

ORIGINAL ARTICLE

## Source Proximity and Meteorological Effects on Residential Ambient Concentrations of PM<sub>2.5</sub>, Organic Carbon, Elemental Carbon, and p-PAHs in Houston and Los Angeles, USA

Jaymin Kwon<sup>\*</sup>, Clifford P. Weisel<sup>1)</sup>, Maria T. Morandi<sup>2)</sup>, Thomas H. Stock<sup>3)</sup>,  
Barbara Turpin<sup>4)</sup>

*California State University, Fresno, Department of Public Health, 2345 E. San Ramon Ave. M/S MH30, Fresno, CA 93740, USA*

<sup>1)</sup>*Rutgers University, Environmental and Occupational Health Science Institute, Department of Environmental and Occupational Medicine, 170 Frelinghuysen Road, Piscataway, NJ 08854, USA*

<sup>2)</sup>*University of Texas, Health Science Center at Houston, School of Public Health, 1200 Pressler Street, Houston, TX 77225, USA*

<sup>3)</sup>*University of Texas, Health Science Center at Houston, School of Public Health, Epidemiology, Human Genetics & Environmental Sciences, 1200 Pressler Street, Houston, TX 77225, USA*

<sup>4)</sup>*University of North Carolina at Chapel Hill, Gillings School of Global Public Health, Environmental Science and Engineering, 135 Dauer Drive, 140 Rosenau Hall, CB #7431, Chapel Hill, NC 27599-7431*

### Abstract

Concentrations of fine particulate matter (PM<sub>2.5</sub>) and several of its particle constituents measured outside homes in Houston, Texas, and Los Angeles, California, were characterized using multiple regression analysis with proximity to point and mobile sources and meteorological factors as the independent variables. PM<sub>2.5</sub> mass and the concentrations of organic carbon (OC), elemental carbon (EC), benzo-[a]-pyrene (BaP), perylene (Per), benzo-[g,h,i]-perylene (BghiP), and coronene (Cor) were examined. Negative associations of wind speed with concentrations demonstrated the effect of dilution by high wind speed. Atmospheric stability increase was associated with concentration increase. Petrochemical source proximity was included in the EC model in Houston. Area source proximity was not selected for any of the PM<sub>2.5</sub> constituents' regression models. When the median values of the meteorological factors were used and the proximity to sources varied, the air concentrations calculated using the models for the eleven PM<sub>2.5</sub> constituents outside the homes closest to influential highways were 1.5-15.8 fold higher than those outside homes furthest from the highway emission sources. When the median distance to the sources was used in the models, the concentrations of the PM<sub>2.5</sub> constituents varied 2 to 82 fold, as the meteorological conditions varied over the observed range. We found different relationships between the two urban areas, illustrating the unique nature of urban sources and suggesting that localized sources need to be evaluated carefully to understand their potential contributions to PM<sub>2.5</sub> mass and its particle constituents concentrations near residences, which influence baseline indoor air concentrations and personal exposures. The results of this study could assist in the appropriate design of monitoring networks for community-level sampling and help improve the accuracy of exposure models linking emission sources with estimated pollutant concentrations at the residential level.

**Key words** : Environmental monitoring, Exposure modeling, PM<sub>2.5</sub>, OC (organic carbon), EC (elemental carbon), PAHs (polycyclic aromatic hydrocarbons), Proximity, Meteorology

---

**Received** 26 September, 2016; **Revised** 6 October, 2016;

**Accepted** 10 October, 2016

**\*Corresponding author** : Jaymin Kwon, California State University, Fresno, Department of Public Health, 2345 E. San Ramon Ave. M/S MH30, Fresno CA 93740, USA  
Phone : +1-559-278-5169  
E-mail : jkwon@csufresno.edu

The Korean Environmental Sciences Society. All rights reserved.  
© This is an Open-Access article distributed under the terms of the Creative Commons Attribution Non-Commercial License (<http://creativecommons.org/licenses/by-nc/3.0>) which permits unrestricted non-commercial use, distribution, and reproduction in any medium, provided the original work is properly cited.

## 1. Introduction

Carbonaceous species are a major contributor to atmospheric fine particulate matter with an aerodynamic diameter of 2.5  $\mu\text{m}$  or smaller ( $\text{PM}_{2.5}$ ). These species can be categorized as organic carbon (OC) and elemental carbon (EC). EC and polycyclic aromatic hydrocarbons (PAHs), a component of OC, are generated as incomplete combustion byproducts from anthropogenic sources including fossil fuel used in power generation, heating, transportation, and industrial processes (Kelly and Fussell, 2012; Noth et al., 2011). OC is also formed in the atmosphere locally and regionally through gas and multiphase photochemical reactions, as are the other major inorganic  $\text{PM}_{2.5}$  constituents, sulfate and nitrate (Kanakidou et al., 2005; Turpin et al., 2000).

Adverse health outcomes have been reported for several endpoints in numerous studies documenting an association between exposure to particulate matter constituents and cardiovascular and respiratory effects, birth outcomes, and premature death by differential toxicity (Kelly and Fussell, 2012; Rohr and Wyzga, 2012). For example, ambient air pollution was positively associated with increased asthmatic emergency hospitalization of senior citizens (Park et al., 2013). A cohort panel study found positive association of gene expression changes in senior citizens with exposure to traffic related air pollutants including EC, OC, and p-PAHs measured in the immediate outdoor environment near retirement communities in Los Angeles (Wittkopp et al., 2016). Exposure to PAHs has been linked to specific adverse outcomes especially in children, including asthma symptoms (Gale et al., 2012), regulatory T-cell function in asthma (Nadeau et al., 2010), intelligent quotient (IQ) development following prenatal exposure (Perera et al., 2009), preterm birth (Padula et al., 2014), and impaired systemic immunity and epigenetic modifications in atopy (Hew et al., 2015).

Health risk assessments of  $\text{PM}_{2.5}$  and its constituents are hampered by exposure measurement error. Frequently, air pollution measured at central monitoring sites is used as a surrogate for community-level air pollution exposure, and measurements of  $\text{PM}_{2.5}$  chemical composition are limited. Capturing spatial variability for individual  $\text{PM}_{2.5}$  constituents is necessary step toward accurate assessment of  $\text{PM}_{2.5}$  exposure at the community level because intra-urban spatial variability can vary greatly for some  $\text{PM}_{2.5}$  constituents (Anastasopoulos et al., 2012; Noth et al., 2011).

In previous Relationship of Indoor, Outdoor and Personal Air (RIOPA) studies, the majority of indoor EC and particle phase PAH concentrations inside RIOPA homes in Houston, Texas, Los Angeles, California, and Elizabeth, New Jersey, were found to be of outdoor origin (Hodas et al., 2012; Naumova et al., 2002; Polidori et al., 2006). Significant enhancement of multiple  $\text{PM}_{2.5}$  constituents were observed indoor of near-highway homes in urban areas (Fuller et al., 2013; Lawson et al., 2011), and near ambient truck emissions (Baxter et al., 2008). Indoor levels of EC and PAHs were strongly affected by outdoor levels in a Los Angeles senior citizen panel study (Hasheminassab et al., 2014).

An increase of outdoor concentrations of  $\text{PM}_{2.5}$  constituents at homes located near emission sources is expected to result in an increase of indoor  $\text{PM}_{2.5}$  constituent concentrations via penetration and would be expected to result in a consequent increase in personal exposure for populations living near the emission sources (Larson et al., 2004). Characterization of the impact of mobile, area, and point emission sources on residential outdoor concentrations (Kwon et al., 2006; Kwon et al., 2016; Polidori et al., 2010) can provide useful information for understanding the contribution of local outdoor sources of VOCs,  $\text{PM}_{2.5}$ , OC, EC, and PAHs to indoor concentrations and to personal exposure levels, particularly for those who

spend most of their time at home, such as infants, toddlers, children, home-bound patients, and senior citizens, especially for those who live near emission sources. The general approach to proximity, meteorology, and statistical analysis applied in previous studies (Kwon et al., 2006; Kwon et al., 2016; Polidori et al., 2010) were also applied to the current Houston and Los Angeles data.

This study was directed at identifying determinants (i.e., meteorological and source factors) of elevated concentrations of PM<sub>2.5</sub> constituents at urban residences in close proximity to sources. The objective of study is to construct statistical models that explain residential outdoor concentrations for distinctively different major urban areas in two states, Houston TX and Los Angeles County CA, and to compare them with those constructed previously for Elizabeth, NJ (Polidori et al., 2010). Understanding determinants of the spatial distribution of PM<sub>2.5</sub>, OC, EC, and particle-bound PAHs in urban settings can assist exposure and risk assessment, and epidemiology research.

