Characterizing Community Exposure to Atmospheric Polycyclic Aromatic Hydrocarbons (PAHs) in The Memphis Tri-State Area

Final Report

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Acronyms

AQS	Air Quality System
ASE	Accelerate Solvent Extraction
CCV	Continuing Calibration Verification
CFR	Code of Federal Register
COC	Chain of Custody
CV	Coefficient of Variance
DFTPP	Decafluorotriphenylphosphine
DQO	Data Quality Objectives
EPA	Environmental Protection Agency
FB	Field Blank
GC/MS	Gas Chromatography/Mass Spectrometry
GIS	Geographical Information Systems
GPS	Geographic Positioning System
IS	Internal Standard
LCS	Laboratory Control Sample
MDL	Method Detection Limit
MQOs	Measurement Quality Objectives
MS	Matrix Spike
MTA	Memphis Tri-State Area
NATA	National Air Toxics Assessment
PAHs	Polycyclic Aromatic Hydrocarbons
PM	Project Manager
POM	Polycyclic Organic Matter
PUF	Polyurethane Foam
PT	Proficiency Test
QA	Quality Assurance
QC	Quality Control
QAM	Quality Assurance Manual
QAPP	Quality Assurance Project Plan
QFF	Quartz Fiber Filter
RPD	Relative Percent Difference
RSD	Relative Standard Deviation
RT	Retention Time
SCHD	Shelby County Health Department
SD	Standard Deviation
SOP	Standard Operating Procedure
VOC	Volatile Organic Compounds
UM	University Of Memphis

Executive Summary

Polycyclic aromatic hydrocarbons (PAHs) in the ambient air are classified as priority pollutants by the U.S. EPA because of their adverse effects on human health, persistence in environmental matrices, and reactivity and ability to transform into more active species. In the U.S., monitoring of PAHs is insufficient despite the ubiquitous presence of this group of chemicals. The objective of this study was to characterize PAHs in the community ambient air in the Memphis Tri-state Area (MTA). Specifically, the data collected in this monitoring campaign were analyzed to understand the pollution levels, spatiotemporal variability, sources, health risks, and environmental justice regarding PAHs.

PAH samples were collected at 19 monitoring sites in Shelby County, TN, Crittenden County, AR, and DeSoto County, MS, every 12 days from March 2018 to May 2019. At each site, particulate- and gas-phase PAHs were collected at a flow rate of 200 L/min to a filter and PUF/XAD-4/PUF cartridge housed in a high volume PUF sampler over 24 hours. A total of 663 samples, 57 collocated duplicate samples, and 71 blanks were collected. In the laboratory, samples were extracted using an accelerated solvent extractor. The extract was then nitrogen blown down to 1 mL and then analyzed on a GC/MS system operated in the SIM mode for 30 target PAH compounds. The sampling and analysis procedures followed the requirements established in the Quality Assurance Project Plan (QAPP) that was reviewed and approved by EPA Region 4 Office.

All the 30 target PAHs were detected in this program, and the low molecular weight (LMW) PAHs were detected in 100% of the samples. The mean (±standard deviation) concentrations of naphthalene and sum of the remaining 29 PAHs were 27.1±45.9 and 45.4±57.1 ng/m^3, respectively. The overall PAH levels were higher than the average urban levels in the U.S. The concentrations displayed a clear seasonal pattern of summer>spring>fall>winter, and a spatial pattern of urban>suburban>rural. The major source was on-road diesel exhausts in this region, according to the diagnostic ratios. The average cancer risk from all the carcinogenic PAHs (cPAHs) was 2.1×10^{-6}, suggesting low cancer risks from inhalation of airborne PAHs in the ambient air. PAH levels showed negative associations with community-level household incomes, indicating low-income populations face higher exposures to ambient PAHs than do high-income populations.

This monitoring program established a community-government-academic partnership and engaged the local communities throughout the project process. The study team delivered over 40 presentations to students, stakeholders, professionals, and regulatory committees via multiple venues over the four year study period. The results of this study will be made publicly available and be disseminated to the general public. The study team will continue the efforts to promote the use of environmental monitoring data by community members.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous air pollutants emitted from any combustion processes of organic materials. They have been classified as priority pollutants by the U.S. EPA (USEPA 1994), because of adverse effects on human health, persistence in environmental matrices, and reactivity and ability to transform into more active species. PAHs pose health concerns to humans, due to their numerous and uneven distributions of sources, high toxicity, lack of monitoring data, and unawareness among the general public. Supported by the U.S. EPA's "Community-Scale Air Toxics Ambient Monitoring Program", we launched this "Characterizing Community Exposure to Atmospheric Polycyclic Aromatic Hydrocarbons (PAHs) in the Memphis Tri-State Area" study in 2016. The monitoring stations used for this study were distributed in the Memphis TN-MS-AR Metropolitan Statistical Area, a tri-state area that comprises West Tennessee, North Mississippi, and the Arkansas Delta (Figure 1). For convenience, we refer this study as "Memphis PAHs Study" and the study area as "Memphis Tri-state Area (MTA)" in this report.

1.1 PAHs in air and the associated health effects

PAHs are a complex mixture of compounds formed during incomplete combustion processes of organic materials (ATSDR 1995a). They are released into the environment as gases or associated with particulate matter (PM), and are ubiquitous in the general environment (Ravindra et al. 2008). Light PAHs (≤4 rings) predominantly exist in the gas phase, while heavy species (>4 rings) are almost exclusively adsorbed onto particles. In the atmosphere, PAHs behave similarly to persistent organic pollutants and can undergo long-range transport (Keyte et al. 2013; Manzetti 2013; Mulder et al. 2015).

Human exposure to polycyclic aromatic hydrocarbons (PAHs) is inevitable, given the widespread presence of PAHs in the air, water, soil, and food (Abdel-Shafy and Mansour 2016; Srogi 2007). The general population is exposed to PAH mainly through inhalation (ATSDR 1995b). Although PAHs exist at low concentrations in the ambient air, epidemiologic studies have linked long-term low-level PAH exposure with many health outcomes. The major toxicities of concern include immunotoxicity, carcinogenicity, and endocrine disruption. Some PAH compounds, such as benzo[a]pyrene and benz[a]anthracene, have been identified as probable human carcinogens by the U.S. EPA (ATSDR 1995a; USEPA 1999a), and in particular, are associated with respiratory tract and bladder cancers (Rota et al. 2014). PAH exposures are also linked to cardiovascular disease (Clark et al. 2012; Xu et al. 2010), birth defects (Langlois et al. 2012; Langlois et al. 2013), early childhood development (Perera et al. 2006; Perera et al. 2011; Perera et al. 2012), childhood obesity (Jung et al. 2014; Rundle et al. 2012; Scinicariello and Buser 2014), and asthma and other respiratory diseases (Jung et al. 2012; Miller et al. 2004; Rosa et al. 2011).

1.2 National PAHs monitoring and data gaps

Nationally, monitoring of ambient PAHs is insufficient despite the ubiquitous presence of this group of chemicals. Our recent analysis shows that there were only 169 different sampling stations that monitored PAHs in 25 years from 1990 to 2014, and they were distributed in 10

EPA Regions (Liu et al. 2017). Regions 4, 5, 6, and 10 had over 20 individual sites per region, while the remaining regions had less than 10 individual sites. There existed substantial heterogeneity in the geographic coverage of PAH monitoring stations: most sites were concentrated in urban areas along the coastlines, while the inland states are not well represented. At most stations, PAHs were monitored every 6 days (58%) or every 12 days (25%), with an average of 8±7 days (median: 6 days, interquartile range (IQR): 6-6). The sampling period at each station varies from a few months to up to 19 years (median: 2 years, IQR: 1-6.5 years). In contrast, at least over 300 stations are monitoring PM in a continuous manner in the U.S. (Seidel and Birnbaum 2015).

The air pollution modeling data also lack details for accurate health risk assessment. In EPA's list of 187 air toxics, PAHs are reported as one air toxic, polycyclic organic matter (POM), without speciation information (USEPA 2014). As a result, EPA's Toxics Release Inventory (TRI) and the National-scale Air Toxics Assessment (NATA) programs only report the emissions and concentrations of composite POM. These estimates are insufficient for risk assessment, as PAH compounds show toxicities that vary over several orders of magnitude (R. Schoeny and Poirier 1993). Hence, data gaps and uncertainties must be resolved to evaluate health risks and to set appropriate standards.

1.3 PAH sources in the Memphis Tri-state Area (MTA)

Memphis, Tennessee, a metropolitan city and historical transportation hub in Mid-South, has numerous major and minor air pollution sources. The city houses many major industries, including transportation carriers, a petroleum refinery, petrochemical storage, and transfer facilities, waste disposal facilities, a power plant (Table S1). The 2012 Toxics Release Inventory (TRI) showed that air toxics emissions in Shelby County remained in the top 100 counties for air toxics emissions in the U.S. (USEPA 2012a). According to the latest TRI database (USEPA 2012b), air toxics with top onsite air emissions (over 100,000 lb/year) included ammonia, hydrochloric acid, methyl methacrylate, and n-hexane.

The major combustion sources in MTA include industries, airports, trains, truck corridors, highways, and Mississippi River (Figure 1). Southwest Memphis houses clustered heavy industries, including a refinery and a coal-fired power plant. Known as the "Distribution Center of America", Memphis is a major Mid-American commercial and transportation hub. Memphis has the busiest airport for cargo traffic (Air Council International 2010), the third-largest rail center, and one of the largest inland ports in the nation. Crittenden County, AR, has the region's largest truck corridor. Historically, excessive dust from operations is notable (Arkansas Department of Environmental Quality 2001). The Lamar Avenue corridor in Memphis is home to the newly enlarged BNSF rail yard used primarily for freight off-loading operations and brings thousands of train cars and trucks daily through the area. Major highways include Interstate 40, 240, 55, and 385, and their inner-city segments all have high traffic volumes (>50,000 vehicles/day) (TDOT 2014). Previous studies have confirmed that industries and transportations are major PAH sources in urban settings (Ravindra et al. 2008). In addition, hundreds of grill-type restaurants and backyard barbecues are releasing combustion-related contaminants, which have been recognized as an important community source of PAHs (Chen et al. 2012; Wexler; and Pinkerton 2012). Hence, exposure to PAHs is a potential health threat to inner-city communities

considering their proximity to possible local industrial sources and increased traffic density. In rural areas, agricultural burning releases large amount of particles and gaseous PAHs that even transport long distances and have broad environmental and health impacts (Keshtkar and Ashbaugh 2007; Korontzi et al. 2008).

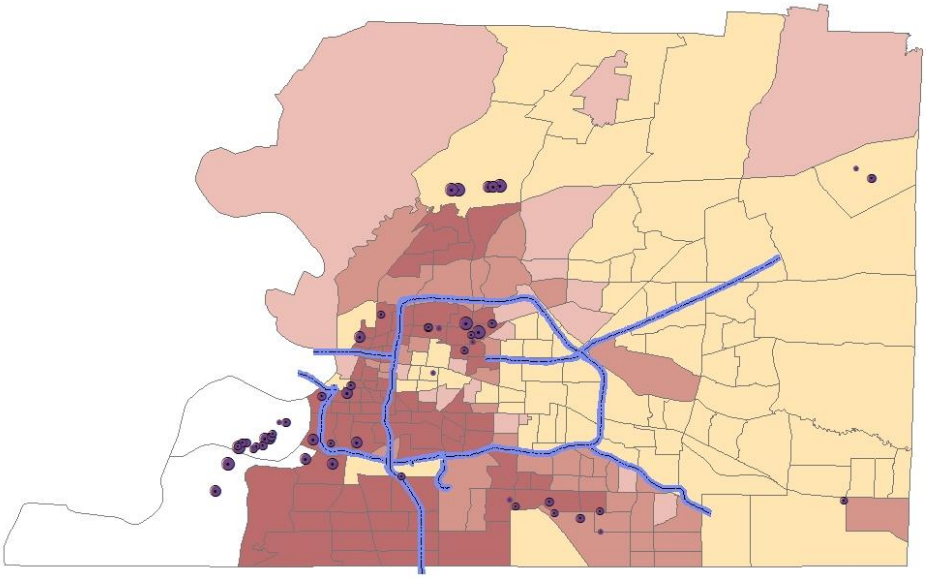


Figure 1. Major stationary and mobile emission sources in the MTA

1.4 Environmental health and justice issues in MTA

Memphis has been challenged by health and poverty issues. The city has a high poverty rate of 32%, and more than half of all children in the county facing economic difficulties. Poverty is accompanied by health issues. Memphis was among the nation's top three "Asthma Capitals" from 2010-2014 (AAFA 2014). Shelby and Crittenden Counties have infant mortality twice the national level (Community Commons 2014). Shelby County has many health indicators ranked top in TN, such as infant mortality (#1), hypertension (#1), obesity (#2), and stroke mortality (#3) (Tennessee Department of Health 2011). Cardiovascular disease and cancer are the top two leading causes of death in Shelby (Tennessee Department of Health 2011). Many schools are located near freeways, which is associated with childhood asthma (Gale et al. 2012). Diseases prevalence also displays strong spatial patterns: mortalities of cardiovascular disease, cancer, and chronic lower respiratory disease are all elevated in the western part of Memphis, an area consisting predominantly of low-income African American residents. The Memphis CANDLE Study found that air pollution impaired fetal neurodevelopment (Loftus et al. 2019). As air pollution is linked to these diseases (Suh et al. 2000), communities have expressed concerns about air pollution and environmental justice.

The environmental justice (EJ) principle states that effects of air pollution exposure on health are differentially distributed by socioeconomic status (SES) and that low SES people often have disproportionately high environmental pollution burdens (O'Neill et al. 2003). A recent environmental disparity study using 2011 NATA data showed that low-income African American neighborhoods tend to reside near mobile sources, and therefore bear higher cancer risks from air toxics (Jia et al. 2014). However, NATA has limitations: it provides only modeling data, does

not report specific PAH compounds, and does not support hot-spot analysis to identify major sources, which are often located in proximity to disadvantaged communities. As a matter of fact, previous air pollution EJ research is mostly focused on criteria pollutants and VOCs, but has never examined PAHs (Payne-Sturges and Gee 2006). A field PAH monitoring will provide valuable data to examine EJ issues related to PAH exposure.

1.5 Need for exposure and risk assessment of atmospheric PAHs in MTA

According to 2005 NATA, Shelby County has an overall cancer risk higher than the 95th percentile risk levels both for Tennessee and the U.S. (Jia et al. 2014). Small monitoring projects at central sites confirmed several air pollution “hot spots” (Greene et al. 2006; Jia and Foran 2013). The current air toxics monitoring program has indicated that the overall cancer risk may be 10 times higher than the national level. Unfortunately, these two local monitoring programs only measured volatile organic compounds (VOCs) not PAHs, as PAH monitoring needs different measurement techniques. To our knowledge, there has been no community-scale PAH study in MTA, hindering our understanding of the health risks from these chemicals.

The public’s understanding of air pollution is often limited to odor, open burning, vehicle exhaust, and visible dust (Ware et al. 2013). They are unfamiliar with PAHs, despite PAHs’ widespread sources and numerous exposure pathways. The public gets exposure to PAHs not only from mobile and industrial sources, but from many other sources, such as grilled food and tobacco smoke (Alomirah et al. 2011). While increasing people’s perception and knowledge is a cornerstone for regulations and interventions, the poor comprehension of risk may impede delivery of the optimal level of pollution control measures. A survey among metropolitan residents shows that most people think air pollution information is beneficial, and >60% of people are seeking air pollution information in their area (Environmental Research Group 2002). All these facts reflect the great demand for air pollution information, including that for PAHs, among residents in MTA.

2. Study Objectives

2.1 Overall objective

The overall objective of the Memphis PAHs Study was to characterize the concentrations and distributions of PAHs in ambient air in MTA, identify major sources, characterize near-source PAH profiles, and assess non-carcinogenic and carcinogenic risks. This study adopted the community-based participatory research (CBPR) approach, in which community residents and organizations were engaged in all phases of the study (Minkler 2010). Figure 2 displays the research framework.

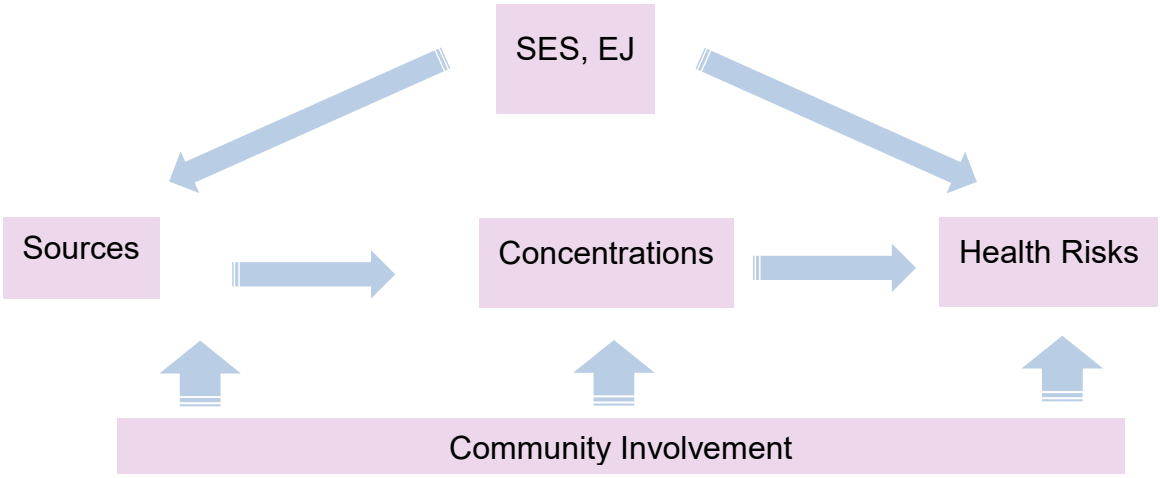


Figure 2. Framework of Memphis PAHs Study

2.2 Specific aims

Under the overall objective, there were five specific aims:

Specific Aim 1: To determine the community-scale concentrations of PAHs and the spatiotemporal variations. This monitoring campaign will yield a rich database of continuous/seasonal PAH measurements at 19 sites in different types of neighborhoods. This database will be used for these sub-aims: (1a) Estimate chronic exposure in the population in MTA, which can then be used for health risk assessment; (1b) Compare PAH levels in MTA with those measured in other regions in the U.S.; (1c) Identify PAH exposure “hot-spots”, where the local residents are the “high-end” exposure sub-populations; (1d) Describe spatial distributions and temporal trends of ambient PAH levels, and examine the influential factors, such as neighborhood, season, day-of-the-week, meteorology, and industrial and agricultural activities.

Specific Aim 2: To assess public health risks potentially associated with exposures to airborne PAHs. The risk assessment will (2a) estimate inhalation cancer risks; (2b) evaluate the non-cancer effects by comparing results from (1a) with the threshold levels; (2c) prioritize those PAHs that might present the highest health risks; and (2d) estimate the contribution of PHAs to the total risks from all carcinogenic air toxics.

Specific Aim 3: To identify major PAH sources and their locations, and to characterize community PAH source profiles. This task will address the following key questions: What

emissions sources contribute to ambient PAHs? How much does each source type contribute? Which sources could be targeted with control measures to reach the highest reduction of PAH concentrations or risks? What are the discrepancies between emission inventories and sources identified by receptor models?

Specific Aim 4: To examine the relation of socioeconomic status (SES) and measured PAH concentrations. A novel aspect of this study is to place PAH exposure in the social context to address potential EJ issues. We will examine how the uneven spatial distribution of PAH sources causes disproportionate environmental exposure among communities in different SES. The hypothesis is that emission sources are often concentrated in these disadvantaged areas and as a result, low SES communities have higher levels of PAH exposures and the associated cancer risks.

Specific Aim 5: To strengthen partnerships among community institutions receptive to learning about and using PAH and air pollution data. We will develop multiple communication venues to engagement communities in the study, ensure widespread dissemination of and access to air pollution data, promote use of these data by community residents to improve daily decisions to reduce harmful effects of air pollutants, and bolster the community's effectiveness in shaping local policies for transportation, development, and construction projects affecting air pollution.

2.3 Target PAHs

The measurement goal of the Memphis PAHs Study was to estimate the concentration, in standard units of nanograms per cubic meter (ng/m^3) of PAHs with 24-hour samples taken at 19 sites in the MTA every 12 days over one year of study period. This was accomplished by collecting filter and PUF sample and performing GC/MS analysis of samples for 32 target compounds. The target PAHs for analysis were chosen based on regulatory requirements, their occurrence in the environment, toxicity, and availability of analytical standards. The final target list consisted of 32 PAHs, which included the 16 EPA priority PAHs and NATTS Tier I core and PT target analytes(Table 1).

