

# CHARACTERIZING SPATIAL VARIATION IN DIESEL PARTICULATE MATTER ACROSS DOWNTOWN PITTSBURGH

Brett J. Tunno, Jessie L.C. Shmool, Drew R. Michanowicz, Sheila Tripathy, Ellen Kinnee, Sara Gillooly, Lauren G. Chubb, Leah Cambal, Courtney Roper, Jane E. Clougherty

University of Pittsburgh Graduate School of Public Health, Department of Environmental and Occupational Health

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## INTRODUCTION:

Diesel-related particulate matter (DPM) has been associated with cancer and other chronic disease outcomes, and thus may pose a significant health risk to residents of Allegheny County. Further, DPM exposures may be particularly high in Downtown Pittsburgh, given a high daytime worker population and heavy traffic activity where a large number of bus lines and truck traffic converge.

It has been unknown, however, the extent to which DPM exposures vary across different parts of the downtown core, and which sources drive spatial patterns in concentrations and exposures. More clearly identifying these key sources can help to tailor cost-effective interventions, to most efficiently reduce population exposures.

Accurately capturing spatial patterns in airborne DPM is complicated by a number of factors:

- First, DPM is a highly complex chemical mixture of both volatile organic and inorganic species. Although black carbon (BC), elemental carbon (EC), and organic carbon (OC) are commonly-used markers, there is no definitive measure, or chemical tracer, for DPM.
- Second, DPM is emitted from many types of sources (e.g., buses, trucks, trains, etc.), at varying temperatures, and in varying combination with other pollution sources (i.e., gasoline vehicle traffic).
- Third, DPM emissions – and thus spatial patterns in exposures -- can differ substantially by time of day (e.g., rush-hours vs. full-week measures).
- Finally, because of the volatility of many DPM components, reactions with other pollutants in ambient air – and thus local concentrations and composition – can differ by season, meteorology, and the local emissions mixture.

Researchers at the University of Pittsburgh's Graduate School of Public Health collaborated with ACHD to investigate the spatial variation in DPM-related fine particulate matter (PM<sub>2.5</sub>) components, within and across the Downtown Pittsburgh core. *We hypothesized that:*

- 1) Pollution concentrations would differ across the downtown area, and this spatial pattern may differ by DPM component.

- 2) Concentrations – and spatial patterns in concentrations -- may differ by season (summer vs. winter), and by time of day (“rush hours” vs. “work week” averages). Few studies have been designed to capture pollutant spatial variation across differing time periods, and our campaign sampled diesel-related pollutants across rush and work-week hours.
- 3) Land use regression (LUR) modeling techniques could help to identify which pollution sources are most strongly associated with patterns in concentrations.
- 4) Source apportionment techniques – which identify pollutants strongly correlated with each other – could help to validate chemical tracers for DPM emissions.

This report summarizes key results of a unique two-year air monitoring campaign across downtown Pittsburgh. Few studies have been able capture spatial variation in diesel-related organic pollutants, or to differentiate spatial patterns by time of day (e.g., rush hours vs. work-week hours), as has been done here. Further information is available in the published and in-press manuscripts cited herein.

## **METHODS:**

The study was designed in two phases:

- *Phase one* (year one) focused on identifying spatial variability (spatial patterns) in the concentrations of multiple pollutants across the downtown area, during full “work-week” hours (Monday – Friday, 7 am – 7 pm).
- *Phase two* (year two) compared pollutant concentration and spatial patterns, during “rush-hour” (Monday - Friday, 5 – 10 am and 3 – 7 pm) vs. “work-week” hours.

**Pollutant selection and analysis:** We opted to focus on the chemical components of fine particulate matter with a diameter of 2.5 micrometers or less (PM<sub>2.5</sub>), which can penetrate deep into the lungs, impacting human health.

Our selection criteria for organic markers required each compound to be: (1) previously identified as markers of diesel exhaust; (2) quantifiable using GC-MS analytic methods, and (3) had relatively lower volatility, and reactivity, compared to other diesel components.

The final organics marker list included:

- nine (9) PAHs (benz[a]anthracene, benzo[a]pyrene, benzo[e]pyrene, benzo[ghi]fluoranthene, benzo[ghi]perylene, chrysene, fluoranthene, indeno[123-cd]pyrene, and pyrene)
- hopanes (homohopane, hopane, norhopane, trisnorhopane)
- steranes (cholestanes).

Based on the results of a prior literature review (Tunno et al., *J Expos Sci Environ Epidem* 2015), we identified three elemental components of PM<sub>2.5</sub> previously associated with diesel emissions -- aluminum (Al), calcium (Ca), and iron (Fe) – and included these in the analyses described here.

**Instrumentation:** Harvard Impactors (HIs) with Teflon™ filters housed in weather tight Pelican boxes for collection of PM<sub>2.5</sub>, BC, and trace metals. In separate boxes, organic compounds were collected on quartz filters using HI-adapted cyclones. Battery-operated vacuum pumps maintained a steady flow rate of 4 liters per minute. The sampling units were originally designed for the New York City Community Air Survey ([www.nyc.gov/health/nyccas](http://www.nyc.gov/health/nyccas)), and were mounted approximately 3 meters above street-level, strictly on metal utility poles to avoid contamination from treated wood poles, as shown in Figure 1.



