# Exploring the Spatiotemporal Variation of Air Pollution Throughout the Urban Landscape of Philadelphia, PA with Mobile Monitoring

Lucas E Cummings<sup>1</sup>, Justin Stewart<sup>1</sup>, Radley Reist<sup>1</sup>, Kabindra M Shakya<sup>1</sup>, and Peleg Kremer<sup>1</sup>

<sup>1</sup>Villanova University

November 24, 2022

#### Abstract

In this study, we implement a mobile monitoring methodology in order to determine the spatiotemporal distribution of particulate matter (PM) and black carbon (BC) in Philadelphia, PA, USA. Over the course of 12 days between June 27, 2019 and July 29, 2019, we measured air pollution concentrations across two replicated 150-mile long routes. Mean concentrations for each pollutant were  $11.25 \pm 5.43$  ug/m3 (PM1),  $11.08 \pm 6.25$  ug/m3 (PM2.5),  $15.57 \pm 8.51$  ug/m3 (PM10), and  $1.27 \pm 0.80$  µg/m3 (BC). We find that finer PM size fractions (PM2.5 and smaller) constitute approximately 71% of PM10. Air pollution hotspots across three size fractions of PM (PM1, PM2.5, and PM10) and BC were present throughout Philadelphia, but were most prevalent in the North Delaware, River Wards, and North planning districts. A plurality of air pollution hotspots found throughout the data collection period (30.19%) occurred between the hours of 8:00 AM – 9:00 AM. Despite significant temporal variation, pollutant concentrations, except for PM10, clustered temporally with a separation before 12 PM. Our approach and findings identify times and places where pollutant concentrations are highest, which is integral to effective air pollution reduction in urban environments.

#### Hosted file

essoar.10503885.1.docx available at https://authorea.com/users/547754/articles/602861-exploring-the-spatiotemporal-variation-of-air-pollution-throughout-the-urban-landscape-of-philadelphia-pa-with-mobile-monitoring

1 Exploring the Spatiotemporal Variation of Air Pollution Throughout the Urban Landscape of

# 2 Philadelphia, PA with Mobile Monitoring

- 3 Lucas E. Cummings<sup>\*1</sup>, Justin D. Stewart<sup>\*1,2</sup>, Radley Reist<sup>1</sup>, Kabindra M. Shakya<sup>1\*\*</sup>, Peleg Kremer<sup>1</sup>
- 4 \*These authors contributed to this work equally
- <sup>5</sup> <sup>1</sup>Department of Geography and the Environment, Villanova University, Pennsylvania, USA
- 6 <sup>2</sup>Department of Ecological Science, Vrije Universiteit Amsterdam, 1081 HV Amsterdam, Netherlands

## 7 **\*\*Corresponding author**

- 8 800 Lancaster Avenue, Villanova, PA 19085, USA
- 9 Department of Geography & the Environment
- 10 Villanova University
- 11 Phone: 610-519-3590
- 12 Email: <u>kabindra.shakya@villanova.edu</u>

## 13 Author Contributions:

- 14 Conceptualization: KMS, PK. Data Collection: JDS, RR, KMS, PK. Formal Data Analysis: JDS, LEC,
- 15 RR. Writing: LEC, JDS, RR, KMS, PK. Supervision & Funding Acquisition: KMS, PK
- 16 **ORCID:**
- 17 LEC: 0000-0002-7685-0525
- 18 JDS: 0000-0002-7812-5095
- 19 PK: 0000-0001-6844-5557
- 20 KMS: 0000-0002-7035-7019
- 21 Sources of Support: NSF Grant # 1832407
- 22 Word count: 5755

#### 23 Abstract

In this study, we implement a mobile monitoring methodology in order to determine 24 the spatiotemporal distribution of particulate matter (PM) and black carbon (BC) in 25 26 Philadelphia, PA, USA. Over the course of 12 days between June 27, 2019 and July 29, 2019, we 27 measured air pollution concentrations across two replicated 150-mile long routes. Mean concentrations for each pollutant were  $11.25 \pm 5.43 \text{ ug/m}^3$  (PM<sub>1</sub>),  $11.08 \pm 6.25 \text{ ug/m}^3$  (PM<sub>2.5</sub>), 28 15.57 ± 8.51 ug/m<sup>3</sup> (PM<sub>10</sub>), and 1.27 ± 0.80  $\mu$ g/m<sup>3</sup> (BC). We find that finer PM size fractions 29 (PM<sub>2.5</sub> and smaller) constitute approximately 71% of PM<sub>10</sub>. Air pollution hotspots across three 30 size fractions of PM (PM<sub>1</sub>, PM<sub>2.5</sub>, and PM<sub>10</sub>) and BC were present throughout Philadelphia, but 31 32 were most prevalent in the North Delaware, River Wards, and North planning districts. A plurality of air pollution hotspots found throughout the data collection period (30.19%) 33 occurred between the hours of 8:00 AM - 9:00 AM. Despite significant temporal variation, 34 pollutant concentrations, except for PM<sub>10</sub>, clustered temporally with a separation before 12 35 PM. Our approach and findings identify times and places where pollutant concentrations are 36 highest, which is integral to effective air pollution reduction in urban environments. 37