Table 1. The target PAH compounds for the Memphis PAHs Study

#	Polycyclic Aromatic Hydrocarbons Total # PAHs = 32	Abbr.	CAS #	No. of Rings	MW (g/mol)	BP (°C, 760 mmHg)	1- EPA-16 16	2-NATTS 21	3 - MDH Priority 15	4 - EPA-RTK,TRI 19	cPAH Lists 5 - OEHHA 5	6 - OEHHA 16	7 - EC "15+1" 14	8 - EPA RPF 17
1	Naphthalene	NAP	91-20-3	2	128	218	*	π						
2	Acenaphthylene	ACY	208-96-8	2	152	280	*	π						
3	Acenaphthene	ACP	83-32-9	2	154	279	*	π						
4	Fluorene	FLR	86-73-7	3	166	295	*	π						
5	9-Fluorenone	9-FL	486-25-9	3	180	342		π						
6	Dibenzothiophene	DBT	132-65-0	3	184	332								
7	Phenanthrene	PHE	85-01-8	3	178	336	*	π						£
8	Anthracene	ANT	120-12-7	3	178	340	*	π						£
9	Fluoranthene	FLT	206-44-0	4	202	375	*	π	#	‡				£
10	Pyrene	PYR	129-00-0	4	202	404	*	π						£
11	Retene	RET	483-65-8	3	234	390		π						
12	Benzo[c]phenanthrene	BcP	195-19-7	4	228	437								
13	Cyclopenta[c,d]pyrene	CPP	27208-37-3	5	226	303			#				¶	£
14	Benz[a]anthracene	BaA	56-55-3	4	228	438	*	π	#	‡		§	¶	£
15	Chrysene	CHR	218-01-9	4	228	448	*	π	#	‡		§	¶	£
16	Benzo[b]fluoranthene	BbF	205-99-2	5	252	480	*	π	#	‡		§	¶	£
17	Benzo[j]fluoranthene	BjF	205-82-3	5	252	480			#	‡		§	¶	£
18	Benzo[k]fluoranthene	BkF	207-08-9	5	252	480	*	π	#	‡		§	¶	£
19	7,12-Dimethylbenz[a]anthracene	DMBA	57-97-6	4	256	480				‡	¥	§		
20	Benzo[e]pyrene	BeP	192-97-2	5	252	468		π						
21	Benzo[a]pyrene	BaP	50-32-8	5	252	495	*	π	#	‡	¥		¶	
22	Perylene	PER	198-55-0	5	252	468		π						
23	3-Methylcholanthrene	3MC	56-49-5	5	268	280				‡	¥	§		

#	Polycyclic Aromatic Hydrocarbons Total # PAHs = 32	Abbr.	CAS #	No. of Rings	MW (g/mol)	BP (°C, 760 mmHg)	1- EPA-16 16	2-NATTS 21	3 - MDH Priority 15	4 - EPA-RTK,TRI 19	cPAH Lists 5 - OEHHA 5	6 - OEHHA 16	7 - EC "15+1" 14	8 - EPA RPF 17
24	Dibenz[a,h]acridine	DhACR	226-36-8	5	279	534				ǂ		§		
25	Dibenz[a,j]acridine	DjACR	224-42-0	5	279	534				ǂ	¥	§		
26	Indeno[1,2,3-cd]pyrene	IcP	193-39-5	6	276	536	*	π	#	ǂ		§	¶	£
27	Dibenz[a,h]anthracene	DhANT	53-70-3	5	278	524	*	π	#	ǂ	¥	§	¶	£
28	7H-Dibenzo[c,g]carbazole	DBC	194-59-2	5	267	401				ǂ		§		
29	Benzo[g,h,i]perylene	BgP	191-24-2	6	276	550	*	π	#	ǂ			¶	£
30	Dibenzo[a,l]pyrene	DlP	191-30-0	6	302	552			#	ǂ		§	¶	£
31	Coronene	COR	191-07-1	7	300	525		π						
32	Dibenzo[a,e]pyrene	DeP	192-65-4	6	302	552			#	ǂ		§	¶	£

Notes:

1. EPA-16. US EPA - PAHs on the Clean Water Act List of Priority Pollutants(USEPA 2014)
2. NATTS TAD Revision 3_FINAL(USEPA 2016)
3. Calibrating Concerns about PAHs in Urban Air QAPP (MPCA 2012)
4. EPA-RTK,TRI. US EPA (2001) - Right-To-Know Act: Polycyclic Aromatic Compounds Category(USEPA 2001a) (EPA 2001)
5. OEHHA CSF. California Office of Health Hazard Assessment PAH Cancer Slope Factors(OEHHA 2009)
6. OEHHA 2015. California Office of Health Hazard Assessment(OEHHA 2015)
7. EC "15+1". European Commission - Commission Regulation (EC) No 1881/2006(EC 2006)
8. EPA Draft RPF - PAHs with Relative Potency Factors from US EPA Draft Document (USEPA 2010)

2.5 Project progress and major activities

This community-scale PAHs monitoring program was implemented in three stages (Table 2):

Stage 1: Preparation. The preparation activities included formation of the study team, engagement of communities, site recruitment and establishment, laboratory instrumentation, preliminary monitoring, and QAPP development.

Stage 2: Field sampling and laboratory analysis. The activities included routine field sample collections every 12 days over 15 months, laboratory analysis of PAH samples, data organization and checking, preliminary data analysis, and community outreach.

Stage 3: Data validation, analysis, and archiving. The major activities included data validation following the QAPP requirements, data analysis to understand PAH exposure levels, sources, and health risks, data backup and archiving, and dissemination of results to communities and professionals.

Table 2. Major activities and timeline of the Memphis PAHs Study.

Period	Activities
Preparation	
Oct-Dec, 2016	Contracting and community stakeholders' meeting.
Jan-March, 2017	QAPP development.
Mar 23, 2017	Revised work plan submitted to EPA Region 4 Office.
Apr 21, 2017	QAPP submitted to EPA Region 4 Office.
Sep 08, 2017	QAPP approved.
Sep 2017-Jan 2018	Monitoring method optimization.
Monitoring	
Feb 2018	UM Laboratory in place for receiving and analyzing samples.
Feb-Mar, 2018	Establishment of sites and testing of samplers.
Mar 2018-May 2019	Field sample collection and laboratory analysis.
May 23, 2019	Sampling completed with 100% data completion.
Data analysis and reporting	
May-Sep 2019	Data validation and analysis.
Aug-Oct 2019	Final report drafting.
Dec 15, 2019	Final report submitted.

3. Sampling and Analytical Methods

3.1 Study area

Air samples were collected in three neighboring counties in MTA: Shelby County, TN, DeSoto County, MS, and Crittenden County, AR (Figure 3). According to the 2010 Census, these counties are MTA's central counties that represent 87% of the total population and 41% of the total area of MTA. The center city Memphis is the largest city in TN, and the 20th largest city in America. Radiant from downtown Memphis, the land-use type displays a clear industrial-urban-suburban-rural gradient: Memphis is an industrial and urban center, Germantown, Bartlett, and Collierville in Shelby, Olive Branch in DeSoto, and West Memphis in Crittenden are suburban areas, and the rest are mostly rural areas. The selected study area was representative of the MTA and logistically reachable for field sample collection.

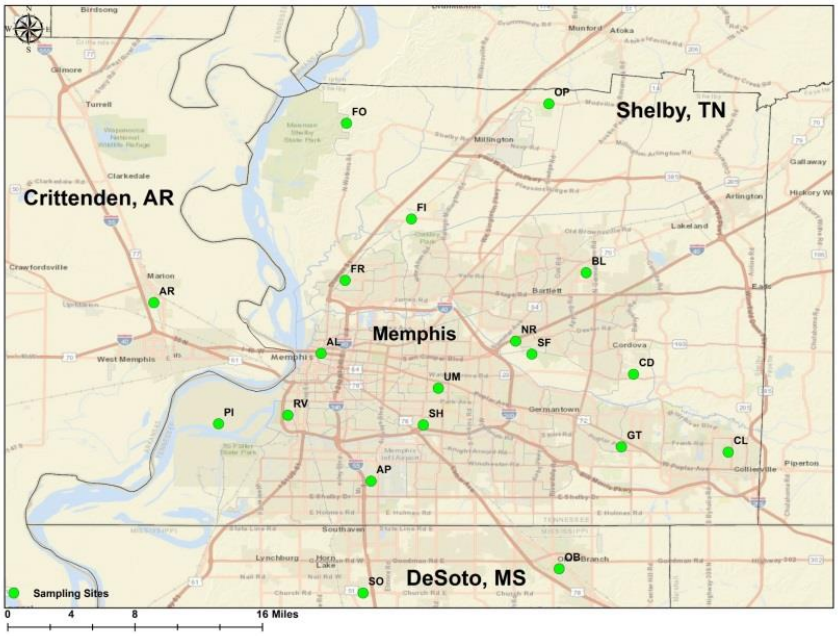


Figure 3. Monitoring sites for the Memphis PAHs Study

3.2 Sampling sites

Analyses of variability of ambient PAHs have shown that PAH concentrations within a small community are quite homogeneous but displayed considerable seasonal variation (Bortnick and Stetzer 2002; Jia CR et al. 2011). Thus, a single sampling location with repeated samples is representative of long-term community exposure.

The sampling sites were selected to have wide spatial and temporal representativeness. It should be noted that PAH sampling requires the availability of electricity, security, access, and a hard base or flooring that is stable and approximately level to secure the sampler. These were the factors that limited the random selection of any sites in this region. Beyond these limitations, the factors listed below were chosen as further decision criteria for sampling locations:

- Presence of past and present industrial and traffic sources;
- Input from community members;
- Sizes of populations with possible exposure and potential for exposure to sensitive receptors;
- Environmental Justice ranking;
- Locations of other measured pollutants;
- Locations of previous permission to place monitors;
- The land use type, including residential areas, parks, farms, and proximity to major emission sources such highways, airports, factories, gas stations, dry cleaning shops and waste disposal sites;
- Obstructions and distance to any traffic and emission sources.

The study team finally recruited 19 monitoring sites in MTA for this study. There were 16 sites in Shelby County, TN, 2 sites in DeSoto County, MS, and 1 site in Crittenden County, AR. The site name, surrounding environments, and addresses are listed in Table 3 and their spatial distribution is displayed in Figure 3. Duplicate samplers were installed at the University of Memphis (UM) site and Alabama Monitoring Station (AL) site. Table 4 summarizes the demographics, socioeconomic status (SES), and nearby sources of the census block group where each site is located. The following is a description of the sites.

(1) UM Site: This urban site was located on the central campus of the University of Memphis. The sampler was set up on the rooftop of the Student Health Center, which is located at the center of the campus. Central Avenue is 300 m north, and a railway is 200 m south.

(2) SH Site: This urban site was located at the Sharpe Monitoring Station. It was in a lawn beside Sharpe Elementary School and 350 m north of Interstate 240. The sampler was placed on a 4-ft scaffolder. The surrounding setting was an African American dominant low-income neighborhood.

(3) AP Site: This site was located in outside of a church about 0.5 mile west of the Memphis International Airport. The sampler was set in lawn north of the church building. This site was in an low-income African American neighborhood.

(4) SO Site: This site was one of the two Mississippi sites. It was located in the lawn of a church property in Southaven, MS. It was 0.5 mile west of the intersection of US-55 and US-69. There were retail stores and an outlets mall nearby.

(5) OB Site: This site was one of the two Mississippi sites. It was located in the lawn of a church property in Oliver Branch, MS. which near the MS-305 and without obvious air pollution sources. The sampler is set on the yard area of the church and located 100 ft from the closest building.

(6) GT Site: This site is located on a Presbyterian church in Germantown, TN, which near the US-72. US-72 travels through Memphis along poplar avenue, one of the city's main roads. The sampler is set on the yard area of the church and located approximately 2,000 ft to the poplar avenue.

(7) CL Site: This site is located on a Christian church in Collierville, TN. It's close to the H.W. Cox park which is the centerpiece of the Collierville Park System and includes the Cox Commnunity Center. The sampler is set on the yard area of the church.

(8) CD Site: This site is located in the city of Cordova, TN, which near the Shelby Farms Park and 3 miles away from the US-40. The sampler is set on the backyard of the recuited participant's house.

(9) BL Site: This site is located in the city of Bartlett, TN, which near the Appling Lake and 1 mile away from the US-79. The sampler is set on the backyard of the recruited participant's house.

(10) NR Site: This site is located in a local community college in the city of Memphis, TN and just aside the US-40. The site is supervised by the Shelby County Health Department and identified as one of the local monitoring sites which belongs to their regional air monitoring network. The sampler is set on the rooftop of the monitoring station.

(11) SF Site: This site is located in the Shelby Farms Park which is the one of the largest urban parks in the country. The site is supervised by the Shelby County Health Department and identified as the National Core Monitoring Station (NCore). The sampler is set insde the monitoring station.

(12) AL Site: This site is located at the intersection of US-40/US-51 and in the downtown of the city of Memphis, TN. The site is supervised by the Shelby County Health Department and identified as one of the local monitoring sites which belongs to their regional air monitoring network. The sampler and a second duplicate one were set at the monitoring station.

(13) FR Site: This site is located in the city of Frayser, TN. The site is 1 mile away from the US-51 and supervised by the Shelby County Health Department and identified as one of the local monitoring sites which belongs to their regional air monitoring network. The sampler is set on the rooftop of the monitoring station.

(14) FI Site: This site is located in an industrial park in the city of Millington, TN. The site is 6 miles away from the US-51 and supervised by the Shelby County Health Department and identified as one of the local monitoring sites which belongs to their regional air monitoring network. The sampler is set on the rooftop of the monitoring station.

(15) OP Site: This site is located in the orgill park in the city of Millington, TN. The site is 1.5 miles away from the US-51 and supervised by the Shelby County Health Department and identified as one of the local monitoring sites which belongs to their regional air monitoring network. The sampler is set on the rooftop of the monitoring station.

(16) FO Site: This site is located in the Meeman-Shelby Forest which bordering the Mississippi river 13 miles north of Memphis,TN. The site is located in the heart of the forest and identified as the urban background site for the PAHs Study. The sampler is set at a yard area of a conference center.

(17) AR Site: This site is located in the city of Marion, AR. The site is 1 mile away from the US-55 and supervised by the Arkansas Department of Environment Quality and identified as one of the local monitoring sites which belongs to their regional air monitoring network. The sampler is set inside the monitoring station.

(18) PI Site: This site is located in an industrial park of the city of Memphis, TN. The site is located in the president's island which isurrounded by the McKellar Lake and nearby the Mississippi River. The site is supervised by the Shelby County Health Department and identified as one of the local monitoring sites which belongs to their regional air monitoring network. The sampler is set on the top of the scaffold which located inside the mornitoring station.

(19) RV Site: This site is located in an urban community of the city of Memphis, TN. The site is half a mile away from the US-55 and located in between an elementary school and the Riverview Park. The site is supervised by the Shelby County Health Department and identified as one of the local monitoring sites which belongs to their regional air monitoring network. The sampler is set inside the monitoring sation.

Table 3. Sites of the Memphis PAHs Study.

No.	Site Code	Site Description	Sampler (SN)	Surrounding Setting	Address
1	UM	University of Memphis Campus	1014 1038	Urban community	3825 DeSoto Ave, Memphis, TN 38152
2	SH	Sharpe Monitoring Station	1026	Urban community	3431 Sharpe Ave, Memphis, TN 38111
3	AP	Episcopal Church near Memphis Airport	1045	Near airport	4150 Boeingshire Dr, Memphis, TN 38116
4	SO	Catholic Church in Southaven	1023	Suburban community	785 Church Rd W, Southaven, MS 38671
5	OB	Presbyterian Church in Olive Branch	1042	Suburban community	8161 Germantown Rd, Olive Branch, MS 38654
6	GT	Presbyterian Church in Germantown	1025	Suburban community	8816 Poplar Pike, Germantown, TN 38138
7	CL	Christian Church in Collierville	1027	Suburban community	300 W Powell Rd, Memphis, TN 38017
8	CD	Residential backyard in Cordova	1039	Suburban community	9162 Old Brook Cove, Memphis, TN 38018
9	BL	Residential backyard in Bartlett	1033	Suburban community	7872 Jills Creek Dr, Memphis, TN 38133
10	NR	Near Road Monitoring Station	1041	Near road	5767 Macon Cove, Memphis, TN 38134
11	SF	National Core Monitoring Station	1040	Urban background	6359 Haley Rd, Memphis, TN 38134
12	AL	Alabama Rd Monitoring Station	1028 1044	Near road	416 Alabama Ave, Memphis, TN 38105
13	FR	Frayser Monitoring Station	1024	Urban community	1330 Frayser Blvd, Memphis, TN 38127
14	FI	Fite Monitoring Station	1022	Suburban community	3065 Fite Rd, Millington, TN 38053
15	OP	Orgill Park Monitoring Station	1031	Suburban community	6855 Mudville Rd, Millington, TN 38053
16	FO	Meeman Forest Monitoring Station	1030	Background	1236 Cuba Millington Rd, Millington, TN 38053
17	AR	Monitoring Station in Arkansas	1032	Suburban community	395 L H Polk Dr, Marion, AR 72364
18	PI	President's Island Monitoring Station	1029	Industrial Park	2816 Harbor Ave, Memphis, TN 38106
19	RV	Riverview Monitoring Station	1043	Urban community	260 Joubert Ave, Memphis, TN 38109

Table 4. Demographic, social, economic, and environmental conditions of sites.

Site Name	Pop density* (Per km^2)	Median household income# (US $)	Median house value# (US $)	% of low income* (a)	% of Minority*	Traffic Proximity* (b)	Diesel PM* (µg/m^3)
UM	1,377	23,023	315,400	59.9	43.0	62	1.1
SH	2,245	23,160	59,000	64.4	95.6	166	1.3
AP	1,299	32,342	86,100	51.6	100.0	451	1.2
SO	153	53,442	170,100	32.5	39.2	94	0.5
OB	213	70,393	175,900	21.8	42.1	74	0.9
GT	1,018	129,135	369,500	12.5	27.7	376	0.8
CL	1,560	97,621	230,500	34.4	10.6	5	0.8
CD	366	82,132	213,500	20.7	44.5	5	0.5
BL	563	74,944	160,200	14.1	15.6	12	1.2
NR	511	-	-	0.0	69.5	133	1.2
SF	27	-	-	0.0	91.7	32	0.7
AL	731	16,389	44,400	71.5	81.2	3,074	2.1
FR	3,173	17,045	59,600	94.6	100.0	42	0.7
FI	67	56,528	148,500	26.2	34.4	68	0.6
OP	65	55,459	88,400	37.5	22.9	23	0.5
FO	36	63,712	172,000	28.6	14.3	0	0.6
AR	115	78,348	146,100	13.2	37.5	32	1.3
PI	0	-	-	0.0	0.0	0	0.5
RV	1,154	19,653	50,100	67.8	100.0	717	0.8

Note:

a. The % of low income was defined lower than 2x poverty level
b. Traffic proximity was defined as the count of vehicles at major roads within 500 meters
*: Data were obtained for the census block group from the U.S. EPA's EJScreen site (EPA 2018)
#: Data were obtained for the census tracts group from the U.S. Census Bureau's ACS.

3.3 Sampling schedule

EPA recommended prioritizing time resolution on the data over the number of samples collected as a better approach for characterizing exposures. EPA believes that collecting enough samples at each site to calculate a longer-term average concentration that could be compared to chronic/lifetime health screening levels would be more useful to the local community. Following EPA's suggestions, the sampling scheme of this study was designed as follows.

3.3.1 Sampling frequency

At each site, we collected one sample every 12 days. The 12-day cycle was adopted primarily to capture all the days of a week. We did not use a more frequent schedule, e.g., every 6 days, due to the logistic limitations. Table 5 shows the schedule of a 12-day cycle.

3.3.2 Sampling duration

EPA's NATTS Program requires that PAHs sample collection must be performed for 24 ± 1 hours beginning at midnight and concluding on midnight of the following day, local time unadjusted for daylight savings time, per the national sampling calendar. However, we were unable to follow the midnight-to-midnight sampling schedule due to the concerns for the unreliability of samplers' timers and the loss of PAHs on collection media due to high temperature in summer. At each site, we started the sampling whenever the field technician arrived at the site, and the sampling lasted for 24 hours.

3.3.3 Routine sampling sequence

Within each sampling cycle, the field staff visited the sites in the following sequence:

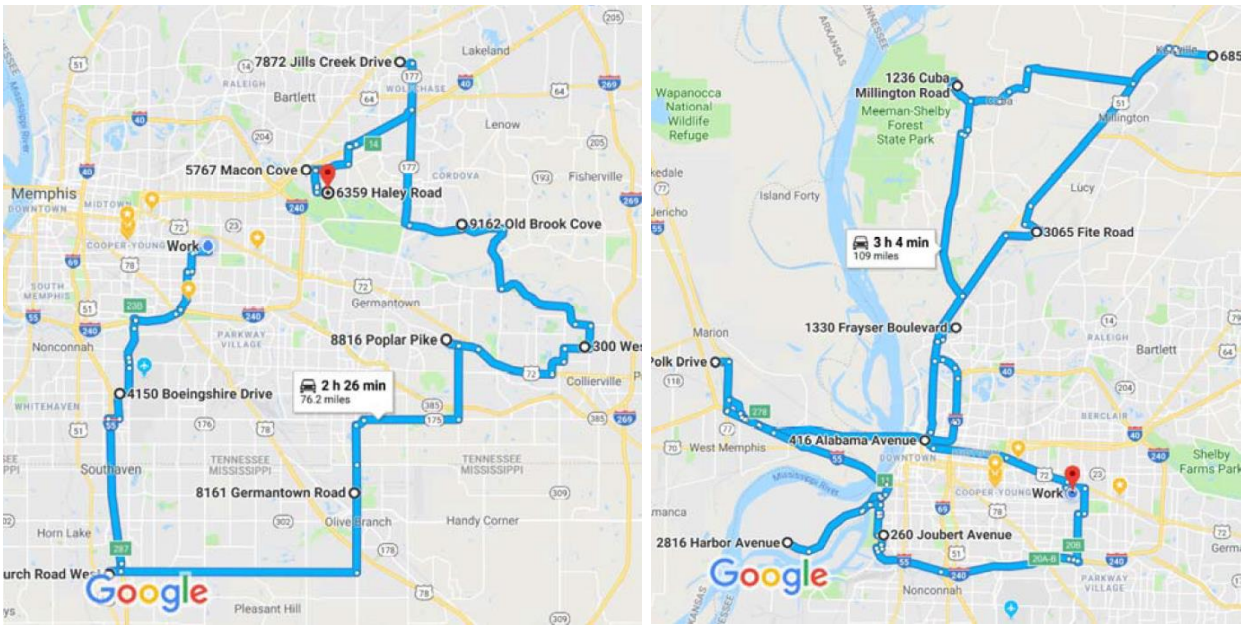
Day 1: Deployment at *UM and SH
Day 2: Deployment at east sites: AP, *SO, OB, *GT, CL, *CD, *BL, NR, and SF
Collection at UM and SH
Day 3: Collection at east sites: AP, SO, OB, GT, CL, CD, BL, NR, and SF
Day 4: Deployment at north and west sites: AL, FR, FI, *OP, *FO, *AR, *PI, RV
Day 5: Collection at north and west sites: AL, FR, FI, OP, FO, AR, PI, RV

*: a portable weather station was installed at this site.

This schedule allowed 7 days for sample analysis and sampling preparation. Figure 4 displays the typical driving routes for site visits.

3.3.4 Sample size

For a 1-year sampling period, the number of samples was calculated as 30 samples/site × 19 sites + 10% duplicates + 10% blanks = 684 samples in total.



1A. Day 2 & 3 site visit routs

1B. Day 4 & 5 site visit routes

Figure 4. Typical site visit routes

Table 5. Sampling schedule and completed cycles.