*Figure 1: Air sampling units mounted on metal utility pole.*

**Sample analysis:** Total PM<sub>2.5</sub> mass concentrations were determined by gravimetric (filter-weighing) methods inside a temperature- and humidity-controlled chamber. BC absorbance was measured via reflectometry.

Organic carbon and elemental carbon concentrations were determined via thermal-optical analysis at Desert Research Institute (DRI) (Reno, NV). Other organic compounds (PAHs, hopanes, steranes) were assessed via thermal desorption gas chromatography mass spectrometry (TD-GC-MS) at DRI.

Elemental constituent concentrations were assessed via inductively-coupled plasma mass spectrometry (ICP-MS), at the Wisconsin State Laboratory of Hygiene, following documented protocols.

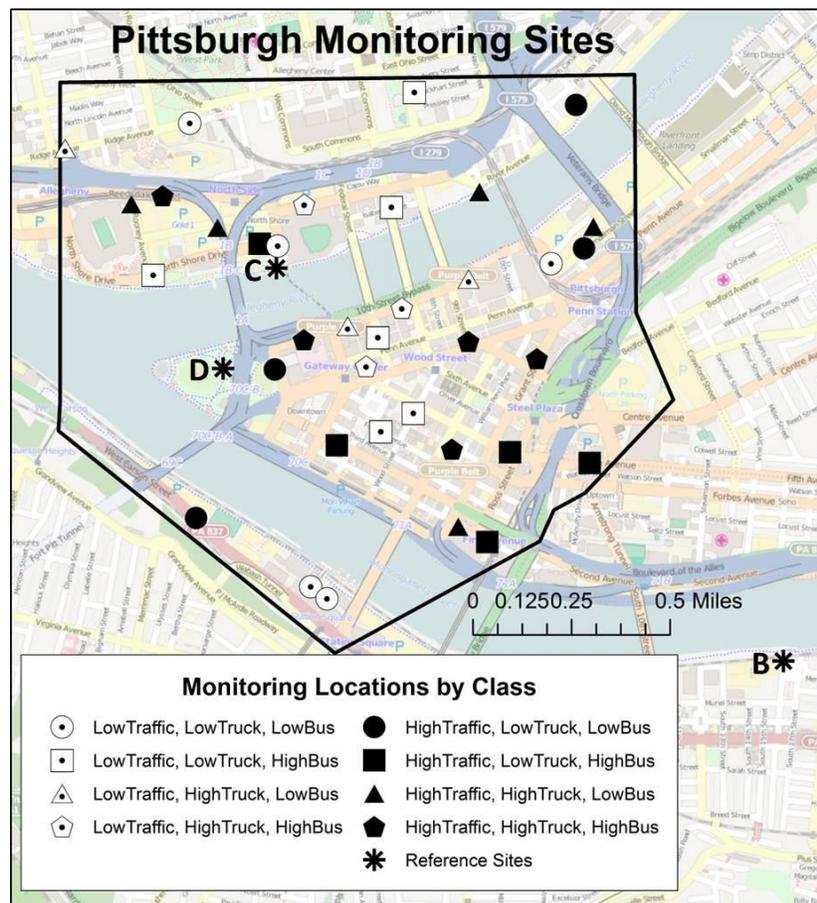
**Sampling domain and site selection:** The study domain (~2.8 km<sup>2</sup>) included the entirety of the downtown core, and a portion of the surrounding areas across the Allegheny and Monongahela rivers (Figure 2).

Prior to selecting sampling sites, we characterized the full domain using spatial software known as geographic information systems (GIS). In GIS, we mapped, and compare patterns in, a wide range of diesel-related sources (e.g., bus routes, heavy truck traffic, active railways) and indicators of gasoline traffic (e.g., total traffic density, parking lots).

Because we aimed to disentangle the effects of key diesel sources (i.e., truck traffic, bus traffic) from gasoline source (i.e., vehicular traffic), we characterized each 50 m x 50 m grid cell within our domain as “high” or “low” in each of these sources. Merging these indicators created eight possible categories, or “source classes.” We randomly selected four (4) cells from each class; these 32 selected sites (shown in Figure 2) capture a wide range of densities in total traffic, truck traffic, and bus route densities, and every combination thereof. Four additional sites were also incorporated to capture locations of interest, incl. those with intensive bus activity (e.g., 6<sup>th</sup> Street, Wood Street, Station Square).

During phase two (Year Two), owing to increased demand on sampling equipment (needing four samplers per site per week, instead of two), we reduced the number of sampling sites to 24, retaining the same balance across source classes, and coverage across the full study area.

Figure 2: Downtown Pittsburgh 36 monitoring locations, plus reference sites B, C, and D. Upwind reference site A at Settlers Cabin Park is not shown.



