journal of Physical Chemistry A, 2003. **107**(34): p. 6603-6608.

[51] Atkinson, R., et al., *Kinetics and Products of the Gas-Phase Reactions of Oh Radicals and N2o5 with Naphthalene and Biphenyl.* Environmental Science & Technology, 1987. **21**(10): p. 1014-1022.

[52] Atkinson, R. and S.M. Aschmann, *Kinetics of the Reactions of Naphthalene, 2-Methylnaphthalene, and 2,3-Dimethylnaphthalene with Oh Radicals and with O-3 at 295 +/- 1 K.* International Journal of Chemical Kinetics, 1986. **18**(5): p. 569-573.

[53] Atkinson, R., et al., *Rate Constants for the Gas-Phase Reactions of Oh Radicals and O3 with Pyrrole at 295+/-1k and Atmospheric-Pressure.* Atmospheric Environment, 1984. **18**(10): p. 2105-2107.

[54] Atkinson, R. and S.M. Aschmann, *Kinetics of the Reactions of Acenaphthene and Acenaphthylene and Structurally-Related Aromatic-Compounds with Oh and No3 Radicals, N2o5 and O-3 at 296+/-2 K.* International Journal of Chemical Kinetics, 1988. **20**(7): p. 513-539.

[55] Carter, W.P.L., *The SAPRC-99 Chemical Mechanism and Updated VOC reactivity Scales*. 2003, California Air Resources Board.

[56] Zhang, H.L. and Q. Ying, *Secondary Organic Aerosol from Polycyclic Aromatic Hydrocarbons in Southeast Texas.* Atmospheric Environment, 2012. **55**: p. 279-287.

[57] Odabasi, M., E. Cetin, and A. Sofuoglu, *Determination of Octanol-air Partition Coefficients and Supercooled Liquid Vapor Pressures of PAHs as A Function of Temperature: Application to Gas-particle Partitioning in An Urban Atmosphere.* Atmospheric Environment, 2006. **40**(34): p. 6615-6625.

[58] Li, X.H., et al., *The Fragment Constant Method for Predicting Octanol-air Partition Coefficients of Persistent Organic Pollutants at Different Temperatures.* Journal of Physical and Chemical Reference Data, 2006. **35**(3): p. 1365-1384.

[59] Dachs, J., et al., *Response to the Comment on "Influence of Soot Carbon on the Soil-air Partitioning of Polycyclic Aromatic Hydrocarbons".* Environmental Science & Technology, 2004. **38**(5): p. 1624-1625.

[60] van Noort, P.C.M., *A Thermodynamics-based Estimation Model for Adsorption of Organic Compounds by Carbonaceous Materials in Environmental Sorbents.* Environmental Toxicology and Chemistry, 2003. **22**(6): p. 1179-1188.

[61] Huson, D.H., *User Manual for MEGAN V4.70.4*. 2012.

[62] Boylan, J.W. and A.G. Russell, *PM and Light Extinction Model Performance Metrics, Goals, and Criteria for Three-dimensional Air Quality Models.* Atmospheric Environment, 2006. **40**(26): p. 4946-4959.

[63] San Jose, R., et al., *BaP (PAH) Air Quality Modelling Exercise over Zaragoza (Spain) Using An Adapted Version of WRF-CMAQ Model.* Environmental Pollution, 2013. **183**: p. 151-158.

[64] Bedjanian, Y. and M.L. Nguyen, *Kinetics of the Reactions of Soot Surface-bound Polycyclic Aromatic Hydrocarbons with O-3.* Chemosphere, 2010. **79**(4): p. 387-393.

[65] Esteve, W., H. Budzinski, and E. Villenave, *Relative Rate Constants for the Heterogeneous Reactions of OH, NO2 and NO Radicals with Polycyclic Aromatic Hydrocarbons Adsorbed on Carbonaceous Particles. Part 1: PAHs Adsorbed on 1-2 mu m Calibrated Graphite Particles.* Atmospheric Environment, 2004. **38**(35): p. 6063-6072.

[66] Esteve, W., H. Budzinski, and E. Villenave, *Relative Rate Constants for the Heterogeneous Reactions of NO2 and OH Radicals with Polycyclic Aromatic Hydrocarbons Adsorbed on Carbonaceous Particles. Part 2: PAHs Adsorbed on Diesel Particulate Exhaust SRM 1650a.* Atmospheric Environment, 2006. **40**(2): p. 201-211.

[67] Miet, K., H. Budzinski, and E. Villenave, *Heterogeneous Reactions of Oh Radicals with Particulate-Pyrene and 1-Nitropyrene of Atmospheric Interest.* Polycyclic Aromatic Compounds, 2009. **29**(5): p. 267-281.

[68] Kahan, T.F., N.O.A. Kwamena, and D.J. Donaldson, *Heterogeneous Ozonation Kinetics of Polycyclic Aromatic Hydrocarbons on Organic Films.* Atmospheric Environment, 2006. **40**(19): p. 3448-3459.

[69] Kwamena, N.O.A., et al., *Kinetic and Product Yield Study of the Heterogeneous Gas-surface Reaction of Anthracene and Ozone.* Journal of Physical Chemistry A, 2006. **110**(10): p. 3638-3646.

[70] Kwamena, N.O.A., et al., *Role of the Aerosol Substrate in the Heterogeneous Ozonation Reactions of Surface-bound PAHs.* Journal of Physical Chemistry A, 2007. **111**(43): p. 11050-11058.

[71] Kwamena, N.O.A., J.A. Thornton, and J.P.D. Abbatt, *Kinetics of Surface-bound Benzo[a]pyrene and Ozone on Solid Organic and Salt Aerosols.* Journal of Physical Chemistry A, 2004. **108**(52): p. 11626-11634.

[72] Miet, K., et al., *Heterogeneous Reactions of Ozone with Pyrene, 1-hydroxypyrene and 1-nitropyrene Adsorbed on Particles.* Atmospheric Environment, 2009. **43**(24): p. 3699-3707.

[73] Mmereki, B.T., et al., *Kinetics and Products of the Reaction of Gas-phase Ozone with Anthracene Adsorbed at the Air-aqueous Interface.* Atmospheric Environment, 2004. **38**(36): p. 6091-6103.

[74] Perraudin, E., H. Budzinski, and E. Villenave, *Kinetic Study of the Reactions of Ozone with Polycyclic Aromatic Hydrocarbons Adsorbed on Atmospheric Model Particles.* Journal of Atmospheric Chemistry, 2007. **56**(1): p. 57-82.

[75] Liu, C.G., et al., *Kinetic Studies of Heterogeneous Reactions of Polycyclic Aromatic Hydrocarbon Aerosols with NO3 Radicals.* Environmental Science & Technology, 2012. **46**(14): p. 7575-7580.

[76] Oros, D.R. and B.R.T. Simoneit, *Identification and Emission Rates of Molecular Tracers in Coal Smoke Particulate Matter.* Fuel, 2000. **79**(5): p. 515-536.

APPENDIX

Table S1 Speciation profiles

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
0000	TOG	ACY	2.00E-04	152.2	2.00E-04
0000	TOG	CHRY	1.00E-04	228.3	1.00E-04
0000	TOG	FTH	1.00E-04	202.26	1.00E-04
0000	TOG	FLU	1.00E-04	166.22	1.00E-04
0000	TOG	NAPH	1.80E-03	128.17	1.80E-03
0000	TOG	PHE	6.00E-04	178.24	6.00E-04
0000	TOG	PYR	1.00E-04	202.26	1.00E-04
0000	NONHAPTOG	ACY	2.14E-04	152.2	2.14E-04
0000	NONHAPTOG	CHRY	1.07E-04	228.3	1.07E-04
0000	NONHAPTOG	FTH	1.07E-04	202.26	1.07E-04
0000	NONHAPTOG	FLU	1.07E-04	166.22	1.07E-04
0000	NONHAPTOG	NAPH	1.93E-03	128.17	1.93E-03
0000	NONHAPTOG	PHE	6.42E-04	178.24	6.42E-04
0000	NONHAPTOG	PYR	1.07E-04	202.26	1.07E-04
0008	TOG	NAPH	1.40E-02	128.17	1.40E-02
0008	TOG	ACY	7.34E-05	152.2	7.34E-05
0008	TOG	ACE	4.52E-05	154.21	4.52E-05
0008	TOG	FLU	1.69E-04	166.22	1.69E-04
0008	TOG	ANT	2.61E-05	178.24	2.61E-05
0008	TOG	PHE	7.32E-04	178.24	7.32E-04
0008	TOG	FTH	3.93E-05	202.26	3.93E-05
0008	TOG	PYR	3.26E-05	202.26	3.26E-05
0008	TOG	BAA	2.58E-07	228.3	2.58E-07
0008	TOG	CHRY	4.30E-07	228.3	4.30E-07
0008	TOG	BGHIP	1.72E-07	276.34	1.72E-07
0008	NONHAPTOG	NAPH	1.43E-02	128.17	1.43E-02
0008	NONHAPTOG	ACY	7.48E-05	152.2	7.48E-05
0008	NONHAPTOG	ACE	4.61E-05	154.21	4.61E-05
0008	NONHAPTOG	FLU	1.72E-04	166.22	1.72E-04
0008	NONHAPTOG	ANT	2.67E-05	178.24	2.67E-05
0008	NONHAPTOG	PHE	7.46E-04	178.24	7.46E-04
0008	NONHAPTOG	FTH	4.01E-05	202.26	4.01E-05
0008	NONHAPTOG	PYR	3.32E-05	202.26	3.32E-05
0008	NONHAPTOG	BAA	2.63E-07	228.3	2.63E-07
0008	NONHAPTOG	CHRY	4.39E-07	228.3	4.39E-07
0008	NONHAPTOG	BGHIP	1.75E-07	276.34	1.75E-07
0009	TOG	NAPH	1.40E-02	128.17	1.40E-02
0009	TOG	ACY	7.34E-05	152.2	7.34E-05
0009	TOG	ACE	4.52E-05	154.21	4.52E-05
0009	TOG	FLU	1.69E-04	166.22	1.69E-04
0009	TOG	ANT	2.61E-05	178.24	2.61E-05

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
0009	TOG	PHE	7.32E-04	178.24	7.32E-04
0009	TOG	FTH	3.93E-05	202.26	3.93E-05
0009	TOG	PYR	3.26E-05	202.26	3.26E-05
0009	TOG	BAA	2.58E-07	228.3	2.58E-07
0009	TOG	CHRY	4.30E-07	228.3	4.30E-07
0009	TOG	BGHIP	1.72E-07	276.34	1.72E-07
0009	NONHAPTOG	NAPH	1.43E-02	128.17	1.43E-02
0009	NONHAPTOG	ACY	7.48E-05	152.2	7.48E-05
0009	NONHAPTOG	ACE	4.61E-05	154.21	4.61E-05
0009	NONHAPTOG	FLU	1.72E-04	166.22	1.72E-04
0009	NONHAPTOG	ANT	2.67E-05	178.24	2.67E-05
0009	NONHAPTOG	PHE	7.46E-04	178.24	7.46E-04
0009	NONHAPTOG	FTH	4.01E-05	202.26	4.01E-05
0009	NONHAPTOG	PYR	3.32E-05	202.26	3.32E-05
0009	NONHAPTOG	BAA	2.63E-07	228.3	2.63E-07
0009	NONHAPTOG	CHRY	4.39E-07	228.3	4.39E-07
0009	NONHAPTOG	BGHIP	1.75E-07	276.34	1.75E-07
0079	TOG	ACE	1.24E-09	154.21	1.24E-09
0079	TOG	ACY	5.10E-10	152.2	5.10E-10
0079	TOG	ANT	2.37E-12	178.24	2.37E-12
0079	TOG	BAA	4.99E-13	228.3	4.99E-13
0079	TOG	BAP	8.21E-13	252.32	8.21E-13
0079	TOG	BGHIP	6.27E-12	276.34	6.27E-12
0079	TOG	BKF	4.68E-13	252.32	4.68E-13
0079	TOG	CHRY	2.37E-10	228.3	2.37E-10
0079	TOG	DAHA	1.82E-12	278.36	1.82E-12
0079	TOG	FLU	1.64E-09	166.22	1.64E-09
0079	TOG	FTH	6.53E-09	202.26	6.53E-09
0079	TOG	ICDP	3.81E-12	276.34	3.81E-12
0079	TOG	NAPH	1.42E-07	128.17	1.42E-07
0079	TOG	PHE	1.92E-08	178.24	1.92E-08
0079	TOG	PYR	1.40E-10	202.26	1.40E-10
0079	NONHAPTOG	ACE	1.49E-09	154.21	1.49E-09
0079	NONHAPTOG	ACY	6.12E-10	152.2	6.12E-10
0079	NONHAPTOG	ANT	2.84E-12	178.24	2.84E-12
0079	NONHAPTOG	BAA	5.99E-13	228.3	5.99E-13
0079	NONHAPTOG	BAP	9.85E-13	252.32	9.85E-13
0079	NONHAPTOG	BGHIP	7.52E-12	276.34	7.52E-12
0079	NONHAPTOG	BKF	5.62E-13	252.32	5.62E-13
0079	NONHAPTOG	CHRY	2.84E-10	228.3	2.84E-10
0079	NONHAPTOG	DAHA	2.19E-12	278.36	2.19E-12