2018 Calendar **2019 Calendar**

Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb	Mar	Apr	May
1 Th	1 Th	1 Su	1 Tu	1 Fr	1 Su *	1 We	1 Sa *	1 Mo	1 Th *	1 Sa	1 Tu *	1 Fr	1 Fr *	1 Mo	1 We *
2 Fr	2 Fr	2 Mo	2 We	2 Sa	2 Mo *	2 Th	2 Su *	2 Tu	2 Fr *	2 Su	2 We	2 Sa	2 Sa *	2 Tu	2 Th
3 Sa	3 Sa	3 Tu	3 Th	3 Su	3 Tu *	3 Fr	3 Mo *	3 We	3 Sa	3 Mo	3 Th	3 Su *	3 Su	3 We	3 Fr
4 Su	4 Su	4 We	4 Fr	4 Mo	4 We *	4 Sa	4 Tu	4 Th	4 Su	4 Tu	4 Fr	4 Mo *	4 Mo	4 Th *	4 Sa
5 Mo	5 Mo	5 Th	5 Sa	5 Tu	5 Th *	5 Su	5 We	5 Fr	5 Mo	5 We *	5 Sa	5 Tu *	5 Tu	5 Fr *	5 Su
6 Tu	6 Tu	6 Fr	6 Su	6 We	6 Fr	6 Mo *	6 Th	6 Sa *	6 Tu	6 Th *	6 Su	6 We *	6 We	6 Sa *	6 Mo
7 We	7 We	7 Sa	7 Mo	7 Th *	7 Sa	7 Tu *	7 Fr	7 Su *	7 We	7 Fr *	7 Mo	7 Th	7 Th	7 Su *	7 Tu
8 Th	8 Th	8 Su *	8 Tu	8 Fr *	8 Su	8 We *	8 Sa	8 Mo *	8 Th	8 Sa *	8 Tu	8 Fr	8 Fr	8 Mo	8 We
9 Fr	9 Fr	9 Mo *	9 We	9 Sa *	9 Mo	9 Th *	9 Su	9 Tu *	9 Fr	9 Su	9 We	9 Sa	9 Sa	9 Tu	9 Th
10 Sa	10 Sa	10 Tu *	10 Th	10 Su *	10 Tu	10 Fr *	10 Mo	10 We	10 Sa	10 Mo	10 Th *	10 Su	10 Su	10 We	10 Fr *
11 Su	11 Su	11 We *	11 Fr	11 Mo *	11 We	11 Sa	11 Tu *	11 Th	11 Su *	11 Tu	11 Fr *	11 Mo	11 Mo *	11 Th	11 Sa *
12 Mo	12 Mo	12 Th *	12 Sa	12 Tu	12 Th	12 Su	12 We *	12 Fr	12 Mo *	12 We	12 Sa *	12 Tu	12 Tu *	12 Fr	12 Su *
13 Tu	13 Tu *	13 Fr	13 Su	13 We	13 Fr *	13 Mo	13 Th *	13 Sa	13 Tu *	13 Th	13 Su *	13 We	13 We *	13 Sa	13 Mo *
14 We	14 We *	14 Sa	14 Mo *	14 Th	14 Sa *	14 Tu	14 Fr *	14 Su	14 We *	14 Fr	14 Mo	14 Th	14 Th *	14 Su	14 Tu
15 Th	15 Th *	15 Su	15 Tu *	15 Fr	15 Su *	15 We	15 Sa *	15 Mo	15 Th	15 Sa	15 Tu	15 Fr *	15 Fr	15 Mo	15 We
16 Fr	16 Fr *	16 Mo	16 We *	16 Sa	16 Mo *	16 Th	16 Su	16 Tu	16 Fr	16 Su	16 We	16 Sa *	16 Sa	16 Tu *	16 Th
17 Sa	17 Sa	17 Tu	17 Th *	17 Su	17 Tu *	17 Fr	17 Mo	17 We	17 Sa	17 Mo *	17 Th	17 Su *	17 Su	17 We *	17 Fr
18 Su	18 Su *	18 We	18 Fr *	18 Mo	18 We	18 Sa *	18 Tu	18 Th *	18 Su	18 Tu *	18 Fr	18 Mo *	18 Mo	18 Th *	18 Sa
19 Mo	19 Mo *	19 Th	19 Sa	19 Tu *	19 Th	19 Su *	19 We	19 Fr *	19 Mo	19 We *	19 Sa	19 Tu	19 Tu	19 Fr *	19 Su
20 Tu	20 Tu	20 Fr *	20 Su	20 We *	20 Fr	20 Mo *	20 Th	20 Sa *	20 Tu	20 Th *	20 Su	20 We	20 We	20 Sa	20 Mo
21 We *	21 We	21 Sa *	21 Mo	21 Th *	21 Sa	21 Tu *	21 Fr	21 Su *	21 We	21 Fr	21 Mo	21 Th	21 Th	21 Su	21 Tu
22 Th *	22 Th	22 Su *	22 Tu	22 Fr *	22 Su	22 We *	22 Sa	22 Mo	22 Th	22 Sa	22 Tu *	22 Fr	22 Fr	22 Mo	22 We *
23 Fr	23 Fr	23 Mo *	23 We	23 Sa *	23 Mo	23 Th	23 Su	23 Tu	23 Fr *	23 Su	23 We *	23 Sa	23 Sa *	23 Tu	23 Th *
24 Sa	24 Sa	24 Tu *	24 Th	24 Su	24 Tu	24 Fr	24 Mo *	24 We	24 Sa *	24 Mo	24 Th *	24 Su	24 Su *	24 We	24 Fr *
25 Su	25 Su	25 We	25 Fr	25 Mo	25 We *	25 Sa	25 Tu *	25 Th	25 Su *	25 Tu	25 Fr *	25 Mo	25 Mo *	25 Th	25 Sa *
26 Mo	26 Mo	26 Th	26 Sa *	26 Tu	26 Th *	26 Su	26 We *	26 Fr	26 Mo *	26 We	26 Sa	26 Tu	26 Tu *	26 Fr	26 Su
27 Tu	27 Tu *	27 Fr	27 Su *	27 We	27 Fr *	27 Mo	27 Th *	27 Sa	27 Tu	27 Th	27 Su	27 We *	27 We	27 Sa	27 Mo
28 We	28 We *	28 Sa	28 Mo *	28 Th	28 Sa *	28 Tu	28 Fr	28 Su	28 We	28 Fr	28 Mo	28 Th *	28 Th	28 Su *	28 Tu
	29 Th *	29 Su	29 Tu *	29 Fr	29 Su *	29 We	29 Sa	29 Mo	29 Th	29 Sa *	29 Tu		29 Fr	29 Mo *	29 We
	30 Fr *	30 Mo	30 We *	30 Sa	30 Mo	30 Th *	30 Su	30 Tu *	30 Fr	30 Su *	30 We		30 Sa	30 Tu *	30 Th
	31 Sa *		31 Th		31 Tu	31 Fr *		31 We *		31 Mo *	31 Th		31 Su		31 Fr

3.4 Overview of the PAHs monitoring method

PAHs in air were collected and analyzed following the methods described in EPA Method TO-13A (USEPA 1999b) and the latest Technical Assistance Document for the NATTS Program (USEPA 2016). The collection media consisted of a quartz fiber filter (QFF) and glass thimble containing polyurethane foam (PUF) and styrene-divinylbenzene polymer resin sorbent (XAD-2 or equivalent) to collect PAHs from ambient air. Approximately 200 to 350 m^3 of ambient air is drawn through a QFF and cartridge containing a "sandwich" of PUF/XAD/PUF over 24 hours. The QFF and contents of the cartridge are extracted together in an accelerated solvent extraction (ASE) system, and the extract is then nitrogen blown down to 1 ml on a automatic evaporator, and the final extract is analyzed for the target PAHs by gas chromatography/mass spectrometry (GC/MS) in a select-ion-monitoring (SIM) mode.

3.5 Sampling method

3.5.1 High volume PUF samplers

The high volume PUF sampler (TE-1000, Tisch Environmental Inc., Cleves, OH) was used for the collection of volatile organic compounds (VOCs) and particulates. The PUF sampler houses a 102-mm circular QFF followed by a glass cartridge containing PUF/XAD/PUF sorbents to capture particulate- and gas-phase PAHs. This sampler is designed to meet the requirements for PAH sampling by U.S. EPA methods TO-4A, TO-9A, and TO-13A. Before field sample collection, the PUF sampler was calibrated following the procedure described in the Operations Manual for the PUF Sampler. In addition, PUF Samplers were calibrated after motor maintenance, at least once every three months, and after 360 sampling hours. The calibration records for all the PUF samplers were kept in a dedicated log book.

3.5.2 Sampling media

Sampling media: This study used 4" diameter, 2 µm pore size QFFs (Part # TE-QMA4, Tisch Environmental Inc., Cleves, OH), pre-cleaned 3/8" diameter polyurethane foams (PUFs, Part #24295, Restek Corporation, Bellefonte, PA), and ultra-clean XAD-4 resin (Part #24230, Restek Corporation, Bellefonte, PA). Approximately 200 g of XAD-4 was sandwiched between two layers of PUF plugs to prevent loss during sampling and extraction.

Sampling media preparation: QFFs were baked at 450 °C in a Muffle furnace (Model: F30420C, Thermo Fisher Scientific, Waltham, MA, U.S.A.) for 5 hours to remove the potential contaminants. PUFs and XAD-4 sorbents were cleaned and dried in an Accelerated Solvent Exactor (Model: ASE 350, Dionex / Thermo Scientific, Waltham, MA, U.S.A.) following the sequence as below (Table 6).

Table 6. The ASE operating sequence for cleaning PUFs and XAD-4 sorbents

Seq.	Solvent	Temp (°C)	Static (min)	Static Cycles	Rinse Vol (%)	Purge Time (sec)
1	Acetone	100	5	3	50	60
2	Hexane	100	5	3	50	60
3	Acetone	100	5	3	50	60

Sampling media storage: The cleaned filters were inspected for holes or uneven texture. The acceptable filters were then placed in an aluminum lined box (Grainger, Glass Petri Dish, Part # 8UX51) and kept in a desiccator with silica gel inside for future use. All the cleaned PUF plugs and XAD-4 sorbents were air dried in the hood to remove the residual solvent from the extraction, sealed separately in pre-cleaned glass containers, and then stored in a freezer at -20°C. All the cleaned sampling media were labeled with the date of cleaning and used within two weeks after the date of cleaning.

3.5.3 Field sample collection

On each sampling day, a 24±1 hours integrated sample was collected at each site in order to obtain average daily levels of airborne PAHs. The sampling collection consisted of the following procedures.

Preparation before field sampling. In the laboratory, the lab specialist spiked the PUF with 50 µL of field surrogate solution that contained 10 ng/µl of fluoranthene-d10 and benzo(a)pyrene-d12. The PUF/XAD/PUF sorbents were then sealed the cartridge with Teflon end caps and placed in a Ziploc bag. The filter was loaded onto the sampling head and covered with a cleaned aluminum cover. One extra sampling set was prepared as the field blank. The prepared sampling head and cartridge were labled and shipped in a cooler with icepacks inside. The field technicians deployed the cartridges and sampling heads in the field within 3 hours after the lab preparation.

Sample deployment. In the field, the field technician turned on the sampler and kept it running for at least 10 min to warm up the sampler. Then the technician installed the sampling head, turned on the sampler, and set the sampling flow rate at approxiamately 200 L/min, yielding individual sample volumes of about 288 m^3 over the 24-h sampling. The technician filled the Chain-of-Custody form that recorded site ID, sample name, deployment date, start time, and flow rate (in Magn), and weather conditions.

Sample retrieval. On the sample retrieval day, the sampler should stop automatically. The field technician arrived at the site within 2 hours after the sampler stopped. The technician sealed cartridge containing the PUF plugs and XAD-2 with silicone end caps and the QFF with the filter holder protected by the aluminum cover. The cartridge and filter holder were then sealed in zip-lock bags. All samples were kept at ≤4 °C in a cooler, shipped back to the laboratory, and stored in a freezer until extraction and analysis. The technician also filled the Chain-of-Custody form that recorded site ID, sample name, retrieval date, end time, flow rate (in Magn), and weather conditions. The technician signed the form and returned it with samples.

Sample storage. Collected samples were stored at -18°C until extraction, and were extracted within 14 days of collection.

Sample volumes. The beginning and ending flows were averaged to calculate the collected air volume. Flows on a sampling unit were calibrated at the standard condition, so conversion from local conditions to standard flows were not necessary.

3.5.4 Weather data collection

A portable weather station (Davis Weather Station Vantage VUE, Davis Instruments Corporation, Hayward, California) was installed with the PUF sampler at 10 sites. The weather station measured temperature, relative humidity, wind speed, wind direction, and rain.

In addition, meteorological data from the Memphis International Airport were used to support this project. The UofM researchers downloaded meteorological data that match each sample set-up, collection, and retrieval day.

3.6 Laboratory analytical method

3.6.1 Equipment and reagents

In the laboratory, the filters and sorbents were extracted and the extracts were concentrated and analyzed on a GC/MS system for the target PAHs. The process required multiple reagents and instruments, as summarized in Table 7.

Table 7. Equipment and reagents for laboratory PAHs analysis.

Item Description	Vendor	Part#
Equipment		
Accelerated Solvent Extractor	Thermo Scientific	ASE 350
Concentrator	Biotage	Turbo Vap II
GC/MS Analytical System	Agilent Technology	7890B/5977A
Gases		
Helium, UHP	Airgas	HE UHP300
Nitrogen, UHP	Airgas	NI UHP300
Solvents		
Dichloromethane (DCM)	Fisher Scientific	D37-4
Acetone	Fisher Scientific	A18-4
Hexane	Fisher Scientific	H292-4
Standard Solutions		
PAHs Standard Mix	AccuStandard Inc.	H-QME-01
EPA 8270 Semivolatile Internal Standards	SigmaAldrich	CRM48902
PAH Addictions	AccuStandard Inc.	M-8100-R
Surrogate Standard Mix	Restek	31826
Perylene	AccuStandard Inc.	H-121S
Coronene	AccuStandard Inc.	H-116S
Cyclopenta(c,d)pyrene	AccuStandard Inc.	H-242S
Dibenzothiophene	AccuStandard Inc.	H-117S

3.6.3 Spiking lab surrogates

On the analysis day, all the collection media were taken out of the freezer and kept at room temperature for 1 hour. All samples and blanks were spiked with 50 µL of laboratory surrogate solution that contained 10 ng/µl of fluorene-d10 and pyrene-d10. Each analyzed sample was evaluated to ensure the recovery of each surrogate compound was within 60-120% of the nominal spiked value. Results falling outside of these limits indicated potential analyte loss or enhancement either through sample collection and handling and/or extraction process and must be qualified appropriately when reported to AQS.

3.6.4 Extraction of sampling media

For each sample, the QFF and PUF/XAD/PUF were loaded together in a 100mL stainless-steel extraction cell. Two 30mm disposable cellulose filters were installed at the bottom of the cell before loading sampling media to prevent blockages of the frit in the bottom end cap. The cells were then placed on the autosampler of the ASE350, and the samples were extracted sequentially following the optimized parameters (Table 8). Each extraction took about 30 min and yielded about 120 mL of extract in the collection bottle. After extraction, the collection bottle was sealed with a new cap and then stored in a freezer at −18 °C, if the extracts were not concentrated and analyzed immediately. Extracts were analyzed within 40 days of extraction.

Table 8. Dionex ASE 350 parameters for PAH sample extraction

Parameter	Set point/Value
Solvent Ratio	Hexane: Acetone (v:v)=3:1
Temperature	60 °C
Cycles	3
Purge	60 sec
Static time	5 min
Flush	50%

3.6.5 Filtration of extracts

The frozen extracts were moved out of the freezer and equilibrated to room temperature. All the extracts were filtered to remove the water content, given the high humidity of air in this region. A filtration funnel was prepared by adding a small plug of deactivated glass wool at the neck and 50-60g of anhydrous sodium sulfate (10-60 mesh, Fisher Scientific Inc., Waltham, MA) on the top. Each extract was eluted through the filtration materials, as displayed in Figure 5. After the elution, 30 mL of hexane was eluted to ensure all the analytes were washed out of the filtration materials. The extracts and hexane were collected in a 250 ml pre-cleaned evaporation tube for the final concentration.

Figure 5. The cleanup column

3.6.6 Concentration of extracts

The filtered extract was concentrated in an automated solvent evaporation system (TurboVap II, Biotage, Charlotte, NC) with a 0.5 mL endpoint stem. The TurboVap evaporator blew the extract down to 0.3 mL (Figure 6) with a gentle nitrogen flow following an optimized procedure (Table 9). After extraction, the extract was transferred to a 2-mL GC autosampler amber vial and added up to the 1mL marker with hexane.

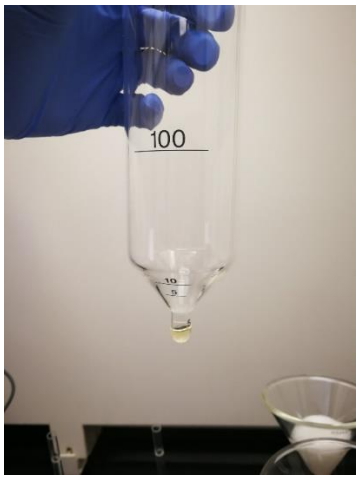


Figure 6. The final extract in tube

Table 9. TurboVap evaporator parameters for PAH extract concentration

Parameter	Set point/Value
Bath temperature	40 °C
Flow rate	Start at 2.5mL/min, then up to 3.0mL/min in 20 min
Total run time	30 mins or more depending on the moisture of the extract

3.6.7 Adding internal standard solution

Internal standards (ISs) were added to all the final extracts before GC/MS analyses to correct for MS variability and potential matrix effects. Each extract was added with 10 uL of IS solution containing naphthalene-d8, acenaphthene-d10, perylene-d12, phenanthrene-d10, and chrysene-d12.

3.6.8 GC/MS analysis

GC/MS Instrumentation. This study used an Agilent 7890B/5977A GC/MS system with a 7963A Autosampler Tray. The GC housed an HP-5ms Ultra Inert column (30m × 0.25mm ID × 0.25 µm film), capable of separating the target PAHs, surrogates, and ISs with appropriate resolution. The carrier gas was helium.

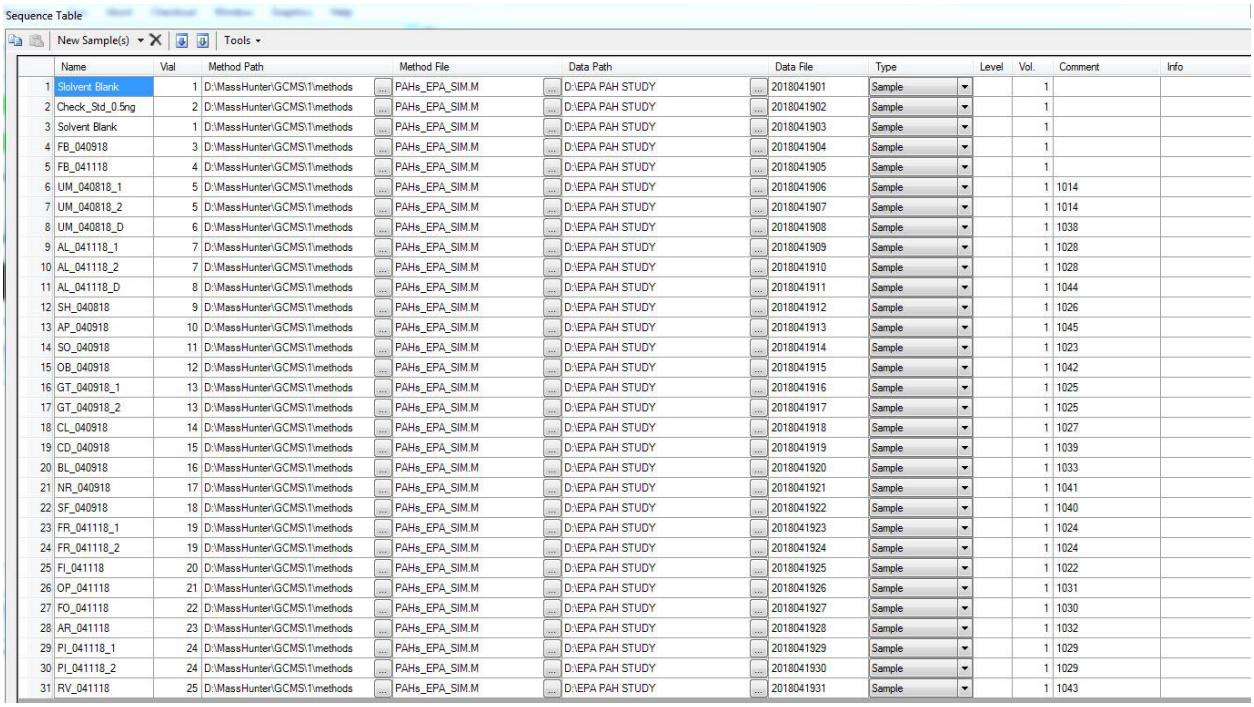
MS tuning. The MS was tuned prior to the analyses of samples using the “eTune” program.

GC/MS parameters. The GC/MS parameters are summarized in Table 10. The GC injection volume was 1.0 µL, and the MS was operated in SIM mode to maximize sensitivity to ions of the target PAHs.

Table 10. GC/MS operating conditions for PAHs analysis.

Parameters	Conditions
Gas Chromatography	
Column	Agilent Technology, DB-5ms (0-325°C; 30m*250µm*0.25µm)
Carrier Gas	Helium
Injection Volume	1 µL, Splitless
Flow Rate	1 mL/min, Constant Flow
Temperature Program	
Initial Temperature	70 °C, hold 4min
Final Temperature	300 °C (20 °C/min to 120 °C, and then 10 °C/min to 300 °C), hold 10 min.
Total Run Time	34.5 min
Mass Spectrometer	
Transfer Line Temperature	290 °C
Source Temperature	230 °C
Electron Energy	70 volts
Ionization Mode	Electron ionization (EI)
Mass Range	40-500 amu, SIM Mode, Time Segments

GC/MS sequence. A typical GC/MS analysis sequence started with analyses of a solvent blank and a check standard solution that contained 0.5 ug/ml of each PAH. Duplicated injections were made every 5 different samples. For each round of sampling, there were 21 samples (including two duplicates), 2 field blanks, 2 solvent blanks, 1 check standard, and 5 duplicate GC injections, as illustrated in Figure 7. A typical sequence consisted of 31 injections/analyses.