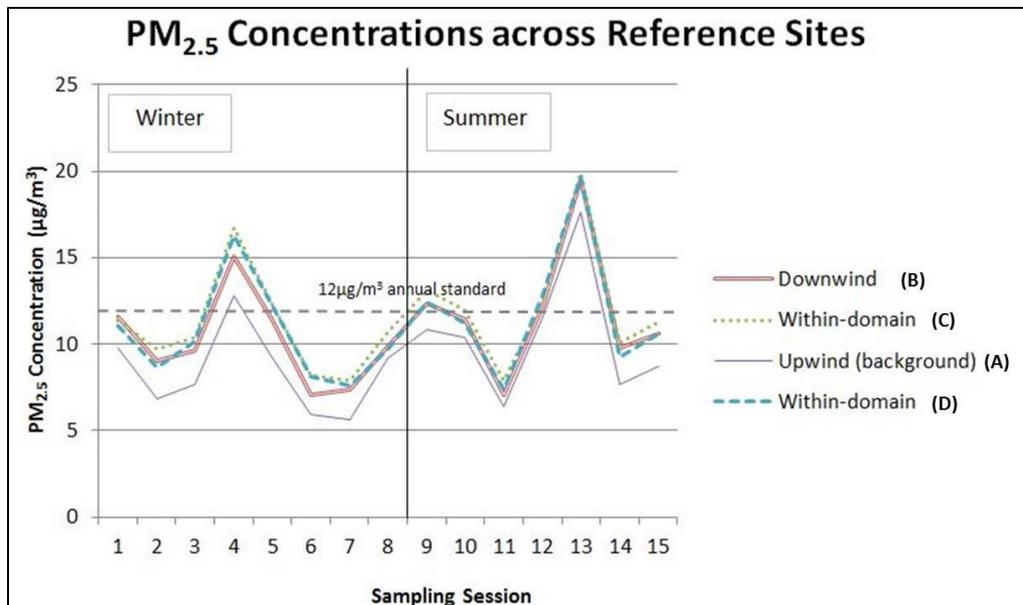
**Reference Sites:** Due to equipment limitations, all monitoring sites could not be sampled during the same week. For this reason, we selected four (4) reference sites, to be monitored every week; this data enabled us to understand pollution trends over time, and to “adjust” samples collected across different weeks. Three reference sites (B, C, D) are shown in Figure 2. Reference Site A is located upwind, in Settlers Cabin Park.

During Year One, we compared trends in pollutant concentrations measured at all four reference monitors, across all study weeks, as shown in Figure 3. All four sites revealed similar trends, with the lowest concentrations observed upwind at Settlers Cabin Park, as expected.

During phase two (Year Two), having already established comparable temporal trends across all four (4) reference sites during both summer and winter seasons, we eliminated two (2) reference

sites, retaining only the upwind Site A in Settlers Cabin Park and Site D in Point State Park, the largest park in the study area, at the nexus of the three rivers.

Figure 3: Temporal trend in PM<sub>2.5</sub> across four reference sites, Year 1



**Diurnal sampling programs:** For Phase One, samplers were programmed to run continuously each weekday (Monday - Friday) 7am to 7pm, simultaneously across all monitoring locations. Winter sampling was performed from January 14<sup>th</sup> to March 3<sup>rd</sup>, and summer sampling from June 10<sup>th</sup> to August 2<sup>nd</sup>, 2013.

During Phase Two, 24 sites were retained, and monitored for contemporaneous “work-week” and “rush-hour” samples. This approach enabled site-specific comparisons of full-week (Monday - Friday, 7am - 7pm) and “rush-hour” concentrations (Monday - Friday, 5 - 10am and 3 - 7pm).

The “rush” hours were selected to capture hours of peak diesel activity, based on Pennsylvania Department of Transportation (PennDOT) hourly traffic counts, and Allegheny County Port Authority bus schedules. Phase two of winter sampling was performed from January 13<sup>th</sup> to February 7<sup>th</sup>, and summer sampling from July 7<sup>th</sup> to August 1<sup>st</sup>, 2014.

**Temporal adjustment:** To estimate seasonally-representative concentrations, researchers applied a temporal adjustment to account for time-varying meteorology between sampling sessions in each season. Specifically, the observed concentration at a given site was divided by the session-specific average concentration from the four reference sites, then multiplied by the overall seasonal average from the four reference sites. All concentrations reported in tables and maps in this document, and in manuscripts prepared for publication, have been temporally adjusted in this manner.

**Land use regression (LUR) modeling:** LUR examines how measured pollution concentrations vary across space, in relation to traffic, buildings, railroads, and other sources. By comparing correlations between concentrations and a wide variety of source indicators (listed in Table 1), the method identifies those source indicators which best explain spatial variability in a pollutant of interest. Season-specific LUR models were developed for PM<sub>2.5</sub>, black carbon (BC), total elemental carbon (EC), organic carbon (OC), polycyclic aromatic hydrocarbons (PAHs), hopanes, steranes, aluminum (Al), calcium (Ca), and iron (Fe).

LUR models rely on indicators of air pollution sources (e.g., traffic density, proximity to industry), created in GIS. We created a wide suite of GIS-based covariates to represent an array of source indicator categories (Table 1). All covariates were summarized within circular buffers surrounding each monitoring location (25 to 200 meters), and we examined correlations between each source term and each pollutant.

*Table 1: GIS-based source covariates created and tested in LUR models:*

Source category for LUR modeling	Covariates examined (25m to 200m buffers)	Data source
Traffic density indicators	Mean density of annualized average traffic Mean density traffic (primary and secondary roads) Number of signaled intersections  Annualized average traffic/ Aspect ratio	Pennsylvania Spatial Data Access (PASDA, 2014) Southwestern Pennsylvania Commission (SPC, 2011)
Road-specific measures	Mean beta index of road complexity and connectivity Distance to nearest intersection Number of intersections Distance to nearest major road Summed length of primary roadways Summed length of primary and secondary roadways Width of roadways	TeleAtlas StreetMap (2014)  PASDA (2014)
Truck, Bus, and Diesel	Mean density of bus traffic Distance to nearest bus route Distance to nearest bus stop Bus stop use (total number of trips) Mean density of heavy truck traffic on nearest primary roadway	Google Transit Feed (7/14)  PASDA (2014)
Industrial emissions	Mean density of SO <sub>2</sub> emissions Mean density of PM <sub>2.5</sub> emissions Mean density of NO <sub>x</sub> emissions Mean density of VOC emissions	National Emissions Inventory (NEI, 2011)
Land use/ Built environment	Total area of commercial parcels Total area of industrial parcels Total area of industrial and commercial parcels Distance to nearest park Summed area of parks Building counts Distance to nearest building Mean percentage of imperviousness	Allegheny County Office of Property Assessments (AC OPA, 2013)  SPC (2011)  Allegheny County Department of Public Works (DPW) National Land Cover Database (NLCD, 2011)