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
0079	NONHAPTOG	FLU	1.97E-09	166.22	1.97E-09
0079	NONHAPTOG	FTH	7.83E-09	202.26	7.83E-09
0079	NONHAPTOG	ICDP	4.57E-12	276.34	4.57E-12
0079	NONHAPTOG	NAPH	1.70E-07	128.17	1.70E-07
0079	NONHAPTOG	PHE	2.30E-08	178.24	2.30E-08
0079	NONHAPTOG	PYR	1.68E-10	202.26	1.68E-10
1007	TOG	NAPH	6.54E-02	128.17	6.54E-02
1007	NONHAPTOG	NAPH	6.54E-02	128.17	6.54E-02
1064	TOG	NAPH	1.34E-05	128.17	1.34E-05
1064	NONHAPTOG	NAPH	1.37E-05	128.17	1.37E-05
1084	TOG	ACE	1.06E-04	154.21	1.06E-04
1084	TOG	ACY	9.77E-04	152.2	9.77E-04
1084	TOG	ANT	1.81E-04	178.24	1.81E-04
1084	TOG	BGHIP	4.30E-06	276.34	4.30E-06
1084	TOG	FTH	1.60E-04	202.26	1.60E-04
1084	TOG	FLU	2.33E-04	166.22	2.33E-04
1084	TOG	NAPH	1.19E-02	128.17	1.19E-02
1084	TOG	PHE	8.24E-04	178.24	8.24E-04
1084	TOG	PYR	9.82E-05	202.26	9.82E-05
1084	NONHAPTOG	ACE	1.30E-04	154.21	1.30E-04
1084	NONHAPTOG	ACY	1.20E-03	152.2	1.20E-03
1084	NONHAPTOG	ANT	2.23E-04	178.24	2.23E-04
1084	NONHAPTOG	BGHIP	5.29E-06	276.34	5.29E-06
1084	NONHAPTOG	FTH	1.97E-04	202.26	1.97E-04
1084	NONHAPTOG	FLU	2.87E-04	166.22	2.87E-04
1084	NONHAPTOG	NAPH	1.46E-02	128.17	1.46E-02
1084	NONHAPTOG	PHE	1.01E-03	178.24	1.01E-03
1084	NONHAPTOG	PYR	1.21E-04	202.26	1.21E-04
1095	TOG	NAPH	3.70E-03	128.17	3.70E-03
1095	NONHAPTOG	NAPH	3.70E-03	128.17	3.70E-03
1096	TOG	NAPH	1.63E-02	128.17	1.63E-02
1096	NONHAPTOG	NAPH	1.63E-02	128.17	1.63E-02
1101	TOG	NAPH	1.78E-03	128.17	1.78E-03
1101	TOG	ACY	1.56E-04	152.2	1.56E-04
1101	TOG	ACE	3.43E-05	154.21	3.43E-05
1101	TOG	FLU	6.95E-05	166.22	6.95E-05
1101	TOG	ANT	2.88E-05	178.24	2.88E-05
1101	TOG	PHE	1.84E-04	178.24	1.84E-04
1101	TOG	FTH	4.82E-05	202.26	4.82E-05
1101	TOG	PYR	5.50E-06	202.26	5.50E-06
1101	TOG	BAA	4.64E-06	228.3	4.64E-06

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
1101	TOG	CHRY	5.20E-06	228.3	5.20E-06
1101	TOG	BAP	2.53E-07	252.32	2.53E-07
1101	TOG	BBF	3.45E-06	252.32	3.45E-06
1101	TOG	BKF	3.45E-06	252.32	3.45E-06
1101	NONHAPTOG	NAPH	1.82E-03	128.17	1.82E-03
1101	NONHAPTOG	ACY	1.59E-04	152.2	1.59E-04
1101	NONHAPTOG	ACE	3.50E-05	154.21	3.50E-05
1101	NONHAPTOG	FLU	7.09E-05	166.22	7.09E-05
1101	NONHAPTOG	ANT	2.94E-05	178.24	2.94E-05
1101	NONHAPTOG	PHE	1.88E-04	178.24	1.88E-04
1101	NONHAPTOG	FTH	4.91E-05	202.26	4.91E-05
1101	NONHAPTOG	PYR	5.61E-06	202.26	5.61E-06
1101	NONHAPTOG	BAA	4.74E-06	228.3	4.74E-06
1101	NONHAPTOG	CHRY	5.31E-06	228.3	5.31E-06
1101	NONHAPTOG	BAP	2.58E-07	252.32	2.58E-07
1101	NONHAPTOG	BBF	3.52E-06	252.32	3.52E-06
1101	NONHAPTOG	BKF	3.52E-06	252.32	3.52E-06
1167	TOG	ACE	1.22E-02	154.21	1.22E-02
1167	TOG	ACY	7.19E-02	152.2	7.19E-02
1167	TOG	BAP	9.00E-03	252.32	9.00E-03
1167	TOG	BBF	1.14E-02	252.32	1.14E-02
1167	TOG	BGHIP	1.30E-02	276.34	1.30E-02
1167	TOG	CHRY	2.39E-02	228.3	2.39E-02
1167	TOG	DAHA	4.00E-04	278.36	4.00E-04
1167	TOG	FTH	3.63E-02	202.26	3.63E-02
1167	TOG	FLU	2.59E-02	166.22	2.59E-02
1167	TOG	ICDP	6.60E-03	276.34	6.60E-03
1167	TOG	NAPH	4.37E-01	128.17	4.37E-01
1167	TOG	PHE	1.68E-01	178.24	1.68E-01
1167	TOG	PYR	3.35E-02	202.26	3.35E-02
1167	NONHAPTOG	ACE	1.23E-02	154.21	1.23E-02
1167	NONHAPTOG	ACY	7.25E-02	152.2	7.25E-02
1167	NONHAPTOG	BAP	9.08E-03	252.32	9.08E-03
1167	NONHAPTOG	BBF	1.15E-02	252.32	1.15E-02
1167	NONHAPTOG	BGHIP	1.31E-02	276.34	1.31E-02
1167	NONHAPTOG	CHRY	2.41E-02	228.3	2.41E-02
1167	NONHAPTOG	DAHA	4.04E-04	278.36	4.04E-04
1167	NONHAPTOG	FTH	3.66E-02	202.26	3.66E-02
1167	NONHAPTOG	FLU	2.61E-02	166.22	2.61E-02
1167	NONHAPTOG	ICDP	6.66E-03	276.34	6.66E-03
1167	NONHAPTOG	NAPH	4.40E-01	128.17	4.40E-01

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
1167	NONHAPTOG	PHE	1.70E-01	178.24	1.70E-01
1167	NONHAPTOG	PYR	3.38E-02	202.26	3.38E-02
1178	TOG	NAPH	3.70E-04	128.17	3.70E-04
1178	NONHAPTOG	NAPH	3.70E-04	128.17	3.70E-04
1186	TOG	NAPH	1.78E-03	128.17	1.78E-03
1186	TOG	ACY	1.56E-04	152.2	1.56E-04
1186	TOG	ACE	3.43E-05	154.21	3.43E-05
1186	TOG	FLU	6.95E-05	166.22	6.95E-05
1186	TOG	ANT	2.88E-05	178.24	2.88E-05
1186	TOG	PHE	1.84E-04	178.24	1.84E-04
1186	TOG	FTH	4.82E-05	202.26	4.82E-05
1186	TOG	PYR	5.50E-06	202.26	5.50E-06
1186	TOG	BAA	4.64E-06	228.3	4.64E-06
1186	TOG	CHRY	5.20E-06	228.3	5.20E-06
1186	TOG	BAP	2.53E-07	252.32	2.53E-07
1186	TOG	BBF	3.45E-06	252.32	3.45E-06
1186	TOG	BKF	3.45E-06	252.32	3.45E-06
1186	NONHAPTOG	NAPH	1.82E-03	128.17	1.82E-03
1186	NONHAPTOG	ACY	1.59E-04	152.2	1.59E-04
1186	NONHAPTOG	ACE	3.50E-05	154.21	3.50E-05
1186	NONHAPTOG	FLU	7.09E-05	166.22	7.09E-05
1186	NONHAPTOG	ANT	2.94E-05	178.24	2.94E-05
1186	NONHAPTOG	PHE	1.88E-04	178.24	1.88E-04
1186	NONHAPTOG	FTH	4.91E-05	202.26	4.91E-05
1186	NONHAPTOG	PYR	5.61E-06	202.26	5.61E-06
1186	NONHAPTOG	BAA	4.74E-06	228.3	4.74E-06
1186	NONHAPTOG	CHRY	5.31E-06	228.3	5.31E-06
1186	NONHAPTOG	BAP	2.58E-07	252.32	2.58E-07
1186	NONHAPTOG	BBF	3.52E-06	252.32	3.52E-06
1186	NONHAPTOG	BKF	3.52E-06	252.32	3.52E-06
1189	TOG	NAPH	6.92E-03	128.17	6.92E-03
1189	NONHAPTOG	NAPH	6.92E-03	128.17	6.92E-03
1192	TOG	NAPH	1.00E-03	128.17	1.00E-03
1192	NONHAPTOG	NAPH	1.00E-03	128.17	1.00E-03
1193	TOG	NAPH	3.50E-03	128.17	3.50E-03
1193	NONHAPTOG	NAPH	3.50E-03	128.17	3.50E-03
1194	TOG	NAPH	1.46E-02	128.17	1.46E-02
1194	NONHAPTOG	NAPH	3.05E-04	128.17	3.05E-04
1195	TOG	NAPH	3.00E-04	128.17	3.00E-04
1195	NONHAPTOG	NAPH	5.05E-04	128.17	5.05E-04
1196	TOG	NAPH	5.00E-04	128.17	5.00E-04

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
1196	NONHAPTOG	NAPH	5.00E-04	128.17	5.00E-04
1202	TOG	NAPH	5.75E-02	128.17	5.75E-02
1202	TOG	ACE	6.10E-03	154.21	6.10E-03
1202	TOG	ACY	1.01E-01	152.2	1.01E-01
1202	TOG	ANT	7.51E-02	178.24	7.51E-02
1202	TOG	BAA	1.60E-03	228.3	1.60E-03
1202	TOG	BAP	1.20E-03	252.32	1.20E-03
1202	TOG	CHRY	1.40E-03	228.3	1.40E-03
1202	TOG	FTH	3.85E-02	202.26	3.85E-02
1202	TOG	FLU	3.50E-02	166.22	3.50E-02
1202	TOG	PHE	7.31E-02	178.24	7.31E-02
1202	TOG	PYR	2.80E-02	202.26	2.80E-02
1202	NONHAPTOG	NAPH	5.78E-02	128.17	5.78E-02
1202	NONHAPTOG	ACE	6.13E-03	154.21	6.13E-03
1202	NONHAPTOG	ACY	1.02E-01	152.2	1.02E-01
1202	NONHAPTOG	ANT	7.55E-02	178.24	7.55E-02
1202	NONHAPTOG	BAA	1.61E-03	228.3	1.61E-03
1202	NONHAPTOG	BAP	1.21E-03	252.32	1.21E-03
1202	NONHAPTOG	CHRY	1.41E-03	228.3	1.41E-03
1202	NONHAPTOG	FTH	3.87E-02	202.26	3.87E-02
1202	NONHAPTOG	FLU	3.52E-02	166.22	3.52E-02
1202	NONHAPTOG	PHE	7.35E-02	178.24	7.35E-02
1202	NONHAPTOG	PYR	2.81E-02	202.26	2.81E-02
2420	TOG	NAPH	5.06E-02	128.17	5.06E-02
2420	NONHAPTOG	NAPH	5.06E-02	128.17	5.06E-02
2480	TOG	NAPH	1.40E-02	128.17	1.40E-02
2480	TOG	ACY	7.34E-05	152.2	7.34E-05
2480	TOG	ACE	4.52E-05	154.21	4.52E-05
2480	TOG	FLU	1.69E-04	166.22	1.69E-04
2480	TOG	ANT	2.61E-05	178.24	2.61E-05
2480	TOG	PHE	7.32E-04	178.24	7.32E-04
2480	TOG	FTH	3.93E-05	202.26	3.93E-05
2480	TOG	PYR	3.26E-05	202.26	3.26E-05
2480	TOG	BAA	2.58E-07	228.3	2.58E-07
2480	TOG	CHRY	4.30E-07	228.3	4.30E-07
2480	TOG	BGHIP	1.72E-07	276.34	1.72E-07
2480	NONHAPTOG	NAPH	1.43E-02	128.17	1.43E-02
2480	NONHAPTOG	ACY	7.48E-05	152.2	7.48E-05
2480	NONHAPTOG	ACE	4.61E-05	154.21	4.61E-05
2480	NONHAPTOG	FLU	1.72E-04	166.22	1.72E-04
2480	NONHAPTOG	ANT	2.67E-05	178.24	2.67E-05