## 2. Materials and Methods

### 2.1. Data sources

As part of the RIOPA study, 48-hour integrated samples of air pollutants were collected in Houston, TX, Los Angeles, CA, and Elizabeth, NJ, during different seasons between the summer of 1999 and the spring of 2001 (Weisel et al., 2005). Homes near outdoor emissions were oversampled based on residential distances from various mobile, area, and point emission sources in order to estimate their contributions to residential outdoor and indoor air and to personal exposures (Weisel et al., 2005). The RIOPA study design, measurement of air pollutants, and quality assurance and control results are described elsewhere (Turpin et al., 2007; Weisel et al., 2005). The RIOPA database is publicly available at <https://riopa.aer.com/login.php>. The current analysis evaluated PM<sub>2.5</sub>, OC, EC, and four particle-bound PAHs (p-PAHs) to examine the influence of proximity to emission sources and local meteorological factors on

**Table 1.** Outdoor concentrations of the chemical species examined in this study

Houston	N	Mean	Std. Dev.	Percentiles			Max
				25	50	75	
PM <sub>2.5</sub> (µg/m <sup>3</sup> )	110	14.73	5.75	10.68	13.25	17.65	34.00
OC (µg/m <sup>3</sup> )*	63	3.37	3.16	1.29	2.54	4.67	18.25
EC (µg/m <sup>3</sup> )*	63	0.70	0.35	0.44	0.65	0.89	2.00
BaP (ng/m <sup>3</sup> )	41	0.058	0.067	0.008	0.027	0.099	0.292
Per (ng/m <sup>3</sup> )	38	0.012	0.012	0.003	0.009	0.024	0.038
BghiP (ng/m <sup>3</sup> )	41	0.167	0.313	0.036	0.072	0.181	1.927
Cor (ng/m <sup>3</sup> )	41	0.124	0.186	0.024	0.045	0.117	0.958
Los Angeles	N	Mean	Std. Dev.	Percentiles			Max
PM <sub>2.5</sub> (µg/m <sup>3</sup> )	101	19.23	13.26	12.00	16.10	22.60	94.90
OC (µg/m <sup>3</sup> )*	45	4.05	1.92	2.59	3.59	5.42	10.06
EC (µg/m <sup>3</sup> )*	45	1.40	0.90	0.74	1.14	1.82	4.64
BaP (ng/m <sup>3</sup> )	53	0.109	0.175	0.022	0.048	0.114	1.046
Per (ng/m <sup>3</sup> )	53	0.023	0.036	0.003	0.010	0.026	0.212
BghiP (ng/m <sup>3</sup> )	53	0.481	0.626	0.114	0.270	0.542	3.123
Cor (ng/m <sup>3</sup> )	53	0.520	0.840	0.060	0.246	0.595	4.684

\*µg of carbon/m<sup>3</sup> 0

**Table 2.** Distribution of the meteorological data during sample collection for the Houston Texas and Los Angeles California study

Description	Mean	Std. Deviation	Minimum	25 <sup>th</sup> Percentile	Median	75 <sup>th</sup> Percentile	Maximum
Houston, TX							
Temperature, °C	22.2	7.5	4.7	15.9	24.7	28.4	32.5
RH, %	73.9	12.3	39.1	65.9	75.0	82.8	103.9
Atmospheric Stability, time fraction	0.58	0.15	0.06	0.50	0.58	0.67	0.94
Dewpoint Temperature, °C	16.5	7.2	-2.0	9.7	19.8	22.4	24.4
Wind speed, m/s	3.54	1.10	1.63	2.62	3.39	4.26	6.29
Atmospheric pressure, inHg	29.97	0.14	29.65	29.87	29.96	30.04	30.42
Precipitation total	0.006	0.011	0.000	0.000	0.000	0.004	0.060
Los Angeles, CA							
Temperature, °C	18.1	3.9	10.0	15.1	18.3	21.3	26.0
RH, %	67.6	13.6	27.9	63.9	71.2	77.2	87.3
Atmospheric Stability, time fraction	0.71	0.14	0.44	0.61	0.67	0.83	1.00
Dewpoint Temperature, °C	11.1	6.0	-4.8	7.9	13.3	15.6	18.4
Wind speed, m/s	1.16	0.39	0.26	0.85	1.20	1.44	2.26
Atmospheric pressure, inHg	29.78	0.11	29.55	29.70	29.76	29.87	30.04
Precipitation total	0.002	0.009	0.000	0.000	0.000	0.000	0.081

the residential outdoor concentrations ( $C_{out}$ ). The four p-PAHs included were: benzo-[a]-pyrene (BaP), perylene (Per), benzo-[g,h,i]-perylene (BghiP), and coronene (Cor). Other sampled p-PAHs were excluded due to too the high proportion of concentrations below method detection limits (MDLs).  $PM_{2.5}$  mass, OC, and EC concentrations were all above their respective MDLs. p-PAH concentrations below their respective MDLs were included as reported rather than replacing these values with one-half of the MDLs before natural log transformation for the data analysis (Turpin et al., 2007). Descriptive statistics for the  $PM_{2.5}$ , OC, EC, and the four selected p-PAHs concentrations in Houston and Los Angeles are shown in the Table 1.

## 2.2. Study area

Houston has the largest density of petrochemical facilities in the world for the production and storage of fuels, chemical precursors, plastics, and solvents. The residential areas around the petrochemical complexes were targeted for recruitment included: Houston Ship Channel, Pasadena, Galena Park, Channelview, Baytown, Deer Park, La Porte and, as a comparison

area, the Texas Medical Center (Weisel et al., 2005). There were four sampling areas in Los Angeles County: West Los Angeles, Pico Rivera, Burbank, and Santa Clarita. Los Angeles areas were influenced by emissions from at least one major highway (Weisel et al., 2005). Elizabeth is a city adjacent to a major petrochemical industrial complex in Linden, NJ, to its south, Newark International Airport and the port of Newark to its north. Residential areas are close to commercial areas and major highways (Weisel et al., 2005). Maps of the three RIOPA study areas can be found in a previous study (Kwon et al., 2016).

## 2.3. Meteorological data

To examine the influence of meteorological factors on residential outdoor  $PM_{2.5}$ , OC, EC, and p-PAHs concentrations ( $C_{out}$ ), temperature, dew-point temperature, relative humidity (RH), wind speed, atmospheric pressure, and precipitation data were downloaded from the RIOPA database (HEI and NUATRC, 2008) as candidate explanatory variables. Descriptive statistics for the meteorological variables by study location are summarized in Table 2. Computed atmospheric Pasquill

stability classes, with a time resolution of 3-h, were retrieved from the Air Resource Laboratory, National Oceanic and Atmospheric Administration (NOAA), real-time environmental applications and display (READY) system (NOAA, 2012). Each 3-hour atmospheric stability class was assigned a code of “1” if the Pasquill category was “stable” or “neutral” (i.e., classes D, E, F, and G), or “0” when it was “unstable” (i.e., classes A, B, and C) to derive the fraction of each 48-hour sampling period with “stable” or “neutral” stability (Kwon et al., 2006).

#### 2.4. Emission sources

Emission sources of PM<sub>2.5</sub> and its constituents around the homes were identified by geographical information system (GIS) mapping. Emission sources of interest were: 1) point sources listed in the National Emission Inventory for year 1999 (1999 NEI), i.e., refineries and solvent production facilities (USEPA, 2003); and 2) mobile sources, i.e., highways and major arterial roadways identified from the 2000 TIGER (topologically integrated geographic encoding and referencing) line files obtained from the U.S. Census Bureau (USCB, 2012). To avoid redundant selections of the same roadways when the distances were measured from the RIOPA homes, line segments of a roadway that had the same feature class and street name were merged into a single segment. Area sources such as gas stations, scrap metal recyclers, and dry cleaning facilities used for a previous VOCs proximity study (Kwon et al., 2016) were tested for assuring these VOC sources are not significant contributors in the PM<sub>2.5</sub> models. Detailed descriptions of the mapping processes of emission sources are provided in the previous work (Kwon et al., 2016).

#### 2.5. Proximity data

The direct distances between sampling locations and emission sources were calculated using the ArcScript extension referred to as “the nearest features” with

distances and bearings (version 3.8b, Jenness Enterprises, Flagstaff, AZ) on ArcView 3.2 (ESRI Inc., Redland, CA). To obtain more generalized and consistent model outcomes, the proximity variables under the same category for each specific component were estimated as the sum of all distances (km) from the first to the fifth closest facilities or roadways in the same class (Kwon et al., 2016). This combined distance approach was developed to accommodate the wide distribution of every distance between surrounding emission clusters and the homes sampled. The inverse of the sum of distances (km<sup>-1</sup>) was used as the source proximity variable. The distributions of the proximity variables are shown in Table 3.

#### 2.6. Statistical analysis

All statistical analyses were performed using SAS (version 9.3, SAS Institute Inc. Cary, NC), and SPSS (version 23, SPSS Inc. Chicago, IL). The PM<sub>2.5</sub>, OC, EC, and four p-PAHs concentrations were consistent with a log normal distribution; therefore natural-log transformed residential outdoor concentrations (lnC<sub>out</sub>) were used as the dependent variables. Bivariate Pearson’s correlations between the lnC<sub>out</sub> and proximity and meteorological variables were used to explore the correlations at  $\alpha=0.05$  ( $p<0.05$ ).