Transportation facilities	Distance to nearest active railroad Summed line length of active railroads Distance to nearest bus depot Summed area of parking lots and garages Distance to river centerline	SPC (2011)  Google Transit (2014) AC OPA (2013) PASDA (National Hydrography Dataset, 2014)
<b>Potential modifying factors</b>		
Structural modifiers	Aspect ratio: building height/ roadway width Mean building heights	DPW
Topography	Average elevation Average slope Mean percentage of tree canopy	National Elevation Dataset (NED, 2013) NLCD (2011)
Meteorology	Temperature Relative humidity Frequency of inversions  Wind direction Wind speed	Obtained from sampler  Univ. of Wyoming, Dept. of Atmos. Science (2013) National Oceanic and Atmospheric Association (NOAA, 2013)

The first step in our LUR model-building was to account for temporal variation, by incorporating the mean concentration from the four reference sites each session into the model. The GIS-based source terms with the strongest correlation with the pollutant were then incorporated, individually, in descending order by the strength of correlation. Source covariates were retained in the model only if it improved overall model fit, by the coefficient of determination ( $R^2$ ). Any source indicator which became non-significant upon inclusion of a new term was removed, in order of descending statistical significance (p-value), until all model terms were significant ( $p < 0.05$ ). A number of statistical sensitivity tests were used to ensure robustness and reproducibility of final models.

**Source apportionment/ Factor analysis:** A two-stage “source apportionment” analysis was performed, using all pollutant measures, each season. This approach is based on factor analysis, a statistical method which identifies groups of highly-correlated pollutants.

In this type of source apportionment, a “factor” is a strongly-correlated group of pollutants which share an underlying distribution (in this case, a common spatial pattern) and are generally distinct from other groups. The number of factors (groups) allowed was determined using statistical criteria (e.g., eigenvalue-one criteria, scree plots). Only factors explaining at least 2% of total concentrations variance were retained in final models. To interpret each factor, we identified those constituents for which that factor explained greater than 60% of variance, and compared these to our existing literature review.

A “factor score” is a location-specific indicator roughly proportional to the sum of concentrations for each pollutant loading strongly onto that factor, weighted by its strength of association with the underlying pattern that each factor reflects.

We calculated and mapped these “factor scores” for each monitoring site, each season. Finally, using the LUR modeling approach described above, we identified key local sources explaining

variance in each factor. We performed this factor analysis only for Year One (n = 36 sites), as the Year Two dataset (n = 24 sites) was statistically under-powered for this approach.

## RESULTS - Phase One (Spatial variation in multiple pollutants):

Notably, greater variation in PM<sub>2.5</sub> and most constituents was observed across sites than between seasons – even within this relatively small study area. Accordingly, mean PM<sub>2.5</sub> across all sites was similar between seasons. Of all constituents examined, only steranes displayed greater seasonal than spatial variance.

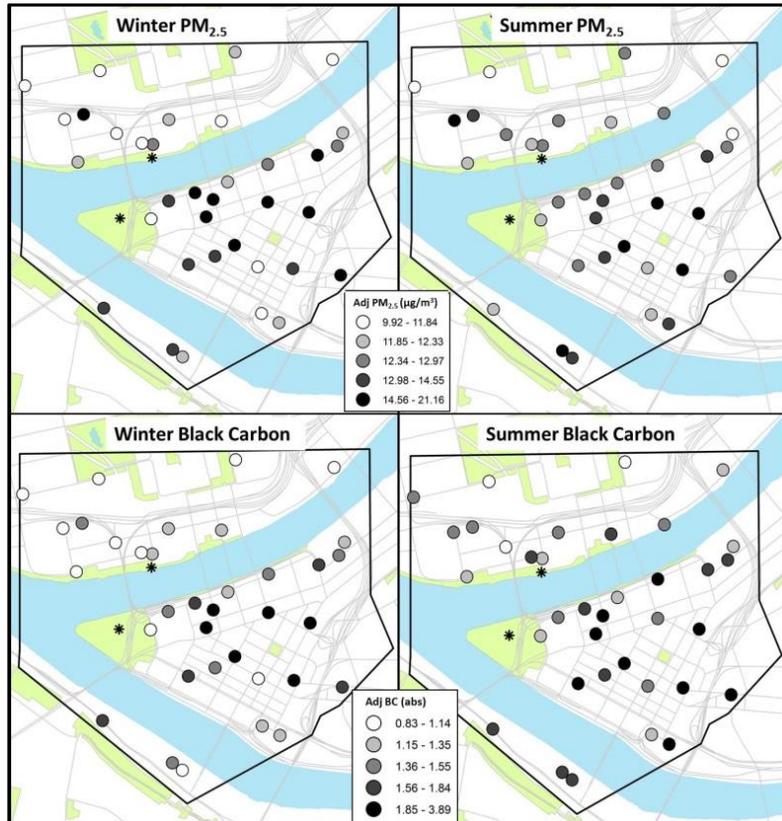
Diesel-related elemental concentrations (i.e., Al, Ca, Fe), likewise, did not significantly differ between seasons. For BC, EC, and OC, we observed significantly higher mean concentrations during summer than winter.