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
2480	NONHAPTOG	PHE	7.46E-04	178.24	7.46E-04
2480	NONHAPTOG	FTH	4.01E-05	202.26	4.01E-05
2480	NONHAPTOG	PYR	3.32E-05	202.26	3.32E-05
2480	NONHAPTOG	BAA	2.63E-07	228.3	2.63E-07
2480	NONHAPTOG	CHRY	4.39E-07	228.3	4.39E-07
2480	NONHAPTOG	BGHIP	1.75E-07	276.34	1.75E-07
2485	TOG	NAPH	1.16E-03	128.17	1.16E-03
2485	NONHAPTOG	NAPH	1.24E-03	128.17	1.24E-03
2508	TOG	NAPH	1.40E-02	128.17	1.40E-02
2508	TOG	ACY	7.34E-05	152.2	7.34E-05
2508	TOG	ACE	4.52E-05	154.21	4.52E-05
2508	TOG	FLU	1.69E-04	166.22	1.69E-04
2508	TOG	ANT	2.61E-05	178.24	2.61E-05
2508	TOG	PHE	7.32E-04	178.24	7.32E-04
2508	TOG	FTH	3.93E-05	202.26	3.93E-05
2508	TOG	PYR	3.26E-05	202.26	3.26E-05
2508	TOG	BAA	2.58E-07	228.3	2.58E-07
2508	TOG	CHRY	4.30E-07	228.3	4.30E-07
2508	TOG	BGHIP	1.72E-07	276.34	1.72E-07
2508	NONHAPTOG	NAPH	1.43E-02	128.17	1.43E-02
2508	NONHAPTOG	ACY	7.48E-05	152.2	7.48E-05
2508	NONHAPTOG	ACE	4.61E-05	154.21	4.61E-05
2508	NONHAPTOG	FLU	1.72E-04	166.22	1.72E-04
2508	NONHAPTOG	ANT	2.67E-05	178.24	2.67E-05
2508	NONHAPTOG	PHE	7.46E-04	178.24	7.46E-04
2508	NONHAPTOG	FTH	4.01E-05	202.26	4.01E-05
2508	NONHAPTOG	PYR	3.32E-05	202.26	3.32E-05
2508	NONHAPTOG	BAA	2.63E-07	228.3	2.63E-07
2508	NONHAPTOG	CHRY	4.39E-07	228.3	4.39E-07
2508	NONHAPTOG	BGHIP	1.75E-07	276.34	1.75E-07
3002	TOG	NAPH	1.00E-03	128.17	1.00E-03
3002	NONHAPTOG	NAPH	1.00E-03	128.17	1.00E-03
3066	TOG	NAPH	1.12E-02	128.17	1.12E-02
3066	NONHAPTOG	NAPH	1.12E-02	128.17	1.12E-02
3127	TOG	NAPH	3.00E-04	128.17	3.00E-04
3127	NONHAPTOG	NAPH	3.00E-04	128.17	3.00E-04
3131	TOG	NAPH	3.00E-04	128.17	3.00E-04
3131	NONHAPTOG	NAPH	3.00E-04	128.17	3.00E-04
3134	TOG	NAPH	3.00E-04	128.17	3.00E-04
3134	NONHAPTOG	NAPH	3.00E-04	128.17	3.00E-04
3135	TOG	NAPH	3.00E-04	128.17	3.00E-04

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
3135	NONHAPTOG	NAPH	3.00E-04	128.17	3.00E-04
3137	TOG	NAPH	3.00E-04	128.17	3.00E-04
3137	NONHAPTOG	NAPH	3.00E-04	128.17	3.00E-04
3138	TOG	NAPH	3.00E-04	128.17	3.00E-04
3138	NONHAPTOG	NAPH	3.00E-04	128.17	3.00E-04
3139	TOG	NAPH	2.00E-04	128.17	2.00E-04
3139	NONHAPTOG	NAPH	2.00E-04	128.17	2.00E-04
3140	TOG	NAPH	2.00E-04	128.17	2.00E-04
3140	NONHAPTOG	NAPH	2.06E-04	128.17	2.06E-04
3141	TOG	NAPH	2.50E-03	128.17	2.50E-03
3141	NONHAPTOG	NAPH	2.50E-03	128.17	2.50E-03
3142	TOG	NAPH	1.12E-02	128.17	1.12E-02
3142	NONHAPTOG	NAPH	1.12E-02	128.17	1.12E-02
3144	TOG	NAPH	1.12E-02	128.17	1.12E-02
3144	NONHAPTOG	NAPH	1.19E-02	129.17	1.19E-02
3145	TOG	NAPH	1.12E-02	128.17	1.12E-02
3145	NONHAPTOG	NAPH	1.12E-02	128.17	1.12E-02
3146	TOG	NAPH	1.12E-02	128.17	1.12E-02
3146	NONHAPTOG	NAPH	1.12E-02	128.17	1.12E-02
3147	TOG	NAPH	1.12E-02	128.17	1.12E-02
3147	NONHAPTOG	NAPH	1.12E-02	128.17	1.12E-02
3149	TOG	NAPH	3.00E-04	128.17	3.00E-04
3149	NONHAPTOG	NAPH	3.00E-04	128.17	3.00E-04
3150	TOG	NAPH	1.78E-03	128.17	1.78E-03
3150	TOG	ACY	1.56E-04	152.2	1.56E-04
3150	TOG	ACE	3.43E-05	154.21	3.43E-05
3150	TOG	FLU	6.95E-05	166.22	6.95E-05
3150	TOG	ANT	2.88E-05	178.24	2.88E-05
3150	TOG	PHE	1.84E-04	178.24	1.84E-04
3150	TOG	FTH	4.82E-05	202.26	4.82E-05
3150	TOG	PYR	5.50E-06	202.26	5.50E-06
3150	TOG	BAA	4.64E-06	228.3	4.64E-06
3150	TOG	CHRY	5.20E-06	228.3	5.20E-06
3150	TOG	BAP	2.53E-07	252.32	2.53E-07
3150	TOG	BBF	3.45E-06	252.32	3.45E-06
3150	TOG	BKF	3.45E-06	252.32	3.45E-06
3150	NONHAPTOG	NAPH	1.94E-03	128.17	1.94E-03
3150	NONHAPTOG	ACY	1.70E-04	152.2	1.70E-04
3150	NONHAPTOG	ACE	3.74E-05	154.21	3.74E-05
3150	NONHAPTOG	FLU	7.57E-05	166.22	7.57E-05
3150	NONHAPTOG	ANT	3.14E-05	178.24	3.14E-05

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
3150	NONHAPTOG	PHE	2.01E-04	178.24	2.01E-04
3150	NONHAPTOG	FTH	5.25E-05	202.26	5.25E-05
3150	NONHAPTOG	PYR	6.00E-06	202.26	6.00E-06
3150	NONHAPTOG	BAA	5.06E-06	228.3	5.06E-06
3150	NONHAPTOG	CHRY	5.67E-06	228.3	5.67E-06
3150	NONHAPTOG	BAP	2.76E-07	252.32	2.76E-07
3150	NONHAPTOG	BBF	3.76E-06	252.32	3.76E-06
3150	NONHAPTOG	BKF	3.76E-06	252.32	3.76E-06
3161	TOG	NAPH	1.40E-02	128.17	1.40E-02
3161	TOG	ACY	7.34E-05	152.2	7.34E-05
3161	TOG	ACE	4.52E-05	154.21	4.52E-05
3161	TOG	FLU	1.69E-04	166.22	1.69E-04
3161	TOG	ANT	2.61E-05	178.24	2.61E-05
3161	TOG	PHE	7.32E-04	178.24	7.32E-04
3161	TOG	FTH	3.93E-05	202.26	3.93E-05
3161	TOG	PYR	3.26E-05	202.26	3.26E-05
3161	TOG	BAA	2.58E-07	228.3	2.58E-07
3161	TOG	CHRY	4.30E-07	228.3	4.30E-07
3161	TOG	BGHIP	1.72E-07	276.34	1.72E-07
3161	NONHAPTOG	NAPH	1.85E-02	128.17	1.85E-02
3161	NONHAPTOG	ACY	9.68E-05	152.2	9.68E-05
3161	NONHAPTOG	ACE	5.97E-05	154.21	5.97E-05
3161	NONHAPTOG	FLU	2.22E-04	166.22	2.22E-04
3161	NONHAPTOG	ANT	3.45E-05	178.24	3.45E-05
3161	NONHAPTOG	PHE	9.66E-04	178.24	9.66E-04
3161	NONHAPTOG	FTH	5.19E-05	202.26	5.19E-05
3161	NONHAPTOG	PYR	4.30E-05	202.26	4.30E-05
3161	NONHAPTOG	BAA	3.41E-07	228.3	3.41E-07
3161	NONHAPTOG	CHRY	5.68E-07	228.3	5.68E-07
3161	NONHAPTOG	BGHIP	2.27E-07	276.34	2.27E-07
4458	TOG	NAPH	2.02E-03	128.17	2.02E-03
4458	NONHAPTOG	NAPH	2.02E-03	128.17	2.02E-03
4547	TOG	NAPH	1.78E-03	128.17	1.78E-03
4547	TOG	ACY	1.56E-04	152.2	1.56E-04
4547	TOG	ACE	3.43E-05	154.21	3.43E-05
4547	TOG	FLU	6.95E-05	166.22	6.95E-05
4547	TOG	ANT	2.88E-05	178.24	2.88E-05
4547	TOG	PHE	1.84E-04	178.24	1.84E-04
4547	TOG	FTH	4.82E-05	202.26	4.82E-05
4547	TOG	PYR	5.50E-06	202.26	5.50E-06
4547	TOG	BAA	4.64E-06	228.3	4.64E-06

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
4547	TOG	CHRY	5.20E-06	228.3	5.20E-06
4547	TOG	BAP	2.53E-07	252.32	2.53E-07
4547	TOG	BBF	3.45E-06	252.32	3.45E-06
4547	TOG	BKF	3.45E-06	252.32	3.45E-06
4547	NONHAPTOG	NAPH	1.79E-03	128.17	1.79E-03
4547	NONHAPTOG	ACY	1.56E-04	152.2	1.56E-04
4547	NONHAPTOG	ACE	3.44E-05	154.21	3.44E-05
4547	NONHAPTOG	FLU	6.97E-05	166.22	6.97E-05
4547	NONHAPTOG	ANT	2.89E-05	178.24	2.89E-05
4547	NONHAPTOG	PHE	1.85E-04	178.24	1.85E-04
4547	NONHAPTOG	FTH	4.83E-05	202.26	4.83E-05
4547	NONHAPTOG	PYR	5.52E-06	202.26	5.52E-06
4547	NONHAPTOG	BAA	4.66E-06	228.3	4.66E-06
4547	NONHAPTOG	CHRY	5.22E-06	228.3	5.22E-06
4547	NONHAPTOG	BAP	2.54E-07	252.32	2.54E-07
4547	NONHAPTOG	BBF	3.46E-06	252.32	3.46E-06
4547	NONHAPTOG	BKF	3.46E-06	252.32	3.46E-06
4553	TOG	CHRY	1.15E-05	228.3	1.15E-05
4553	TOG	NAPH	3.13E-04	128.17	3.13E-04
4553	TOG	ANT	3.83E-06	178.24	3.83E-06
4553	TOG	FLU	1.39E-05	166.22	1.39E-05
4553	TOG	PHE	4.21E-05	178.24	4.21E-05
4553	TOG	PYR	1.99E-05	202.26	1.99E-05
4553	NONHAPTOG	CHRY	1.25E-05	228.3	1.25E-05
4553	NONHAPTOG	NAPH	3.41E-04	128.17	3.41E-04
4553	NONHAPTOG	ANT	4.18E-06	178.24	4.18E-06
4553	NONHAPTOG	FLU	1.52E-05	166.22	1.52E-05
4553	NONHAPTOG	PHE	4.59E-05	178.24	4.59E-05
4553	NONHAPTOG	PYR	2.16E-05	202.26	2.16E-05
4642	TOG	ACE	1.06E-04	154.21	1.06E-04
4642	TOG	ACY	9.77E-04	152.2	9.77E-04
4642	TOG	ANT	1.81E-04	178.24	1.81E-04
4642	TOG	BGHIP	4.30E-06	276.34	4.30E-06
4642	TOG	FTH	1.60E-04	202.26	1.60E-04
4642	TOG	FLU	2.33E-04	166.22	2.33E-04
4642	TOG	NAPH	1.19E-02	128.17	1.19E-02
4642	TOG	PHE	8.24E-04	178.24	8.24E-04
4642	TOG	PYR	9.82E-05	202.26	9.82E-05
4642	NONHAPTOG	ACE	1.27E-04	154.21	1.27E-04
4642	NONHAPTOG	ACY	1.17E-03	152.2	1.17E-03
4642	NONHAPTOG	ANT	2.17E-04	178.24	2.17E-04