Multiple stepwise linear regression analysis was conducted to select a group of variables ( $X_i$ ) to explain lnC<sub>out</sub> ( $Y_i$ ). The default entry and inclusion criteria were set at  $p<0.15$ . However, an entry criterion of  $p<0.10$  was used for BghiP and Cor in Houston, and PM<sub>2.5</sub> in Los Angeles to avoid model over-specification and problems related to potential co-linearity among the selected variables. Because of the logarithmic transformation, the additive effects of the independent variables on the regression model predictions become multiplicative. The regression equation can be written as:

**Table 3.** Distribution of the local, mobile, and point emission sources proximity variables (the sum of distance (km) to the closest five facilities or roadways from participated homes)

Description	Mean	Std. Deviation	Minimum	25 <sup>th</sup> Percentile	Median	75 <sup>th</sup> Percentile	Maximum
Houston Texas							
Highway (A10, A20)	24.0	5.1	13.0	21.2	23.9	25.5	42.2
Arterial (A30)	9.6	5.3	2.0	5.5	8.6	16.0	23.8
PM <sub>2.5</sub>	9.6	5.4	1.8	5.3	7.8	11.4	26.2
POM (15-PAHs)	42.2	14.6	24.1	35.9	39.1	41.5	130.0
POM (7-PAHs)	53.8	16.3	38.8	43.8	51.5	53.4	136.2
POM (Non 15-PAHs)	60.8	19.5	41.6	46.2	58.4	62.6	139.3
petrochemical facilities	24.4	10.5	10.6	19.6	22.9	25.7	70.1
Los Angeles California							
Highway (A10, A20)	20.5	3.3	12.7	19.5	20.0	22.7	29.3
Arterial (A30)	18.8	7.0	3.2	16.7	19.6	20.8	31.8
POM (15-PAHs)	34.6	13.5	9.4	22.8	43.0	43.8	48.5
POM (7-PAHs)	34.7	13.4	9.4	22.8	43.0	44.0	48.9
POM (Non 15-PAHs)	152.0	72.9	102.6	108.1	110.8	165.0	302.0

$$C_{outi} = \exp^{\beta_0} \cdot \exp^{\beta_1 X_{i1}} \cdot \exp^{\beta_2 X_{i2}} \cdot \exp^{\beta_{P-1} X_{i,P-1}} \cdot \exp^{\epsilon} \quad (1)$$

where  $C_{outi}$  is residential outdoor concentration ( $\mu\text{g}/\text{m}^3$  for PM<sub>2.5</sub>, OC, EC, and  $\text{ng}/\text{m}^3$  for p-PAHs),  $\beta_0, \beta_1 \dots \beta_{P-1}$  are the regression coefficients,  $X_{i1}, X_{i2} \dots X_{i, P-1}$  are the selected variables, and  $\epsilon$  is the error term (Kwon et al., 2016).

To reduce Type I errors under multiple testing scenarios, preliminary regression analysis was performed for each compound to determine the relative importance of variables within the same type of independent variables, either proximity or meteorological variables separately. The best-fit models selected by the stepwise selection methods and the corresponding statistical results were evaluated for satisfying the major assumptions of linear regression analysis (Polidori et al., 2010).

The effect of an individual variable ( $X_i$ ) explaining the variability of residential outdoor concentrations ( $C_{out}$ ) in the multiple regression models are simulated by holding constant other variables in each model at

their respective observed median values, similar to simulations performed in previous studies (Kwon et al., 2006; Kwon et al., 2016; Polidori et al., 2010). To avoid misleading interpretations of model explanatory power, simulations of explanatory effects were limited to the PM<sub>2.5</sub> constituents with adjusted model  $R^2$  larger than 0.25 and individual variables with  $p < 0.05$ .

### 3. Results

#### 3.1. Descriptive summary

Bivariate Pearson's correlations between the  $\ln C_{out}$  of PM<sub>2.5</sub>, OC, EC, p-PAHs and the meteorological and proximity parameters are presented in Table 4. Variables with statistically significant correlations at  $\alpha = 0.05$  ( $p < 0.05$ ) are sorted in ascending order of the significance of  $p$ -values. Correlations between  $\ln C_{out}$  and proximity and meteorological variables were in the expected direction. Proximity variables, the inverse distances to emission sources, and atmospheric stability were positively correlated with the  $\ln C_{out}$  of PM<sub>2.5</sub>, EC, and p-PAHs concentrations. Wind speed, RH, and precipitation were negatively

**Table 4.** Bivariate Pearson's correlations between ln-transformed outdoor concentrations and variables for correlations with p<.05 (CC= Pearson's coefficient of correlation; P= p-value; N=sample size)

RIOPA TX	Houston				RIOPA CA	Los Angeles				
Pollutant	Variable	CC	P	N	Pollutant	Variable	CC	P	N	
LnPM <sub>2.5</sub>	U*	-0.34	0.0002	110	LnPM <sub>2.5</sub>	Precip*	-0.28	0.003	108	
	Precip*	-0.31	0.001	110		Press	-0.26	0.0069	108	
	RH*	-0.31	0.001	110		Temp	0.24	0.0112	108	
	Stab	0.25	0.009	110		U*	-0.22	0.0214	108	
LnOC	DewC*	-0.41	0.001	63	LnOC	DewC*	0.21	0.0285	108	
	RH*	-0.35	0.005	63		A125Inv*	0.19	0.0381	121	
	Temp	-0.33	0.008	63		U*	-0.45	0.0028	42	
	Press	0.32	0.010	63		RH	-0.38	0.0127	42	
LnEC	Petro5Inv*	0.41	0.001	63	LnEC	A125Inv*	0.40	0.007	44	
	RH*	-0.38	0.002	63		RH	-0.38	0.0118	42	
	Stab*	0.36	0.004	63		DewC	-0.34	0.0269	42	
	U*	-0.35	0.005	63		LnBaP	Press	0.49	0.0003	49
Press	0.26	0.037	63	RH	-0.48		0.0005	49		
LnBaP	DewC	-0.39	0.011	41	Temp		-0.34	0.0169	49	
	Press	0.34	0.028	41	LnPer		DewC	-0.47	0.0006	49
	LnPer	DewC*	-0.52	0.001		38	RH	-0.46	0.0009	49
		Temp	-0.45	0.004		38	Press	0.38	0.0069	49
Press		0.40	0.012	38		RH	-0.40	0.014	38	
RH		-0.40	0.014	38	LnBghiP	DewC*	-0.50	0.001	41	
LnBghiP	DewC*	-0.50	0.001	41		RH	-0.49	0.0003	49	
	Temp	-0.39	0.011	41		Press	0.48	0.0005	49	
	RH	-0.37	0.019	41		Pn15_5inv	0.40	0.0028	53	
	Press	0.31	0.049	41	Temp	-0.30	0.0369	49		
LnCor	DewC*	-0.47	0.002	41	LnCor	DewC	-0.50	0.0003	49	
	RH	-0.41	0.008	41		RH	-0.48	0.0004	49	
	Temp	-0.34	0.032	41		Stab*	0.49	0.0004	47	
	Stab*	0.31	0.047	41		Press	0.43	0.0023	49	

\* Variables included in the model. U= wind speed (m/s), Temp= temperature °C, RH=relative humidity (%), DewC=dew point temperature °C, Press= atmospheric pressure (inHg), Precip= precipitation, Stab= fraction of time atmospheric stability was stable, A125inv= inverse distance to the closest 5 highways (A10, A20), Petro5inv= inverse distance to the closest 5 petrochemical point sources, Pn15\_5inv= inverse distance to the closest 5 Non-15 PAHs point sources

**Table 5.** The summary of the best-fit models of PM<sub>2.5</sub>, OC, EC of Houston and Los Angeles (Ln-transformed concentrations, µg/m<sup>3</sup>)

Pollutant	Y		Intercept	X <sub>1st</sub>	X <sub>2nd</sub>	X <sub>3rd</sub>	X <sub>4th</sub>	X <sub>5th</sub>
Houston								
PM <sub>2.5</sub>	lnPM		β <sub>0</sub>	U <sup>1</sup>	PRECIP <sup>1</sup>	RH		
Adjusted Model R <sup>2</sup> =	0.24	β <sub>i</sub>	3.584	<b>-0.061</b>	<b>-10.508</b>	-0.007		
Enter p<	0.150	Standard Error	0.212	<b>0.016</b>	<b>3.338</b>	0.003		
Model d.f.	3	Partial R <sup>2</sup>		<b>0.119</b>	<b>0.101</b>	0.044		
Model p-value	<.0001	p-value		<b>0.0002</b>	<b>0.0003</b>	0.013		
Organic Carbon	lnOC		β <sub>0</sub>	DEWC	PRECIP	RH	STAB	
Adjusted Model R <sup>2</sup> =	0.28	β <sub>i</sub>	2.197	<b>-0.058</b>	29.414	-0.022	<i>1.657</i>	
Enter p<	0.15	Standard Error	0.818	<b>0.022</b>	11.932	0.010	<i>0.839</i>	
Model d.f.	4	Partial R <sup>2</sup>		<b>0.170</b>	0.059	0.051	<i>0.045</i>	
Model p-value	0.0001	p-value		<b>0.0008</b>	0.036	0.046	<i>0.053</i>	
Elemental Carbon	lnEC		β <sub>0</sub>	Petro5INV	U <sup>1</sup>	RH	STAB	<i>A125INV<sup>d</sup></i>
Adjusted Model R <sup>2</sup> =	0.40	β <sub>i</sub>	-0.640	<b>6.805</b>	<b>-0.049</b>	-0.012	<i>1.010</i>	<i>10.604</i>
Enter p<	0.15	Standard Error	0.559	<b>2.593</b>	<b>0.032</b>	0.004	<i>0.442</i>	<i>5.649</i>
Model d.f.	5	Partial R <sup>2</sup>		<b>0.169</b>	<b>0.139</b>	0.066	<i>0.039</i>	<i>0.034</i>
Model p-value	<.00011	p-value		<b>0.0008</b>	<b>0.001</b>	0.015	<i>0.056</i>	<i>0.066</i>
Los Angeles								
PM <sub>2.5</sub>	lnPM		β <sub>0</sub>	PRECIP <sup>1</sup>	A305INV	DEWC	U <sup>1</sup>	A125INV
Adjusted Model R <sup>2</sup> =	0.22	β <sub>i</sub>	2.319	<b>-13.107</b>	1.984	0.026	-0.178	8.420
Enter p<	0.10	Standard Error	0.257	<b>5.659</b>	0.753	0.009	0.067	4.021
Model d.f.	5	Partial R <sup>2</sup>		<b>0.080</b>	0.057	0.039	0.047	0.034
Model p-value	<.0001	p-value		<b>0.004</b>	0.012	0.036	0.017	0.039
Organic Carbon	lnOC		β <sub>0</sub>	U	A125INV			
Adjusted Model R <sup>2</sup> =	0.22	β <sub>i</sub>	1.268	-0.283	13.504			
Enter p<	0.15	Standard Error	0.350	0.087	6.229			
Model d.f.	2	Partial R <sup>2</sup>		0.168	0.099			
Model p-value	0.0044	p-value		0.011	0.037			
Elemental Carbon	lnEC		β <sub>0</sub>	A125INV <sup>1</sup>	U <sup>1</sup>			
Adjusted Model R <sup>2</sup> =	0.26	β <sub>i</sub>	-0.530	27.604	-0.302			
Enter p<	0.15	Standard Error	0.462	8.220	0.115			
Model d.f.	2	Partial R <sup>2</sup>		0.162	0.138			
Model p-value	0.002	p-value		0.012	0.013			