Table 2: Season-specific concentrations of PM<sub>2.5</sub>, BC, EC, and OC (Year 1)

Pollutant	Winter (n = 35)			Summer (n = 36)			p-value for difference between seasons (paired samples)
	Mean (SD)	Min	Max	Mean (SD)	Min	Max	
PM <sub>2.5</sub> (µg/m <sup>3</sup> )	13.2 (2.33)	9.92	21.2	13.3 (1.99)	11.1	20.5	0.84
BC (abs)	1.49 (0.58)	0.88	3.07	1.68 (0.64)	0.83	3.89	0.002
EC (µg/m <sup>3</sup> )	1.30 (0.53)	0.59	3.96	1.89 (1.09)	0.90	7.32	0.01
OC (µg/m <sup>3</sup> )	1.89 (0.55)	1.04	4.02	2.46 (0.62)	1.48	3.78	0.0001

In both seasons, we found similar spatial patterning in PM<sub>2.5</sub>, BC, EC, and OC, with higher concentrations near the center of downtown (Figure 4). Some PAHs, hopanes, and steranes, however, displayed somewhat different spatial patterns.

Figure 4:  $PM_{2.5}$  and BC concentrations differed substantially across downtown Pittsburgh.



**LUR modeling results:** In season-specific LUR models, a substantial portion of the total variability in measured  $PM_{2.5}$  was explained by temporal variance, comprised of meteorology and week-to-week differences in regional concentrations.  $PM_{2.5}$  was also influenced by mean bus density near each site during winter, and by mean bus density and total area of nearby parking garages during summer.

Bus density accounted for a greater difference in  $PM_{2.5}$  mass during winter than summer; a one-interquartile range (IQR) increase (25<sup>th</sup> to 75<sup>th</sup> percentile) increase in bus density conferred an increase of  $2.01 \mu\text{g}/\text{m}^3$  in  $PM_{2.5}$  during winter, vs. only  $0.89 \mu\text{g}/\text{m}^3$  during summer).

For BC and EC, both winter and summer LUR models included a significant effect of bus density. For OC, bus stop use (total number of scheduled stops per day) was significant in both models.

Substantial differences were observed in season-specific models for organic compounds. For winter, we found greater temporal contributions to models for total PAHs, total hopanes, and total steranes; no spatial source terms explained the remaining variability.

In contrast, bus density terms were significant in summer models for organic compounds. The spatial pattern in total PAHs was predicted by mean bus density. Mean bus density and mean truck density explained variation in total hopanes. Total steranes were predicted by bus stop use.

Substantial seasonal differences were also observed in LUR models for trace elements. During winter, temporal contributions were minimal, and bus density explained some spatial variability only in Al. During summer, bus terms were significant in each of the trace element models. For Al and Ca, the spatial pattern was predicted by bus stop use and summed area of parking garages. For Fe, terms for mean bus density and total area of parking garages were significant.

**Source apportionment results:** Source apportionment analyses, merging all diesel-related constituents described here, plus a larger suite of elemental source tracers, identified diesel-related factors in both seasons.

An example of our factor analysis output is shown in Figure 5. For these wintertime samples, a 5-factor solution explained 88% of overall variability in constituent concentrations. Factor one was characterized by several organic compounds elsewhere associated with *traffic density*. Factor two was characterized by several elemental constituents associated with *traffic*. Factor three included pollutants associated with *diesel* (BC, fluoranthene, NO<sub>2</sub>, pyrene, total carbon). Factor four included commonly-used tracers for *fuel oil* (Ni, V). Factor five included pollutants associated with *motor vehicle emissions* (benzene, Cd, La, toluene).