38 39

#### 40 Introduction

Air pollution is a major environmental threat for urban populations, affecting the health 41 of 9 out of 10 urban residents<sup>1</sup>. Within urban environments, locally high concentrations of air 42 43 pollutants are common<sup>2</sup>. As populations continue to migrate to urban areas<sup>3</sup>, we can expect air 44 pollution to continue to present health risks for human populations. In order to attenuate negative health impacts<sup>1</sup> of air pollution in the future, it is imperative that we are able 45 accurately assess the spatiotemporal distribution of air pollution in urban environments. 46 Comprehensive air pollution monitoring is crucial to understanding where and how to focus 47 48 efforts to attenuate air pollution and its associated health risks in the urban environment.

49 Particulate matter (PM) consists of heterogeneous mixtures of organic<sup>4</sup> and inorganic components<sup>5</sup> that vary in size, shape, composition, and origin within the urban environment<sup>6</sup>. 50 Coarse size fractions (PM<sub>10</sub> - PM<sub>2.5</sub>) of PM largely originate from crustal sources, whereas fine 51 PM (PM<sub>2.5</sub> - PM<sub>0.1</sub>) derive mainly from industrial emissions, non-renewable power generation, 52 and vehicle exhausts<sup>7</sup>. Black carbon (BC) is a major component of PM that results from the 53 incomplete combustion of fossil fuel and other organic matter. As such, the presence of BC is 54 often used as an indicator of urban traffic pollution<sup>8</sup>. Quantifying the abundance and 55 56 distribution of various PM sizes in urban environments is of particular interest to public health<sup>9</sup>, as prolonged exposure to PM is associated with increased rates of mortality<sup>10</sup>; small particles 57 58 easily deposit in the lungs<sup>11</sup>, leading to a number of observed negative health outcomes including reduced lung function<sup>12</sup>, asthma<sup>13</sup>, cardiovascular and respiratory disease<sup>14</sup>, and 59 60 pathogen exposure<sup>15</sup>.

61 Many studies have investigated urban air quality, but these studies tend to rely on a small number of stationary points of measurement<sup>16,17</sup> and interpolation<sup>18,19</sup> to characterize air 62 63 pollution across an entire city. While these methods are effective at capturing temporal trends 64 in local pollutant concentrations, they are unable to capture fine-scale spatial variation in air 65 pollution throughout urban environments. In recent years, mobile monitoring has emerged as a 66 novel method with which to study the spatial and temporal distribution of air pollutants<sup>6,20-22</sup>. As mobile monitoring methods are capable of collecting data at finer spatial scales than is 67 feasible with stationary monitoring<sup>6,23</sup>, mobile monitoring can provide more accurate 68 information about air quality within the city. In this study, we employ vehicular mobile 69 70 monitoring of PM across 24 different size fractions between 10 – 0.25  $\mu$ m and BC throughout the urban landscape of Philadelphia, Pennsylvania and identify statistically significant hotspots 71 72 of air pollution in time and space during the summer of 2019. With mobile monitoring, we can 73 observe the spatiotemporal distribution of air pollutants and discern patterns in variation at a 74 fine spatial scale. By doing so, it is possible to identify locations in urban environments where high concentrations of air pollutants are common<sup>24</sup> and provide more holistic assessments of 75 76 risks associated with air pollution in urban areas.

77

#### 78 Methods

79 Site Description:

Philadelphia, Pennsylvania, USA had an estimated population of 1,584,138 in 2018. Located in the Mid-Atlantic region, Philadelphia is dominated by a dense urban center surrounded by predominantly low-rise residential and commercial districts, city parks, and

83 industrial sectors. The city's eastern border is defined by the Delaware River, which flows southward to the Delaware Bay and Atlantic Ocean, while the city's other major river, the 84 Schuylkill River, flows southward to the Delaware through the western neighborhoods of 85 Philadelphia. The southern and eastern parts of the city house heavy industry along both 86 riverbanks (Planning Districts Lower Southwest, Lower South, and River Wards), while large 87 park areas are found in the western and northern areas of the city (Planning Districts Lower 88 Northwest, Upper Northwest, and Central Northeast) (Figure 1). Philadelphia consistently ranks 89 as one of the most polluted metropolitan areas in the United States<sup>14</sup>. 90



91

Figure 1. Map of study area, including routes traveled and Philadelphia planning districts.