<sup>1</sup>: Variable commonly selected in the specific models in Houston and Los Angeles. The variable is bold when the individual p-values are less than 0.01. The variable is italic when individual p-values are higher than 0.05



**Table 6.** The summary of the best-fit models of selected particle bound PAHs of Houston and Los Angeles (Ln-transformed concentrations, ng/m<sup>3</sup>)

Pollutant	Y		Intercept	X <sub>1st</sub>	X <sub>2nd</sub>	X <sub>3rd</sub>
Houston						
Benzo(a)Pyrene	lnBaP		$\beta_0$	<b>RH</b>	<i>PRECIP</i>	<i>A125INV</i> <sup>1</sup>
Adjusted Model R <sup>2</sup> =	0.38	$\beta_i$	0.191	<b>-0.068</b>	<i>31.759</i>	<i>27.854</i>
Enter p<	0.15	Standard Error	1.241	<b>0.014</b>	<i>18.069</i>	<i>17.952</i>
Model d.f.	3	Partial R <sup>2</sup>		<b>0.337</b>	<i>0.049</i>	<i>0.038</i>
Model p-value	0.0001	p-value		<b>&lt;.0001</b>	<i>0.087</i>	<i>0.129</i>
Perylene	lnPer		$\beta_0$	<b>DEWC</b>	<i>PRECIP</i>	
Adjusted Model R <sup>2</sup> =	0.30	$\beta_i$	-3.067	<b>-0.132</b>	<i>41.692</i>	
Enter p<	0.15	Standard Error	0.583	<b>0.032</b>	<i>21.537</i>	
Model d.f.	2	Partial R <sup>2</sup>		<b>0.271</b>	<i>0.071</i>	
Model p-value	0.0007	p-value		<b>0.0002</b>	<i>0.061</i>	
Benzo(ghi)Perylene	lnBghiP		$\beta_0$	<b>DEWC</b>	<i>STAB</i> <sup>1</sup>	<i>A125INV</i> <sup>1</sup>
Adjusted Model R <sup>2</sup> =	0.36	$\beta_i$	-3.847	<b>-0.097</b>	<i>2.694</i>	<i>31.507</i>
Enter p<	0.10	Standard Error	1.009	<b>0.024</b>	<i>1.087</i>	<i>15.295</i>
Model d.f.	3	Partial R <sup>2</sup>		<b>0.247</b>	<i>0.091</i>	<i>0.068</i>
Model p-value	0.0002	p-value		<b>0.0009</b>	<i>0.028</i>	<i>0.047</i>
Coronene	lnCor		$\beta_0$	<b>DEWC</b>	<i>STAB</i> <sup>1</sup>	<i>A125INV</i> <sup>1</sup>
Adjusted Model R <sup>2</sup> =	0.36	$\beta_i$	-4.514	<b>-0.098</b>	<i>3.104</i>	<i>34.603</i>
Enter p<	0.10	Standard Error	1.063	<b>0.025</b>	<i>1.146</i>	<i>16.115</i>
Model d.f.	3	Partial R <sup>2</sup>		<b>0.225</b>	<i>0.109</i>	<i>0.074</i>
Model p-value	0.0002	p-value		<b>0.0018</b>	<i>0.017</i>	<i>0.038</i>
Los Angeles						
Benzo(a)Pyrene	lnBaP		$\beta_0$	<b>STAB</b>	U	<i>A125INV</i> <sup>1</sup>
Adjusted Model R <sup>2</sup> =	0.50	$\beta_i$	-7.880	<b>6.579</b>	<i>-0.545</i>	<i>31.142</i>
Enter p<	0.15	Standard Error	1.865	<b>1.604</b>	<i>0.261</i>	<i>16.787</i>
Model d.f.	3	Partial R <sup>2</sup>		<b>0.434</b>	<i>0.064</i>	<i>0.041</i>
Model p-value	<.0001	p-value		<b>&lt;.0001</b>	<i>0.030</i>	<i>0.071</i>
Perylene	lnPer		$\beta_0$	<b>STAB</b>	U	<i>A125INV</i>
Adjusted Model R <sup>2</sup> =	0.43	$\beta_i$	-10.247	<b>6.862</b>	<i>-0.809</i>	<i>49.599</i>
Enter p<	0.15	Standard Error	2.588	<b>2.226</b>	<i>0.363</i>	<i>23.295</i>
Model d.f.	3	Partial R <sup>2</sup>		<b>0.320</b>	<i>0.085</i>	<i>0.062</i>
Model p-value	<.0001	p-value		<b>&lt;.0001</b>	<i>0.022</i>	<i>0.040</i>
Benzo(ghi)Perylene	lnBghiP		$\beta_0$	U	<i>STAB</i> <sup>1</sup>	<i>A125INV</i> <sup>1</sup>
Adjusted Model R <sup>2</sup> =	0.49	$\beta_i$	-5.081	<b>-0.670</b>	<i>4.892</i>	<i>37.589</i>
Enter p<	0.15	Standard Error	1.758	<b>0.246</b>	<i>1.513</i>	<i>15.827</i>
Model d.f.	3	Partial R <sup>2</sup>		<b>0.361</b>	<i>0.094</i>	<i>0.069</i>
Model p-value	<.0001	p-value		<b>&lt;.0001</b>	<i>0.012</i>	<i>0.023</i>
Coronene	lnCor		$\beta_0$	U	<i>A125INV</i> <sup>1</sup>	<i>STAB</i> <sup>1</sup>
Adjusted Model R <sup>2</sup> =	0.43	$\beta_i$	-6.098	<b>-1.101</b>	<i>61.369</i>	<i>5.514</i>
Enter p<	0.15	Standard Error	2.722	<b>0.382</b>	<i>24.502</i>	<i>2.342</i>
Model d.f.	3	Partial R <sup>2</sup>		<b>0.343</b>	<i>0.056</i>	<i>0.075</i>
Model p-value	<.0001	p-value		<b>&lt;.0001</b>	<i>0.062</i>	<i>0.024</i>

<sup>1</sup>: Variable commonly selected in the specific models in Houston and Los Angeles. The variable is **bold** when the individual p-values are less than 0.01. The variable is italic when individual p-values are higher than 0.05

correlated with the  $\ln C_{out}$  of  $PM_{2.5}$ , OC, EC, and p-PAHs. Temperature and dew point were negatively correlated with  $\ln C_{out}$  of  $PM_{2.5}$ , OC, EC, and p-PAHs concentrations, except for  $PM_{2.5}$  in Los Angeles, which showed positive correlation with temperature and dew point.

In all cases, the best-fitting multiple regression models were obtained through stepwise selection. Summaries of regression models for Houston and Los Angeles including adjusted  $R^2$ , degrees of freedom, and  $p$ -values for the overall model, and partial  $R^2$  and  $p$ -values for individual variables are shown in Table 5 for  $PM_{2.5}$ , OC, EC and in Table 6 for the four p-PAHs. The variables included in the models are sorted by the order of selection ( $X_{ith}$ ). A variable enters the model based on the significance of its association with  $\ln C_{out}$ . Most of the predictors included in the models were the variables significantly correlated with  $\ln C_{out}$  (Table 4). The F-statistics for all models and the  $p$ -values for all parameter estimates were statistically significant ( $p < 0.001$ ,  $P_{model} < F$ ) except for OC and EC in Los Angeles ( $p = 0.004$  and  $0.002$ , respectively). The  $p$ -values for parameter estimates of the first selected independent variables ( $X_{1st}$ ) were statistically significant. Based on the adjusted model  $R^2$ , the models explained 24 to 40 % of the total variability of  $\ln C_{out}$  of  $PM_{2.5}$ , OC, and EC in Houston, and 22 to 26 % of the total variability of  $\ln C_{out}$  of  $PM_{2.5}$ , OC, and EC in Los Angeles. Models explained 30 to 38% of the total variability of  $\ln C_{out}$  of p-PAHs in Houston, and 43 to 50% of the total variability of  $\ln C_{out}$  of p-PAHs in Los Angeles. Although the models and individual variables were statistically significant, still a larger proportion (50–78%) of the  $C_{out}$  variability remains unexplained by these models.

### 3.2. Meteorological variables

Wind speed was selected as a significant explanatory variable in models for  $PM_{2.5}$  and EC in Houston, and

$PM_{2.5}$ , OC, EC, BaP, Per, BghiP, and Cor in Los Angeles. Wind speed entered as the  $X_{1st}$  in four of the models:  $PM_{2.5}$  in Houston, OC, BghiP, and Cor in Los Angeles with partial  $R^2$ 's explaining 12, 17, 36, and 34% of overall variability in residential outdoor concentrations respectively. Wind speed entered as the second variable selected in three models: EC in Houston, EC and Per in Los Angeles with partial  $R^2$ 's explaining 14, 14, and 9% of overall variability in residential outdoor concentrations respectively. Wind speed was included as a variable in all seven models in Los Angeles, while it was only included in  $PM_{2.5}$  and EC models in Houston. Wind speed was the most often variable selected among the meteorological variables. As expected, wind speed was inversely associated with the  $\ln C_{out}$  in all models where it was included.