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
4642	NONHAPTOG	BGHIP	5.16E-06	276.34	5.16E-06
4642	NONHAPTOG	FTH	1.92E-04	202.26	1.92E-04
4642	NONHAPTOG	FLU	2.80E-04	166.22	2.80E-04
4642	NONHAPTOG	NAPH	1.43E-02	128.17	1.43E-02
4642	NONHAPTOG	PHE	9.89E-04	178.24	9.89E-04
4642	NONHAPTOG	PYR	1.18E-04	202.26	1.18E-04
4651	TOG	ACY	1.34E-04	152.2	1.34E-04
4651	TOG	ANT	2.89E-05	178.24	2.89E-05
4651	TOG	FTH	8.67E-05	202.26	8.67E-05
4651	TOG	NAPH	2.12E-03	128.17	2.12E-03
4651	TOG	PHE	4.33E-04	178.24	4.33E-04
4651	TOG	PYR	5.42E-05	202.26	5.42E-05
4651	NONHAPTOG	ACY	1.71E-04	152.2	1.71E-04
4651	NONHAPTOG	ANT	3.70E-05	178.24	3.70E-05
4651	NONHAPTOG	FTH	1.11E-04	202.26	1.11E-04
4651	NONHAPTOG	NAPH	2.72E-03	128.17	2.72E-03
4651	NONHAPTOG	PHE	5.55E-04	178.24	5.55E-04
4651	NONHAPTOG	PYR	6.93E-05	202.26	6.93E-05
4652	TOG	CHRY	4.33E-05	228.3	4.33E-05
4652	TOG	NAPH	2.93E-03	128.17	2.93E-03
4652	TOG	ACY	1.65E-04	152.2	1.65E-04
4652	TOG	ANT	5.20E-05	178.24	5.20E-05
4652	TOG	FLU	1.65E-04	166.22	1.65E-04
4652	TOG	PHE	7.19E-04	178.24	7.19E-04
4652	TOG	PYR	1.65E-04	202.26	1.65E-04
4652	NONHAPTOG	CHRY	4.33E-05	228.3	4.33E-05
4652	NONHAPTOG	NAPH	2.93E-03	128.17	2.93E-03
4652	NONHAPTOG	ACY	1.65E-04	152.2	1.65E-04
4652	NONHAPTOG	ANT	5.20E-05	178.24	5.20E-05
4652	NONHAPTOG	FLU	1.65E-04	166.22	1.65E-04
4652	NONHAPTOG	PHE	7.19E-04	178.24	7.19E-04
4652	NONHAPTOG	PYR	1.65E-04	202.26	1.65E-04
4659	TOG	ACE	1.35E-04	154.21	1.35E-04
4659	TOG	ACY	2.32E-04	152.2	2.32E-04
4659	TOG	FLU	8.84E-05	166.22	8.84E-05
4659	TOG	NAPH	1.52E-03	128.17	1.52E-03
4659	NONHAPTOG	ACE	1.49E-04	154.21	1.49E-04
4659	NONHAPTOG	ACY	2.57E-04	152.2	2.57E-04
4659	NONHAPTOG	FLU	9.81E-05	166.22	9.81E-05
4659	NONHAPTOG	NAPH	1.69E-03	128.17	1.69E-03
4674	TOG	NAPH	1.40E-02	128.17	1.40E-02

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
4674	TOG	ACY	7.34E-05	152.2	7.34E-05
4674	TOG	ACE	4.52E-05	154.21	4.52E-05
4674	TOG	FLU	1.69E-04	166.22	1.69E-04
4674	TOG	ANT	2.61E-05	178.24	2.61E-05
4674	TOG	PHE	7.32E-04	178.24	7.32E-04
4674	TOG	FTH	3.93E-05	202.26	3.93E-05
4674	TOG	PYR	3.26E-05	202.26	3.26E-05
4674	TOG	BAA	2.58E-07	228.3	2.58E-07
4674	TOG	CHRY	4.30E-07	228.3	4.30E-07
4674	TOG	BGHIP	1.72E-07	276.34	1.72E-07
4674	NONHAPTOG	NAPH	1.88E-02	128.17	1.88E-02
4674	NONHAPTOG	ACY	9.83E-05	152.2	9.83E-05
4674	NONHAPTOG	ACE	6.06E-05	154.21	6.06E-05
4674	NONHAPTOG	FLU	2.26E-04	166.22	2.26E-04
4674	NONHAPTOG	ANT	3.50E-05	178.24	3.50E-05
4674	NONHAPTOG	PHE	9.81E-04	178.24	9.81E-04
4674	NONHAPTOG	FTH	5.27E-05	202.26	5.27E-05
4674	NONHAPTOG	PYR	4.37E-05	202.26	4.37E-05
4674	NONHAPTOG	BAA	3.46E-07	228.3	3.46E-07
4674	NONHAPTOG	CHRY	5.76E-07	228.3	5.76E-07
4674	NONHAPTOG	BGHIP	2.30E-07	276.34	2.30E-07
4730	TOG	NAPH	1.22E-03	128.17	1.22E-03
4730	NONHAPTOG	NAPH	1.23E-03	128.17	1.23E-03
5560	TOG	NAPH	2.74E-03	128.17	2.74E-03
5560	TOG	ACE	1.64E-04	154.21	1.64E-04
5560	TOG	ACY	4.01E-04	152.2	4.01E-04
5560	TOG	ANT	7.67E-05	178.24	7.67E-05
5560	TOG	FTH	2.77E-05	202.26	2.77E-05
5560	TOG	FLU	5.35E-04	166.22	5.35E-04
5560	TOG	PHE	8.57E-04	178.24	8.57E-04
5560	TOG	PYR	1.75E-05	202.26	1.75E-05
5560	NONHAPTOG	NAPH	3.39E-03	128.17	3.39E-03
5560	NONHAPTOG	ACE	2.04E-04	154.21	2.04E-04
5560	NONHAPTOG	ACY	4.97E-04	152.2	4.97E-04
5560	NONHAPTOG	ANT	9.51E-05	178.24	9.51E-05
5560	NONHAPTOG	FTH	3.44E-05	202.26	3.44E-05
5560	NONHAPTOG	FLU	6.63E-04	166.22	6.63E-04
5560	NONHAPTOG	PHE	1.06E-03	178.24	1.06E-03
5560	NONHAPTOG	PYR	2.16E-05	202.26	2.16E-05
5565	TOG	NAPH	1.40E-02	128.17	1.40E-02
5565	TOG	ACY	7.34E-05	152.2	7.34E-05

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
5565	TOG	ACE	4.52E-05	154.21	4.52E-05
5565	TOG	FLU	1.69E-04	166.22	1.69E-04
5565	TOG	ANT	2.61E-05	178.24	2.61E-05
5565	TOG	PHE	7.32E-04	178.24	7.32E-04
5565	TOG	FTH	3.93E-05	202.26	3.93E-05
5565	TOG	PYR	3.26E-05	202.26	3.26E-05
5565	TOG	BAA	2.58E-07	228.3	2.58E-07
5565	TOG	CHRY	4.30E-07	228.3	4.30E-07
5565	TOG	BGHIP	1.72E-07	276.34	1.72E-07
5565	NONHAPTOG	NAPH	1.74E-02	128.17	1.74E-02
5565	NONHAPTOG	ACY	9.10E-05	152.2	9.10E-05
5565	NONHAPTOG	ACE	5.61E-05	154.21	5.61E-05
5565	NONHAPTOG	FLU	2.09E-04	166.22	2.09E-04
5565	NONHAPTOG	ANT	3.24E-05	178.24	3.24E-05
5565	NONHAPTOG	PHE	9.08E-04	178.24	9.08E-04
5565	NONHAPTOG	FTH	4.87E-05	202.26	4.87E-05
5565	NONHAPTOG	PYR	4.04E-05	202.26	4.04E-05
5565	NONHAPTOG	BAA	3.20E-07	228.3	3.20E-07
5565	NONHAPTOG	CHRY	5.33E-07	228.3	5.33E-07
5565	NONHAPTOG	BGHIP	2.13E-07	276.34	2.13E-07
5674	PM2_5	PBAA	1.35E-03	1	1.35E-03
5674	PM2_5	PBAP	5.07E-04	1	5.07E-04
5674	PM2_5	PBGHIP	1.13E-04	1	1.13E-04
5674	PM2_5	PFTH	8.14E-03	1	8.14E-03
5674	PM2_5	PFLU	6.28E-02	1	6.28E-02
5674	PM2_5	PICDP	9.62E-05	1	9.62E-05
5674	PM2_5	PPHE	1.10E-01	1	1.10E-01
5674	PM2_5	PPYR	1.24E-02	1	1.24E-02
5674	PM2_5	PDAHA	2.30E-05	1	2.30E-05
5674	PM2_5	PBKF	8.84E-04	1	8.84E-04
5674	PM2_5	PBBF	3.20E-04	1	3.20E-04
8520	TOG	ACE	3.00E-04	154.21	3.00E-04
8520	TOG	NAPH	1.37E-02	128.17	1.37E-02
8520	NONHAPTOG	ACE	4.53E-04	154.21	4.53E-04
8520	NONHAPTOG	NAPH	2.07E-02	128.17	2.07E-02
8744	TOG	NAPH	2.00E-04	128.17	2.00E-04
8744	NONHAPTOG	NAPH	2.02E-04	128.17	2.02E-04
8745	TOG	NAPH	6.50E-04	128.17	6.50E-04
8745	NONHAPTOG	NAPH	6.76E-04	128.17	6.76E-04
8750	TOG	NAPH	1.78E-03	128.17	1.78E-03
8750	TOG	ACY	1.56E-04	152.2	1.56E-04

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
8750	TOG	ACE	3.43E-05	154.21	3.43E-05
8750	TOG	FLU	6.95E-05	166.22	6.95E-05
8750	TOG	ANT	2.88E-05	178.24	2.88E-05
8750	TOG	PHE	1.84E-04	178.24	1.84E-04
8750	TOG	FTH	4.82E-05	202.26	4.82E-05
8750	TOG	PYR	5.50E-06	202.26	5.50E-06
8750	TOG	BAA	4.64E-06	228.3	4.64E-06
8750	TOG	CHRY	5.20E-06	228.3	5.20E-06
8750	TOG	BAP	2.53E-07	252.32	2.53E-07
8750	TOG	BBF	3.45E-06	252.32	3.45E-06
8750	TOG	BKF	3.45E-06	252.32	3.45E-06
8762	TOG	NAPH	1.78E-03	128.17	1.78E-03
8762	TOG	ACY	1.56E-04	152.2	1.56E-04
8762	TOG	ACE	3.43E-05	154.21	3.43E-05
8762	TOG	FLU	6.95E-05	166.22	6.95E-05
8762	TOG	ANT	2.88E-05	178.24	2.88E-05
8762	TOG	PHE	1.84E-04	178.24	1.84E-04
8762	TOG	FTH	4.82E-05	202.26	4.82E-05
8762	TOG	PYR	5.50E-06	202.26	5.50E-06
8762	TOG	BAA	4.64E-06	228.3	4.64E-06
8762	TOG	CHRY	5.20E-06	228.3	5.20E-06
8762	TOG	BAP	2.53E-07	252.32	2.53E-07
8762	TOG	BBF	3.45E-06	252.32	3.45E-06
8762	TOG	BKF	3.45E-06	252.32	3.45E-06
8762	NONHAPTOG	NAPH	1.79E-03	128.17	1.79E-03
8762	NONHAPTOG	ACY	1.56E-04	152.2	1.56E-04
8762	NONHAPTOG	ACE	3.44E-05	154.21	3.44E-05
8762	NONHAPTOG	FLU	6.97E-05	166.22	6.97E-05
8762	NONHAPTOG	ANT	2.89E-05	178.24	2.89E-05
8762	NONHAPTOG	PHE	1.85E-04	178.24	1.85E-04
8762	NONHAPTOG	FTH	4.83E-05	202.26	4.83E-05
8762	NONHAPTOG	PYR	5.52E-06	202.26	5.52E-06
8762	NONHAPTOG	BAA	4.66E-06	228.3	4.66E-06
8762	NONHAPTOG	CHRY	5.22E-06	228.3	5.22E-06
8762	NONHAPTOG	BAP	2.54E-07	252.32	2.54E-07
8762	NONHAPTOG	BBF	3.46E-06	252.32	3.46E-06
8762	NONHAPTOG	BKF	3.46E-06	252.32	3.46E-06
8774	TOG	NAPH	1.40E-02	128.17	1.40E-02
8774	TOG	ACY	7.34E-05	152.2	7.34E-05
8774	TOG	ACE	4.52E-05	154.21	4.52E-05
8774	TOG	FLU	1.69E-04	166.22	1.69E-04

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
8774	TOG	ANT	2.61E-05	178.24	2.61E-05
8774	TOG	PHE	7.32E-04	178.24	7.32E-04
8774	TOG	FTH	3.93E-05	202.26	3.93E-05
8774	TOG	PYR	3.26E-05	202.26	3.26E-05
8774	TOG	BAA	2.58E-07	228.3	2.58E-07
8774	TOG	CHRY	4.30E-07	228.3	4.30E-07
8774	TOG	BGHIP	1.72E-07	276.34	1.72E-07
8774	NONHAPTOG	NAPH	1.68E-02	128.17	1.68E-02
8774	NONHAPTOG	ACY	8.80E-05	152.2	8.80E-05
8774	NONHAPTOG	ACE	5.43E-05	154.21	5.43E-05
8774	NONHAPTOG	FLU	2.02E-04	166.22	2.02E-04
8774	NONHAPTOG	ANT	3.14E-05	178.24	3.14E-05
8774	NONHAPTOG	PHE	8.78E-04	178.24	8.78E-04
8774	NONHAPTOG	FTH	4.72E-05	202.26	4.72E-05
8774	NONHAPTOG	PYR	3.91E-05	202.26	3.91E-05
8774	NONHAPTOG	BAA	3.10E-07	228.3	3.10E-07
8774	NONHAPTOG	CHRY	5.16E-07	228.3	5.16E-07
8774	NONHAPTOG	BGHIP	2.06E-07	276.34	2.06E-07
87710	TOG	NAPH	1.40E-02	128.17	1.40E-02
87710	TOG	ACY	7.34E-05	152.2	7.34E-05
87710	TOG	ACE	4.52E-05	154.21	4.52E-05
87710	TOG	FLU	1.69E-04	166.22	1.69E-04
87710	TOG	ANT	2.61E-05	178.24	2.61E-05
87710	TOG	PHE	7.32E-04	178.24	7.32E-04
87710	TOG	FTH	3.93E-05	202.26	3.93E-05
87710	TOG	PYR	3.26E-05	202.26	3.26E-05
87710	TOG	BAA	2.58E-07	228.3	2.58E-07
87710	TOG	CHRY	4.30E-07	228.3	4.30E-07
87710	TOG	BGHIP	1.72E-07	276.34	1.72E-07
87710	NONHAPTOG	NAPH	1.70E-02	128.17	1.70E-02
87710	NONHAPTOG	ACY	8.88E-05	152.2	8.88E-05
87710	NONHAPTOG	ACE	5.47E-05	154.21	5.47E-05
87710	NONHAPTOG	FLU	2.04E-04	166.22	2.04E-04
87710	NONHAPTOG	ANT	3.16E-05	178.24	3.16E-05
87710	NONHAPTOG	PHE	8.86E-04	178.24	8.86E-04
87710	NONHAPTOG	FTH	4.76E-05	202.26	4.76E-05
87710	NONHAPTOG	PYR	3.94E-05	202.26	3.94E-05
87710	NONHAPTOG	BAA	3.12E-07	228.3	3.12E-07
87710	NONHAPTOG	CHRY	5.20E-07	228.3	5.20E-07
87710	NONHAPTOG	BGHIP	2.08E-07	276.34	2.08E-07
5565B	TOG	NAPH	1.40E-02	128.17	1.40E-02