Sequence Table

New Sample(s) | Tools

	Name	Vial	Method Path	Method File	Data Path	Data File	Type	Level	Vol.	Comment	Info
1	Solvent Blank	1	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041901	Sample		1		
2	Check_Std_0.5ng	2	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041902	Sample		1		
3	Solvent Blank	1	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041903	Sample		1		
4	FB_040918	3	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041904	Sample		1		
5	FB_041118	4	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041905	Sample		1		
6	UM_040818_1	5	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041906	Sample		1	1014	
7	UM_040818_2	5	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041907	Sample		1	1014	
8	UM_040818_D	6	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041908	Sample		1	1038	
9	AL_041118_1	7	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041909	Sample		1	1028	
10	AL_041118_2	7	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041910	Sample		1	1028	
11	AL_041118_D	8	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041911	Sample		1	1044	
12	SH_040818	9	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041912	Sample		1	1026	
13	AP_040918	10	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041913	Sample		1	1045	
14	SO_040918	11	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041914	Sample		1	1023	
15	OB_040918	12	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041915	Sample		1	1042	
16	GT_040918_1	13	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041916	Sample		1	1025	
17	GT_040918_2	13	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041917	Sample		1	1025	
18	CL_040918	14	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041918	Sample		1	1027	
19	CD_040918	15	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041919	Sample		1	1039	
20	BL_040918	16	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041920	Sample		1	1033	
21	NR_040918	17	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041921	Sample		1	1041	
22	SF_040918	18	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041922	Sample		1	1040	
23	FR_041118_1	19	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041923	Sample		1	1024	
24	FR_041118_2	19	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041924	Sample		1	1024	
25	FI_041118	20	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041925	Sample		1	1022	
26	OP_041118	21	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041926	Sample		1	1031	
27	FO_041118	22	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041927	Sample		1	1030	
28	AR_041118	23	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041928	Sample		1	1032	
29	PI_041118_1	24	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041929	Sample		1	1029	
30	PI_041118_2	24	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041930	Sample		1	1029	
31	RV_041118	25	D:\MassHunter\GCMS\1\methods	PAHs_EPA_SIM.M	D:\EPA PAH STUDY	2018041931	Sample		1	1043	

Figure 7. Typical GC/MS analysis sequence

3.6.9 GC/MS calibration

Preparation of calibration standard solutions. The initial calibration established 7-point calibration curve for each target PAH. The standard solutions were prepared in hexane at 7 concentrations: 0.02, 0.1, 0.25, 0.5, 1.25, 2.5 and 5.0 µg/mL, equivalent to 0.02, 0.1, 0.25, 0.5, 1.25, 2.5 and 5.0 ng, respectively, with 1-µL GC injection. All the standard solutions also contained surrogate and IS compounds.

Analyses of calibration solutions. To establish calibration curves, each concentration was analyzed twice following the GC/MS analysis methods described in Section 3.6.8.

Establishment of calibration curves. Each compound was assigned to the IS compound with the nearest retention time. The MS abundance of a compound was normalized by the abundance of the corresponding IS, yielding an abundance ratio. A linear regression curve was then established by plotting abundance ratios and concentrations. The R^2 for the linear regression should be ≥0.995 for target compounds.

Determination of method detection limits (MDLs). The spiking level of 20 ng was chosen for preparing the MDL spiked samples. This spiking level was carefully selected following the NATTS guidelines. A MDL sample was prepared by injecting 10 µL of 2 µg/ml PAH mix solution to the sample media. Seven or more separate MDL samples and seven or more method blank samples were then analyzed following the same analytical procedure as a regular sample. As all the blanks were clean, MDLs were calculated for the spiked samples (MDLsp) by multiplying SDsp by the one-sided student's T value at 99% confidence corresponding to the number of spikes analyzed.

$$MDL_{sp} = SD_{sp} \cdot T_{(n,99\%)} \quad (2)$$

MDLs were determined annually or when changes to the instrument or preparation procedure resulted in significant changes to the sensitivity of the instrument and/or procedure. sample quantitation limit (SQL) is defined as 3.18 times the MDL.

Calibration verification. A check standard solution containing XX ng/ml of each target PAH were analyzed before the analysis of each batch of samples (See the GC/MS sequence in Section 3.6.9.1) to verify the initial calibration. The analysis recovered within ± 30% of the nominal concentration.

3.6.10 Calculations of PAH concentrations

MS data analysis. The target PAHs were identified by referring to a combination of the compound's retention time, the MS library, and the analyst's experience and judgment. The masses were calculated using the calibration curves from the MS.

Calculation of concentrations. The final air concentration of each target PAH was determined by multiplying the concentration in the extract by the final extract volume and dividing by the collected sample air volume at standard conditions of 25°C and 760 mm Hg:

$$C_A = \frac{1000 \times Ct \times Ve}{V_A}$$

where:

C_A = concentration of the target compound in the air (ng/m^3)
Ct = concentration of the unknown sample in the extract (µg/mL)
Ve = final volume of extract (mL)
V_A = volume of collected air volume at STP (m^3)

3.7 Quality assurance and quality control (QA/QC)

3.7.1 Attainment of data quality objectives (DQOs)

The study design and monitoring methods of this 15-month PAHs monitoring campaign met the following data quality objectives.

(1) Measured concentrations of 30 target PAH compounds (Table 1) in ambient air at each monitoring site.

(2) Generated data of sufficiently high and known quality that are nationally consistent. This monitoring program implemented and maintained a robust and functional quality system, executed the latest NATTS monitoring methods for ambient PAHs, and provided sufficient method sensitivity to obtain a limit of detection at or lower than that at which adverse health effects have been determined.

(3) Collected sufficient data to represent the annual average ambient concentrations of PAHs at each monitoring site. This study collected one sample at each site every 12 days over 15 consecutive months, resulting 34-36 samples at each site.

(4) Complemented existing programs. This study was integrated with existing programs in MTA, including criteria pollutant monitoring, near road monitoring, and National Core (NCore).

(5) Reflected community-oriented population exposure. Stationary monitors were sited to be representative of average concentrations within a 0.5- to 4-kilometer area (i.e., the neighborhood scale). These neighborhood-scale measurements were more reflective of typical population exposure and could be incorporated in the estimation of long-term population risk.

(6) Represented geographic variability. The selected monitoring sites represented a variety of conditions and environments that would allow the characterization of different emissions sources and meteorological conditions. The total of 19 sites

- included neighborhoods with high population risk;
- covered a gradient of industrial, urban, suburban, and rural areas;
- reflected the variability among pollutant patterns across communities; and
- included background monitoring in the Meeman Forest State Park that had no localized sources.

3.7.2 Measurement quality objectives (MQOs)

Measurement quality objectives (MQOs), or acceptance criteria are designed to evaluate and control various phases (i.e., sampling, preparation, and analysis) of the measurement process to ensure that total measurement uncertainty is within the range prescribed by the DQOs. The

specific MQOs of this study included completeness, representativeness, precision, bias, and sensitivity. The key paramters and criteria are summarized in Table

(1) Completeness: The entire study should obtain at least 85% of valid data compared to the amount that was expected to be obtained under correct, normal conditions.

(2) Representativeness: The monitoring site locations should be reflective of exposure to estimate long-term risk among all the populations in the industrial, urban, suburban and rural areas in MTA. Sampling must occur at one-in-twelve day frequency over 24 ± 1 hours. If a sample had run for less than 18 hours, the sample was considered void and were not analyzed at the UofM laboratory.

(3) Comparability: Ambient PAHs should be collected and analyzed following EPA TO-13A method and NATTS guidelines. The methods and procedures used in this project should be consistent with existing national, state, and local monitoring programs.

(4) Precision: The percent difference of duplicate co-located samples should be within 30% and that of duplicate laboratory analyses should be within 15% for concentrations above the sample quantitation limits (SQLs).

(5) Bias: Measurement error must be no more than 30%.

(6) Sensitivity: Methods used to characterize PAHs should have the sensitivity to monitor at concentrations likely to be of health and/or regulatory concern if at all possible.

The quality of the data were evaluated and controlled to ensure that it was maintained within the established acceptance criteria. The data were considered of sufficient quantity and quality for the decision-making to commence if the above MQOs were met.

Table 11. Summary of MQOs and acceptance criteria for PAHs analysis

Parameter	Description and Details	Required Frequency	Acceptance Criteria
Solvent Blank (SB)	Aliquot of Solvent (without IS) analyzed to ensure the GC/MS is free of interferences and of compounds of interest (target PAHs, internal standards, and surrogates)	Prior to each DFTPP tune check	No target compound, IS, or surrogates qualitatively detected
Method Blank (MB)	Blank cartridge and QFFs taken through all extraction and analysis procedures	One with every extraction batch of 20 or fewer field-collected samples	Target analyte amounts ≤2x MDL
Field Blank (FB)	Blank cartridge and QFF assembly exposed to ambient atmosphere for minimally five minutes	One per month	Target analyte amounts ≤5x MDL

Parameter	Description and Details	Required Frequency	Acceptance Criteria
Cartridge Batch Blank	A cartridge (and QFF) selected for analysis to ensure acceptable background levels in the batch of cartridges	One cartridge for each batch of 20 or fewer prepared cartridges	All target compounds each ≤10 ng/cartridge
Initial Calibration (ICAL)	Analysis of a minimum of five calibration levels covering approximately 0.1 to 2 µg/ml	Initially, following failed DFTPP tune check, failed CCV, or when changes to the instrument affect calibration response. Recommended every six weeks.	Average RRF ≤30% RSD and each calibration level must be within ±30% of nominal For quadratic or linear regression, r≥0.995, each calibration level must be within ±30% of nominal
Continuing Calibration Verification (CCV)	Analysis of a known standard at the mid-range of the calibration curve to verify ongoing instrument calibration	Following each DFTPP tune check not followed by ICAL and recommended at the conclusion of each sample sequence	Recovery within ±30% of nominal of RRF with 30% of mean ICAL RRF
Field Surrogate Compounds	Deuterated PAHs which access recovery during sample collection, handling, and analysis	Added to every cartridge prior to field deployment	Recovery 60-120% of nominal spiked amount
Extraction Surrogate Compounds	Deuterated PAHs which access recovery during sample extraction and analysis	Added to media before extraction	Recovery 60-120% of nominal spiked amount
Internal Standards (IS)	Deuterated PAHs added to extracts to assess the impact of and correct for variability in instrument response	Added to all calibration standards, QC samples, and field sample extracts except the SB	Area response within 50-200% of the response of the mid-level calibration standard in the ICAL
Laboratory Control Sample (LCS)	Cartridge spiked with known amount of target analyte	Minimally quarterly. Recommended as one with every extraction batch of 20 or fewer field-collected samples	Recovery 60-120% of nominal spiked amount
Laboratory Control Sample Duplicate (LCSD)	Duplicate cartridge spiked with known amount of target analyte	Minimally quarterly. Recommended as one with every extraction batch of 20 or fewer field-collected samples	Recovery 60-120% of nominal spiked amount and precision ≤20% RPD compared to LCS
Replicate Analysis	Replicate analysis of a field sample extract	Once with every analysis sequence	Precision ≤10% RPD for concentrations ≥0.5 µg/ml
Collocated samples	Sample collected concurrently with the primary sample	10% of primary samples for sites conducting collocated sampling (as required by workplan)	Precision ≤20% RPD for concentrations ≥0.5 µg/ml

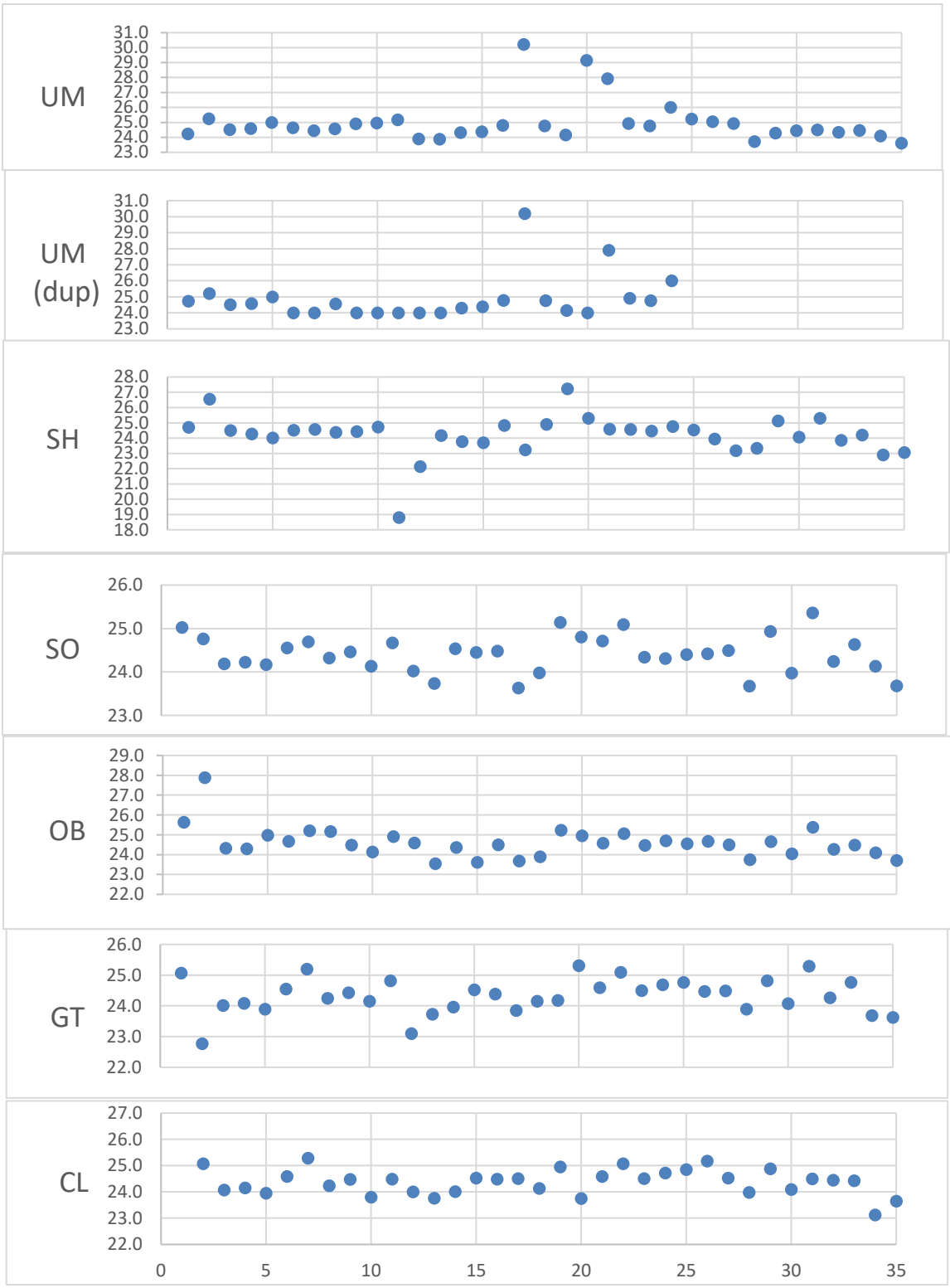
Parameter	Description and Details	Required Frequency	Acceptance Criteria
Retention Time (RT)	RT of each target PAH, surrogate compound, and internal standard	All qualitatively identified compounds	Target analytes within ±0.06 RRT units of mean ICAL RRT Internal standards within ±0.33 minutes of mean ICAL RT

3.7.3 Completeness

The study team had completed 36 cycles of sampling by May 26th, 2019, and collected a total of 818 samples. It was concluded that the study team completed all the planned sampling cycles. The original plan required 504 samples, the revised plan required 720 samples, and we actually completed 818 samples, by keeping the original budget. The sample completeness was over 100%: it was 120% in terms of sampling cycle and 114% in terms of sample size.

3.7.4 Representativeness

The sample representativeness was characterized by spatial representativeness, sampling frequency, and sampling duration. Requirements for spatial representativeness and sampling frequency were met, as described in Section 3. This section presents the results of sampling durations. Figure 8 summarized sampling durations (in hours) in control charts by site. Using the criterion of 23-25 hour sampling, 17.6% of samples went out of the range.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29.0
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27.0
26.0
25.0
24.0
23.0
UM (dup)
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29.0
28.0
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23.0
SH
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19.0
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23.0
OB
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28.0
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26.0
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24.0
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22.0
GT
26.0
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24.0
23.0
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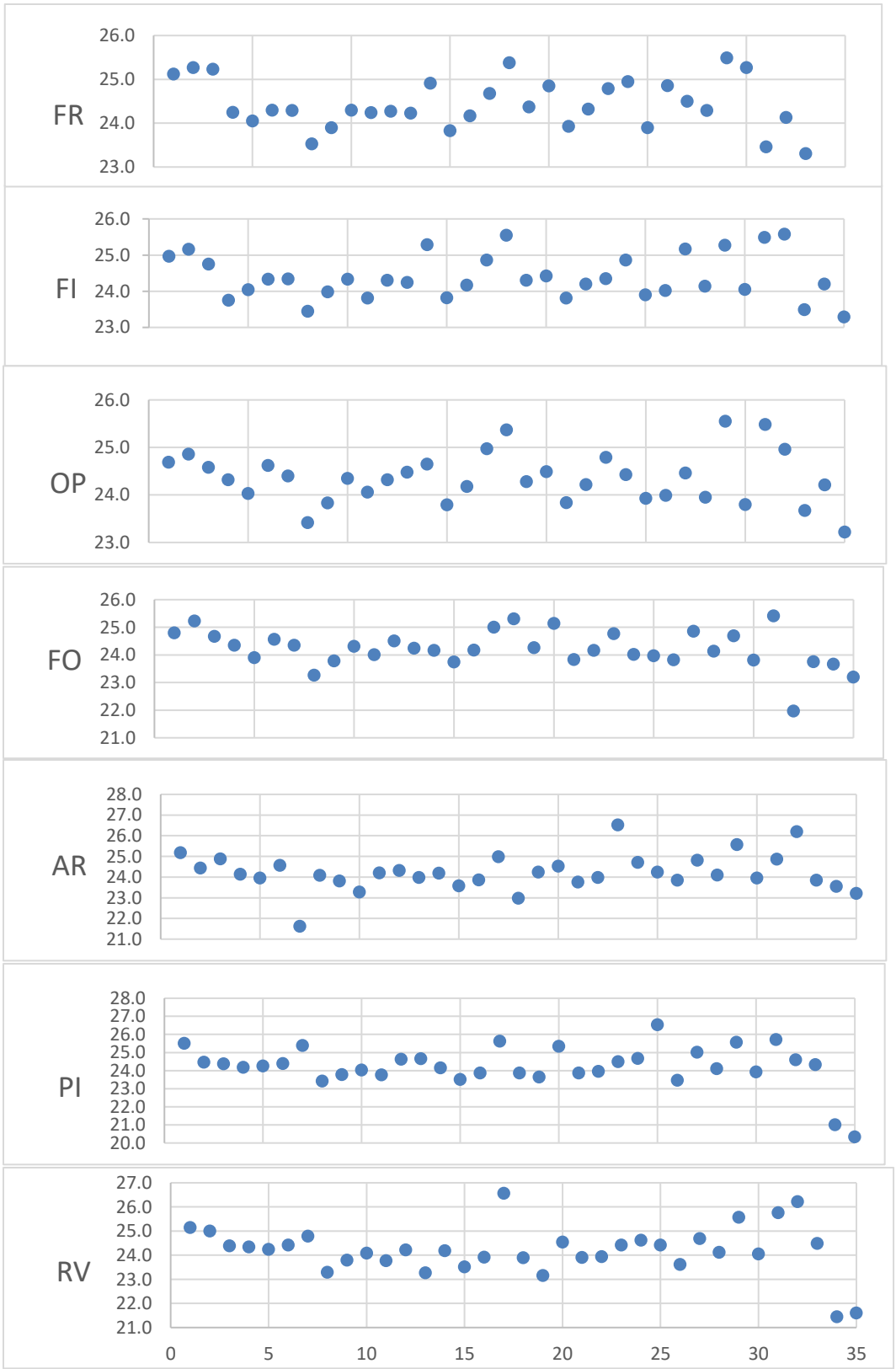
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21.0
PI
28.0
27.0
26.0
25.0
24.0
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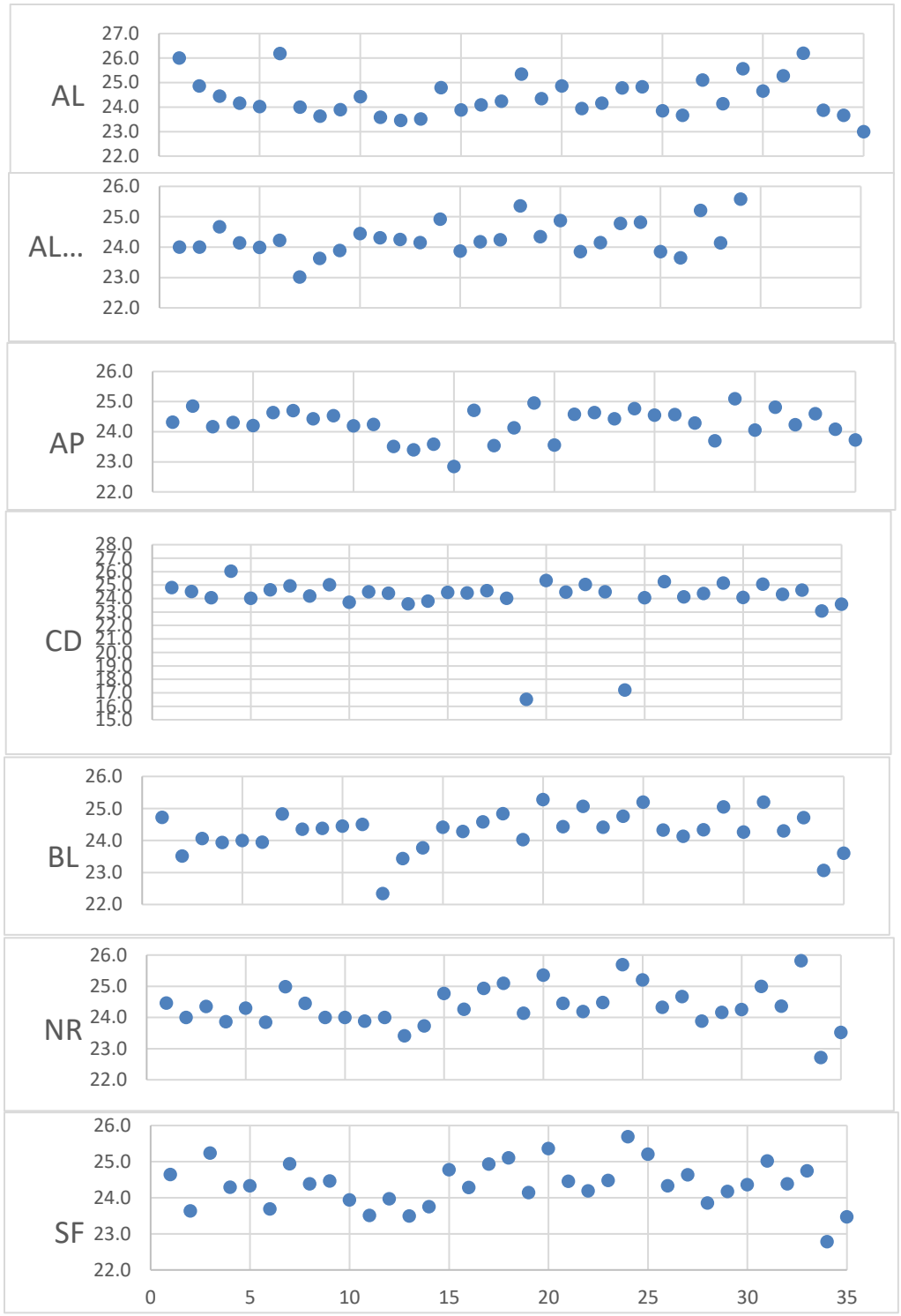


Figure 8. Sampling duration (in hours) of all the sampling events by site

3.7.5 QA performance of GC/MS calibrations

Two full calibrations were performed before and during the study period (03/27/2018 - 05/24/2019). The first full calibration was performed on 02/15/2018, and second on 09/06/2018. Laboratory GC/MS calibration performance measures included solvent blank, cartridge blank, instrument precision, analytical method precision, linearity of the calibration curve, relative standard deviation (RSD) of relative response factors (RRFs), and the method detection limits (MDLs).