Atmospheric stability was selected as a variable for 8 models: OC, EC, BghiP, and Cor in Houston, and for all four p-PAHs, BaP, Per, BghiP, and Cor in Los Angeles. Atmospheric stability was the first variable selected in 2 models: BaP and Per in Los Angeles with partial  $R^2$ 's explaining 43 and 32% of overall variability in residential outdoor concentrations respectively. Atmospheric stability was the second variable selected in 3 models: BghiP and Cor in Houston, and BghiP in Los Angeles with partial  $R^2$ 's explaining 9, 11, and 9% of overall variability in residential outdoor concentrations respectively. The partial  $R^2$  of stability in the other 3 models in which it was selected as third and fourth variables explained 4 to 7% of overall variability in residential outdoor concentrations. Atmospheric stability was positively associated with the concentrations.

Dew point was included in models of OC, Per, BghiP, and Cor in Houston as the first variable selected with partial  $R^2$ 's explaining 17, 27, 25, and 23% of the overall variability in the residential outdoor concentrations, respectively, and in the  $PM_{2.5}$  model in Los Angeles as the third variable selected with partial  $R^2$  explaining 4% of overall variability in residential

outdoor concentrations. Dew point temperature was associated negatively in all four Houston models, while it was associated positively with PM<sub>2.5</sub> concentration of Los Angeles.

RH was selected in four models in Houston: PM<sub>2.5</sub>, OC, EC, and BaP with partial R<sup>2</sup>s explaining 4, 5, 7, and 34% of the overall variability in residential outdoor concentrations, respectively. RH was associated negatively with lnC<sub>out</sub>. RH was not selected in models for Los Angeles. All the Houston models included either dew point or RH as a predictor of lnC<sub>out</sub>. For OC and the four p-PAHs of Houston, dew point and RH were the most influential variables selected, while wind speed and atmospheric stability were the most influential variables selected in Los Angeles.

Precipitation was included in the four models in Houston and one model in Los Angeles. The models that included precipitation were PM<sub>2.5</sub>, OC, BaP, and Per in Houston with the partial R<sup>2</sup>s explaining 10, 6, 5, and 7% of overall variability in residential outdoor concentrations, respectively. Precipitation was included in the PM<sub>2.5</sub> model in Los Angeles with the partial R<sup>2</sup> explaining of 8% of overall variability in residential outdoor concentrations. Precipitation was associated negatively with PM<sub>2.5</sub> in Houston and in Los Angeles. In contrast, positive associations were observed in the OC, BaP, and Per models in Houston,

indicating that higher concentrations were measured on sampling days with higher precipitation. It is important to note that sampling was not performed during days of heavy precipitation.

### 3.3. Source proximity

The lnC<sub>out</sub> of EC had strong correlations with source proximity to petrochemical facilities in Houston (Table 7). Because it had the strongest association with EC, proximity to the petrochemical facilities was chosen to represent the general petrochemical refineries in Houston. Source proximity to petrochemical emissions was selected as the first and the most influential variable selected explaining 17% of overall variability in residential outdoor EC concentrations in Houston. Other than EC in Houston, source proximity was not selected for any PM<sub>2.5</sub> constituent models in Houston. Information on the point sources of the PM<sub>2.5</sub> constituents was not available for each species in the 1999 NEI. PAHs were listed as mixtures of polycyclic organic matter (POM) in three categories of 15-PAHs, 7-PAHs, and Non-15-PAHs. Area sources of VOCs were tested in order to determine if any of them were selected for models and thus detect false associations. As expected, the proximity to VOCs facilities was not significant and not selected in the PM models, reassuring that each regression model was selective

**Table 7.** The enhancement in close to far proximity for outdoor residential concentrations estimated from models using median values for other variables from dataset of each city

Highway proximity (Close/Far)		
	Houston	Los Angeles
PM <sub>2.5</sub>	n.a.	1.46
OC	n.a.	1.84
EC	1.75	3.46
BaP	4.38	4.61
Per	n.a.	9.32
BghiP	5.31	5.43
Cor	6.26	15.83
Petrochemical point source proximity (Close/Far)		
	Houston	Los Angeles
EC	1.73	n.a.

n.a.: Not available because the proximity variable was not selected in the model

only for influential variables explaining the residential outdoor levels of the specific air contaminant.

Highway proximity was included in models of Cor, BghiP, BaP, and EC in Houston with the partial  $R^2$ s explaining 7, 7, 4, and 3% of overall variability in residential outdoor concentrations, respectively. Highway proximity was a significant predictor in all seven models in Los Angeles for EC, OC, BghiP, Per, Cor, BaP, and  $PM_{2.5}$  mass with partial  $R^2$ s explaining 16, 10, 7, 6, 6, 4, and 3% of overall variability in residential outdoor concentrations, respectively. Both highway and arterial roadway proximity were selected as variables in the  $PM_{2.5}$  mass model in Los Angeles. Among the 14 models in Houston and Los Angeles, 11 models included highway proximity, and one model included arterial roadway proximity. Highway proximity was the first variable selected in the EC model in Los Angeles, and the second variable selected in OC, BaP, and Cor in Los Angeles. Arterial roadway proximity was the second variable selected in the  $PM_{2.5}$  mass model in Los Angeles with a partial  $R^2$  explaining 6% of overall variability in residential outdoor concentrations. Highway proximity and arterial roadway proximity were positively associated with  $\ln C_{out}$  in all models. Therefore models confirm that close proximity to traffic increased  $\ln C_{out}$ , and that the concentrations decreased as the distance from the highways or arterial roadways increased.

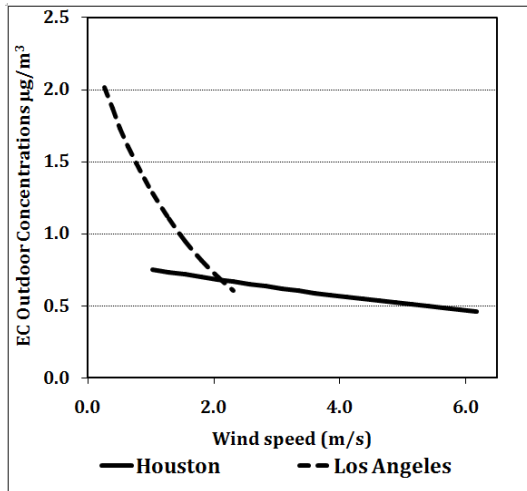
#### 4. Discussion

We have previously found that the majority of indoor  $PM_{2.5}$ , PAHs, and EC concentrations inside RIOPA homes originated from outdoor sources (Meng et al., 2007; Naumova et al., 2002; Polidori et al., 2006). Thus, the characterization of the impact of mobile and point emission sources on residential outdoor concentrations is important for understanding the outdoor source contribution of  $PM_{2.5}$  constituents to population exposure (Kwon et al., 2006; Polidori et al.,

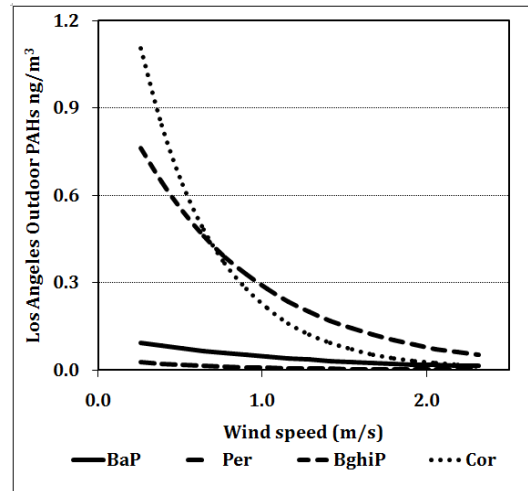
2010).

This study demonstrates the significant influence of source proximity on outdoor residential concentrations of eleven  $PM_{2.5}$  constituents measured in Houston and Los Angeles, and the influence of site-specific meteorological conditions for all  $PM_{2.5}$  constituents. Because  $PM_{2.5}$  mass and OC include major constituents that are secondarily formed regionally through atmospheric chemistry, it was expected that the enhancement of  $PM_{2.5}$  mass and OC are not nearly as large as the enhancement of PM components such as EC and p-PAHs that are mainly emitted directly from the primary sources (Table 7). Not only smaller enhancements were observed, but also the  $PM_{2.5}$  mass and OC models in Houston did not include any emission sources.  $PM_{2.5}$  mass and OC models in Los Angeles included mobile sources, however, overall variability explained by these models were relatively smaller (<28%) for  $PM_{2.5}$  and OC in Houston and Los Angeles (Table 5).

In Houston, the partial  $R^2$  value for the source proximity to refineries and highways accounted for 46% of the  $R^2$  for EC model while wind speed, RH, and atmospheric stability accounted for 54%. This suggests that the impact of emissions from petrochemical facilities and refineries on the  $\ln C_{out}$  of EC near the Houston Ship Channel and surrounding areas, where the Houston homes were mostly located, can be significant. In Elizabeth, NJ, proximity to refinery and truck loading areas were previously shown to be associated with elevated  $\ln C_{out}$  of EC (Polidori et al., 2010). The impact of emission from highways on EC concentrations in Los Angeles was significant. However, the impact of emission from point sources, such as refineries, on EC concentrations in Los Angeles was not significant because the homes in Los Angeles were not near refineries. In Los Angeles, the model partial  $R^2$  for highway proximity was larger than for wind speed, accounting for 16 and 14% of overall variability respectively. In a prior study, highway



**Fig. 1.** Simulated effect of wind speed on residential ambient air concentrations of EC in Houston and Los Angeles -estimated from the best fit models using the median values for other variables in each model.