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
5565B	TOG	ACY	7.34E-05	152.2	7.34E-05
5565B	TOG	ACE	4.52E-05	154.21	4.52E-05
5565B	TOG	FLU	1.69E-04	166.22	1.69E-04
5565B	TOG	ANT	2.61E-05	178.24	2.61E-05
5565B	TOG	PHE	7.32E-04	178.24	7.32E-04
5565B	TOG	FTH	3.93E-05	202.26	3.93E-05
5565B	TOG	PYR	3.26E-05	202.26	3.26E-05
5565B	TOG	BAA	2.58E-07	228.3	2.58E-07
5565B	TOG	CHRY	4.30E-07	228.3	4.30E-07
5565B	TOG	BGHIP	1.72E-07	276.34	1.72E-07
5565B	NONHAPTOG	NAPH	1.74E-02	128.17	1.74E-02
5565B	NONHAPTOG	ACY	9.10E-05	152.2	9.10E-05
5565B	NONHAPTOG	ACE	5.61E-05	154.21	5.61E-05
5565B	NONHAPTOG	FLU	2.09E-04	166.22	2.09E-04
5565B	NONHAPTOG	ANT	3.24E-05	178.24	3.24E-05
5565B	NONHAPTOG	PHE	9.08E-04	178.24	9.08E-04
5565B	NONHAPTOG	FTH	4.87E-05	202.26	4.87E-05
5565B	NONHAPTOG	PYR	4.04E-05	202.26	4.04E-05
5565B	NONHAPTOG	BAA	3.20E-07	228.3	3.20E-07
5565B	NONHAPTOG	CHRY	5.33E-07	228.3	5.33E-07
5565B	NONHAPTOG	BGHIP	2.13E-07	276.34	2.13E-07
8750a	TOG	NAPH	1.78E-03	128.17	1.78E-03
8750a	TOG	ACY	1.56E-04	152.2	1.56E-04
8750a	TOG	ACE	3.43E-05	154.21	3.43E-05
8750a	TOG	FLU	6.95E-05	166.22	6.95E-05
8750a	TOG	ANT	2.88E-05	178.24	2.88E-05
8750a	TOG	PHE	1.84E-04	178.24	1.84E-04
8750a	TOG	FTH	4.82E-05	202.26	4.82E-05
8750a	TOG	PYR	5.50E-06	202.26	5.50E-06
8750a	TOG	BAA	4.64E-06	228.3	4.64E-06
8750a	TOG	CHRY	5.20E-06	228.3	5.20E-06
8750a	TOG	BAP	2.53E-07	252.32	2.53E-07
8750a	TOG	BBF	3.45E-06	252.32	3.45E-06
8750a	TOG	BKF	3.45E-06	252.32	3.45E-06
8750a	NONHAPTOG	NAPH	1.90E-03	128.17	1.90E-03
8750a	NONHAPTOG	ACY	1.67E-04	152.2	1.67E-04
8750a	NONHAPTOG	ACE	3.67E-05	154.21	3.67E-05
8750a	NONHAPTOG	FLU	7.44E-05	166.22	7.44E-05
8750a	NONHAPTOG	ANT	3.08E-05	178.24	3.08E-05
8750a	NONHAPTOG	PHE	1.97E-04	178.24	1.97E-04
8750a	NONHAPTOG	FTH	5.15E-05	202.26	5.15E-05

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
8750a	NONHAPTOG	PYR	5.89E-06	202.26	5.89E-06
8750a	NONHAPTOG	BAA	4.97E-06	228.3	4.97E-06
8750a	NONHAPTOG	CHRY	5.57E-06	228.3	5.57E-06
8750a	NONHAPTOG	BAP	2.71E-07	252.32	2.71E-07
8750a	NONHAPTOG	BBF	3.69E-06	252.32	3.69E-06
8750a	NONHAPTOG	BKF	3.69E-06	252.32	3.69E-06
8750aE	NONHAPTOG	NAPH	1.90E-03	128.17	1.90E-03
8750aE	NONHAPTOG	ACY	1.67E-04	152.2	1.67E-04
8750aE	NONHAPTOG	ACE	3.67E-05	154.21	3.67E-05
8750aE	NONHAPTOG	FLU	7.44E-05	166.22	7.44E-05
8750aE	NONHAPTOG	ANT	3.08E-05	178.24	3.08E-05
8750aE	NONHAPTOG	PHE	1.97E-04	178.24	1.97E-04
8750aE	NONHAPTOG	FTH	5.15E-05	202.26	5.15E-05
8750aE	NONHAPTOG	PYR	5.89E-06	202.26	5.89E-06
8750aE	NONHAPTOG	BAA	4.97E-06	228.3	4.97E-06
8750aE	NONHAPTOG	CHRY	5.57E-06	228.3	5.57E-06
8750aE	NONHAPTOG	BAP	2.71E-07	252.32	2.71E-07
8750aE	NONHAPTOG	BBF	3.69E-06	252.32	3.69E-06
8750aE	NONHAPTOG	BKF	3.69E-06	252.32	3.69E-06
877P0	TOG	NAPH	1.40E-02	128.17	1.40E-02
877P0	TOG	ACY	7.34E-05	152.2	7.34E-05
877P0	TOG	ACE	4.52E-05	154.21	4.52E-05
877P0	TOG	FLU	1.69E-04	166.22	1.69E-04
877P0	TOG	ANT	2.61E-05	178.24	2.61E-05
877P0	TOG	PHE	7.32E-04	178.24	7.32E-04
877P0	TOG	FTH	3.93E-05	202.26	3.93E-05
877P0	TOG	PYR	3.26E-05	202.26	3.26E-05
877P0	TOG	BAA	2.58E-07	228.3	2.58E-07
877P0	TOG	CHRY	4.30E-07	228.3	4.30E-07
877P0	TOG	BGHIP	1.72E-07	276.34	1.72E-07
877P0	NONHAPTOG	NAPH	1.68E-02	128.17	1.68E-02
877P0	NONHAPTOG	ACY	8.80E-05	152.2	8.80E-05
877P0	NONHAPTOG	ACE	5.43E-05	154.21	5.43E-05
877P0	NONHAPTOG	FLU	2.02E-04	166.22	2.02E-04
877P0	NONHAPTOG	ANT	3.14E-05	178.24	3.14E-05
877P0	NONHAPTOG	PHE	8.78E-04	178.24	8.78E-04
877P0	NONHAPTOG	FTH	4.72E-05	202.26	4.72E-05
877P0	NONHAPTOG	PYR	3.91E-05	202.26	3.91E-05
877P0	NONHAPTOG	BAA	3.10E-07	228.3	3.10E-07
877P0	NONHAPTOG	CHRY	5.16E-07	228.3	5.16E-07
877P0	NONHAPTOG	BGHIP	2.06E-07	276.34	2.06E-07

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
91102	PM2_5	PANT	4.96E-04	1.0	4.96E-04
91102	PM2_5	PBAA	6.27E-04	1.0	6.27E-04
91102	PM2_5	PBAP	9.88E-05	1.0	9.88E-05
91102	PM2_5	PBBF	1.70E-04	1.0	1.70E-04
91102	PM2_5	PBGHIP	3.34E-04	1.0	3.34E-04
91102	PM2_5	PBKF	1.71E-04	1.0	1.71E-04
91102	PM2_5	PCHRY	6.27E-04	1.0	6.27E-04
91102	PM2_5	PFTH	5.88E-04	1.0	5.88E-04
91102	PM2_5	PICDP	1.93E-04	1.0	1.93E-04
91102	PM2_5	PPHE	4.96E-04	1.0	4.96E-04
91102	PM2_5	PPYR	6.13E-04	1.0	6.13E-04
91103	PM2_5	PNAPH	4.20E-04	1.0	4.20E-04
91103	PM2_5	PACE	2.00E-05	1.0	2.00E-05
91103	PM2_5	PANT	2.00E-05	1.0	2.00E-05
91103	PM2_5	PBAA	1.20E-04	1.0	1.20E-04
91103	PM2_5	PBAP	1.30E-04	1.0	1.30E-04
91103	PM2_5	PBGHIP	2.00E-05	1.0	2.00E-05
91103	PM2_5	PCHRY	1.70E-04	1.0	1.70E-04
91103	PM2_5	PFTH	5.00E-04	1.0	5.00E-04
91103	PM2_5	PPHE	3.00E-05	1.0	3.00E-05
91103	PM2_5	PPYR	2.90E-04	1.0	2.90E-04
91103	PM2_5	PDAHA	8.00E-05	1.0	8.00E-05
91103	PM2_5	PBKF	5.00E-05	1.0	5.00E-05
91103	PM2_5	PBBF	1.30E-04	1.0	1.30E-04
91105	PM2_5	PANT	9.72E-06	1.0	9.72E-06
91105	PM2_5	PBAA	1.05E-04	1.0	1.05E-04
91105	PM2_5	PBBF	6.67E-05	1.0	6.67E-05
91105	PM2_5	PBKF	5.45E-05	1.0	5.45E-05
91105	PM2_5	PBGHIP	3.32E-05	1.0	3.32E-05
91105	PM2_5	PBAP	5.27E-05	1.0	5.27E-05
91105	PM2_5	PCHRY	1.13E-04	1.0	1.13E-04
91105	PM2_5	PFTH	2.35E-04	1.0	2.35E-04
91105	PM2_5	PFLU	2.05E-04	1.0	2.05E-04
91105	PM2_5	PPHE	3.08E-05	1.0	3.08E-05
91105	PM2_5	PPYR	2.37E-04	1.0	2.37E-04
91105	PM2_5	PICDP	3.73E-05	1.0	3.73E-05
91106	PM2_5	PFLU	1.34E-04	1.0	1.34E-04
91106	PM2_5	PANT	6.63E-05	1.0	6.63E-05
91106	PM2_5	PPHE	1.07E-03	1.0	1.07E-03
91106	PM2_5	PFTH	1.21E-04	1.0	1.21E-04
91106	PM2_5	PPYR	1.16E-04	1.0	1.16E-04

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
91106	PM2_5	PBAA	1.99E-06	1.0	1.99E-06
91106	PM2_5	PCHRY	6.24E-06	1.0	6.24E-06
91106	PM2_5	PBAP	8.24E-06	1.0	8.24E-06
91106	PM2_5	PBBF	3.49E-06	1.0	3.49E-06
91106	PM2_5	PBKF	3.49E-06	1.0	3.49E-06
91106	PM2_5	PBGHIP	4.99E-07	1.0	4.99E-07
91106	PM2_5	PICDP	1.25E-06	1.0	1.25E-06
91106	PM2_5	PDAHA	2.50E-06	1.0	2.50E-06
91106	PMFINE	PFLU	0.000273507	1.0	2.74E-04
91106	PMFINE	PANT	0.000135051	1.0	1.35E-04
91106	PMFINE	PPHE	0.002178973	1.0	2.18E-03
91106	PMFINE	PFTH	0.000247404	1.0	2.47E-04
91106	PMFINE	PPYR	0.00023719	1.0	2.37E-04
91106	PMFINE	PBAA	4.06E-06	1.0	4.06E-06
91106	PMFINE	PCHRY	1.27E-05	1.0	1.27E-05
91106	PMFINE	PBAP	1.68E-05	1.0	1.68E-05
91106	PMFINE	PBBF	7.12E-06	1.0	7.12E-06
91106	PMFINE	PBKF	7.12E-06	1.0	7.12E-06
91106	PMFINE	PBGHIP	1.02E-06	1.0	1.02E-06
91106	PMFINE	PICDP	2.54E-06	1.0	2.54E-06
91106	PMFINE	PDAHA	5.08E-06	1.0	5.08E-06
91108	PM2_5	PBAA	9.80E-07	1.0	9.80E-07
91108	PM2_5	PFTH	1.79E-06	1.0	1.79E-06
91108	PM2_5	PPYR	9.40E-07	1.0	9.40E-07
91109	PM2_5	PANT	4.96E-04	1.0	4.96E-04
91109	PM2_5	PBAA	6.27E-04	1.0	6.27E-04
91109	PM2_5	PBAP	9.88E-05	1.0	9.88E-05
91109	PM2_5	PBBF	1.70E-04	1.0	1.70E-04
91109	PM2_5	PBGHIP	3.34E-04	1.0	3.34E-04
91109	PM2_5	PBKF	1.71E-04	1.0	1.71E-04
91109	PM2_5	PCHRY	6.27E-04	1.0	6.27E-04
91109	PM2_5	PFTH	5.88E-04	1.0	5.88E-04
91109	PM2_5	PICDP	1.93E-04	1.0	1.93E-04
91109	PM2_5	PPHE	4.96E-04	1.0	4.96E-04
91109	PM2_5	PPYR	6.13E-04	1.0	6.13E-04
91110	PM2_5	PACE	6.04E-08	1.0	6.04E-08
91110	PM2_5	PACY	8.81E-08	1.0	8.81E-08
91110	PM2_5	PANT	1.88E-08	1.0	1.88E-08
91110	PM2_5	PBAA	1.42E-07	1.0	1.42E-07
91110	PM2_5	PBAP	5.10E-06	1.0	5.10E-06
91110	PM2_5	PBBF	1.65E-06	1.0	1.65E-06