Figure 5: The winter season Year One 5-factor solution

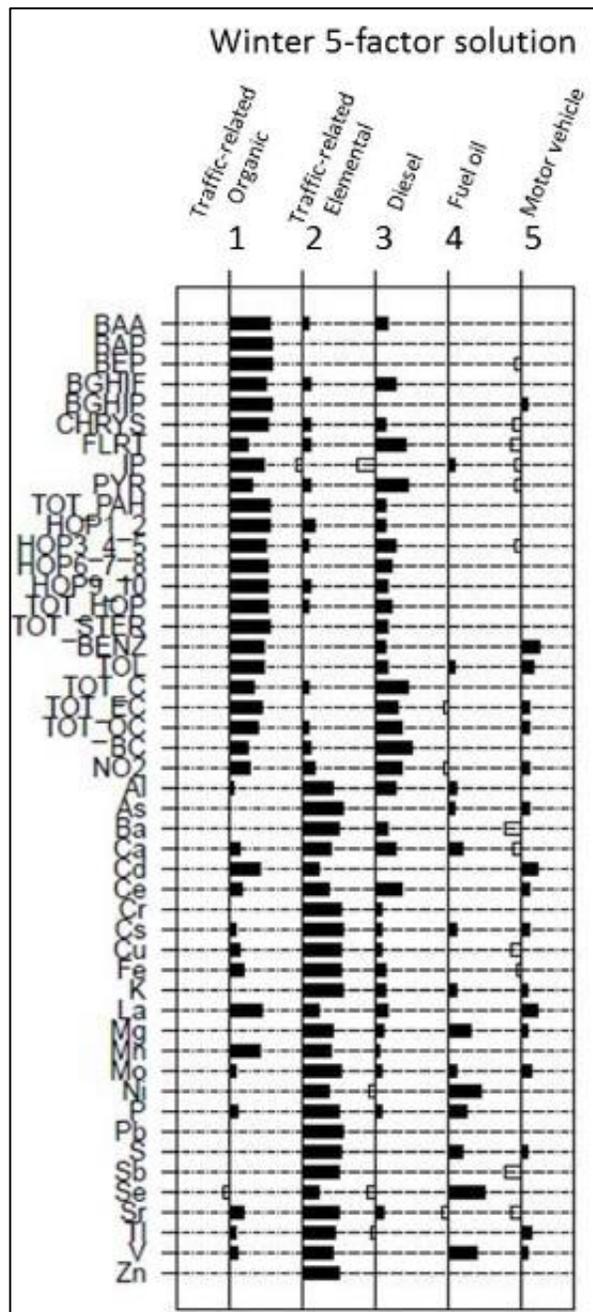
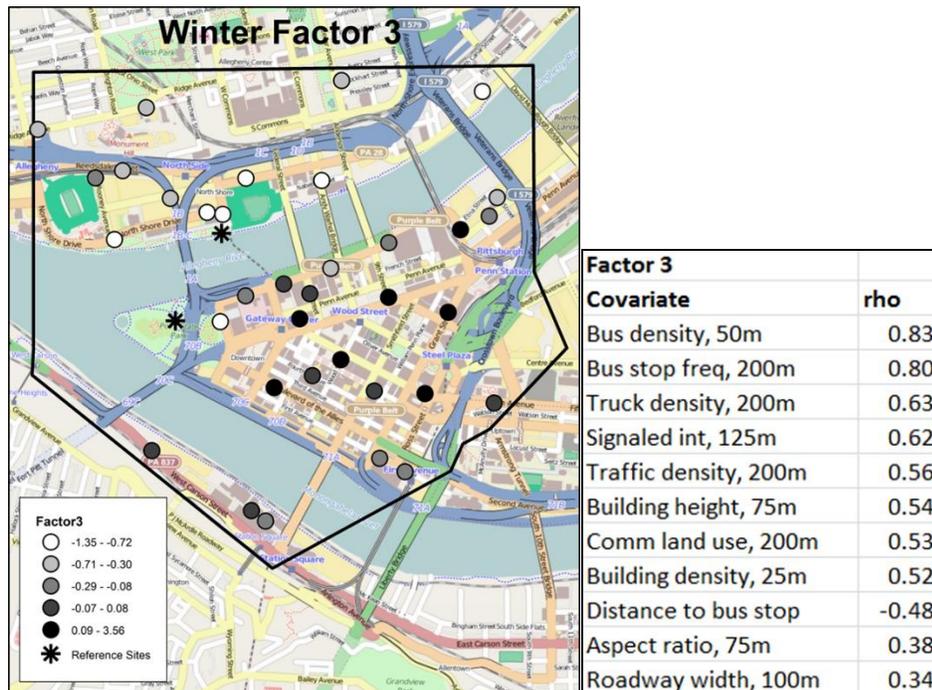


Figure 6. Spatial patterns and correlations with source indicators for Factor 3 (“diesel”) from the winter Year One factor analysis shown in Figure 5.



During winter, high scores for factor 3 (“diesel”) were clustered in the center of the downtown core, and were strongly correlated with several bus and traffic-related covariates (listed in Figure 6). LUR modeling identified bus density within 50 m and truck density within 200 m as significant predictors ( $R^2 = 0.75$ ).

For summer samples, a 9-factor solution explained 88% of variability in constituents. Factor 1 was characterized by *traffic-related elements and fuel oil* (Ni, V). Factor 2 suggested *diesel* sources (benzo[ghi]fluoranthene, chrysene, Cr, fluoranthene, Mn, pyrene, total carbon, total EC, Zn). Factor 3 also included *diesel-related constituents* (benz[a]anthracene, BC, hopanes, NO<sub>2</sub>, total PAHs, total steranes). Factor 4 was characterized by *brake/ tire wear* markers (Cu, Mo, Sb). Factor 5 included *benzene and norhopane*, whereas factor 6 included *benzo[a]pyrene and indeno[123-cd]pyrene* – all previously associated with diesel exhaust. Factor 7 included *toluene and OC*. Factor eight included *benzo[e]pyrene*. Factor nine included a *coal emissions* tracer (Se).

In addition to total carbon, both fluoranthene and pyrene were consistently associated with other diesel tracers, regardless of season or method – suggesting that these PAHs may be useful tracers for diesel-related activities and sources.