Solvent blanks. A solvent blank not fortified with IS was analyzed prior to calibration to ensure the instrument was sufficiently clean to continue analysis. All solvent blanks were clean with no target compounds detected.

Method blanks. A total of seven method blanks were prepared and analyzed for the full calibrations (Table 12). NAP showed masses of 0.094 – 0.887 ng in the method blanks. Considering a nominal sample volume of 288 m3, the background levels were equivalent to up to 0.003 ng/m^3 air concentrations, which were negligible in comparison to the typical NAP concentrations. A few 3-4 ring PAHs had low levels in the method blanks, but mostly were below 0.02 ng. The total mass was below 1 ng in all the method blanks except one that had a total mass of 1.070 ng. Overall, the background contamination of sampling materials and analytical solvents met the criterion.

Table 12. Masses of target PAHs in method blanks analyzed for full calibrations.

PAHs		Masses of Target PAHs (ng)					
	#1	#2	#3	#4	#5	#6	#7
NAP	0.306	0.342	0.887	0.547	0.634	0.359	0.094
ACY	0.003	0.005	0.006	0.005	0.006	0.004	0.002
ACP	0.005	0.006	0.022	0.009	0.017	0.006	0.002
FLR	0.009	0.011	0.035	0.015	0.028	0.011	0.005
9-FL	0.012	0.015	0.025	0.014	0.025	0.013	0.000
DBT	0.000	0.000	0.007	0.000	0.005	0.002	0.001
PHE	0.012	0.015	0.058	0.018	0.045	0.015	0.011
ANT	0.001	0.002	0.002	0.001	0.002	0.002	0.001
FLT	0.003	0.003	0.005	0.003	0.006	0.004	0.000
RET	0.003	0.003	0.004	0.002	0.004	0.004	0.002
PYR	0.005	0.006	0.008	0.006	0.006	0.006	0.000
BcP	0.000	0.001	0.001	0.001	0.001	0.001	0.000
CPP	0.000	0.000	0.001	0.001	0.001	0.001	0.000
BaA	0.000	0.002	0.002	0.002	0.002	0.002	0.000
CHR	0.002	0.002	0.002	0.003	0.002	0.002	0.000
BbjkF	0.000	0.000	0.000	0.000	0.000	0.000	0.000
DMBA	0.000	0.000	0.000	0.000	0.000	0.000	0.000
BeP	0.000	0.000	0.000	0.000	0.000	0.000	0.000
BaP	0.000	0.000	0.000	0.000	0.000	0.000	0.000
PER	0.000	0.000	0.000	0.000	0.000	0.000	0.000
3MC	0.000	0.000	0.000	0.000	0.000	0.000	0.000

PAHs		Masses of Target PAHs (ng)					
	#1	#2	#3	#4	#5	#6	#7
DhACR	0.000	0.000	0.000	0.000	0.000	0.000	0.000
DjACR	0.000	0.000	0.000	0.000	0.000	0.000	0.000
IcP	0.000	0.000	0.000	0.000	0.000	0.000	0.000
DhANT	0.000	0.000	0.000	0.000	0.000	0.000	0.000
BgP	0.000	0.000	0.000	0.000	0.000	0.000	0.000
DBC	0.000	0.000	0.000	0.000	0.000	0.000	0.000
DlP	0.000	0.000	0.000	0.000	0.000	0.000	0.000
COR	0.000	0.000	0.000	0.000	0.000	0.000	0.000
DeP	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Total	**0.365**	**0.415**	**1.070**	**0.633**	**0.788**	**0.435**	**0.135**

Precision of duplicate GC/MS analyses. In the first calibration, the percent differences of duplicate injections averaged 2.0% over all the PAHs, with a range from 0.4% to 4.4%. In the second calibration, the percent differences of duplicate injections averaged 2.0% over all the PAHs, with a range from 0.3% to 4.4%. The precision was stable between two calibrations, indicating the GC/MS had high and stable reproducibility.

Precision of laboratory control sample duplicate. In the first calibration, the percent differences of duplicate lab control samples averaged 4.6% over all the PAHs, with a range from 0.1% to 17.5%. In the second calibration, the percent differences of duplicate lab control samples averaged 2.0% over all the PAHs, with a range from 0.5% to 74.8%. The second calibration had a few compounds that showed poor precisions, including FLR-d10, PHE, DBC, and DeP. Overall, the precision of laboratory control sample duplicates met the criteria of $\leq$15%.

Linearity indicated by R^2. In the first calibration, R^2 of calibration curves ranged from 0.9659 to 1.0000. The compounds with R^2<0.995 were DjA, DBC, DlP, and DeP, all being the late eluting heavy PAHs. In the second calibration, R^2 of calibration curves ranged from 0.9897 to 1.0000. Only 9-FL had a slightly low R^2 of 0.9897. Overall, the linearity of calibration curves was good in the range of 0.02 – 5 ng, with only a few exceptions.

Linearity indicated by RSD. Following data acquisition for the calibration standards, the relative response factor (RRF) of each surrogate and target compound in each calibration level was determined as follows:

$$RRF = (A_s/A_{IS}) / (C_s/C_{IS}) \quad (1)$$

where:

A_s = peak area for quantitation ion of the surrogate or target compound
A_{IS} = peak area for quantitation ion of the assigned internal standard compound
C_s = concentration of the surrogate or target compound
C_{IS} = concentration of the assigned internal standard compound

The RSD was calculated as the quotient of the standard deviation of 7 RRFs by the average of 7 RRFs, and was expressed as a percentage. In the first calibration, the 25 PAHs eluting before

retention time of 26 min had RSDs <30%, but 8 out of the later 9 PAHs had RSDs >30%. In the second calibration, the compounds before retention time of 25 min generally had RSDs <30%, but the later eluting PAHs had RSDs >30%. Overall, the RSDs data indicated that early-eluting light PAHs had good linearity, but late eluting heavy PAHs had inconsistent RRFs over the calibration concentration range.

Method detection limits (MDLs). The MDLs ranged 0.0015–1.2674 ng and 0.0021–0.1639 ng in the first and second calibrations, respectively. Based on a nominal sampling flow rate of 200 L/min, 24 hr sampling resulted in a nominal sample volume of 288 m^3. Then the MDLs, in terms of air concentrations, ranged 0.0054–4.4006 ng/m^3 and 0.0072–3.0378 ng/m^3 in the first and second calibrations, respectively. The two calibrations yielded similar MDLs, indicating the consistency in laboratory analyses of PAH samples. The MDLs were lower than the cancer and non-cancer criteria (Table 14), meaning the monitoring method was sensitive enough to monitor concentrations likely to be of health and/or regulatory concern.

Table 13. Performance of the first GC/MS calibration

#	Name	Ret Time (min)	MS Pre (%)	Metd Pre (%)	Slope	R^2	RSD (%)	MDL (ng)	MDL (ng/m^3)
IS	**NAP-d8**	**8.41**							
1	NAP	8.45	0.4	1.9	1.7819	0.9951	2.0	1.2674	4.4006
IS	**ACP-d10**	**12.04**							
2	ACY	11.69	0.5	17.5	2.9262	0.9993	15.3	0.0085	0.0295
3	ACP	12.11	1.0	16.3	2.2004	0.9999	1.1	0.0345	0.1197
LS	FLR-d10	13.17	0.7	16.4	1.9991	1.0000	1.3	0.0029	0.0102
4	FLR	13.24	0.9	15.7	2.7207	1.0000	4.8	0.0545	0.1892
IS	**PHE-d10**	**15.36**							
5	9-FL	14.95	1.6	0.9	1.0616	0.9971	27.5	0.0368	0.1276
6	DBT	15.12	1.3	0.9	1.9203	0.9999	2.2	0.0121	0.0419
7	PHE	15.41	1.1	0.6	2.1027	1.0000	2.2	0.0915	0.3176
8	ANT	15.51	2.1	0.4	1.9061	0.9993	20.2	0.0030	0.0105
FS	FLT-d10	18.18	1.7	1.0	2.0300	0.9994	9.4	0.0031	0.0109
9	FLT	18.21	1.8	0.9	2.3355	0.9998	13.0	0.0083	0.0287
10	RET	19.59	1.7	2.5	1.0456	0.9984	17.6	0.0054	0.0188
IS	**CHR-d12**	**21.63**							
LS	PYR-d10	18.68	1.7	1.1	2.2990	0.9990	8.2	0.0056	0.0193
11	PYR	18.72	1.6	0.2	2.3432	0.9987	8.5	0.0086	0.0299
12	BcP	21.13	0.8	1.1	1.5076	0.9998	5.4	0.0031	0.0107
13	CPP	21.57	1.0	2.2	1.7444	0.9997	31.0	0.0030	0.0103
14	BaA	21.59	0.7	1.4	2.0442	0.9991	23.9	0.0041	0.0141
15	CHR	21.69	1.0	1.0	2.1405	0.9994	4.4	0.0037	0.0130
IS	**PER-d12**	**24.78**							
16	BbjkF	24.04	3.4	13.7	2.1648	0.9969	6.0	0.0153	0.0533
17	DMBA	24.07	1.5	0.5	0.8071	0.9960	11.5	0.0028	0.0096
18	BeP	24.55	2.1	0.1	2.4056	0.9947	19.7	0.0212	0.0737
FS	BaP-d12	24.61	1.0	2.5	2.2094	0.9992	18.3	0.0088	0.0306
19	BaP	24.66	2.7	0.5	2.0889	0.9993	18.0	0.0035	0.0122
20	PER	24.83	4.4	1.0	2.1616	0.9994	5.2	0.0031	0.0108
21	3MC	25.25	2.9	2.7	2.2055	0.9982	27.0	0.0025	0.0085
22	DhACR	26.80	3.1	1.3	0.8813	0.9922	46.1		
23	DjACR	26.91	3.5	2.6	0.8280	0.9870	57.4		
24	IcP	27.34	2.3	11.7	1.5213	0.9980	40.0	0.0057	0.0199
25	DhANT	27.44	2.4	11.1	1.5023	0.9976	46.7	0.0035	0.0122
26	BgP	28.05	2.8	11.4	1.9807	0.9986	17.4	0.0043	0.0150
27	DBC	28.16	3.0	0.6	0.4136	0.9810	40.3		
28	DlP	32.75	4.3	3.6	0.4583	0.9659	33.1	0.0021	0.0072
29	COR	33.89	2.6	11.3	1.0203	0.9925	27.8	0.0015	0.0054
30	DeP	34.01	3.1	3.2	0.2752	0.9824	36.6		

Table 14. Performance of the second GC/MS calibration

#	Name	Ret Time (min)	MS Pre (%)	Metd Pre (%)	Slope	R^2	RSD (%)	MDL (ng)	MDL (ng/m^3)
IS	**NAP-d8**	**8.40**							
1	NAP	8.43	0.3	34.0	2.3726	0.9999	1.7	0.1639	0.5690
IS	**ACP-d10**	**12.01**							
2	ACY	11.67	1.6	4.0	3.8916	0.9989	12.9	0.0087	0.0301
3	ACP	12.08	0.4	17.4	2.4808	0.9999	4.4	0.0088	0.0305
LS	FLR-d10	13.15	1.5	74.8	2.0620	0.9999	32.9	0.0279	0.0969
4	FLR	13.21	2.2	21.9	3.1230	0.9999	36.8	0.0067	0.0234
IS	**PHE-d10**	**15.35**							
5	9-FL	14.97	1.4	4.2	1.9517	0.9897	23.0	0.0034	0.0119
6	DBT	15.11	0.4	8.6	2.3453	0.9998	2.1	0.0023	0.0078
7	PHE	15.39	1.3	33.5	2.2716	1.0000	7.4	0.0253	0.0880
8	ANT	15.51	2.6	8.7	2.4942	0.9998	8.0	0.0042	0.0146
FS	FLT-d10	18.16	0.8	6.3	2.1931	0.9998	7.8	0.0023	0.0078
9	FLT	18.20	0.4	13.1	2.7723	0.9999	10.6	0.0081	0.0282
10	RET	19.57	0.9	0.8	1.2063	0.9998	13.1	0.0026	0.0091
IS	**CHR-d12**	**21.60**							
LS	PYR-d10	18.66	1.2	0.5	1.8984	0.9998	2.9	0.0036	0.0124
11	PYR	18.70	0.8	5.5	2.5334	1.0000	4.0	0.0132	0.0459
12	BcP	21.10	0.5	0.7	1.6086	0.9998	2.2	0.0034	0.0119
13	CPP	21.55	1.4	0.7	2.5979	0.9980	28.9	0.0029	0.0100
14	BaA	21.57	1.5	2.9	2.4051	0.9998	17.4	0.0021	0.0072
15	CHR	21.66	0.4	5.2	2.3452	1.0000	7.8	0.0058	0.0201
IS	**PER-d12**	**24.74**							
16	BbjkF	24.01	2.2	3.6	8.8363	0.9968	5.4	0.0080	0.0277
17	DMBA	24.03	2.3	0.5	1.1063	0.9984	5.6	0.0066	0.0229
18	BeP	24.51	2.6	5.6	2.2505	0.9986	3.2	0.0158	0.0547
FS	BaP-d12	24.57	2.6	1.8	1.9001	0.9991	10.0	0.0124	0.0431
19	BaP	24.62	4.2	11.1	2.3371	0.9993	19.9	0.0113	0.0393
20	PER	24.79	1.8	10.5	2.4121	0.9996	2.6	0.0071	0.0245
21	3MC	25.47	3.0	13.9	2.4482	0.9990	75.3	0.0064	0.0222
22	DhACR	26.76	4.1	15.3	1.8316	0.9979	61.2	0.0132	0.0457
23	DjACR	26.88	3.3	27.0	1.8398	0.9989	72.9	0.0117	0.0405
24	IcP	27.29	2.1	1.7	2.1615	0.9988	97.2	0.0211	0.0734
25	DhA	27.38	2.5	6.3	2.2570	0.9995	84.7	0.0189	0.0656
26	BgP	27.98	2.3	17.8	2.3764	0.9991	3.5	0.0069	0.0239
27	DBC	27.93	4.4	28.5	1.4057	0.9997	46.3	0.0113	0.0392
28	DlP	32.24	3.2	8.6	1.4921	0.9988	20.5	0.0026	0.0091
29	COR	33.77	3.4	11.9	0.9995	0.9995	8.7	0.0037	3.0378
30	DeP	33.91	3.4	30.2	0.9982	0.9982	21.8	0.0026	2.0019

Table 15. Comparison of MDLs with the health criteria of target PAHs

PAHs	IURs (CAL) 10^{-6} per ng/m^3	Con at 10^{-6} ng/m^3	RfC (IRIS) ng/m^3	Con at HQ=0.1 ng/m^3	MDL1 ng/m^3	MDL2 ng/m^3	Meet Y/N
NAP	0.034	29.4	3000	300	4.401	0.569	Y
ACY			100	10	0.030	0.030	Y
ACP			100	10	0.120	0.031	Y
FLR			1000	100	0.189	0.023	Y
9-FL					0.128	0.012	N/A
DBT					0.042	0.008	N/A
PHE			50	5	0.318	0.088	Y
ANT			50	5	0.011	0.015	Y
FLT			50	5	0.029	0.028	Y
RET					0.019	0.009	N/A
PYR			50	5	0.030	0.046	Y
BcP					0.011	0.012	N/A
CPP					0.010	0.010	N/A
BaA	0.11	9.1	50	5	0.014	0.007	Y
CHR	0.011	90.9	50	5	0.013	0.020	Y
BbjkF	0.11	9.1	50	5	0.053	0.028	Y
DMBA	71	0			0.010	0.023	Y
BeP					0.074	0.055	N/A
BaP	1.1	0.9	3	0.3	0.012	0.039	Y
PER					0.011	0.025	N/A
3MC	6.3	0.2			0.009	0.022	Y
DhACR	0.11	9.1			0.000	0.046	Y
DjACR	0.11	9.1			0.000	0.041	Y
IcP	0.11	9.1	50	5	0.020	0.073	Y
DhANT	1.2	0.8	50	5	0.012	0.066	Y
BgP			50	5	0.015	0.024	Y
DBC	1.1	0.9			0.000	0.039	Y
DlP	11	0.1			0.007	0.009	
COR					0.005	3.038	Y
DeP	1.1	0.9			0.000	2.002	Y

Notes: IURs=Inhalation Unit Risks; RfC = Reference Concentration (in µg/m^3); 10^{-4} risk: The concentration level (in µg/m^3) that causes a cancer of 10^{-4}. Sources: IRIS = Integrated Risk Information System; CAL= California EPA; Criteria are retrieved from U.S. EPA's Integrated Risk Information System at http://www.epa.gov/IRIS/. N/A: Not applicable.

3.7.6 QA performance of field samples

Solvent blanks. Two solvent blanks were analyzed before the analyses of each batch of field samples (Figure 7) to make sure the analytical system was sufficiently clean for analysis. All the analyses did not detect any target PAHs, IS compounds, or surrogate compounds.

Method blanks. Method blanks were occasionally prepared and analyzed to check background PAH levels associated with all solvents and materials used for sample preparation and extraction. A total of nine methods blanks were used for the entire monitoring campaign. Results

showed that method blanks had trace levels of some 2~4-ring PAHs, including NAP, ACP, FLR, PHE, ANT, and RET. The levels of NAP were below 0.36 ng and those of other PAHs were mostly below 0.01 ng (Table 16). All the method blanks had sum PAHs below 1 ng, meeting the criterion.

Field blanks. Two field blanks were collected and analyzed along with each batch of the actual field samples. Field blanks had very low levels of some 3-ring and 4-ring PAHs, but most being below 0.1 ng. Only one blank sample had the sum of all compounds ≥ 1 ng (Table 16).

Table 16. Masses (ng) of PAHs found in method blanks and field blanks.

Name	Field Blanks (n=71)				Method Blanks (n=9)			
	Mean	Med	Min	Max	Mean	Med	Min	Max
NAP	0.29	0.26	0.02	1.11	0.13	0.10	0.03	0.36
ACY	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.00
ACP	0.03	0.01	0.00	0.54	0.01	0.00	0.00	0.02
FLR	0.05	0.03	0.00	0.33	0.01	0.01	0.00	0.03
9-FL	0.01	0.01	0.00	0.05	0.00	0.00	0.00	0.01
DBT	0.01	0.01	0.00	0.06	0.00	0.00	0.00	0.00
PHE	0.12	0.05	0.01	0.54	0.02	0.02	0.01	0.05
ANT	0.01	0.00	0.00	0.06	0.01	0.00	0.00	0.04
FLT	0.01	0.01	0.00	0.07	0.00	0.00	0.00	0.00
RET	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.00
PYR	0.02	0.01	0.00	0.09	0.01	0.00	0.00	0.02
BcP	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.00
CPP	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
BaA	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00
CHR	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.02
BbjkF	0.00	0.00	0.00	0.06	0.00	0.00	0.00	0.00
DMBA	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.00
BeP	0.00	0.00	0.00	0.06	0.00	0.00	0.00	0.00
BaP	0.00	0.00	0.00	0.03	0.00	0.00	0.00	0.00
PER	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3MC	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
DhACR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
DjACR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
IcP	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00
DhANT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
BgP	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00
DBC	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
DlP	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00
COR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
DeP	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Total	**0.58**	**0.40**	**0.06**	**2.12**	**0.20**	**0.17**	**0.05**	**0.52**

Calibration checks. A quality control (QC) check sample was analyzed to confirm the calibration curves before analyzing each batch of actual samples. One µL of 0.5 ng/ µL check sample was injected into the equipment and the analyses results were compared with the original concentrations. The calculated masses of most target PAHs were within ±30% of the actual

concentrations (Table 17). A few later eluting heavy PAHs, including IcP, DhANT, DlP, and COR, showed lower values in comparison to the expected concentrations, possibly due to the storage losses of the stock solution.

Table 17. Performance of check standard analyses

PAHs	Actual mass (ng)	Calculated mass (ng)	Ratio
NAP	0.5	0.6	1.2
ACY	0.5	0.5	1.0
ACP	0.5	0.5	1.0
FLR	0.5	0.5	1.0
9-FL	0.5	0.5	0.9
DBT	0.5	0.5	1.1
PHE	0.5	0.5	1.0
ANT	0.5	0.5	0.9
FLT	0.5	0.5	0.9
RET	0.5	0.4	0.9
PYR	0.5	0.6	1.1
BcP	0.5	0.5	1.1
CPP	0.5	0.4	0.9
BaA	0.5	0.4	0.9
CHR	0.5	0.5	1.0
BbjkF	1.5	2.0	1.3
DMBA	0.5	0.6	1.2
BeP	0.5	0.5	1.0
BaP	0.5	0.4	0.9
PER	0.5	0.5	0.9
3MC	0.5	0.4	0.7
IcP	0.5	0.3	0.6
DhANT	0.5	0.3	0.6
BgP	0.5	0.3	0.7
DlP	0.5	0.3	0.6
COR	0.5	0.3	0.5

Precision of duplicate analyses of the same extract. The instrumental precision was measured though repeated analyses of the same final extract. Duplicate analyses were performed for 10% of the samples (Figure 7), and a total of 171 duplicate pairs were analyzed during the whole sampling year (Table 18). The mean precisions of duplicate analyses ranged from 0.3% to 18.1%, most below 10% except the compound BeP. Some high percent differences were observed duo to the very low concentrations of certain PAHs.

Table 18. Precision of duplicate analyses of the same extract (n=171).