**Fig. 2.** Simulated effect of wind speed on residential ambient air concentrations of BaP, Per, BghiP, and Cor in Los Angeles -estimated from the best fit models using the median values for other variables in each model.

proximity was included in models of PM<sub>2.5</sub> mass, OC, BghiP, and Cor in Elizabeth, NJ (Polidori et al., 2010).

Overall, the partial R<sup>2</sup> for the meteorological variables was typically larger than that for the proximity variables for most of the Houston and Los Angeles models with exception of the EC in Houston, implying that a greater percentage of the explanatory power was due to changes in the meteorological conditions rather than to the distance from emission sources. Although the p-values for highway proximity were higher than p-values of other individual variables included in models, 11 of the 14 models in Houston and Los Angeles included the roadway proximity. Overall, the multiple regression models explained 22~50% of the total variability of C<sub>out</sub> of PM<sub>2.5</sub> constituents.

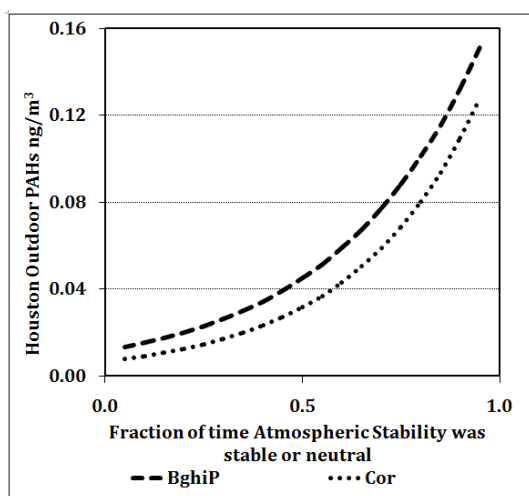
#### 4.1. Wind speed and stability

The most frequently selected meteorological variable explaining the variability of C<sub>out</sub> of PM<sub>2.5</sub> and selected constituents was wind speed. Seven of the 9 models included wind speed as either first or second

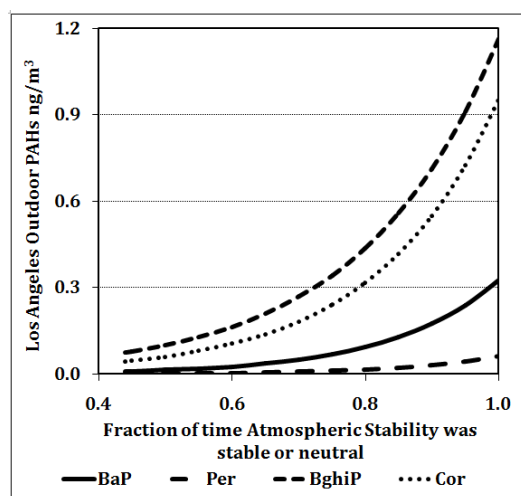
predictor variable with higher than 50% of model R<sup>2</sup> values for 3 models. The consistent inclusion of wind speed confirmed the importance of wind speed on the airborne concentration of PM in cities and the effect of horizontal dispersion on the dilution of air pollutants. Wind speed was also selected and associated negatively with concentration of PM<sub>2.5</sub>, Cor, and BghiP in a previous analysis of RIOPA data in Elizabeth, NJ (Polidori et al., 2010). Atmospheric stability was included in eight models including four p-PAHs models in Los Angeles together with the wind speed. Atmospheric stability and wind speed accounted for more than 86% of model R<sup>2</sup> for p-PAHs in Los Angeles explaining greater than 40% of overall variability in residential outdoor concentrations. Stability was included in the PM<sub>2.5</sub> mass, OC, EC, BghiP, and Cor models in Elizabeth NJ with positive coefficients (Polidori et al., 2010).

#### 4.2. Effect of wind

The calculated C<sub>out</sub> for EC estimated from the multiple regression models are plotted against wind



**Fig. 3.** Simulated effect of atmospheric stability on residential ambient air concentrations of BghiP and Cor in Houston estimated from the best fit models using the median values for other variables in each model.



**Fig. 4.** Simulated effect of atmospheric stability on residential ambient air concentrations of BaP, Per, BghiP, and Cor in Los Angeles estimated from the best fit models using the median values for other variables in each model.

speed in Houston and Los Angeles in Fig. 1 and for the Los Angeles p-PAHs in Fig. 2. The regression model results were based on the range of wind speeds measured in each city with all other variables in each model being held constant at the observed median values. The calculated  $C_{out}$  of the  $PM_{2.5}$  constituents decreased with increasing wind speed. The wind speed ranged from 1.0 to 6.2 m/s in Houston, while it ranged from 0.26 to 2.3 m/s in Los Angeles. The explained  $C_{out}$  in Los Angeles decreased more rapidly than that of Houston per unit increase of wind speed as shown in Fig. 1. Across the  $PM_{2.5}$  components, the  $C_{out}$  estimated at the minimum wind speed ranged from 2- to 82-fold higher in Los Angeles, and was 1.6-fold higher in Houston than the  $C_{out}$  estimated at the maximum wind speed for each site using the median distance between the home and sources. The enhancement of the calculated  $C_{out}$  of p-PAHs between the minimum and the maximum wind speed in Los Angeles was more significant (7- to 82-fold) than for the other PM constituents (2- to 3.3-fold) probably because p-PAHs

were primarily emitted from local sources compared to other constituents of  $PM_{2.5}$  and OC.

#### 4.3. Effect of atmospheric stability

The relationship between the calculated  $C_{out}$  of p-PAHs and the effect of atmospheric stability in Houston and Los Angeles estimated from the multiple regression models is illustrated in Fig. 3 and 4. The regression models were run by varying atmospheric stability across the observed range and using the respective median values for all other variables. The calculated  $C_{out}$  of p-PAHs increased with increasing amount of time with a stable or neutral atmospheric stability class during the sampling period. Atmospheric stability ranged from 0.05 to 0.95 in Houston, and from 0.44 to 1.0 in Los Angeles for the  $PM_{2.5}$  constituents.  $C_{out}$ 's estimated at the maximum atmospheric stability were 11- to 16-fold higher for p-PAHs in Houston (Fig. 3), and 15- to 47-fold higher in p-PAHs in Los Angeles (Fig. 4), respectively, than the  $C_{out}$ 's estimated at the minimum atmospheric stability. Increase between

minimum and maximum atmospheric stability for BghiP and Cor in Houston were 11- and 16- fold respectively, which were comparable to the increases observed for BghiP and Cor in Los Angeles, 16- and 22-fold respectively. The effect of atmospheric stability was relatively more significant for the C<sub>out</sub> of p-PAHs compared to the effect of atmospheric stability on the C<sub>out</sub> of PM<sub>2.5</sub>, OC, and EC concentrations.

#### 4.4. Dewpoint, RH, and precipitation

In the previous analysis on PM<sub>2.5</sub> constituents in Elizabeth, NJ (Polidori et al., 2010), temperature was associated negatively with the concentrations of BghiP and Cor. One possible explanation is the seasonal difference in gas/particle partitioning, and the increase of incomplete combustion byproducts from mobile source emissions at colder temperatures. Rather than temperature, dewpoint was included in four models in Houston and one model in Los Angeles. Dewpoint was negatively associated with the lnC<sub>out</sub> of OC, Per, BghiP, and Cor in Houston, while it was positively associated with the lnC<sub>out</sub> of PM<sub>2.5</sub> in Los Angeles. RH was included in models of PM<sub>2.5</sub>, OC, EC, and BaP in Houston, but RH was not included in any Los Angeles model. RH was associated negatively with lnC<sub>out</sub> for all four models in Houston. RH and dewpoint were included concurrently in the OC model with negative coefficients in Houston. Dewpoint, and RH are indicators of the moisture in the atmosphere along with the interaction with temperature. It seems likely that the higher water vapor content in the atmosphere is associated with decreased C<sub>out</sub> of PM<sub>2.5</sub> constituents in Houston. The results are also similar to VOCs analysis of the RIOPA study by Su et al. (2013), who reported that RH was negatively associated with residential outdoor concentrations of benzene, ethylbenzene, *m,p*-xylene, *o*-xylene, MTBE, styrene, and β-pinene. The inclusion of RH and/or dewpoint in Houston may imply a potentially greater impact of the humid climate in Houston on C<sub>out</sub> compared to Los Angeles or

Elizabeth. The seasonality observed in Elizabeth may reflect the colder temperatures in the winter that are not as extreme in Houston or Los Angeles.

Precipitation was associated negatively with PM<sub>2.5</sub> in Houston and in Los Angeles, which indicating decreased lnC<sub>out</sub> with increased precipitation. The scavenging of PM<sub>2.5</sub> by precipitation was observed in PM<sub>2.5</sub> both in Houston and Los Angeles suggesting that it is an important removal mechanism (Akyuz and Cabuk, 2009). In contrast to PM<sub>2.5</sub>, positive associations were observed for precipitation in the OC, BaP, and Per models in Houston. This inconsistent association of precipitation may indicate that either emissions of the p-PAHs were increased or unfavorable conditions for dispersion of p-PAHs occurred in Houston in comparatively more rainy days. It should be noted that the most of the RIOPA samples were not collected during heavy rain, therefore impact of precipitation in this analysis cannot be accurately generalized for rainy climate. Although the scavenging of PM<sub>2.5</sub> constituents by precipitation is expected (Akyuz and Cabuk, 2009), positive correlation of precipitation on PAHs are not uncommon. A significant positive correlation between PAHs and precipitation was observed in a site of Flanders (Ravindra et al., 2006), and significant inconsistency of the model estimated coefficients for precipitation were observed even among the adjacent sites for simultaneous time periods (Kim et al., 2013).