94 Sampling Description:

A driving route was developed using a stratified random selection of points representing 95 different combinations of urban structure to provide a representative sample of Philadelphia 96 97 for mobile monitoring. Additionally, selected points of interest, such as industrial sites, United 98 States Environmental Protection Agency (U.S. EPA) Toxics Release Inventory (TRI) sites, and EPA 99 air pollution monitoring station sites, were included in route development. The optimized driving route, which passed through the selected sample points, was created using ESRI ArcGIS 100 10.7.1 Network Analyst, and the resulting ~300 mile route was then split into two near-equal 101 102 segments of approximately 150 miles each, with each segment being drivable in a single day.

103 A van, equipped with two global positioning system (GPS) units (Trimble Juno 3B fitted with Trimble R1 GNSS receivers) and instrumentation measuring PM (GRIMM Portable Laser 104 Aerosol Spectrometer, Model 11-C) and BC (MicroAeth MA200), was driven along the two 105 predetermined routes in Philadelphia. The GRIMM spectrometer was factory calibrated prior to 106 the monitoring campaign. Air pollution instrumentation was placed inside a box attached to the 107 roof of a van (~1.5 meters), and the inlets of the instrumentation were connected to an 108 109 isokinetic sampling probe of diameter 1.5 mm. Measurements were conducted over a period of 12 day between June 27, 2019 and July 29, 2019. Measurement would begin between the 110 111 hours of 6:00 AM and 7:00 AM on one of the two routes and continued until the entirety of the 112 route was travelled. In order to maintain continuous measurements in the face of satellite reception issues and equipment malfunction, the two GPS units were used simultaneously. 113 114 Occasional road closures in Philadelphia created slight variability in the routes traveled from 115 day to day (SI-1). Data was captured at different temporal resolutions; GPS data was recorded

116 for every one second interval, while BC data was recorded every five seconds and PM data was117 recorded every six seconds.

118

119 Data Processing/Analysis:

Air pollution and GPS data were joined by time to create a database of geolocated air pollution data. Histograms and quantile-quantile plots were used to remove the top and bottom 0.5% of air pollution measurements. Pollution data lacking geolocation information due to instrument error was not considered for spatial analysis in this paper. One day (July 15, 2019) is entirely excluded from spatial analysis as a result of GPS malfunction that resulted in a significant amount of missing geolocation data. Vector (point) datasets representing air pollution along the routes were created and used for representation and spatial analysis.

127 Spatial analysis was conducted in ESRI ArcGIS Pro 2.4. Point datasets were projected 128 into the Pennsylvania State Plane South projected coordinate system. Air pollution data was spatially joined to a systematic grid of 120 m<sup>2</sup> fishnet cells overlaid on Philadelphia, which has 129 previously been used to generalize and characterize urban landscape and ecosystem 130 function<sup>6,25,26</sup>. All points falling within a given cell were averaged to determine the average 131 132 concentration of pollutants in that 120 m<sup>2</sup> area. For each day of data collection, PM<sub>1</sub>, PM<sub>2.5</sub>, PM<sub>10</sub>, and BC hotspots with statistical significance at a 95% confidence level were identified 133 using the Hot Spot Analysis (Getis-Ord Gi<sup>\*</sup>) tool. Hotspot analysis allows for the identification of 134 135 statistically significant locations in a study area where features with high or low values cluster within the context of its neighborhood<sup>27</sup>. The neighborhood threshold radius for all hotspot 136 137 analyses was set at the minimum distance to ensure that for each day, observations for all

pollutants measured had at least one other feature designated as a neighbor (615 m). The 138 inverse distance squared conceptualization of spatial relationships was used for this analysis, 139 which sees the influence of an observation on its spatial neighbors decrease significantly with 140 141 increasing distance. False discovery rate correction was applied to correct for false positives. In 142 order to compare the locations of hotspots across the days of data collection, significant 143 hotspots (p < 0.05) for each day were spatially joined with the fishnet grid. Hot spots within a given cell were averaged to determine the mean pollution concentration of the hotspots in 144 each cell for each day. Data for PM and BC concentrations and GPS coordinates for hotspots 145 146 can be accessed at <u>https://github.com/Shakya-Kremer-Lab/AirPollution</u>.

147 Statistical analysis was conducted in R (3.6.1). Combination violin and boxplots were produced to show the range and distribution of air pollutants across all days. Pairwise Mann-148 149 Whitney tests with Bonferroni correction on mass concentrations for the PM<sub>1</sub>, PM<sub>2.5</sub>, PM<sub>10</sub> size fractions - chosen as representatives of the fine-to-coarse PM size fraction gradient - and BC 150 were conducted to test if pollution levels differed between days. The relationship between BC 151 and PM<sub>2.5</sub> was tested at the univariate level with Bonferroni corrected Spearman correlations. 152 Multivariate relationships between BC and PM<sub>2.5</sub> were assessed using permutational (n=999) 153 Procrustes rotations. This test compares a collection of multidimensional shapes by 154 transforming them into a state of maximal superimposition and resulting in a correlation 155 156 coefficient, m<sup>2</sup>.