PAHs	Percent differences of repeated injections (%)					
	Mean	Min	Median	P75	P90	Max
NAP	0.9	0.0	0.3	0.7	2.2	9.9
ACY	5.1	0.0	2.8	6.8	12.7	32.2
ACP	1.9	0.0	0.7	1.7	5.1	19.3

FLR	1.6	0.0	0.6	1.4	3.2	33.6
9-FL	2.2	0.0	0.8	2.3	6.5	17.8
DBT	1.1	0.0	0.6	1.3	2.4	17.7
PHE	0.8	0.0	0.4	0.7	1.6	16.9
ANT	8.2	0.1	4.7	10.8	21.7	45.9
FLT	1.5	0.0	0.4	1.1	3.6	21.0
RET	3.6	0.1	1.6	3.5	8.7	29.4
PYR	1.9	0.0	0.5	1.4	3.5	24.6
BcP	3.0	0.0	1.7	3.5	7.6	23.1
CPP	6.3	0.0	4.7	8.3	14.6	37.7
BaA	4.8	0.0	3.6	7.2	10.4	18.7
CHR	3.1	0.0	0.8	2.7	7.8	43.6
BbjkF	8.9	0.0	5.5	13.2	23.4	45.6
DMBA	13.3	1.4	11.4	19.2	26.5	35.2
BeP	8.1	0.0	4.6	10.5	23.5	41.8
BaP	12.9	0.2	6.9	16.1	29.6	75.6
PER	7.7	0.3	4.2	7.1	23.3	39.6
3MC	7.6	4.2	5.3	9.3	11.6	13.2
DhACR	9.6	1.4	3.2	12.2	24.0	33.1
DjACR	N/A	N/A	N/A	N/A	N/A	N/A
IcP	9.5	0.0	5.6	11.8	24.0	74.1
DhANT	9.2	0.1	6.1	11.4	19.7	62.7
BgP	8.4	0.0	5.3	11.7	21.5	43.4
DBC	15.5	0.1	10.7	26.2	38.3	56.6
DlP	11.6	0.5	9.2	13.8	18.3	42.6
COR	13.0	0.6	10.8	16.0	24.5	42.7
DeP	18.1	16.1	18.1	19.1	19.7	20.1

Recoveries of surrogates. Surrogate compounds were spiked to each sample prior to the filed deployment and the laboratory analyses to check the recoveries of analyztes. Almost all the recovery results fell between 60%-120%, which met the crietion as required (Table 19).

Table 19. Recoveries of field and laboratory surrogates

Surrogate	N	Mean	STD	Min	Median	P90	Max
Fluorene-d10	968	72.8	21.9	30.0	72.0	93.0	141.3
Fluoranthene-d10	968	86.9	13.8	45.3	86.2	100.0	179.1
Pyrene-d10	968	89.2	16.0	45.6	86.6	107.6	168.2
Benzo(a)pyrene-d12	968	95.3	21.0	34.1	95.8	119.6	180.5

Precisions of collocated samples. Two pairs of collocated samples were collected in each sampling cycle to check the duplicate precision. In the later sampling stage, 11 duplicate samples at the UM site and 6 duplicates at the AL site were missed due to the missing of a sampler. A total of 53 duplicate samples were collected during the routine sampling. We also conducted additional replicate sampling tests between Cycles 17 and 18 on October 21-23, 2018, resulting in 4 triplicate samples. In total, we collected 57 duplicate or triplicate samples, reaching 10% of samples as required.

Duplicate sample analyses provide a check on sampling and analytical precision. Good precision was realized for the LMW PAHs such as NAP, ACY, ACP, and all the HMW PAHs. The percent differences ranged from 1.2-37.1% in median concentrations. High percent differences (e.g., >50%) were observed for the 3-4 ring PAHs.

Table 20. Precision of collocated samples.

Name	Percent differences (%) of duplicates (n=53)				Relative standard deviation (%) of collated triplicates			
	Mean	Min	Med	Max	UM (10/20)	AL (10/20)	UM (10/21)	AL (10/21)
NAP	18.2	0.3	13.1	83.9	28.7	9.7	23.8	19.2
ACY	44.0	2.5	33.4	141.0	36.2	28.2	13.5	4.1
ACP	33.2	1.6	29.4	117.1	41.8	10.7	33.9	8.5
FLR	78.6	6.6	91.1	146.5	45.1	31.9	55.9	52.7
9-FL	68.7	2.8	64.7	151.7	47.6	17.3	54.6	42.0
DBT	83.0	0.9	71.0	172.7	50.6	44.8	79.9	73.4
PHE	81.0	2.1	81.1	157.1	50.2	44.1	70.9	67.6
ANT	129.2	25.0	128.6	191.3	56.8	85.7	86.7	89.6
FLT	81.9	2.1	65.3	166.7	63.1	45.5	81.7	75.4
RET	62.8	1.3	69.6	132.3	58.3	40.4	20.6	21.8
PYR	77.4	0.3	55.8	170.5	63.1	39.5	80.6	62.5
BcP	51.9	0.1	16.8	164.3	45.0	23.7	45.7	1.8
CPP	54.2	1.4	18.8	177.0	36.8	15.3	34.0	19.5
BaA	59.2	0.3	19.6	177.7	32.9	73.2	39.3	0.7
CHR	55.7	1.3	32.5	163.1	39.5	43.7	39.7	5.7
BbjkF	19.9	0.5	12.5	69.7	5.9	22.7	3.8	1.9
DMBA	35.5	2.0	18.4	103.4	28.6	6.9	23.2	39.7
BeP	19.5	1.4	13.8	85.0	18.0	17.8	2.7	2.4
BaP	31.2	0.9	23.5	85.3	19.2	22.3	5.9	8.3
PER	49.4	1.7	37.1	143.9	23.4	50.6	1.5	12.3
3MC	1.2	0.7	1.2	1.7	N/A	N/A	N/A	N/A
DhACR	16.3	16.3	16.3	16.3	N/A	N/A	N/A	N/A
DjACR	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
IcP	19.5	0.5	14.0	84.7	15.3	9.1	5.5	3.9
DhANT	25.2	1.0	16.8	119.0	17.8	16.6	3.3	4.5
BgP	17.3	0.5	13.0	83.2	57.7	43.7	5.5	4.5
DBC	22.4	2.3	27.9	40.9	19.9	16.2	17.8	16.5
DlP	11.4	1.4	9.1	23.6	N/A	N/A	13.2	7.2
COR	25.9	0.2	23.9	86.3	N/A	N/A	10.7	10.0
DeP	31.9	22.1	31.9	41.7	N/A	N/A	12.9	15.3

4. Data Analysis Methods

4.1 Groupings of PAHs

For better interpretation of results, we divided the 30 PAHs into three groups based on their molecular weights (MWs): (1) naphthalene (NAP), which is the most volatile and abundant PAH; (2) low molecular weight (LMW) PAHs, which have MW≤228. There were 14 LMW target PAHs and their sum concentration was defined as C(LMW14); and (3) high molecular weight (HMW) PAHs, which have MW≥252. There were 15 HMW target PAHs and their sum concentration was defined as C(HMW15). The sum concentration of all the target PAHs except NAP was defined as C(PAH29), as NAP predominated the sum PAH concentration. The sum concentration of all the 16 EPA priority PAHs was defined as C(PAH16). As NAP predominated the sum PAH concentration, we also defined C(PAH29) and C(PAH15) as sum concentrations of 29 and 15 PAHs, respectively, that were exclusive of NAP.

4.2 Descriptive statistics

We defined sample detection frequency (DF) for a certain PAH as the percent of measurements above its MDL out of the total sample size. Sample DFs were calculated for seasonal and annual data, and site DFs were calculated for the annual data. We calculated descriptive statistics for seasonal and annual data. Descriptive statistics included mean, standard deviation and percentiles.

4.3 Comparison with national PAH monitoring results

The National Monitoring Programs of U.S. EPA include the Photochemical Assessment Monitoring Stations (PAMS) network, Urban Air Toxics Monitoring Program (UATMP), National Air Toxics Trends Stations (NATTS) network, Community-Scale Air Toxics Ambient Monitoring (CSATAM) Program, and monitoring for other pollutants such as Non-Methane Organic Compounds (NMOCs) (USEPA 2015). PAHs in ambient air are routinely monitored at 6-day or 12-day cycles at selected sites of these programs. Airborne PAHs are collected and analyzed per the guidance given in EPA Method TO-13A (USEPA 1999b) and the latest Technical Assistance Document for the NATTS Program (USEPA 2016). The collection media consists of a quartz fiber filter (QFF) and a glass thimble containing polyurethane foam (PUF) and styrene-divinylbenzene polymer resin sorbent (XAD-2 or equivalent) to collect particulate- and gas-phase PAHs. A high-volume sampler draws approximately 200 to 350 m^3 of ambient air over 24 hours. In laboratory, the QFF, PUF and sorbent are extracted using a soxhelator or accelerated solvent extractor (ASE), and the extract is further concentrated to a small volume, e.g., 1 or 0.5 ml. The final concentrated extract is analyzed by gas chromatograph/mass spectrometer (GC/MS) in a select-ion-monitoring (SIM) mode for some or all sixteen priority PAHs. All the sampling and analytical operations follow strict quality assurance (QA) procedures to appropriate accuracy, precision, and representativeness of data, as required by EPA (USEPA 2001b).

For this study, daily average concentrations of 16 priority PAHs were downloaded from U.S. EPA's Air Quality System (AQS) for the period of 2018-2019 (USEPA 2018). This dataset contained PAH measurements collected at a total of 150 different sites. Below detection limit

concentrations, which was present as zero in the data, were replaced with half of the detection limit.

4.4 Spatial comparisons

To compare PAH levels in different areas, we classified the surrounding settings of sites in two ways. First, we roughly classified the settings as urban, suburban, and rural (Table 21), and then the medians of individual and sum PAHs were compared by urbanicity. Second, we further classified the settings into six categories: suburban community (Sub), urban community (Urban), near-road (NR), industrial (IND), airport (AP), and rural, as listed in Table 21. Then the medians of individual and sum PAHs were compared.

Table 21. Classification of sampling sites in the MTA area

Site	Classification 1	Classification 2
SO	Sub	Sub
OB	Sub	Sub
GT	Sub	Sub
CL	Sub	Sub
CD	Sub	Sub
BL	Sub	Sub
FI	Sub	Sub
AR	Sub	Sub
PI	Urban	IND
RV	Urban	IND
UM	Urban	Urban
AL	Urban	NR
SH	Urban	NR
AP	Urban	AP
NR	Urban	NR
SF	Urban	Urban
FR	Urban	Urban
OP	Rural	Rural
FO	Rural	Rural

4.5 Risk assessment

The cancer risks from exposure to carcinogenic PAHs were estimated using the relative potency factors (RPFs) and inhalation unit risk (IUR) method. The BaP equivalent concentration for each carcinogenic PAHs was calculated by multiplying its measured concentration with the corresponding RPF. The values of RPFs are 0.4, 0.08, 0.4, 0.2, 0.1, 0.8, 0.3, 0.03, 1, 0.07, 10, 0.009, 30 and 0.4 for ACY, FLT, CPP, BaA, CHR, BbF, BjF, BkF, BaP, IcP, DhANT, BgP, DIP and DeP, respectively (USEPA, 2010). The BaP equivalent daily concentrations of these 14 carcinogenic PAHs were summed to obtain the toxic quivalent quotient BaP-TEQ at each site, which were then aggregated sequentially to the annual average based on all sites. The cancer risk was then calculated as the product of BaP-TEQ and BaP's IUR. In this study, we used the values of IUR suggested by WHO and California EPA as upper and lower estimates, which were 8.7×10^{-2} and 1.1×10^{-6}, respectively.

4.6 Identification of emission sources

PAH diagnostic ratio is the preferred method for source identification due to its simplicity and validity by numerous studies (Tobiszewski and Namiesnik 2012; Usenko et al. 2010; Yunker et al. 2002). We adopted four diagnostic ratios: FLT/(FLT+PYR), IP/(IP+BgP), FLR/(FLR+PYR), and BaP/(BaP+BgP) that have been shown to differentiate major source categories (Tobiszewski and Namiesnik 2012). Specifically, FLT/(FLT+PYR) ratios of >0.4 and ≤0.4, or IP/(IP+BgP), ratios of >0.2 and ≤0.2 indicate pyrogenic and petrogenic sources, respectively; while FLR/(FLR+PYR) ratios of >0.5 and ≤0.5 separate diesel and gasoline emissions; and BaP/BgP ratios of >0.6 and ≤0.6 differentiate traffics and non-traffic sources.

4.7 Association between PAH exposure and socioeconomic status (SES)

Census-tract level racial composition and SES data were obtained from the Census 2000, Summary File 3 (U.S. Census Bureau 2012). The following variables were extracted: percent of the population that is African American (shortened as “AA%”), median household income in 1,000s, percent of the population below the poverty level, percent of female headed households with children, percent of the population with less than a high school education, total population, population density (1,000 person /mile^2), and percent of the population aged 65 and older. The data pertaining to PAH exposure and SES data were linked by the census tract number, and the associations were explored using linear regressions.

5. Results and Discussion

5.1 Detection of PAHs

5.1.1 Overall and seasonal detection frequencies

All of the 30 target PAHs were detected in the MTA during the 14-month monitoring period (Table 22). The 15 LMW PAHs (NAP through CHR) were detected in 100% of the samples. In addition, two HMW PAHs were also detected in almost all the samples, including BbjkF (DF=97%) and BeP (DF=95%). Four HMW PAHs were detected in 50-75% of samples, including PER (DF=49%), IcP (DF=63%), DhANT (DF=64%), and BgP (DF=71%). The rest 9 HMW PAHs showed detectable levels in less than 25% of samples. In particular, 3MC, DhACR, DjACR, DIP, and DeP were occasionally detected in only less than 5% of samples.

The detection frequencies were generally consistent across seasons. This consistency implies the consistency in the monitoring methods and the stable sources in this region. A few abnormalities were noted. PER was rarely detected in spring; 3MC was only detected in fall; IcP, DhANT, and BgP were frequently detected in summer; and DBC showed higher DFs in fall and winter.

Table 22. Detection frequencies (%) of target PAHs in MTA.

PAHs	Sampling frequency in each season/DF%				
	All	Spring	Summer	Fall	Winter
NAP	100	100	100	100	100
ACY	100	100	100	100	100
ACP	100	100	100	100	100
FLR	100	100	100	100	100
9-FL	100	100	100	100	100
DBT	100	100	100	100	100
PHE	100	100	100	100	100
ANT	100	100	100	100	100
FLT	100	100	100	100	100
RET	100	100	100	100	100
PYR	100	100	100	100	100
BcP	100	100	100	100	100
CPP	100	100	100	100	100
BaA	100	100	100	100	100
CHR	100	100	100	100	100
BbjkF	97	96	100	94	99
DMBA	25	22	19	16	42
BeP	95	90	100	96	96
BaP	49	40	47	52	63
PER	24	8	36	39	22
3MC	2	0	0	11	0
DhACR	4	9	0	1	2

PAHs	Sampling frequency in each season/DF%				
	All	Spring	Summer	Fall	Winter
DjACR	1	2	0	0	1
IcP	63	62	92	59	43
DhANT	64	52	97	83	33
BgP	71	62	92	70	64
DBC	15	3	5	38	20
DlP	5	4	2	1	13
COR	12	13	14	6	14
DeP	1	1	1	1	0

5.1.2 Sample detection frequencies by site

The detection frequencies of ambient PAHs were generally consistent across the 19 monitoring sites in MTA (Table 23). A few outstanding DFs were noted. DMBA was detected in <15% of samples at FO, AR, PI, and RV sites. BaP and COR were more frequently detected at the three urban sites, UM, AL and SH. DBC was detected in <10% of samples at the four suburban sites, GT, CL, CD, and BL.

Table 23. Sample detection frequencies (%) by site

PAH	UM	AL	SH	AP	SO	OB	GT	CL	CD	BL	NR	SF	FR	FI	OP	FO	AR	PI	RV
NAP	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
ACY	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
ACP	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
FLR	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
9-FL	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
DBT	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
PHE	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
ANT	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
FLT	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
RET	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
PYR	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
BcP	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
CPP	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
BaA	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
CHR	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
BbjkF	100	100	97	100	94	97	97	94	97	100	100	100	100	94	97	91	97	94	91
DMBA	57	29	34	31	29	23	20	29	29	29	26	26	18	20	29	6	9	11	14
BeP	100	97	94	97	94	94	97	91	94	94	100	97	100	91	91	86	91	94	94
BaP	74	89	83	51	40	37	31	43	31	46	60	51	70	46	40	37	37	37	37
PER	43	49	57	37	20	11	14	17	11	20	31	26	27	17	14	14	14	9	20
3MC	3	3	9	3	3	3	0	3	3	0	3	6	0	0	3	3	0	3	0
DhACR	0	3	6	3	0	6	3	6	9	0	6	3	9	6	3	3	3	3	3
DjACR	0	0	0	6	3	0	0	0	3	0	3	3	0	3	0	0	0	0	0
IcP	66	77	77	74	66	63	69	71	54	63	63	66	61	60	57	54	54	54	54
DhANT	69	63	74	74	74	69	74	71	60	63	71	66	64	49	57	54	54	54	54
BgP	80	86	83	86	74	71	71	69	63	63	77	71	73	69	63	54	60	60	69
DBC	14	23	26	29	20	14	6	9	3	3	14	23	18	14	17	11	11	14	11
DlP	11	9	11	9	9	3	6	3	6	3	6	6	6	3	0	0	3	3	0
COR	34	26	34	14	9	6	9	6	6	6	14	17	18	6	3	3	3	6	6
DeP	3	0	3	3	3	0	0	0	0	0	0	0	0	0	0	0	3	0	0

5.2 Concentrations and comparison with national levels

The average C(PAH29) was 45.4±57.1 ng/m^3 and C(NAP) was 27.1±45.9 ng/m^3 in this region (Table 24). Obviously, NAP was the most predominant PAH, accounting for 37% of the total PAH. Other predominant PAHs included PHE (16.08±18.36 ng/m^3), FLR (8.79±17.3 ng/m^3), ACP (5.86±8.36 ng/m^3), FLT (4.25±6.18 ng/m^3), PYR (2.94±4.18 ng/m^3), and ANT (2.48±4.69 ng/m^3). All these are among EPA's 16 priority PAH list. The rest of the compounds all had mean concentrations below 2 ng/m^3. Extreme concentrations were also noted, e.g., the maximum concentrations of NAP, ACP, FLR and PHE were 991.8, 113.9, 362.5, and 116.6 ng/m^3, respectively.

Table 24. Descriptive statistics of ambient air concentrations of PAHs (ng/m^3).

PAHs	N	Mean	SD	Min	Med	P90	Max
NAP	663	27.06	45.92	0.29	18.16	51.65	991.78
ACY	663	0.41	0.82	0.01	0.15	0.99	12.87
ACP	663	5.86	8.36	0.01	2.70	15.26	113.94
FLR	663	8.79	17.29	0.05	3.96	22.33	362.50
9-FL	663	1.74	2.57	0.01	0.85	4.15	21.83
DBT	663	1.84	2.70	0.01	0.84	4.72	17.48
PHE	663	16.08	18.36	0.11	8.89	40.33	116.56
ANT	663	2.48	4.69	0.01	0.61	7.57	51.13
FLT	663	4.25	6.18	0.03	1.77	10.66	45.32
RET	663	0.34	0.39	0.01	0.24	0.70	4.49
PYR	663	2.94	4.18	0.02	1.37	7.51	30.24
BcP	663	0.05	0.09	0.00	0.03	0.10	1.74
CPP	663	0.03	0.05	0.00	0.02	0.06	0.58
BaA	663	0.08	0.15	0.00	0.04	0.18	1.94
CHR	663	0.24	0.28	0.01	0.16	0.47	3.04
BbjkF	663	0.08	0.12	0.00	0.04	0.16	1.79
DMBA	663	0.01	0.02	0.00	0.00	0.02	0.16
BeP	663	0.11	0.11	0.00	0.09	0.22	1.40
BaP	663	0.03	0.07	0.00	0.00	0.09	1.01
PER	663	0.01	0.02	0.00	0.00	0.02	0.19
3MC	663	0.00	0.00	0.00	0.00	0.00	0.05
DhACR	663	0.00	0.00	0.00	0.00	0.00	0.04
DjACR	663	0.00	0.00	0.00	0.00	0.00	0.04
IcP	663	0.02	0.05	0.00	0.01	0.06	0.52
DhANT	663	0.01	0.01	0.00	0.01	0.02	0.22
BgP	663	0.03	0.06	0.00	0.01	0.06	0.88
DBC	663	0.01	0.03	0.00	0.00	0.02	0.51
DlP	663	0.00	0.01	0.00	0.00	0.00	0.14
COR	663	0.00	0.01	0.00	0.00	0.00	0.17
DeP	663	0.00	0.00	0.00	0.00	0.00	0.10
C(LMW14)	663	45.13	56.96	0.30	23.91	117.56	565.37
C(HMW15)	663	0.30	0.41	0.00	0.19	0.64	4.92
C(PAH15)	663	41.30	52.09	0.26	21.97	107.42	557.75
C(PAH29)	663	45.42	57.07	0.30	24.15	117.94	565.88

MTA had higher PAH concentrations compared to those measured in the urban areas in the U.S. (Table 25). The ambient C(PAH15) measured in MTA (41.3±52.1 ng/m^3) was significantly higher than that measured in the U.S. (17.5±34.4ng/m^3), while the lower mean NAP concentration (27.1±45.9 ng/m^3) was observed in MTA. The mean concentrations of ACP, FLR, PHE, ANT, FLT and PYR found in MTA were 5.86, 8.79, 16.08, 2.48, 4.25 and 2.94ng/m^3, respectively, which was 2-10 times higher compared with the U.S. national averages. Most HMW PAH in MTA were slightly lower than or in the same range as the US average levels.

Table 25. Comparison of PAH concentrations between MTA and the U.S.

PAH16	MTA		The U.S.	
	Mean	SD	Mean	SD
NAP	27.06	45.92	36.19	39.51
ACY	0.41	0.82	0.20	0.46
ACP	5.86	8.36	3.80	8.73
FLR	8.79	17.29	3.77	7.42
PHE	16.08	18.36	7.14	15.53
ANT	2.48	4.69	0.32	1.51
FLT	4.25	6.18	1.76	3.57
PYR	2.94	4.18	0.94	1.69
BaA	0.08	0.15	0.10	0.20
CHR	0.24	0.28	0.18	0.30
BbF	0.08	0.12	0.21	0.37
BkF			0.09	0.16
BaP	0.03	0.07	0.10	0.23
IcP	0.02	0.05	0.20	0.59
DhANT	0.01	0.01	0.05	0.08
BgP	0.03	0.06	0.12	0.20
C(PAH15)	41.30	52.09	17.51	34.44

5.3 Seasonal variation

Ambient PAHs displayed significant seasonality in this region. Significantly higher PAH concentrations were found in summer. The mean concentrations of NAP and PAH15 were 47.4±89.1 ng/m^3 and 85.8±60.9 ng/m^3, respectively, 2-8 times higher than those measured in spring, fall and winter (Table 26 and Figures 9 and 10). Two extremely high values of NAP were also found in summer (Figure 9). Concentrations of LMW PAHs were higher in summer and those of HMW PAHs were higher in winter. C(LMW14) was 94.1±67.7 ng/m^3 in summer, compared to 44.1±48.5, 32.0±52.9, and 12.1±14.8 ng/m^3 in spring, fall, winter, respectively (Table 26). Concentrations of HMW PAHs showed small variability in four seasons. These seasonal variations in ambient PAH concentrations reflected the presence of a number of PAH sources, the complexity of the source profiles for individual sources, and the reactivity of the various PAH compounds.