While the association of wind speed (negative) and atmospheric stability (positive) with C<sub>out</sub> of PM<sub>2.5</sub> constituents were consistent across the models, the directions of association with C<sub>out</sub> of PM<sub>2.5</sub> constituents with dewpoint, precipitation, and RH were inconsistent. Wind speed and atmospheric stability represent horizontal and vertical dispersion and dilution of PM<sub>2.5</sub> constituents, while dewpoint, precipitation, and RH may also be related to temporal changes of emissions during sample period which vary in different urban areas. This may be related to regional climate factors

that interact with meteorological variables differently in locations with different climate.

#### 4.5. Effect of point sources

The model-estimated EC concentrations were calculated against the proximity to petrochemical facilities in Houston, while all other variables were held constant using the observed median values. The calculated  $C_{out}$  of EC decreased with increasing distance to petrochemical facilities. The  $C_{out}$  of EC in Houston estimated at the closest distance of 10.6 km is  $0.85 \mu\text{g}/\text{m}^3$ , which is 1.7-fold higher than the estimated  $C_{out}$  of EC of  $0.49 \mu\text{g}/\text{m}^3$ , at the farthest distance of 70.1 km (Table 7). In Elizabeth, NJ, EC concentration was previously found to be associated with proximity to a refinery and a truck loading area, and  $\text{PM}_{2.5}$  was associated with truck loading areas (Polidori et al., 2010). The EC model in Houston included highway proximity concurrently in addition to point source proximity.

#### 4.6. Effect of highways

Model-calculated  $C_{out}$  of p-PAHs,  $\text{PM}_{2.5}$ , OC and EC against proximity to highways in Houston and Los Angeles while all other variables held constant at their observed median values are shown in Table 7. The calculated  $C_{out}$  of BghiP, and Cor in Houston and  $\text{PM}_{2.5}$ , OC, EC, Per, and BghiP in Los Angeles increased with decreasing distances to highways. To avoid misleading model explanatory power, calculation of individual variable effects were limited to  $\text{PM}_{2.5}$  constituents with adjusted model  $R^2$  larger than 0.25 and individual variables with  $p < 0.05$ . Therefore, enhancement effects for EC and BaP in Houston, and BaP and Cor in Los Angeles should be carefully interpreted due to the higher individual  $p$ -values ( $p > 0.05$ ) of the highway proximity. The sum of distances to the 5 closest highways ranged between 13.1 and 42 km in Houston, while it ranged between 12.7 and 29.4 km in Los Angeles. The  $C_{out}$  of EC

estimated at the closest distances were 3.5-fold ( $2.6 \mu\text{g}/\text{m}^2$  vs.  $0.75 \mu\text{g}/\text{m}^3$ ) higher in Los Angeles compared to the  $C_{out}$  estimated at the farthest distances (Table 7). The  $C_{out}$  of  $\text{PM}_{2.5}$  and OC in Los Angeles estimated at the closest distance were 1.5- and 1.8-fold higher respectively compared to the  $C_{out}$  of estimated at the furthest distance. The increase in calculated  $C_{out}$  between maximum and minimum distance to highways for p-PAHs in Houston and Los Angeles were comparable. The increases between maximum and minimum distances for BghiP and Cor in Houston were 5.3- and 6.3-fold respectively, and for Per and BghiP in Los Angeles were 9.3- and 5.4-fold respectively. The enhancements of estimated concentration in closest proximity were greater for primary PM constituents (p-PAHs, EC) that are directly emitted from sources compared to the enhancements for OC and  $\text{PM}_{2.5}$  mass (Table 7). This is expected because  $\text{PM}_{2.5}$  mass and OC have large regional secondary sources meaning particulate matter formed from gaseous emissions through gas and multiphase chemistry (Turpin et al., 2000). A recent Los Angeles Airport study reported large increases, 4- to 11-fold, over local background in particle number concentrations that routinely extended 18 km downwind with the greatest increases observed at locations under landing jet trajectories (Hudda and Fruin, 2016). Likewise, emission sources such as a congested network of highways in urban areas, continuously emit mixtures of pollutants. Their influence was greater in close proximity elevating urban background levels several km downwind especially for the primarily emitted PM constituents such as EC and particle-bound PAHs.

## 5. Conclusions

Multiple regression models identified the meteorological variables and proximity to point, area, and mobile sources that accounted for the greatest amount of variability in outdoor residential  $C_{out}$  of



PM<sub>2.5</sub> and selected constituents in Houston and Los Angeles. The multiple regression models explained 22~50% of the total variability of C<sub>out</sub> of PM<sub>2.5</sub> mass and OC, EC, Per, BaP, BghiP, and Cor constituents.

Difference in emission sources and meteorological conditions between the two urban sites studied led to some differences in variables included in the regression models. Wind speed, atmospheric stability, and RH were included as influential factors for several PM<sub>2.5</sub> constituents with consistent association in Houston and Los Angeles. Stability, wind speed, and highway proximity were the significant factors for p-PAHs in Los Angeles. Highway proximity was included in all seven models in Los Angeles while it was included in four models (EC, Per, BaP, BghiP, and Cor) in Houston. Proximity to petrochemical facilities was significant predictor for EC concentrations in Houston concurrently with highway proximity.

The dilution effect of high wind speed is demonstrated by negative associations of wind speed with concentrations. Atmospheric stability increase was associated with concentration increase in six models in two cities. Dewpoint and precipitation were included as influential factors for several PM<sub>2.5</sub> constituents, however the association varied by different constituents or by sites.

This study characterized residential outdoor concentrations of PM<sub>2.5</sub> constituents in urban residential areas in different cities with distinctively different climates and different local geographic profiles of emission sources in each study area successfully. The findings of this study supports the previous study performed on the VOCs in the same areas (Kwon et al., 2016). The results confirmed that urban site-specific regression model analysis on the community level sampling can be used for improving exposure estimates of the multi-components of PM<sub>2.5</sub> for epidemiological research in urban environments using available information of site-specific meteorology and location of the emission sources.

## Acknowledgements

We gratefully acknowledge the hospitality of the RIOPA participants, the hard work of all the students and technicians involved in the field and laboratories, and Dr. Jim Zhang for valuable contributions. The original RIOPA research was supported by The Mickey Leland National Urban Air Toxics Research Center (NUATRC) (contract # 96-01A/P01818769) and by The Health Effects Institute (HEI, contract # 98-23-3). HEI is jointly funded by the U.S. EPA (EPA: Assistance Agreement R828112) and automotive manufacturers. The NJ data analysis was supported by the U.S. EPA Office of Transportation and Air Quality (Contract #68-C-04-149). The contents of this article do not necessarily reflect the views of the Mickey Leland NUATRC, HEI, (and policies of) U.S. EPA or of motor vehicle and engine manufacturers. Drs. Weisel and Turpin were supported in part by the National Institute of Environmental Health Center for Excellence (ES05022). Dr. Turpin was supported in part by the NJ Agricultural Experiment Station. Dr. Kwon was also supported by an internal grant from the California State University, Fresno. Dr. Kwon is grateful to Bob for providing invaluable intuitions and encouragement.

## Conflict of interest

The authors declare no conflict of interest.

## Abbreviations

1999 NEI: National Emission Inventory of 1999  
C<sub>out</sub>: residential outdoor concentrations  
EC: elemental carbon in PM<sub>2.5</sub>  
BaP: benzo-[a]-pyrene in PM<sub>2.5</sub>  
BghiP: benzo-[g,h,i]-perylene in PM<sub>2.5</sub>  
Cor: coronene in PM<sub>2.5</sub>

OC: organic carbon in PM<sub>2.5</sub>

Per: perylene in PM<sub>2.5</sub>

PM<sub>2.5</sub>: fine particulate matter with an aerodynamic diameter of 2.5 µm or smaller