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
91110	PM2_5	PBGHIP	1.03E-06	1.0	1.03E-06
91110	PM2_5	PBKF	7.33E-08	1.0	7.33E-08
91110	PM2_5	PCHRY	5.03E-06	1.0	5.03E-06
91110	PM2_5	PDAHA	3.84E-07	1.0	3.84E-07
91110	PM2_5	PFLU	1.29E-07	1.0	1.29E-07
91110	PM2_5	PFTH	4.34E-06	1.0	4.34E-06
91110	PM2_5	PICDP	1.41E-06	1.0	1.41E-06
91110	PM2_5	PNAPH	4.16E-05	1.0	4.16E-05
91110	PM2_5	PPHE	1.31E-05	1.0	1.31E-05
91110	PM2_5	PPYR	9.00E-06	1.0	9.00E-06
91112	PM2_5	PCHRY	7.36E-06	1.0	7.36E-06
91112	PM2_5	PFTH	2.64E-05	1.0	2.64E-05
91112	PM2_5	PPHE	4.83E-05	1.0	4.83E-05
91112	PM2_5	PPYR	2.50E-05	1.0	2.50E-05
91112	PM2_5	PFLU	3.87E-05	1.0	3.87E-05
91112	PM2_5	PNAPH	1.26E-02	1.0	1.26E-02
91112	PM2_5	PACE	2.41E-05	1.0	2.41E-05
91112	PM2_5	PACY	9.74E-06	1.0	9.74E-06
91112	PM2_5	PANT	6.58E-06	1.0	6.58E-06
91113	PM2_5	PNAPH	7.17E-05	1.0	7.17E-05
91113	PM2_5	PACY	2.13E-05	1.0	2.13E-05
91113	PM2_5	PANT	2.21E-05	1.0	2.21E-05
91113	PM2_5	PPHE	7.73E-05	1.0	7.73E-05
91113	PM2_5	PFTH	7.80E-05	1.0	7.80E-05
91113	PM2_5	PPYR	8.45E-05	1.0	8.45E-05
91113	PM2_5	PBAA	2.03E-04	1.0	2.03E-04
91113	PM2_5	PCHRY	1.71E-04	1.0	1.71E-04
91113	PM2_5	PBAP	5.08E-04	1.0	5.08E-04
91113	PM2_5	PBBF	2.48E-04	1.0	2.48E-04
91113	PM2_5	PBKF	2.48E-04	1.0	2.48E-04
91113	PM2_5	PBGHIP	1.38E-03	1.0	1.38E-03
91113	PM2_5	PICDP	5.16E-04	1.0	5.16E-04
91113	PM2_5	PDAHA	1.19E-05	1.0	1.19E-05
91113	PMFINE	PNAPH	1.78E-04	1.0	1.78E-04
91113	PMFINE	PACY	5.31E-05	1.0	5.31E-05
91113	PMFINE	PANT	5.50E-05	1.0	5.50E-05
91113	PMFINE	PPHE	1.92E-04	1.0	1.92E-04
91113	PMFINE	PFTH	1.94E-04	1.0	1.94E-04
91113	PMFINE	PPYR	2.10E-04	1.0	2.10E-04
91113	PMFINE	PBAA	5.05E-04	1.0	5.05E-04
91113	PMFINE	PCHRY	4.27E-04	1.0	4.27E-04

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
91113	PMFINE	PBAP	1.26E-03	1.0	1.26E-03
91113	PMFINE	PBBF	6.16E-04	1.0	6.16E-04
91113	PMFINE	PBKF	6.16E-04	1.0	6.16E-04
91113	PMFINE	PBGHIP	3.42E-03	1.0	3.42E-03
91113	PMFINE	PICDP	1.28E-03	1.0	1.28E-03
91113	PMFINE	PDAHA	2.96E-05	1.0	2.96E-05
91114	PM2_5	PACE	9.87E-05	1.0	9.87E-05
91114	PM2_5	PACY	1.28E-04	1.0	1.28E-04
91114	PM2_5	PANT	5.20E-04	1.0	5.20E-04
91114	PM2_5	PBAA	3.21E-04	1.0	3.21E-04
91114	PM2_5	PBGHIP	7.56E-05	1.0	7.56E-05
91114	PM2_5	PBAP	1.24E-04	1.0	1.24E-04
91114	PM2_5	PBBF	1.78E-04	1.0	1.78E-04
91114	PM2_5	PBKF	4.42E-05	1.0	4.42E-05
91114	PM2_5	PCHRY	3.03E-04	1.0	3.03E-04
91114	PM2_5	PFTH	1.03E-03	1.0	1.03E-03
91114	PM2_5	PFLU	2.67E-04	1.0	2.67E-04
91114	PM2_5	PICDP	1.15E-04	1.0	1.15E-04
91114	PM2_5	PNAPH	8.02E-05	1.0	8.02E-05
91114	PM2_5	PPHE	1.73E-03	1.0	1.73E-03
91114	PM2_5	PPYR	9.00E-04	1.0	9.00E-04
91115	PM2_5	PBAP	3.30E-06	1.0	3.30E-06
91115	PM2_5	PFTH	1.06E-05	1.0	1.06E-05
91115	PM2_5	PNAPH	2.77E-02	1.0	2.77E-02
91115	PM2_5	PPYR	9.92E-06	1.0	9.92E-06
91116	PM2_5	PBAA	4.62E-05	1.0	4.62E-05
91116	PM2_5	PBAP	2.98E-05	1.0	2.98E-05
91116	PM2_5	PBGHIP	3.76E-05	1.0	3.76E-05
91116	PM2_5	PBKF	2.59E-05	1.0	2.59E-05
91116	PM2_5	PFTH	3.68E-05	1.0	3.68E-05
91116	PM2_5	PPYR	7.29E-05	1.0	7.29E-05
91117	PM2_5	PACE	2.83E-03	1.0	2.83E-03
91117	PM2_5	PACY	2.29E-06	1.0	2.29E-06
91117	PM2_5	PANT	1.82E-06	1.0	1.82E-06
91117	PM2_5	PBAA	1.49E-05	1.0	1.49E-05
91117	PM2_5	PBGHIP	1.02E-05	1.0	1.02E-05
91117	PM2_5	PCHRY	4.23E-06	1.0	4.23E-06
91117	PM2_5	PDAHA	3.61E-06	1.0	3.61E-06
91117	PM2_5	PFLU	9.81E-06	1.0	9.81E-06
91117	PM2_5	PFTH	8.80E-06	1.0	8.80E-06
91117	PM2_5	PNAPH	3.58E-03	1.0	3.58E-03

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
91117	PM2_5	PPHE	1.91E-05	1.0	1.91E-05
91117	PM2_5	PPYR	8.12E-06	1.0	8.12E-06
91119	PM2_5	PACE	5.31E-08	1.0	5.31E-08
91119	PM2_5	PACY	8.70E-06	1.0	8.70E-06
91119	PM2_5	PANT	1.34E-06	1.0	1.34E-06
91119	PM2_5	PBAA	2.47E-07	1.0	2.47E-07
91119	PM2_5	PBAP	2.51E-08	1.0	2.51E-08
91119	PM2_5	PBBF	8.51E-08	1.0	8.51E-08
91119	PM2_5	PBGHIP	5.07E-08	1.0	5.07E-08
91119	PM2_5	PBKF	3.64E-08	1.0	3.64E-08
91119	PM2_5	PCHRY	2.15E-07	1.0	2.15E-07
91119	PM2_5	PDAHA	3.49E-08	1.0	3.49E-08
91119	PM2_5	PFLU	7.10E-07	1.0	7.10E-07
91119	PM2_5	PFTH	2.35E-06	1.0	2.35E-06
91119	PM2_5	PICDP	2.91E-08	1.0	2.91E-08
91119	PM2_5	PNAPH	8.59E-05	1.0	8.59E-05
91119	PM2_5	PPHE	1.89E-05	1.0	1.89E-05
91119	PM2_5	PPYR	9.76E-07	1.0	9.76E-07
91122	PM2_5	PNAPH	7.17E-05	1.0	7.17E-05
91122	PM2_5	PACY	2.13E-05	1.0	2.13E-05
91122	PM2_5	PANT	2.21E-05	1.0	2.21E-05
91122	PM2_5	PPHE	7.73E-05	1.0	7.73E-05
91122	PM2_5	PFTH	7.80E-05	1.0	7.80E-05
91122	PM2_5	PPYR	8.45E-05	1.0	8.45E-05
91122	PM2_5	PBAA	2.03E-04	1.0	2.03E-04
91122	PM2_5	PCHRY	1.71E-04	1.0	1.71E-04
91122	PM2_5	PBAP	5.08E-04	1.0	5.08E-04
91122	PM2_5	PBBF	2.48E-04	1.0	2.48E-04
91122	PM2_5	PBKF	2.48E-04	1.0	2.48E-04
91122	PM2_5	PBGHIP	1.38E-03	1.0	1.38E-03
91122	PM2_5	PICDP	5.16E-04	1.0	5.16E-04
91122	PM2_5	PDAHA	1.19E-05	1.0	1.19E-05
91122	PMFINE	PNAPH	2.86E-04	1.0	2.86E-04
91122	PMFINE	PACY	8.50E-05	1.0	8.50E-05
91122	PMFINE	PANT	8.81E-05	1.0	8.81E-05
91122	PMFINE	PPHE	3.08E-04	1.0	3.08E-04
91122	PMFINE	PFTH	3.10E-04	1.0	3.10E-04
91122	PMFINE	PPYR	3.37E-04	1.0	3.37E-04
91122	PMFINE	PBAA	8.07E-04	1.0	8.07E-04
91122	PMFINE	PCHRY	6.83E-04	1.0	6.83E-04
91122	PMFINE	PBAP	2.02E-03	1.0	2.02E-03

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
1122	PMFINE	PBBF	9.86E-04	1.0	9.86E-04
91122	PMFINE	PBKF	9.86E-04	1.0	9.86E-04
91122	PMFINE	PBGHIP	5.48E-03	1.0	5.48E-03
91122	PMFINE	PICDP	2.06E-03	1.0	2.06E-03
91122	PMFINE	PDAHA	4.73E-05	1.0	4.73E-05
91125	PM2_5	PACE	6.21E-09	1.0	6.21E-09
91125	PM2_5	PACY	3.79E-09	1.0	3.79E-09
91125	PM2_5	PANT	1.02E-07	1.0	1.02E-07
91125	PM2_5	PBAA	4.14E-08	1.0	4.14E-08
91125	PM2_5	PBAP	1.94E-07	1.0	1.94E-07
91125	PM2_5	PBBF	1.18E-07	1.0	1.18E-07
91125	PM2_5	PBGHIP	5.23E-07	1.0	5.23E-07
91125	PM2_5	PBKF	8.84E-08	1.0	8.84E-08
91125	PM2_5	PCHRY	7.12E-08	1.0	7.12E-08
91125	PM2_5	PDAHA	2.52E-10	1.0	2.52E-10
91125	PM2_5	PFLU	4.62E-08	1.0	4.62E-08
91125	PM2_5	PFTH	8.74E-08	1.0	8.74E-08
91125	PM2_5	PICDP	1.88E-07	1.0	1.88E-07
91125	PM2_5	PNAPH	9.24E-08	1.0	9.24E-08
91125	PM2_5	PPHE	1.67E-07	1.0	1.67E-07
91125	PM2_5	PPYR	8.25E-07	1.0	8.25E-07
91126	PM2_5	PACE	4.40E-07	1.0	4.40E-07
91126	PM2_5	PACY	5.69E-06	1.0	5.69E-06
91126	PM2_5	PANT	4.91E-06	1.0	4.91E-06
91126	PM2_5	PBAA	2.33E-07	1.0	2.33E-07
91126	PM2_5	PBAP	2.46E-08	1.0	2.46E-08
91126	PM2_5	PBBF	1.87E-06	1.0	1.87E-06
91126	PM2_5	PBGHIP	1.55E-07	1.0	1.55E-07
91126	PM2_5	PBKF	1.87E-06	1.0	1.87E-06
91126	PM2_5	PCHRY	5.56E-06	1.0	5.56E-06
91126	PM2_5	PFLU	1.24E-06	1.0	1.24E-06
91126	PM2_5	PFTH	1.16E-05	1.0	1.16E-05
91126	PM2_5	PICDP	4.40E-08	1.0	4.40E-08
91126	PM2_5	PNAPH	2.97E-04	1.0	2.97E-04
91126	PM2_5	PPHE	7.37E-06	1.0	7.37E-06
91126	PM2_5	PPYR	2.20E-06	1.0	2.20E-06
91127	PM2_5	PACE	5.33E-06	1.0	5.33E-06
91127	PM2_5	PACY	3.32E-05	1.0	3.32E-05
91127	PM2_5	PANT	1.39E-05	1.0	1.39E-05
91127	PM2_5	PBAA	7.16E-06	1.0	7.16E-06
91127	PM2_5	PBAP	3.90E-06	1.0	3.90E-06