## RESULTS - Phase Two (Rush-hour vs. work-week concentrations):

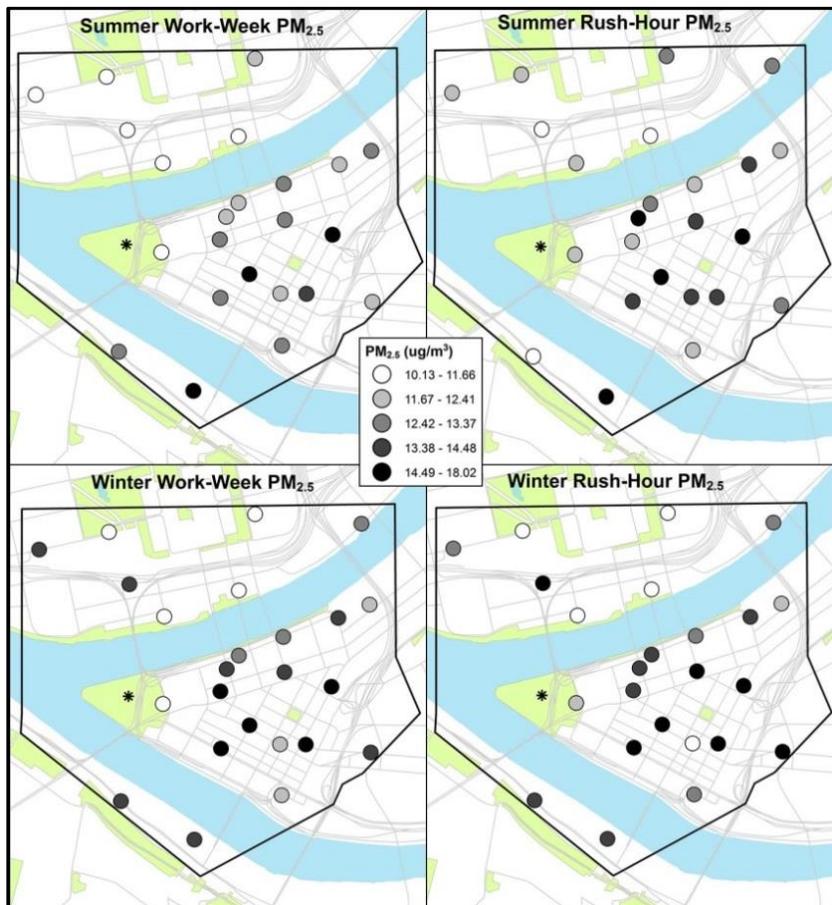
We found slightly higher mean  $PM_{2.5}$ , and significantly higher BC, in rush-hour samples, compared full work-week samples, for both summer and winter. Likewise, higher concentrations of traffic-related trace elements and diesel-related organics were identified in rush-hour samples.

EC concentrations were moderately higher for rush-hour sampling during summer, and significantly so in winter. For OC, concentrations were slightly higher during rush-hour sampling during summer, but not winter.

Many elemental concentrations were non-significantly higher for rush-hour sampling. During winter, we found higher concentrations of traffic and soil/ road dust tracers (i.e., copper, manganese, potassium, zinc) -- and higher concentrations of the diesel tracers benz[a]anthracene, benzo[e]pyrene, and chrysene -- for the rush-hour program, vs. the full work-week.

During summer, we found significantly higher concentrations of fluoranthene, pyrene, and total steranes for the rush-hour program, vs. the full work-week.

Figure 7. Spatial variability in measured  $PM_{2.5}$  concentrations.



### **LUR models – rush hour vs. work week:**

For most pollutants, spatial patterns were similar for both rush-hour and work-week samples (Figure 7), and both sets of LUR models explained proportions of variability in concentrations. Together, these observations suggest that most spatial variance in these pollutants may be attributable to diesel and vehicular source activity during rush hours.

Mean bus density was the strongest spatial predictor in most LURs, regardless of sampling interval. The spatial pattern in winter and summer PM<sub>2.5</sub> was predicted by mean bus density, for both work-week and rush-hour samples.

For winter BC, the LUR models for both sampling schemes included mean bus density; the summer BC models included mean bus density and mean truck density for both sampling schemes.

For winter and summer, the spatial pattern in EC was predicted by mean bus density for both sampling schemes. For winter OC, the model included bus stop use for both schemes, whereas the summer OC work-week model included commercial land use, and the rush-hour model included mean truck density.

For winter, the spatial pattern in total PAHs was predicted by mean bus density for both schemes. For summer total PAHs, mean bus density was the sole predictor for work-week sampling, whereas bus stop use explained spatial variance in rush-hour samples.

For total hopanes in winter, the model included mean bus density for the work-week sampling interval, but no source covariate for the rush-hour scheme. The summer total hopanes model included mean bus density for work-week samples, and mean truck density for rush-hour samples.

The rush-hour spatial pattern in Al, during summer, was predicted by mean truck density; no source covariate explained significant variability in work week samples, in either season.

For summer rush-hours, spatial variance in Ca was predicted by mean truck density; no source covariates significantly predicted spatial variance in work-week samples. In winter, rush-hour models for Ca included distance to nearest primary road, and work-hour models included mean bus density.

For Fe, during summer, the rush-hour model included mean truck density for rush-hour. The work week model included length of primary and secondary roadways. No source covariates were significant during winter.

### **CONCLUSIONS:**

We found substantial spatial variation in PM<sub>2.5</sub> and chemical components; for most pollutants, we found higher concentrations near the center of the downtown core. Notably, spatial variation was greater than seasonal variation for almost all pollutants measured, even across this very small downtown area (~2.8 km<sup>2</sup>).