POM: Polycyclic organic matter

p-PAHs: particle bound polycyclic aromatic hydrocarbons

RH: relative humidity

RIOPA: Relationship among Indoor, Outdoor, and Personal Air

## REFERENCES

- Akyuz, M., Cabuk, H., 2009, Meteorological variations of PM<sub>2.5</sub>/PM<sub>10</sub> concentrations and particle-associated polycyclic aromatic hydrocarbons in the atmospheric environment of Zonguldak, Turkey, *J. Hazard. Mater.*, 170, 13-21.
- Anastasopoulos, A. T., Wheeler, A. J., Karman, D., Kulka, R. H., 2012, Intraurban concentrations, spatial variability and correlation of ambient polycyclic aromatic hydrocarbons (PAH) and PM<sub>2.5</sub>, *Atmos. Environ.*, 59, 272-283.
- Baxter, L. K., Barzyk, T. M., Vette, A. F., Croghan, C., Williams, R. W., 2008, Contributions of diesel truck emissions to indoor elemental carbon concentrations in homes in proximity to Ambassador Bridge, *Atmos. Environ.*, 42, 9080-9086.
- Fuller, C. H., Brugge, D., Williams, P. L., Mittleman, M. A., Lane, K., Durant, J. L., Spengler, J. D., 2013, Indoor and outdoor measurements of particle number concentration in near-highway homes, *J. Expo. Sci. Environ. Epidemiol.*, 23, 506-512.
- Gale, S. L., Noth, E. M., Mann, J., Balmes, J., Hammond, S. K., Tager, I. B., 2012, Polycyclic aromatic hydrocarbon exposure and wheeze in a cohort of children with asthma in Fresno, CA, *J. Expo. Sci. Environ. Epidemiol.*, 22, 386-392.
- Hasheminassab, S., Daher, N., Shafer, M. M., Schauer, J. J., Delfino, R. J., Sioutas, C., 2014, Chemical characterization and source apportionment of indoor and outdoor fine particulate matter (PM<sub>2.5</sub>) in retirement communities of the Los Angeles Basin, *Sci. Total Environ.*, 490, 528-537.
- HEI, NUATRC, 2008, Health Effect Institute & National Urban Air Toxics Research Center, Relationships of Indoor, Outdoor, and Personal Air (RIOPA) Database Part 2008, MA.
- Hew, K. M., Walker, A. I., Kohli, A., Garcia, M., Syed, A., McDonald-Hyman, C., Noth, E. M., Mann, J. K., Pratt, B., Balmes, J., Hammond, S. K., Eisen, E. A., Nadeau, K. C., 2015, Childhood exposure to ambient polycyclic aromatic hydrocarbons is linked to epigenetic modifications and impaired systemic immunity in T cells, *Clin. Exp. Allergy*, 45, 238-248.
- Hodas, N., Meng, Q., Lunden, M. M., Rich, D. Q., Ozkaynak, H., Baxter, L. K., Zhang, Q., Turpin, B. J., 2012, Variability in the fraction of ambient fine particulate matter found indoors and observed heterogeneity in health effect estimates, *J. Expo. Sci. Environ. Epidemiol.*, 22, 448-454.
- Hudda, N., Fruin, S. A., 2016, International airport impacts to air quality: Size and related properties of large increases in ultrafine particle number concentrations, *Environ. Sci. Technol.*, 50, 3362-3370.
- Kanakidou, M., Seinfeld, J. H., Pandis, S. N., Barnes, I., Dentener, F. J., Facchini, M. C., Van Dingenen, R., Ervens, B., Nenes, A., Nielsen, C. J., Swietlicki, E., Putaud, J. P., Balkanski, Y., Fuzzi, S., Horth, J., Moortgat, G. K., Winterhalter, R., Myhre, C. E. L., Tsigaridis, K., Vignati, E., Stephanou, E. G., Wilson, J., 2005, Organic aerosol and global climate modelling: A review, *Atmos. Chem. Phys.*, 5, 1053-1123.
- Kelly, F. J., Fussell, J. C., 2012, Size, source and chemical composition as determinants of toxicity attributable to ambient particulate matter, *Atmos. Environ.*, 60, 504-526.
- Kim, Y., Seo, Y. K., Baek, S. O., 2013, A statistical inference for concentrations of benzo[a]pyrene partially measured in the ambient air of an industrial city in Korea, *Atmos. Environ.*, 81, 92-101.
- Kwon, J., Weisel, C. P., Turpin, B. J., Zhang, J., Korn, L. R., Morandi, M. T., Stock, T. H., Colome, S., 2006, Source proximity and outdoor-residential VOC concentrations: Results from the RIOPA study, *Environ. Sci. & Technol.*, 40, 4074-4082.
- Kwon, J., Weisel, C. P., Morandi, M. T., Stock, T. H., 2016, Source proximity and meteorological effects on residential outdoor VOCs in urban areas: Results from

- the Houston and Los Angeles RIOPA studies, *Sci. Total Environ.*, 573, 954-964.
- Larson, T., Gould, T., Simpson, C., Liu, L. J., Claiborn, C., Lewtas, J., 2004, Source apportionment of indoor, outdoor, and personal PM<sub>2.5</sub> in Seattle, Washington, using positive matrix factorization, *J. Air Waste Manag. Assoc.*, 54, 1175-1187.
- Lawson, S. J., Galbally, I. E., Powell, J. C., Keywood, M. D., Molloy, S. B., Cheng, M., Selleck, P. W., 2011, The effect of proximity to major roads on indoor air quality in typical Australian dwellings, *Atmos. Environ.*, 45, 2252-2259.
- Meng, Q. Y., Turpin, B. J., Lee, J. H., Polidori, A., Weisel, C. P., Morandi, M., Colome, S., Zhang, J., Stock, T., Winer, A., 2007, How does infiltration behavior modify the composition of ambient PM<sub>2.5</sub> in indoor spaces? An analysis of RIOPA data, *Environ. Sci. Technol.*, 41, 7315-7321.
- Nadeau, K., McDonald-Hyman, C., Noth, E. M., Pratt, B., Hammond, S. K., Balmes, J., Tager, I., 2010, Ambient air pollution impairs regulatory T-cell function in asthma, *J. Allergy Clin. Immunol.*, 126, 845-852 e810.
- Naumova, Y. Y., Eisenreich, S. J., Turpin, B. J., Weisel, C. P., Morandi, M. T., Colome, S. D., Totten, L. A., Stock, T. H., Winer, A. M., Alimokhtari, S., Kwon, J., Shendell, D., Jones, J., Maberti, S., Wall, S. J., 2002, Polycyclic aromatic hydrocarbons in the indoor and outdoor air of three cities in the U.S, *Environ. Sci. Technol.*, 36, 2552-2559.
- National Oceanic and Atmospheric Administration, 2012, Air resources laboratory, real-time environmental applications and display (READY) system.
- Noth, E. M., Hammond, S. K., Biging, G. S., Tager, I. B., 2011, A spatial-temporal regression model to predict daily outdoor residential PAH concentrations in an epidemiologic study in Fresno, CA, *Atmos. Environ.*, 45, 2394-2403.
- Padula, A. M., Noth, E. M., Hammond, S. K., Lurmann, F. W., Yang, W., Tager, I. B., Shaw, G. M., 2014, Exposure to airborne polycyclic aromatic hydrocarbons during pregnancy and risk of preterm birth, *Environ. Res.*, 135, 221-226.
- Park, M., Luo, S., Kwon, J., Stock, T. H., Delclos, G., Kim, H., Yun-Chul, H., 2013, Effects of air pollution on asthma hospitalization rates in different age groups in metropolitan cities of Korea, *Air Qual. Atmos. Health*, 6(3), 543-551.
- Perera, F. P., Li, Z., Whyatt, R., Hoepner, L., Wang, S., Camann, D., Rauh, V., 2009, Prenatal airborne polycyclic aromatic hydrocarbon exposure and child IQ at age 5 years, *Pediatrics*, 124, e195-202.
- Polidori, A., Turpin, B., Meng, Q. Y., Lee, J. H., Weisel, C., Morandi, M., Colome, S., Stock, T., Winer, A., Zhang, J., Kwon, J., Alimokhtari, S., Shendell, D., Jones, J., Farrar, C., Maberti, S., 2006, Fine organic particulate matter dominates indoor-generated PM<sub>2.5</sub> in RIOPA homes, *J. Expo Sci. Environ. Epidemiol.*, 16, 321-331.
- Polidori, A., Kwon, J., Turpin, B. J., Weisel, C., 2010, Source proximity and residential outdoor concentrations of PM<sub>2.5</sub>, OC, EC, and PAHs, *J. Expo Sci. Environ. Epidemiol.*, 20, 457-468.
- Ravindra, K., Bencs, L., Wauters, E., de Hoog, J., Deutsch, F., Roekens, E., Bleux, N., Berghmans, P., Van Grieken, R., 2006, Seasonal and site-specific variation in vapour and aerosol phase PAHs over Flanders (Belgium) and their relation with anthropogenic activities, *Atmos. Environ.*, 40, 771-785.
- Rohr, A. C., Wyzga, R. E., 2012, Attributing health effects to individual particulate matter constituents, *Atmos. Environ.*, 62, 130-152.
- Su, F. C., Mukherjee, B., Batterman, S. 2013, Determinants of personal, indoor and outdoor VOC concentrations: an analysis of the RIOPA data, *Environ. Res.*, 126, 192-203.
- Turpin, B. J., Saxena, P., Andrews, E., 2000, Measuring and simulating particulate organics in the atmosphere: Problems and prospects, *Atmos. Environ.*, 34, 2983-3013.
- Turpin, B. J., Weisel, C. P., Morandi, M., Colome, S., Stock, T., Eisenreich, S., Buckley, B., 2007, Relationships of Indoor, Outdoor, and Personal Air (RIOPA): Part II. Analyses of concentrations of particulate matter species, *Res. Rep. Health Eff. Inst.*, 1-77, Discussion 79-92.
- United States Census Bureau, 2012, TIGER Products.
- United States Environmental Protection Agency, 2003, Technology transfer network, clearinghouse for inventories and emission factors (TTN CHIEF) for 1999, National emission inventory documentation and

data - Final version 3.0.

Weisel, C. P., Zhang, J., Turpin, B. J., Morandi, M. T., Colome, S., Stock, T. H., Spektor, D. M., Korn, L., Winer, A. M., Kwon, J., Meng, Q. Y., Zhang, L., Harrington, R., Liu, W., Reff, A., Lee, J. H., Alimokhtari, S., Mohan, K., Shendell, D., Jones, J., Farrar, L., Maberti, S., Fan, T., 2005, Relationships of Indoor, Outdoor, and Personal Air (RIOPA). Part I. Collection methods and descriptive analyses, Res. Rep.

Health Eff. Inst., 1-107, Discussion 109-127.

Wittkopp, S., Staimer, N., Tjoa, T., Stinchcombe, T., Daher, N., Schauer, J. J., Shafer, M. M., Sioutas, C., Gillen, D. L., Delfino, R. J., 2016, Nrf2-related gene expression and exposure to traffic-related air pollution in elderly subjects with cardiovascular disease: An exploratory panel study, J. Expo. Sci. Environ. Epidemiol., 26, 141-149.