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
91127	PM2_5	PBBF	2.40E-06	1.0	2.40E-06
91127	PM2_5	PBGHIP	1.34E-06	1.0	1.34E-06
91127	PM2_5	PBKF	4.73E-09	1.0	4.73E-09
91127	PM2_5	PCHRY	1.06E-05	1.0	1.06E-05
91127	PM2_5	PDAHA	6.48E-07	1.0	6.48E-07
91127	PM2_5	PFLU	1.82E-05	1.0	1.82E-05
91127	PM2_5	PFTH	1.22E-05	1.0	1.22E-05
91127	PM2_5	PICDP	8.03E-07	1.0	8.03E-07
91127	PM2_5	PNAPH	7.15E-05	1.0	7.15E-05
91127	PM2_5	PPHE	3.08E-05	1.0	3.08E-05
91127	PM2_5	PPYR	1.52E-05	1.0	1.52E-05
91132	PM2_5	PACE	3.80E-03	1.0	3.80E-03
91132	PM2_5	PACY	3.71E-04	1.0	3.71E-04
91132	PM2_5	PANT	1.63E-03	1.0	1.63E-03
91132	PM2_5	PBAA	8.80E-04	1.0	8.80E-04
91132	PM2_5	PBAP	3.72E-04	1.0	3.72E-04
91132	PM2_5	PBBF	8.12E-04	1.0	8.12E-04
91132	PM2_5	PBGHIP	2.27E-04	1.0	2.27E-04
91132	PM2_5	PBKF	5.56E-04	1.0	5.56E-04
91132	PM2_5	PCHRY	1.59E-03	1.0	1.59E-03
91132	PM2_5	PDAHA	1.02E-04	1.0	1.02E-04
91132	PM2_5	PFLU	1.12E-03	1.0	1.12E-03
91132	PM2_5	PFTH	5.16E-03	1.0	5.16E-03
91132	PM2_5	PICDP	1.93E-04	1.0	1.93E-04
91132	PM2_5	PNAPH	1.58E-03	1.0	1.58E-03
91132	PM2_5	PPHE	6.51E-03	1.0	6.51E-03
91132	PM2_5	PPYR	3.57E-03	1.0	3.57E-03
91135	PM2_5	PCHRY	4.30E-04	1.0	4.30E-04
91135	PM2_5	PANT	5.71E-05	1.0	5.71E-05
91135	PM2_5	PFTH	2.48E-04	1.0	2.48E-04
91135	PM2_5	PPHE	2.98E-04	1.0	2.98E-04
91135	PM2_5	PPYR	1.78E-04	1.0	1.78E-04
91136	PM2_5	PNAPH	6.54E-02	1.0	6.54E-02
91136	PM2_5	PACY	4.88E-03	1.0	4.88E-03
91136	PM2_5	PBAA	1.51E-02	1.0	1.51E-02
91136	PM2_5	PBAP	2.31E-04	1.0	2.31E-04
91136	PM2_5	PCHRY	2.87E-04	1.0	2.87E-04
91136	PM2_5	PFTH	5.56E-04	1.0	5.56E-04
91136	PM2_5	PICDP	1.00E-04	1.0	1.00E-04
91136	PM2_5	PPHE	2.41E-03	1.0	2.41E-03
91136	PM2_5	PPYR	2.43E-04	1.0	2.43E-04

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
91137	PM2_5	PACE	3.80E-03	1.0	3.80E-03
91137	PM2_5	PACY	3.71E-04	1.0	3.71E-04
91137	PM2_5	PANT	1.63E-03	1.0	1.63E-03
91137	PM2_5	PBAA	8.80E-04	1.0	8.80E-04
91137	PM2_5	PBAP	3.72E-04	1.0	3.72E-04
91137	PM2_5	PBBF	8.12E-04	1.0	8.12E-04
91137	PM2_5	PBGHIP	2.27E-04	1.0	2.27E-04
91137	PM2_5	PBKF	5.56E-04	1.0	5.56E-04
91137	PM2_5	PCHRY	1.59E-03	1.0	1.59E-03
91137	PM2_5	PDAHA	1.02E-04	1.0	1.02E-04
91137	PM2_5	PFLU	1.12E-03	1.0	1.12E-03
91137	PM2_5	PFTH	5.16E-03	1.0	5.16E-03
91137	PM2_5	PICDP	1.93E-04	1.0	1.93E-04
91137	PM2_5	PNAPH	1.58E-03	1.0	1.58E-03
91137	PM2_5	PPHE	6.51E-03	1.0	6.51E-03
91137	PM2_5	PPYR	3.57E-03	1.0	3.57E-03
91138	PM2_5	PACE	5.33E-06	1.0	5.33E-06
91138	PM2_5	PACY	3.32E-05	1.0	3.32E-05
91138	PM2_5	PANT	1.39E-05	1.0	1.39E-05
91138	PM2_5	PBAA	7.16E-06	1.0	7.16E-06
91138	PM2_5	PBAP	3.90E-06	1.0	3.90E-06
91138	PM2_5	PBBF	2.40E-06	1.0	2.40E-06
91138	PM2_5	PBGHIP	1.34E-06	1.0	1.34E-06
91138	PM2_5	PBKF	4.73E-09	1.0	4.73E-09
91138	PM2_5	PCHRY	1.06E-05	1.0	1.06E-05
91138	PM2_5	PDAHA	6.48E-07	1.0	6.48E-07
91138	PM2_5	PFLU	1.82E-05	1.0	1.82E-05
91138	PM2_5	PFTH	1.22E-05	1.0	1.22E-05
91138	PM2_5	PICDP	8.03E-07	1.0	8.03E-07
91138	PM2_5	PNAPH	7.15E-05	1.0	7.15E-05
91138	PM2_5	PPHE	3.08E-05	1.0	3.08E-05
91138	PM2_5	PPYR	1.52E-05	1.0	1.52E-05
91139	PM2_5	PACY	3.70E-03	1.0	3.70E-03
91139	PM2_5	PBAP	4.10E-04	1.0	4.10E-04
91139	PM2_5	PBBF	2.30E-04	1.0	2.30E-04
91139	PM2_5	PBKF	3.10E-04	1.0	3.10E-04
91139	PM2_5	PCHRY	1.60E-04	1.0	1.60E-04
91139	PM2_5	PFLU	1.00E-04	1.0	1.00E-04
91139	PM2_5	PPHE	3.00E-04	1.0	3.00E-04
91140	PM2_5	PACY	7.43E-03	1.0	7.43E-03
91140	PM2_5	PANT	2.14E-05	1.0	2.14E-05

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
91140	PM2_5	PBAA	7.71E-05	1.0	7.71E-05
91140	PM2_5	PBAP	1.32E-03	1.0	1.32E-03
91140	PM2_5	PBBF	4.35E-04	1.0	4.35E-04
91140	PM2_5	PBGHIP	7.04E-06	1.0	7.04E-06
91140	PM2_5	PBKF	4.35E-04	1.0	4.35E-04
91140	PM2_5	PCHRY	1.57E-04	1.0	1.57E-04
91140	PM2_5	PDAHA	6.13E-07	1.0	6.13E-07
91140	PM2_5	PFTH	4.90E-03	1.0	4.90E-03
91140	PM2_5	PICDP	6.13E-07	1.0	6.13E-07
91140	PM2_5	PPHE	1.19E-03	1.0	1.19E-03
91140	PM2_5	PPYR	7.42E-03	1.0	7.42E-03
91141	PM2_5	PANT	4.13E-04	1.0	4.13E-04
91141	PM2_5	PBAA	1.53E-06	1.0	1.53E-06
91141	PM2_5	PBAP	7.77E-03	1.0	7.77E-03
91141	PM2_5	PBGHIP	1.04E-02	1.0	1.04E-02
91141	PM2_5	PFTH	5.14E-04	1.0	5.14E-04
91141	PM2_5	PNAPH	5.37E-06	1.0	5.37E-06
91141	PM2_5	PPHE	9.36E-03	1.0	9.36E-03
91141	PM2_5	PPYR	7.48E-03	1.0	7.48E-03
91145	PM2_5	PNAPH	1.36E-02	1.0	1.36E-02
91145	PM2_5	PACE	3.27E-03	1.0	3.27E-03
91145	PM2_5	PACY	4.90E-03	1.0	4.90E-03
91145	PM2_5	PANT	4.13E-04	1.0	7.60E-06
91145	PM2_5	PBAA	2.79E-03	1.0	2.79E-03
91145	PM2_5	PBAP	7.77E-03	1.0	3.74E-05
91145	PM2_5	PCHRY	3.70E-04	1.0	3.70E-04
91145	PM2_5	PFTH	5.53E-04	1.0	5.53E-04
91145	PM2_5	PFLU	6.63E-04	1.0	6.63E-04
91145	PM2_5	PICDP	1.50E-04	1.0	1.50E-04
91145	PM2_5	PPHE	9.36E-03	1.0	2.41E-03
91145	PM2_5	PPYR	7.48E-03	1.0	2.43E-04
91145	PM2_5	PBGHIP	1.04E-02	1.0	1.04E-02
91146	PM2_5	PANT	2.70E-05	1.0	2.70E-05
91146	PM2_5	PBAA	9.90E-05	1.0	9.90E-05
91146	PM2_5	PBAP	3.60E-05	1.0	3.60E-05
91146	PM2_5	PCHRY	5.50E-05	1.0	5.50E-05
91146	PM2_5	PFLU	3.99E-04	1.0	3.99E-04
91146	PM2_5	PFTH	5.00E-05	1.0	5.00E-05
91146	PM2_5	PICDP	6.00E-06	1.0	6.00E-06
91146	PM2_5	PPHE	2.71E-04	1.0	2.71E-04
91146	PM2_5	PPYR	7.60E-05	1.0	7.60E-05

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
91146	PM2_5	PBKF	1.70E-05	1.0	1.70E-05
91146	PM2_5	PBBF	1.07E-04	1.0	1.07E-04
91147	PM2_5	PACE	3.10E-02	1.0	3.10E-02
91147	PM2_5	PACY	1.14E-02	1.0	1.14E-02
91147	PM2_5	PANT	1.09E-03	1.0	1.09E-03
91147	PM2_5	PBAA	1.12E-03	1.0	1.12E-03
91147	PM2_5	PBAP	1.73E-03	1.0	1.73E-03
91147	PM2_5	PBBF	2.72E-03	1.0	2.72E-03
91147	PM2_5	PBGHIP	1.18E-03	1.0	1.18E-03
91147	PM2_5	PBKF	3.82E-03	1.0	3.82E-03
91147	PM2_5	PCHRY	1.43E-03	1.0	1.43E-03
91147	PM2_5	PDAHA	5.45E-04	1.0	5.45E-04
91147	PM2_5	PFLU	2.33E-03	1.0	2.33E-03
91147	PM2_5	PFTH	3.07E-03	1.0	3.07E-03
91147	PM2_5	PICDP	1.45E-03	1.0	1.45E-03
91147	PM2_5	PNAPH	4.91E-03	1.0	4.91E-03
91147	PM2_5	PPHE	1.62E-03	1.0	1.62E-03
91147	PM2_5	PPYR	1.70E-03	1.0	1.70E-03
91148	PM2_5	PANT	4.34E-04	1.0	4.34E-04
91148	PM2_5	PBAA	2.20E-04	1.0	2.20E-04
91148	PM2_5	PFTH	3.06E-04	1.0	3.06E-04
91148	PM2_5	PPHE	4.34E-04	1.0	4.34E-04
91155	PM2_5	PANT	3.98E-03	1.0	3.98E-03
91155	PM2_5	PBAA	1.46E-03	1.0	1.46E-03
91155	PM2_5	PBAP	1.17E-03	1.0	1.17E-03
91155	PM2_5	PBBF	1.25E-03	1.0	1.25E-03
91155	PM2_5	PBKF	1.25E-03	1.0	1.25E-03
91155	PM2_5	PCHRY	1.42E-03	1.0	1.42E-03
91155	PM2_5	PDAHA	1.87E-03	1.0	1.87E-03
91155	PM2_5	PFLU	9.38E-06	1.0	9.38E-06
91155	PM2_5	PFTH	3.12E-03	1.0	3.12E-03
91155	PM2_5	PICDP	1.25E-03	1.0	1.25E-03
91155	PM2_5	PPHE	5.65E-03	1.0	5.65E-03
91155	PM2_5	PPYR	3.12E-03	1.0	3.12E-03
91156	PM2_5	PBAA	5.46E-05	1.0	5.46E-05
91156	PM2_5	PBAP	4.50E-05	1.0	4.50E-05
91156	PM2_5	PBBF	6.49E-05	1.0	6.49E-05
91156	PM2_5	PBGHIP	7.25E-05	1.0	7.25E-05
91156	PM2_5	PBKF	6.49E-05	1.0	6.49E-05
91156	PM2_5	PCHRY	4.31E-05	1.0	4.31E-05
91156	PM2_5	PDAHA	2.38E-05	1.0	2.38E-05