In land use regression (LUR) models for PM<sub>2.5</sub>, BC, and EC, bus density was the strongest predictor of spatial variation, during both summer and winter -- indicating a strong impact of buses on pollutant concentrations.

For organic compounds, bus sources were identified in models for summer, but not winter. Though vehicle and truck traffic terms were also spatially correlated with several pollutants, buses remained the strongest predictor in most models.

Source apportionment analyses, merging all constituent concentrations, identified diesel-related factors in both seasons. During winter, a factor comprised of BC, fluoranthene, nitrogen dioxide (NO<sub>2</sub>), pyrene, and total carbon was predicted by bus and truck-related source terms. For summer, a factor including of multiple potential diesel tracers (benzo[ghi]fluoranthene, chromium, chrysene, fluoranthene, manganese, pyrene, total carbon, EC, zinc) was predicted by bus-related terms. A factor consisting of benz[a]anthracene, BC, hopanes, NO<sub>2</sub>, total PAHs, and total steranes was associated with truck density.

We found higher concentrations of most pollutants in rush-hour vs. full work-week samples. Similar patterns and comparable LUR models for both rush hour and work week measures in most pollutants suggests that spatial variance in concentrations for most of the pollutants examined may be explained by rush-hour source activity.

Finally, bus traffic and other bus-related terms were the most common spatial predictors in LUR models, regardless of season, sampling scheme, or analytic approach (e.g., LUR vs. source apportionment) -- suggesting that rush-hour diesel emissions may substantially influence overall spatial variance in pollution exposures.

## **DISCUSSION/ FUTURE DIRECTIONS:**

Our study uncovered several important features of patterning in diesel-related pollution concentrations in the Pittsburgh downtown core. Notably, we found substantial spatial variation in concentrations across this small area, which exceeded mean differences between seasons. We also found similar spatial patterns in both seasons, and for both rush-hour and full “work week” samples. A notable exception was in winter vs. summer organics: the former displayed relatively flat patterns, potentially due to meteorological differences or to seasonal differences in fuel composition, as these sources operate year-round.

We have already secured additional funding from the Mascaro Center for Sustainable Innovation at University of Pittsburgh (“Quantifying Reductions in Diesel-Related Air Pollution Exposures across Downtown Pittsburgh”) a pilot-scale study aimed at: (1) incorporating nitrogen isotope signatures into our analyses, to corroborate diesel sources identification. (Isotope measures were collected at all of our sampling sites with Dr. Emily Elliott, University of Pittsburgh Department of Geology and Environmental Sciences.), and (2) to perform a literature review to *identify successful interventions* effective elsewhere in removing bus and/ or vehicle-related exposures in

dense urban areas, then to work with ACHD and other local agencies to identify effective measures appropriate for Pittsburgh.

Nevertheless, patterns did differ for many pollutants – even for pollutants expected to indicate the same diesel-related sources – pointing to the possibility of disentangling the impacts of different types of diesel sources (e.g., buses vs. trucks) across the downtown area. Two key sources of interest we were unable to fully explore – due to current limitations in available source data – were *railroad and barge traffic*. Accessing data with reasonable spatial and temporal variance on these sources (e.g., number and timing of trips per day) may help to further disentangle their effects.

The Port Authority is currently in the process of upgrading the public bus fleet; by 2020, all Port Authority buses will be post-2007 technology or later. In 2017, the largest portion of this changeover is expected, thus *post-fleet changeover monitoring* at the same locations may elucidate benefits of this intervention.

We found comparable spatial patterns in rush- vs. work-week samples – raising the question of whether emissions during non-rush hours have the same spatial pattern as during rush hours, or were simply too low, in relative terms, to substantially change filter loadings and observed patterns. As we have now validated and refined these methods to accurately capture even very low concentrations of organic and elemental constituents, we may consider the utility of monitoring these locations *solely during non-rush hours*, to compare results.

*Street canyons* play an important role in pollutant dispersion and entrapment in urban cores. More explicit examination of entrapment and enrichment of bus-related emissions within downtown street canyons could be a valuable exploration.

Finally, incorporating source-specific *dispersion modeling* into these models, as we have successfully done in prior spatial models (e.g., Michanowicz et al., 2016a; Michanowicz et al., 2016b) may improve our ability to estimate impacts of sources both inside and outside of the sampling domain (e.g., Shenango Coke Works, outside the northern boundary of our sampling domain, was in operation throughout our sampling seasons).

## **PUBLICATIONS RESULTING FROM THIS WORK:**

Tunno BJ, Shmool JLC, Michanowicz D, Tripathy S, Chubb LG, Kinnee E, Cambal L, Roper C, Clougherty JE. Spatial and Seasonal Variation in Diesel-Related Elemental and Organic PM<sub>2.5</sub> Components across Downtown Pittsburgh. *Science of the Total Environment*. 2016; 573:27-38. doi: 10.1016/j.scitotenv.2016.08.011.

Tunno BJ, Michanowicz D, Shmool JLC, Tripathy S, Kinnee E, Chubb LG, Cambal L, Roper C, Clougherty JE. Spatial Patterns in Diesel-Related Pollutants during Rush Hours vs. Work-week Hours in a Downtown Core (*Manuscript in preparation*).

Tunno BJ, Tripathy S, Kinnee E, Clougherty JE. PM<sub>2.5</sub> Elemental and Organic Constituents Source Apportionment using a Super-Saturation Design across Downtown Pittsburgh (*Manuscript in preparation*).

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