Table 26. Comparison of ambient concentrations of PAHs in MTA by season.

PAHs	Spring (n=226)		Summer (n=145)		Fall (n=140)		Winter (n=152)	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD
NAP	25.65	22.53	47.39	89.09	17.90	12.94	18.22	14.53
ACY	0.37	0.71	0.76	0.84	0.35	1.12	0.20	0.44
ACP	5.74	6.35	12.86	9.24	3.77	9.92	1.27	1.50
FLR	8.38	9.69	18.35	13.30	6.98	30.56	1.96	2.13
FL9	1.80	2.52	3.77	3.58	0.96	0.91	0.44	0.44
DBT	1.93	2.79	3.95	3.51	1.06	1.17	0.40	0.63
PHE	16.18	17.21	31.96	22.73	11.59	11.18	4.92	6.52
ANT	2.45	4.38	5.40	7.15	1.54	2.27	0.62	1.38
FLT	3.96	5.53	9.47	8.65	2.71	3.26	1.12	1.57
RET	0.36	0.37	0.60	0.56	0.23	0.18	0.14	0.16
PYR	2.54	3.17	6.30	5.91	2.43	3.48	0.82	1.12
BcP	0.04	0.05	0.09	0.09	0.06	0.16	0.02	0.02
CPP	0.03	0.05	0.04	0.05	0.03	0.05	0.02	0.03
BaA	0.07	0.12	0.15	0.18	0.08	0.18	0.04	0.05
CHR	0.21	0.24	0.40	0.36	0.22	0.31	0.14	0.11
BbjkF	0.08	0.17	0.11	0.09	0.05	0.08	0.05	0.05
DMBA	0.01	0.02	0.01	0.02	0.00	0.01	0.00	0.01
BeP	0.10	0.11	0.16	0.08	0.11	0.13	0.08	0.09
BaP	0.03	0.08	0.02	0.04	0.03	0.09	0.04	0.07
PER	0.00	0.02	0.01	0.02	0.01	0.01	0.01	0.01
MC3	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00
DhACR	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00
DjACR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
IcP	0.02	0.03	0.01	0.01	0.03	0.05	0.04	0.08
DhANT	0.00	0.01	0.01	0.01	0.01	0.02	0.00	0.01
BgP	0.02	0.03	0.02	0.02	0.03	0.05	0.06	0.11
DBC	0.00	0.02	0.01	0.05	0.01	0.02	0.00	0.01
DlP	0.00	0.01	0.00	0.00	0.00	0.01	0.00	0.01
COR	0.00	0.01	0.00	0.01	0.00	0.01	0.01	0.02
DeP	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00
LMW14	44.06	48.50	94.09	67.68	31.98	52.86	12.11	14.73
HMW15	0.27	0.43	0.36	0.25	0.29	0.45	0.30	0.44
PAH15	40.05	43.35	85.81	60.86	29.81	51.46	11.28	13.79
PAH29	44.33	48.62	94.45	67.79	32.27	52.94	12.41	14.97
PAH30	69.97	59.58	141.83	122.30	50.17	57.34	30.63	25.76

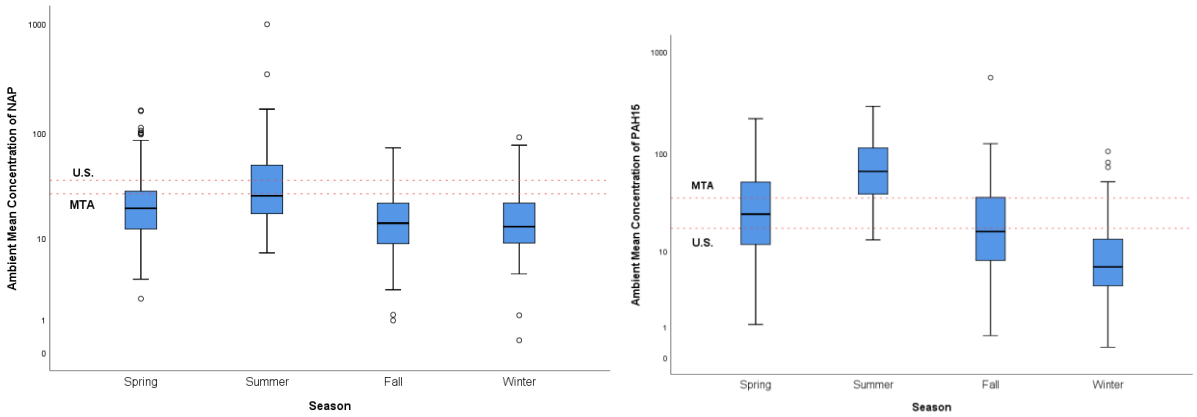


Figure 9. Ambient concentrations of NAP and PAH15 by season.

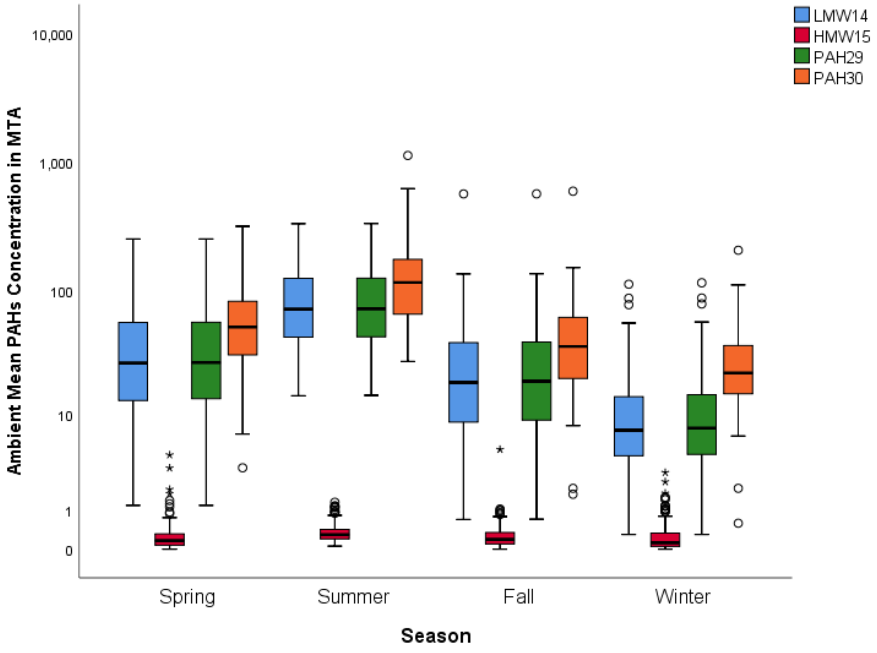


Figure 10. Sum concentrations of PAHs by season.

5.4 Spatial variation

Ambient PAH concentrations displayed obvious spatial patterns in this region. PAH levels were higher at urban sites than those suburban sites, followed by rural sites. For example, the median concentrations of NAP were 25 ng/m^3, 15 ng/m^3, and 11 ng/m^3 in urban, suburban, and rural areas, respectively, and the median concentrations of PAH29 were 55, 39, and 36 ng/m^3 in urban, suburban, and rural areas, respectively (Table 27 and Figure 11).

A further examination revealed that higher concentrations of NAP and HMW PAH were detected at the near-road (NR), near-airport (AP) and industrial park (IND) sites (Table 28 and Figure 12). For LMW PAHs such as ACP, 9-FL, DBT, ANT, RET, PYR, BcP, CPP, BaA and CHR, no differences were observed among the rural, suburban and urban areas (Table 27), but they showed higher levels at the AP, NR and IND sites (Table 28).

Table 27. Descriptive summary of PAH concentrations in MTA by Classification 1.

	Rural (N=70)			Sub (N=280)			Urban (N=313)		
PAH	**Mean**	**SD**	**Median**	**Mean**	**SD**	**Median**	**Mean**	**SD**	**Median**
NAP	13.00	6.79	11.44	21.44	59.59	14.89	35.24	33.91	24.96
ACY	0.64	1.69	0.11	0.29	0.51	0.13	0.47	0.73	0.21
ACP	5.78	14.03	2.32	5.22	6.72	2.45	6.44	7.98	3.06
FLR	15.11	44.50	3.38	7.35	8.69	3.88	8.67	10.88	4.32
9-FL	2.03	3.03	0.60	1.40	1.67	0.82	1.99	3.04	0.89
DBT	2.78	4.32	0.68	1.62	2.11	0.84	1.82	2.67	0.85
PHE	19.96	26.27	6.46	15.15	15.99	9.38	16.04	18.16	8.89
ANT	4.31	7.25	0.48	2.13	3.69	0.75	2.39	4.69	0.59
FLT	5.17	8.02	1.15	3.80	4.76	1.81	4.45	6.80	1.86
RET	0.34	0.45	0.24	0.33	0.37	0.25	0.33	0.40	0.22
PYR	3.70	5.54	0.97	2.51	3.02	1.39	3.17	4.66	1.46
BcP	0.05	0.06	0.02	0.05	0.11	0.03	0.06	0.08	0.03
CPP	0.03	0.03	0.01	0.02	0.04	0.01	0.04	0.05	0.02
BaA	0.09	0.13	0.03	0.06	0.10	0.04	0.10	0.18	0.05
CHR	0.21	0.23	0.11	0.19	0.21	0.14	0.28	0.34	0.18
BbjkF	0.04	0.03	0.02	0.07	0.14	0.03	0.09	0.11	0.05
DMBA	0.01	0.00	0.01	0.01	0.01	0.01	0.01	0.02	0.01
BeP	0.07	0.05	0.06	0.10	0.10	0.07	0.13	0.12	0.10
BaP	0.02	0.02	0.02	0.03	0.07	0.02	0.05	0.08	0.02
PER	0.01	0.00	0.01	0.01	0.01	0.01	0.02	0.02	0.01
3MC	0.01	0.00	0.01	0.01	0.00	0.01	0.01	0.00	0.01
DhACR	0.02	0.00	0.02	0.02	0.00	0.02	0.02	0.00	0.02
DjACR	0.02	0.00	0.02	0.02	0.00	0.02	0.02	0.00	0.02
IcP	0.02	0.02	0.03	0.03	0.04	0.03	0.04	0.06	0.04
DhANT	0.02	0.01	0.01	0.02	0.01	0.01	0.02	0.02	0.01
BgP	0.02	0.02	0.01	0.02	0.04	0.01	0.05	0.08	0.02
DBC	0.02	0.01	0.02	0.02	0.01	0.02	0.03	0.04	0.02
DlP	0.00	0.00	0.01	0.01	0.01	0.01	0.01	0.01	0.01
COR	0.00	0.00	0.00	0.00	0.01	0.00	0.01	0.01	0.00
DeP	0.01	0.00	0.01	0.01	0.00	0.01	0.01	0.01	0.01
LMW14	60.19	95.68	17.90	40.12	43.81	23.75	46.24	55.39	25.32
HMW15	0.15	0.13	0.12	0.24	0.38	0.17	0.38	0.46	0.26
PAH15	55.03	89.87	16.40	36.84	40.06	21.45	42.22	49.75	23.22
PAH29	60.34	95.73	18.01	40.36	43.90	23.88	46.62	55.54	26.01
PAH30	73.34	98.92	35.76	61.80	81.75	39.49	81.86	78.96	54.93

Table 28. Descriptive summary of PAH concentrations in MTA by Classification 2.

PAH/Mean	AP (N=35)	IND (N=70)	NR (N=105)	Rural (N=70)	Sub (N=280)	Urban (N=103)
NAP	29.53	41.21	37.90	13.00	21.44	30.41
ACY	0.31	0.30	0.64	0.64	0.29	0.46
ACP	2.73	8.63	6.21	5.78	5.22	6.45
FLR	2.79	7.09	10.86	15.11	7.35	9.51
9-FL	0.97	0.92	2.44	2.03	1.40	2.60
DBT	0.85	0.88	2.23	2.78	1.62	2.38
PHE	7.47	8.72	20.50	19.96	15.15	19.40
ANT	0.47	1.24	3.16	4.31	2.13	3.03
FLT	1.89	1.81	5.09	5.17	3.80	6.45
RET	0.48	0.19	0.28	0.34	0.33	0.44
PYR	1.48	1.21	3.60	3.70	2.51	4.62
BcP	0.04	0.02	0.06	0.05	0.05	0.08
CPP	0.03	0.01	0.04	0.03	0.02	0.05
BaA	0.07	0.04	0.12	0.09	0.06	0.14
CHR	0.23	0.11	0.32	0.21	0.19	0.36
BbjkF	0.08	0.05	0.13	0.04	0.07	0.09
DMBA	0.01	0.01	0.02	0.01	0.01	0.02
BeP	0.13	0.08	0.17	0.07	0.10	0.13
BaP	0.05	0.02	0.08	0.02	0.03	0.04
PER	0.02	0.01	0.02	0.01	0.01	0.02
3MC	0.01	0.01	0.01	0.01	0.01	0.01
DhACR	0.02	0.02	0.02	0.02	0.02	0.02
DjACR	0.02	0.02	0.02	0.02	0.02	0.02
IcP	0.05	0.03	0.06	0.02	0.03	0.04
DhANT	0.02	0.02	0.02	0.02	0.02	0.02
BgP	0.05	0.02	0.07	0.02	0.02	0.04
DBC	0.03	0.02	0.02	0.02	0.02	0.03
DlP	0.01	0.00	0.01	0.00	0.01	0.01
COR	0.01	0.00	0.01	0.00	0.00	0.01
DeP	0.01	0.01	0.01	0.01	0.01	0.01
LMW14	19.79	31.17	55.55	60.19	40.12	55.96
HMW15	0.39	0.18	0.54	0.15	0.24	0.36
PAH15	17.64	29.23	50.82	55.03	36.84	50.63
PAH29	20.18	31.34	56.09	60.34	40.36	56.32
PAH30	49.70	72.55	93.99	73.34	61.80	86.74

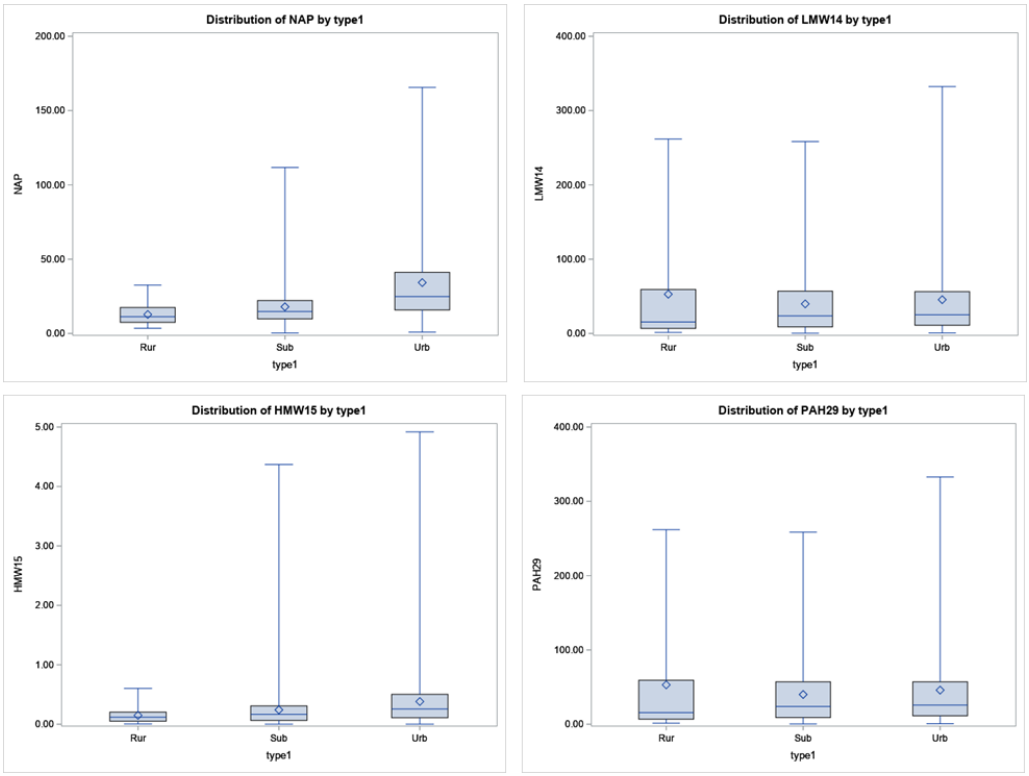


Figure 11. Sum concentrations of PAHs by Type1.

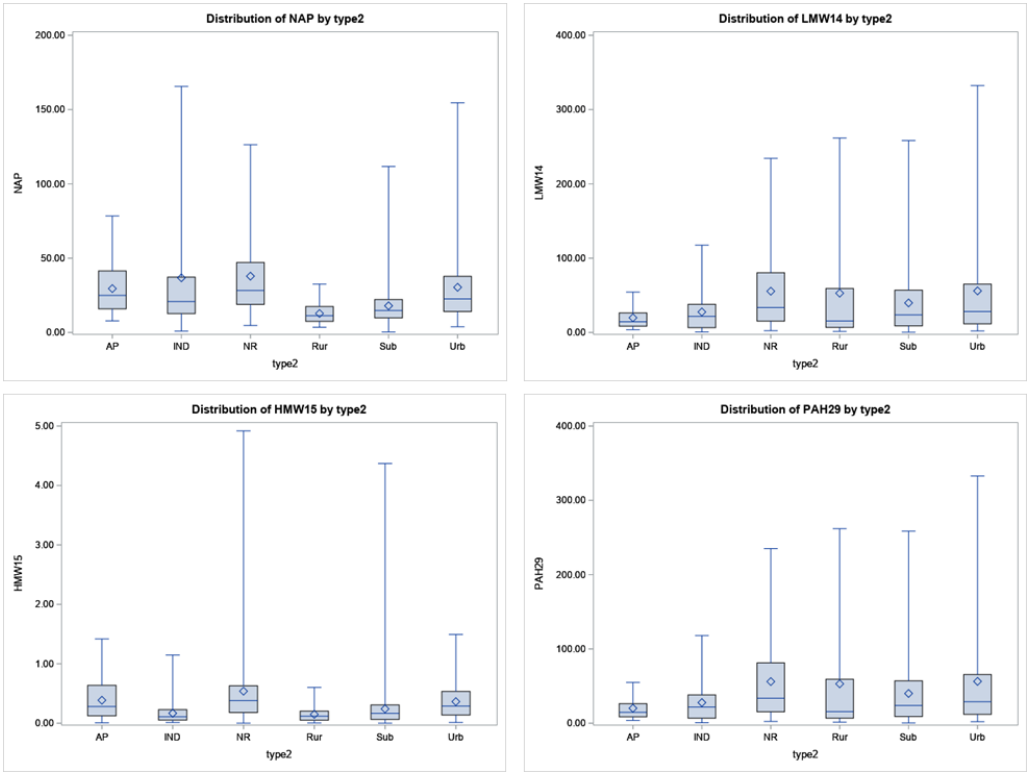


Figure 12. Sum concentrations of PAHs by Type2.

5.5 Risk assessment

The average BaP-TEQ, as the sum of 14 carcinogenic PAHs, was 1.05±1.42 ng/m^3. The average lifetime cancer risk was estimated to be 1.15×10^{-6} based on the TEFs from WHO and 9.09×10^{-5} based on TEFs from Cal EPA (Table 29). These two methods yielded two risks of near 100 times, and we adopted the results based on WHO TEFs as most studies utilized this approach. The cancer risk resulting from NAP exposures was estimated to be 9.20×10^{-7}. Regarding cPAH risks, NATA's estimates were 0.18, 1.62, 1.52, and 0.36×10^{-6} in 1999, 2002, 2005, and 2011, respectively. The PAHs data from a 2007 national analysis showed that the PAH risk was 0.92×10^{-6} (Loh et al. 2007). Regarding NAP cancer risks, NATA's estimates were 2.18, 2.09, 2.31, and 1.36×10^{-6} in 1999, 2002, 2005, and 2011, respectively. The results from our study were comparable to previous national estimates.

Table 29. Cancer risk estimates for carcinogenic PAHs (cPAHs) and NAP

Measures	Mean	Std Dev	Min	Median	Max
cPAHs-WHO	1.15E-06	1.56E-06	2.28E-07	8.09E-07	1.97E-05
cPAHs-CalEPA	9.09E-05	1.23E-04	1.80E-05	6.39E-05	1.56E-03
NAP	9.20E-07	1.56E-06	9.86E-09	6.17E-07	3.37E-05

5.6 Source identification

The two PAH diagnostic ratios depicted the predominant sources of PAHs in ambient air (Figure 13). Overall, almost all the sites had FLR/(FLR+PYR) ratios of >0.5, indicating that diesel sources dominated. As far as emission activities are concerned, there seems to be an approximately even split between the sites with traffic and non-traffic PAH sources, based on the BaP/BgP ratio threshold of 0.6. There were no discernable source patterns between urban suburban and rural areas.

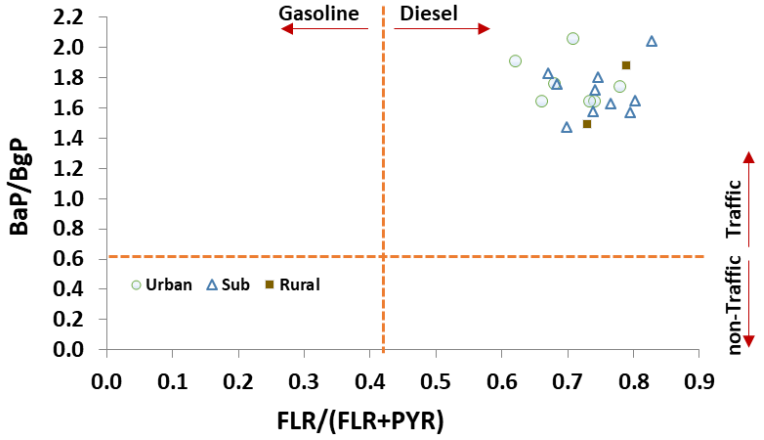


Figure 13. Major sources based on two selected diagnostic ratios.

Notes: FlR/(FLR+PYR) ratios of >0.5 and ≤0.5 indicate diesel and gasoline emissions, while BaP/BgP ratios of >0.6 and ≤0.6 separate traffic and non-traffic sources.