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
91156	PM2_5	PFTH	2.69E-05	1.0	2.69E-05
91156	PM2_5	PICDP	3.51E-05	1.0	3.51E-05
91156	PM2_5	PPHE	5.25E-05	1.0	5.25E-05
91156	PM2_5	PPYR	3.63E-05	1.0	3.63E-05
91157	PM2_5	PACE	9.91E-09	1.0	9.91E-09
91157	PM2_5	PACY	5.26E-09	1.0	5.26E-09
91157	PM2_5	PANT	3.08E-08	1.0	3.08E-08
91157	PM2_5	PBAA	6.64E-07	1.0	6.64E-07
91157	PM2_5	PBAP	3.32E-08	1.0	3.32E-08
91157	PM2_5	PBBF	2.41E-07	1.0	2.41E-07
91157	PM2_5	PBGHIP	2.74E-07	1.0	2.74E-07
91157	PM2_5	PBKF	2.04E-07	1.0	2.04E-07
91157	PM2_5	PCHRY	3.28E-07	1.0	3.28E-07
91157	PM2_5	PDAHA	4.16E-08	1.0	4.16E-08
91157	PM2_5	PFLU	6.84E-09	1.0	6.84E-09
91157	PM2_5	PFTH	1.69E-06	1.0	1.69E-06
91157	PM2_5	PICDP	2.72E-07	1.0	2.72E-07
91157	PM2_5	PNAPH	1.45E-08	1.0	1.45E-08
91157	PM2_5	PPHE	3.01E-07	1.0	3.01E-07
91157	PM2_5	PPYR	1.24E-06	1.0	1.24E-06
91158	PM2_5	PACE	3.80E-03	1.0	3.80E-03
91158	PM2_5	PACY	3.71E-04	1.0	3.71E-04
91158	PM2_5	PANT	1.63E-03	1.0	1.63E-03
91158	PM2_5	PBAA	8.80E-04	1.0	8.80E-04
91158	PM2_5	PBAP	3.72E-04	1.0	3.72E-04
91158	PM2_5	PBBF	8.12E-04	1.0	8.12E-04
91158	PM2_5	PBGHIP	2.27E-04	1.0	2.27E-04
91158	PM2_5	PBKF	5.56E-04	1.0	5.56E-04
91158	PM2_5	PCHRY	1.59E-03	1.0	1.59E-03
91158	PM2_5	PDAHA	1.02E-04	1.0	1.02E-04
91158	PM2_5	PFLU	1.12E-03	1.0	1.12E-03
91158	PM2_5	PFTH	5.16E-03	1.0	5.16E-03
91158	PM2_5	PICDP	1.93E-04	1.0	1.93E-04
91158	PM2_5	PNAPH	1.58E-03	1.0	1.58E-03
91158	PM2_5	PPHE	6.51E-03	1.0	6.51E-03
91158	PM2_5	PPYR	3.57E-03	1.0	3.57E-03
91159	PM2_5	PACE	2.84E-07	1.0	2.84E-07
91159	PM2_5	PACY	2.42E-06	1.0	2.42E-06
91159	PM2_5	PANT	3.18E-07	1.0	3.18E-07
91159	PM2_5	PBAA	2.38E-08	1.0	2.38E-08
91159	PM2_5	PBAP	2.14E-09	1.0	2.14E-09

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
91159	PM2_5	PBBF	1.21E-08	1.0	1.21E-08
91159	PM2_5	PBGHIP	9.07E-09	1.0	9.07E-09
91159	PM2_5	PBKF	8.83E-09	1.0	8.83E-09
91159	PM2_5	PCHRY	4.14E-08	1.0	4.14E-08
91159	PM2_5	PDAHA	6.28E-10	1.0	6.28E-10
91159	PM2_5	PFLU	1.88E-06	1.0	1.88E-06
91159	PM2_5	PFTH	2.74E-06	1.0	2.74E-06
91159	PM2_5	PICDP	1.70E-09	1.0	1.70E-09
91159	PM2_5	PNAPH	2.56E-05	1.0	2.56E-05
91159	PM2_5	PPHE	5.92E-06	1.0	5.92E-06
91159	PM2_5	PPYR	3.26E-06	1.0	3.26E-06
91162	PM2_5	PFLU	1.34E-04	1.0	1.34E-04
91162	PM2_5	PANT	6.63E-05	1.0	6.63E-05
91162	PM2_5	PPHE	1.07E-03	1.0	1.07E-03
91162	PM2_5	PFTH	1.21E-04	1.0	1.21E-04
91162	PM2_5	PPYR	1.16E-04	1.0	1.16E-04
91162	PM2_5	PBAA	1.99E-06	1.0	1.99E-06
91162	PM2_5	PCHRY	6.24E-06	1.0	6.24E-06
91162	PM2_5	PBAP	8.24E-06	1.0	8.24E-06
91162	PM2_5	PBBF	3.49E-06	1.0	3.49E-06
91162	PM2_5	PBKF	3.49E-06	1.0	3.49E-06
91162	PM2_5	PBGHIP	4.99E-07	1.0	4.99E-07
91162	PM2_5	PICDP	1.25E-06	1.0	1.25E-06
91162	PM2_5	PDAHA	2.50E-06	1.0	2.50E-06
91162	PMFINE	PFLU	1.12E-03	1.0	1.12E-03
91162	PMFINE	PANT	5.54E-04	1.0	5.54E-04
91162	PMFINE	PPHE	8.93E-03	1.0	8.93E-03
91162	PMFINE	PFTH	1.01E-03	1.0	1.01E-03
91162	PMFINE	PPYR	9.73E-04	1.0	9.73E-04
91162	PMFINE	PBAA	1.67E-05	1.0	1.67E-05
91162	PMFINE	PCHRY	5.21E-05	1.0	5.21E-05
91162	PMFINE	PBAP	6.89E-05	1.0	6.89E-05
91162	PMFINE	PBBF	2.92E-05	1.0	2.92E-05
91162	PMFINE	PBKF	2.92E-05	1.0	2.92E-05
91162	PMFINE	PBGHIP	4.17E-06	1.0	4.17E-06
91162	PMFINE	PICDP	1.04E-05	1.0	1.04E-05
91162	PMFINE	PDAHA	2.08E-05	1.0	2.08E-05
91168	PM2_5	PACE	3.80E-03	1.0	3.80E-03
91168	PM2_5	PACY	3.71E-04	1.0	3.71E-04
91168	PM2_5	PANT	1.63E-03	1.0	1.63E-03
91168	PM2_5	PBAA	8.80E-04	1.0	8.80E-04

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
91168	PM2_5	PBAP	3.72E-04	1.0	3.72E-04
91168	PM2_5	PBBF	8.12E-04	1.0	8.12E-04
91168	PM2_5	PBGHIP	2.27E-04	1.0	2.27E-04
91168	PM2_5	PBKF	5.56E-04	1.0	5.56E-04
91168	PM2_5	PCHRY	1.59E-03	1.0	1.59E-03
91168	PM2_5	PDAHA	1.02E-04	1.0	1.02E-04
91168	PM2_5	PFLU	1.12E-03	1.0	1.12E-03
91168	PM2_5	PFTH	5.16E-03	1.0	5.16E-03
91168	PM2_5	PICDP	1.93E-04	1.0	1.93E-04
91168	PM2_5	PNAPH	1.58E-03	1.0	1.58E-03
91168	PM2_5	PPHE	6.51E-03	1.0	6.51E-03
91168	PM2_5	PPYR	3.57E-03	1.0	3.57E-03
91175	PM2_5	PCHRY	6.87E-04	1.0	6.87E-04
91175	PM2_5	PANT	1.53E-04	1.0	1.53E-04
91175	PM2_5	PFLU	7.63E-05	1.0	7.63E-05
91175	PM2_5	PPHE	1.53E-04	1.0	1.53E-04
91175	PM2_5	PPYR	7.63E-05	1.0	7.63E-05
91177	PM2_5	PACE	2.10E-07	1.0	2.10E-07
91177	PM2_5	PACY	3.65E-09	1.0	3.65E-09
91177	PM2_5	PANT	7.27E-08	1.0	7.27E-08
91177	PM2_5	PBAA	5.64E-07	1.0	5.64E-07
91177	PM2_5	PBAP	4.64E-07	1.0	4.64E-07
91177	PM2_5	PBBF	6.36E-08	1.0	6.36E-08
91177	PM2_5	PBGHIP	3.65E-08	1.0	3.65E-08
91177	PM2_5	PBKF	5.55E-07	1.0	5.55E-07
91177	PM2_5	PCHRY	6.55E-06	1.0	6.55E-06
91177	PM2_5	PFLU	4.01E-06	1.0	4.01E-06
91177	PM2_5	PFTH	5.64E-05	1.0	5.64E-05
91177	PM2_5	PICDP	9.09E-08	1.0	9.09E-08
91177	PM2_5	PNAPH	2.47E-02	1.0	2.47E-02
91177	PM2_5	PPHE	4.01E-05	1.0	4.01E-05
91177	PM2_5	PPYR	1.64E-06	1.0	1.64E-06
91178	PM2_5	PACE	3.80E-03	1.0	3.80E-03
91178	PM2_5	PACY	3.71E-04	1.0	3.71E-04
91178	PM2_5	PANT	1.63E-03	1.0	1.63E-03
91178	PM2_5	PBAA	8.80E-04	1.0	8.80E-04
91178	PM2_5	PBAP	3.72E-04	1.0	3.72E-04
91178	PM2_5	PBBF	8.12E-04	1.0	8.12E-04
91178	PM2_5	PBGHIP	2.27E-04	1.0	2.27E-04
91178	PM2_5	PBKF	5.56E-04	1.0	5.56E-04
91178	PM2_5	PCHRY	1.59E-03	1.0	1.59E-03

Table S1 Continued

Speciation profile number	Pollutant ID	Species ID	Split factor	Divisor	Mass Fraction
91178	PM2_5	PDAHA	1.02E-04	1.0	1.02E-04
91178	PM2_5	PFLU	1.12E-03	1.0	1.12E-03
91178	PM2_5	PFTH	5.16E-03	1.0	5.16E-03
91178	PM2_5	PICDP	1.93E-04	1.0	1.93E-04
91178	PM2_5	PNAPH	1.58E-03	1.0	1.58E-03
91178	PM2_5	PPHE	6.51E-03	1.0	6.51E-03
91178	PM2_5	PPYR	3.57E-03	1.0	3.57E-03
92018	PM2_5	PANT	6.97E-05	1.0	6.97E-05
92018	PM2_5	PBAA	2.48E-05	1.0	2.48E-05
92018	PM2_5	PCHRY	6.15E-05	1.0	6.15E-05
92018	PM2_5	PFTH	8.72E-05	1.0	8.72E-05
92018	PM2_5	PPHE	2.39E-04	1.0	2.39E-04
92018	PM2_5	PPYR	9.17E-05	1.0	9.17E-05
92018	PM2_5	PFLU	1.30E-05	1.0	1.30E-05
92018	PM2_5	PNAPH	1.44E-05	1.0	1.44E-05

Table S2 Annual estimation of 16 PAHs emission from each major sector (unit: Mg/year)

Sector	NAPH	ACE	ACY	FLU	PHE	ANT	FTH	PYR	CHRY	BaA
rwc	9719.44	102.27	1033.83	317.87	1337.83	311.09	248.90	187.14	20.81	24.33
mobile	3464.62	15.70	42.02	124.69	371.34	55.15	187.48	254.02	60.68	93.67
egu	298.61	19.51	7.18	2.68	7.09	1.47	4.18	3.94	2.02	1.00
nonroad	1852.78	10.34	33.22	88.84	268.26	42.61	144.09	192.56	50.52	75.79
oilgas	224.76	50.40	75.49	10.25	144.15	6.37	8.55	115.15	5.71	43.09
rail	812.06	2.62	4.25	241.39	479.61	4.02	36.21	51.00	0.26	4.96
other	5992.40	441.86	715.09	291.07	989.65	308.49	484.58	415.96	165.50	260.01
total	22364.66	642.70	1911.08	1076.79	3597.93	729.19	1114.00	1219.76	305.50	502.85

Table S2 Continued

Annual	BbF	BkF	BaP	BghiP	IcdP	DahA	7-PAH	16-PAH
rwc	12.72	12.72	11.25	7.73	5.17	60.47	147.46	13413.56
mobile	17.24	8.84	47.17	39.43	16.72	1.63	245.96	4800.40
egu	1.88	2.09	1.83	0.90	1.11	0.35	10.27	355.83
nonroad	20.26	14.01	49.86	69.66	27.50	1.65	239.60	2941.96
oilgas	0.00	0.01	119.58	160.05	2.31	0.00	170.71	965.89
rail	1.29	3.32	2.14	0.89	0.39	0.18	12.54	1644.58
other	122.98	82.68	170.28	141.94	48.48	23.78	873.72	10654.76
total	176.38	123.66	402.11	420.60	101.68	88.06	1700.24	34776.97

Table S3 Species added to the gas phase mechanism

Species	Name in Model	k ($cm^3 molecules^{-1} sec^{-1}$)
Naphthalene	NAPH	1.07E-12@-895
		2.50E-19
Acenaphthylene	ACY	1.10E-10
		5.50E-16
Acenaphthene	ACE	7.33E-11
		5.00E-19
Fluorene	FLU	1.40E-11
Phenanthrene	PHE	2.74E-11
		4.00E-19
Anthracene	ANT	1.30E-10
Fluoranthene	FTH	1.10E-11
Pyrene	PYR	5.00E-11
Benzo[a]Anthracene	BAA	5.00E-11
Chrysene	CHRY	5.00E-11
Benzo[b]Fluoranthene	BBF	1.86E-11
Benzo[k]Fluoranthene	BKF	5.36E-11
Benzo[a]Pyrene	BAP	3.85E-10
Dibenz(ah)Anthracene	DAHA	5.00E-11
Benzo[ghi]Perylene	BGHIP	5.00E-11
Indeno(1,2,3-cd)Pyrene	ICDP	4.47E-10