The finding of traffic and non-traffic related fuel combustions as predominant PAH sources are consistent with PAHs source inventory in the U.S. The source inventory compiled for the calendar year of 2004 showed the major sources, not counting consumer product usage which contributed primarily to NAP emissions (USEPA 1998; Zhang and Tao 2009), were traffic oil combustion (23.0%), waste incineration (9.5%), biofuel combustion (9.1%) and petroluem refinery (8.7%) (Zhang and Tao 2009). More detailed source types may be derived from various cutoffs and diagnostic ratios. In this study, both IP/(IP+BgP) and FLR/(FLR+PYR) ratios indicated that diesel combustion source dominated and gasoline source contribution was minimal. This likely reflects the fact that diesel engines produce higher PM emissions than gasoline engines, especially those in the fine (≤2.5 μm) and ultrafine (≤0.1 μm) size fractions that have large surface areas to absorb PAHs (Borras et al. 2009; Matti Maricq 2007). In addition, diesel-fueled generators as an emergency power supply are widely used in commercial and residential areas, and become an important non-traffic PAH source.

The contributing diesel sources could also be further confirmed by the correlations between airborne PAH concentrations and the diesel PM concentrations. As seen in Figure 14, PAH concentrations in the ambient air showed positive correlations with diesel PM concentrations, implying that diesel emissions were a contributor to PAHs.

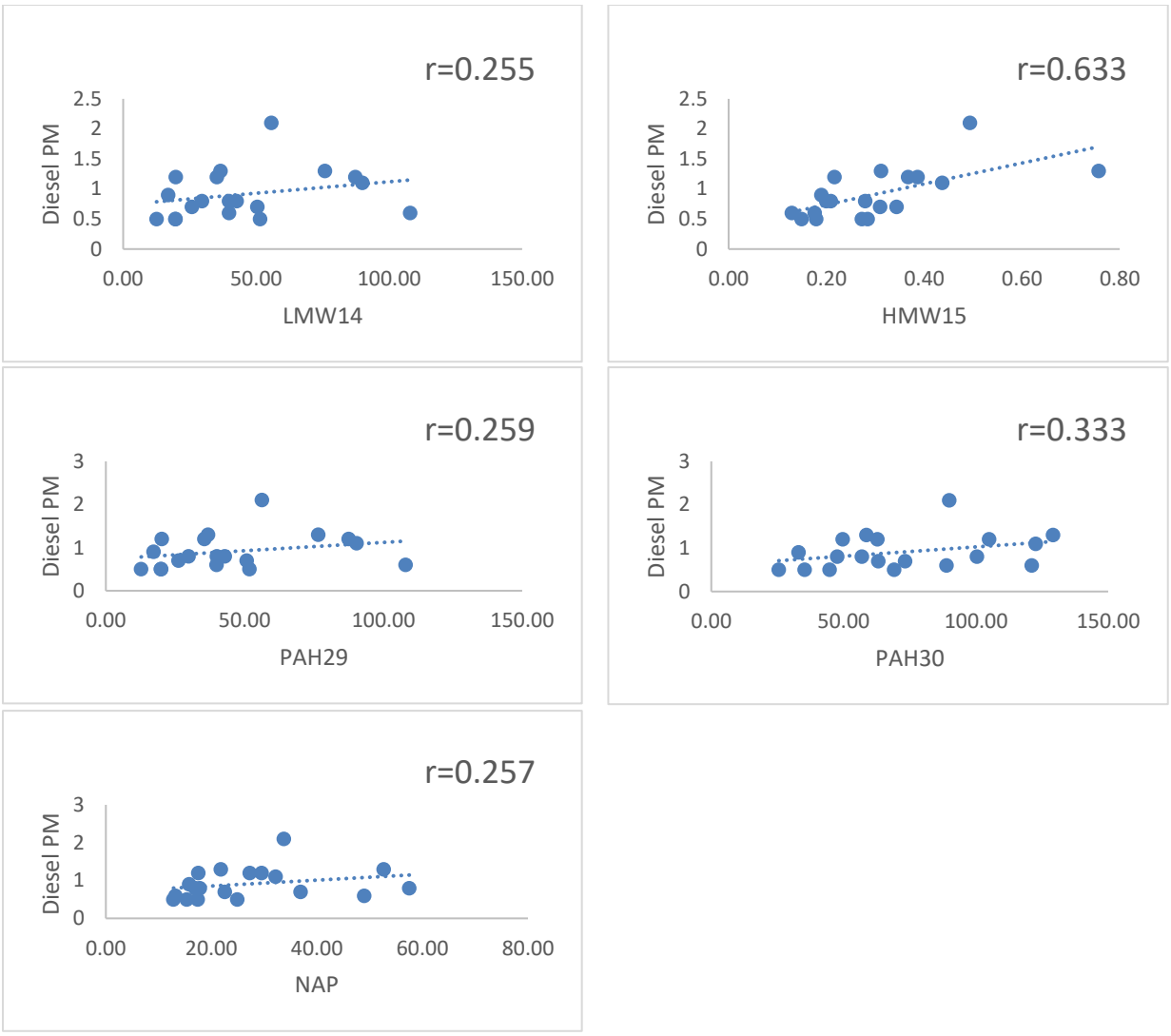


Figure 14. Association between PAH exposure and diesel PM

5.7 Environmental disparities in PAH exposures

Higher environmental exposure existed in low-income areas in MTA. As displayed in Figure 15, there was a clear negative association between PAH concentrations and household income. The negative association persisted for light, heavy, and all PAHs. These facts indicated that exposure increased with decreasing household income, or poorer populations were bearing higher environmental exposures.

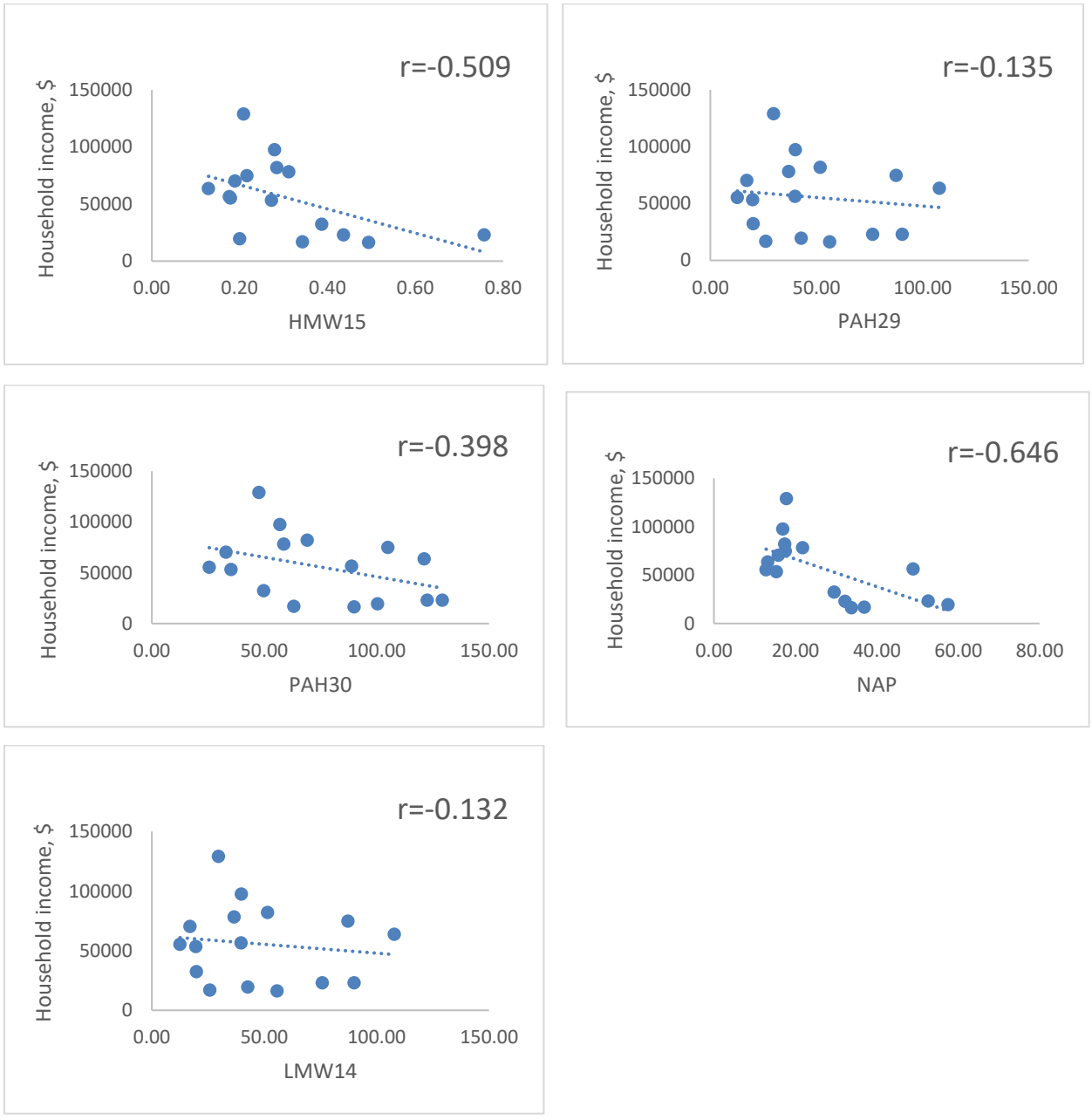


Figure 15. Association between PAH exposure and median household income

6. Community Involvement

6.1 Objectives of community involvement

The goal of community engagement was to strengthen partnerships among community institutions receptive to learning about and using PAH and air pollution data. There were five specific aims for community engagement in the Memphis PAHs Study:

Aim 1: Build a community-government-academic partnership focusing on environmental pollution and environmental justice in the Memphis area;

Aim 2: Develop multiple communication venues to improve the public's knowledge of air pollution, public health, environmental justice, and environmental policy and regulations;

Aim 3: Promote the use of environmental monitoring data by community residents to improve daily decisions to reduce the harmful effects of air pollutants; and

Aim 4: Increase public trust and satisfaction with the Health Department as an expert and credible source of information about regulated products.

This study engaged community members in this region using the community-based participatory research (CBPR) strategy. Through CBPR, the Health Department, University of Memphis researchers, non-profit organizations, and community members established a government-academic-community partnership that involved the communities in all aspects of the research process and potential benefits communities through future interventions or policy changes.

6.2 Community involvement activities

The study team created many channels, venues, and opportunities for public participation, comment and input. The following listed the community outreach activities in chronological order.

6.2.1 Preparation stage

The major tasks in the preparatory stage were partnership building, creating multiple communication venues, and obtaining the public's perception of and input to the project.

Year 2015

10/14/2015. A public meeting on air toxics studies was held in the University Center at the University of Memphis. Dr. Jia presented the study objectives and design to a group of EPA experts, SCHD staff, UM researchers, and the general public.

11/17/2015. Dr. Jia and Larry Smith presented the Memphis PAHs Study in the National EJ Conference at the University of Memphis.

Year 2016

01/14/2016. Pollution Control staff did a short 30-minute radio interview and the PAH study was discussed.

02/17/2016. Pollution Control staff did an interview on local radio regarding environmental issues in general and discussed the PAH study in some detail.

02/29/2016. Pollution Control staff provided an air quality program and the PAH study was discussed with the members of the Fern Society.

03/14/2016. EPA led an environmental jobs fair at Lemoyne Owen College. The University of Memphis and Shelby County Pollution Control Section provided a program for over 200 children specifically focused on the PAH study. In fact, a little jingle was used to get each group of children to shout the entire phrase.

03/15/2016. Larry Smith attended the TDEC yearly environmental conference in Kingsport TN along with Jim Holt. No formal presentation was done but the upcoming PAH study was discussed informally with a number of the participants.

04/27/2016. The Shelby County Pollution Control Section populated a table at an environmental jobs fair at the Frayser Business Academy, a Memphis Charter School, for grades 5th to 8th grade. Over 100 students and teachers were informed about the PAH study.

05/25/2016. Larry Smith attended a Shelby County grants working group and one of the grants discussed was the PAH grant.

06/22/2016. Shelby County staff met with the Memphis Chamber of Commerce staff and briefed them on regional air quality issues and the past REACT study and the upcoming PAH study. We also mentioned the City Space PM 2.5 study as well.

07/21/2016. Shelby County Pollution Control staff gave a program to a group of students enrolled in the Pre Environmental Engineering Program through the U of M. The PAH study was discussed with the group.

08/10/2016. Dr. Jia, Mr. Holt, and Mr. Smith Attended EPA National Air Monitoring conference in St. Louis. During the course of the conference Dr. Jia, Mr. Holt and Larry Smith discussed the PAH study with a number of conference participants. Dr. Jia made several good contacts.

09/01/2016. The study team developed a poster and a flyer for the project.

09/27/2016. Dr. Jia and Pollution Control populated a table at the University of Memphis “Tiger Goes Green” event. The event was well attended and generated a lot of interest in the upcoming PAH project.

11/14/2016. A professional meeting was held in SCHD involving SCHD staff, Drs. John Spengler and Gary Adamkiewicz from Harvard School of Public Health, the Cityspace project staff Dr. Siobhan T. Whitlock, and other staff from EPA Region 4 office, and Dr. Jia from the University of Memphis. The project team gave an overview of this PAH study. The Cityspace team and Memphis PAHs study team agreed it would be more efficient if the two teams hold the public meetings jointly in the future.

Year 2017

02/16/2017. A public meeting for the Memphis PAHs Study was held in the Memphis Central Library. The meeting notice was posted on Facebook and sent to over 100 individuals via email. Ten people attended the meeting. The project team gave an overview of this project, and attendees made valuable comments on the project.

03/01/2017. The study team launched the website for this project: https://memphisair.org/pahs/.

03/07/2017. Project Manager Larry Smith debriefed the Memphis PAHs Study with Shelby County Air Board's monthly meeting.

03/14/2017. Dr. Jia and Larry Smith presented "Memphis Air Toxics Studies" in the 2017 Tennessee Environmental Conference in Kingsport, TN. Over 40 people attended the presentation and provided feedback on the upcoming PAHs study. The project information was well received as the audience represented government agencies, non-profit organizations, industries, consulting companies, academia, and interested individuals.

04/20/2017. Larry Smith attended a meeting of the Memphis 3.0 working group. This group was working on a strategic plan for Memphis. Larry Smith gave a short presentation on the Memphis PAH Study to about 30 people.

05/07/2017. Larry Smith attended a meeting of the Midtown Unitarian church and presented a program on the Memphis PAH Study.

05/02/2017. Larry Smith briefed the University of Tennessee Medical school's pediatrics students on the Memphis PAH Study.

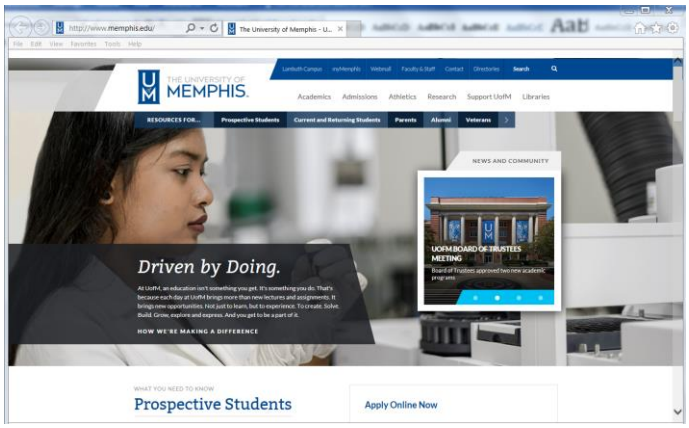
06/16/2017. Larry Smith provided a lunch program for the local chapter of the American Society of Safety Engineers.

08/13/2017. Larry Smith provided a program to 25 science students at LeMoyne Owen College regarding the PAH program.

08/25/2017. Dr. Jia met Jennifer Richardson from Clean Memphis and discussed the plan to give lectures on air pollution in schools through Clean Memphis' environmental education program.

08/25/2017. Larry Smith spoke with 5 area residents of the Riverview Community regarding the PAH project and the station that resides at Riverview school.

08/30/2017. The photo of Dr. Jia's lab and in particular, the GC/MS for this project is used on the main page of the University of Memphis website: www.memphis.edu.



10/01-12/15/2017. There were multiple site recruitment trips in this period. Larry Smith and Dr. Jia distributed the study flyer and talked to property owners about the project.

10/14/2017. Larry Smith and Dr. Jia gave a presentation on the Memphis PAHs Study, titled "Monitoring for Combustion Related Air Toxics in the Memphis Area" in the 15th Annual National Environmental Justice Conference, Memphis, TN.

10/25/2017. Dr. Jia, in collaboration with Jenna Richardson from Clean Memphis, gave guest lectures to 5 science classes in Cordova Middle School and talked about the Memphis PAH Study.

12/13/207. Shelby County Health Department helped EPA Region 4 held two CitySpace Community Meetings. Larry Smith and Dr. Jia reported the study design and preliminary data to the attendees.

6.2.2 Monitoring stage

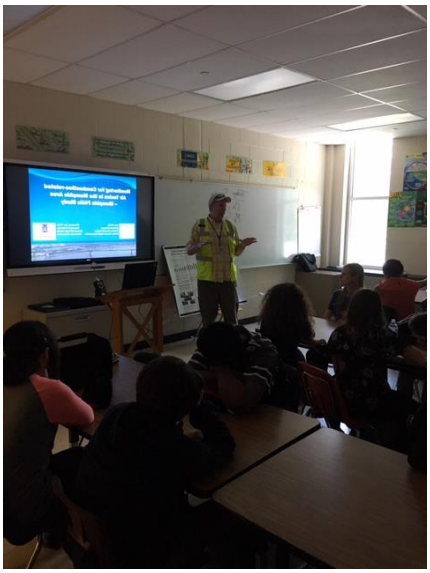
During the monitoring stage, the study team continued community involvement by informing and communicating with the community and its leaders.

Year 2018

02/14/2018. Dr. Jia informed over 20 middle school students of the Memphis PAHs Study in Lausanne Collegiate School, Memphis when he gave a lecture on air pollution.

03/26/2018. The study team member Fariha Sultana presented the study information to over 100 faculty, staff, and students of the University of Memphis in the 2018 UM Student Research Forum. The title was "Characterizing Polycyclic Aromatic Hydrocarbons (PAHs) in Ambient Air in the Memphis Tri-state Area". The presentation won the second-place award in this event.

04/02/2018. Dr. Jia presented the project information in several of his lectures to 40 graduate students in the Lecture of "Air Pollution" of the School of Public Health core course "Environmental Health".



05/16/2018. Larry Smith and Dr. Jia gave lectures on the Memphis PAHs Study to two classes of students in White Station High School, the best public high school in Memphis. A total of approximately 80 students were informed of the basics of air pollution and human health, polycyclic aromatic hydrocarbons, and particulate matter pollution. The information and progress of the Memphis PAH study were presented.

08/09/2018. Larry Smith and Chunrong Jia attended TN Environmental Literacy Plan Meeting - An Update and Opportunity for Collaboration. An audience of over 40 environmental educators was briefed of the Memphis PAHs Study.

10/09/2018. The Memphis PAHs Study information and preliminary results were presented in the University of Memphis “Tiger Blue Goes Green” event.

11/08/2018. Dr. Jia attended TN Environmental Literacy Plan Meeting - An Update and Opportunity for Collaboration. An audience of over 40 environmental educators was briefed of the Memphis PAHs Study.

11/15/2018. Dr. Jia presented the study information in a forum titled “Sustainability in Memphis Colleges and Universities”, organized by Sierra Club.

Year 2019

03/20/2019. The Memphis PAHs Study information and preliminary results were presented in the Harvard JPB Environmental Health Fellows Program Workshop.

03/25/2019. The Memphis PAHs Study information and preliminary results were presented in the U of M 31st Annual Student Research Forum event.

03/25/2019. The Memphis PAHs study was presented to a group of public health students at the University of Memphis.

05/15/2019. Project Manager Larry Smith presented the Memphis PAHs Study information and preliminary results in the 48th Annual Environmental Show of the South, Chattanooga, TN.

6.2.3 Post-monitoring stage

The community engagement was focused on information dissemination and risk communication in the post-monitoring stage. The research team and communities worked together to translate and disseminate research findings to promote positive changes in air quality and the public’s health.

08/30/2019. Dr. Jia presented the Memphis PAHs Study information to the science class of the University of Memphis Middle School.

10/16/2019. Dr. Jia and Larry Smith presented the results of the Memphis PAHs Study to the Shelby County Air Board. The experts and the public discussed the levels and sources of PAHs in this region.

6.3 Outputs of community involvement

The tens of community involvement activities yielded outputs in response to the aims set in Section 7.1.

(1) Established a community-government-academic partnership, in response to Aim 1. The study team outreached many community partners during the study period. SCHD and U of M established institutional relationships with various organizations, initiatives, and community leaders, including (a) individuals and community leaders who have interest and concerns with environmental issues in Memphis; (b) non-profit and faith-based organizations, such as the Sierra Club, Westwood Neighborhood Association, Bridges, Memphis Botanical Garden, and Engineers' Club of Memphis, and individual churches; (c) government agencies, such as Shelby County Schools, Memphis & Shelby County Office of Sustainability, White House Council on Strong Cities, and Strong Communities; and (d) academia including researchers at the University of Memphis. Dr. Jia established the "Memphis Environmental Health Research Community (MEHRC)," and obtained funding from the FedEx Institute of Technology to support the research and community activities relating to air pollution research. This unique community-government-academic partnership fostered mutual respect, understanding, trust, and environmental education in the local community, and could enhance the acceptability, effectiveness, and sustainability of pollution control to reduce health disparities.

(2) Developed multiple communication venues to disseminate the project information, in response to Aim 2. The venues included: (a) A project flyer and a web site for the project; (b) Stakeholders' meetings; (c) National, regional, and local conferences and events, e.g., the National EJ Conference, the TN Environmental Conference, and U of M Tiger Blue Goes Green; (d) Government meetings, e.g., Memphis and Shelby County Air Pollution Board Meetings; and (e) Classroom connections with public and private schools. (f) Radio interviews and broadcasting. These activities improved the public's knowledge of air pollution, public health, environmental justice, and environmental policy and regulations.

We have not disseminated the project results to the public as the final dataset and findings have not been reviewed by the experts. For the next steps, we will share information with the communities about pollution levels and the associated health risks through multiple channels as described. It should be a two-way exchange of information: The project team informs the audience of interest, and then gather information from the people that could possibly be affected by the risk at hand. The team will also communicate the information to the other small communities such as the environmental, policymaking, academic, and regulatory communities.

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