157 Temporal variation in mass values for PM across size fractions during core times (where 158 data overlaps on all days) was visualized using heatmaps on a log<sub>10</sub> scale at 4-minute intervals. 159 Heatmaps were annotated with PM<sub>2.5</sub> hotspots that cover times greater than a 2-minute period.

- 160 K-means clustering was employed to identify clusters of air pollution. The number of clusters (2
- 161 for all pollutants) were identified through a variance-by-number-of-cluster plots, where a bend
- 162 in the plot indicate that a suitable number of clusters are defined to explain the data.

## 163 Results and Discussion



Figure 2. (A) Heatmap showing the log<sub>10</sub> concentration of BC across time (x-axis) and different days (y-axis). The black line reflects the
log<sub>10</sub> mean concentration of BC averaged across all days. (B) Procrustes rotation ordination of correlation between BC and PM<sub>2.5</sub> on
all days with correlation coefficient (m<sup>2</sup>) and a p-value. (C) Plot of log<sub>10</sub> mean BC concentration over time, colored by cluster,
determined by k-means clustering. (D) Plot of log<sub>10</sub> mean PM<sub>2.5</sub> concentration over time, colored by cluster, determined by k-means
clustering. (E) Plot of log<sub>10</sub> mean PM<sub>1</sub> concentration over time, colored by cluster, determined by k-means clustering. (F) Plot of log<sub>10</sub>

171 Observed PM and BC concentrations had roughly Poisson distributions, (Figure 3A-D) as 172 lower concentrations were observed much more frequently than higher concentrations. The mean concentrations observed throughout the measurement period were  $11.25 \pm 5.43$  ug/m<sup>3</sup> 173 174 for PM<sub>1</sub>, 11.08 ± 6.25 ug/m<sup>3</sup> for PM<sub>2.5</sub>, 15.57 ± 8.51 ug/m<sup>3</sup> for PM<sub>10</sub>, and 1.27 ± 0.80  $\mu$ g/m<sup>3</sup> for 175 BC. The mean PM<sub>2.5</sub> concentration over the sampling period is slightly greater than, but not significantly different from, the 2018 annual mean PM<sub>2.5</sub> concentrations found at seven EPA 176 PM<sub>2.5</sub> monitoring stations in Philadelphia, which ranged from 8.0 – 9.8 μg/m<sup>3 28</sup>. Variability in 177 PM<sub>10</sub> concentrations seems to be largely influenced by variation in finer PM concentrations; we 178 179 find that PM<sub>2.5</sub> comprises approximately 71% of the observed PM<sub>10</sub> in Philadelphia. These 180 results are similar to the findings of a previous study on air pollution in Philadelphia, where  $PM_{2.5}$  made up 75% of the  $PM_{10}$  in the city<sup>29</sup>. Overall, BC accounts for 11.4% of the observed 181  $PM_{2.5}$  in Philadelphia. The ratio of BC /  $PM_{2.5}$  in Philadelphia is comparable to the BC /  $PM_{2.5}$ 182 ratios of other large cities, which range from 5% - 20%<sup>30</sup>. Pairwise comparison of BC and PM<sub>25</sub> 183 concentrations revealed that the relationship between the two pollutants was generally 184 variable from day to day (Table S4). BC was strongly correlated with PM<sub>2.5</sub> concentrations at the 185 186 multivariate level when taking into account their relationships across all days (Figure 3B, 187 Procrustes,  $m^2 = 0.9249$ , p = 0.043), and significant (p < 0.05) positive correlations between BC and PM<sub>2.5</sub> were observed on 10 of the 12 days of data collection (Figure S2). Among days where 188 189 we found a significant correlation between PM<sub>2.5</sub> and BC, weak to moderate relationships were observed (Figure SI-2, Spearman's p: 0.215-0.616); variation in this relationship from day to day 190 191 is due in part to the heterogeneity of emission sources and the urban landscape<sup>23</sup>. The high

- $PM_{2.5}$  /  $PM_{10}$  ratio and abundance of BC in Philadelphia are indicative of the significant impact
- 193 that traffic-related emissions have on air pollution concentrations throughout Philadelphia<sup>31-33</sup>.

## 194 Spatial Variation



195

Figure 3. Violin plots and maps for (A) PM<sub>1</sub>, (B) PM<sub>2.5</sub>, (C) PM<sub>10</sub>, and (D) BC. Violin plots show the distribution of all observed air pollutant concentrations on a log<sub>10</sub> scale, while maps show the overall average concentration of each pollutant in each 120 m<sup>2</sup> cell sampled over the data collection period.

We found that PM<sub>1</sub>, PM<sub>2.5</sub>, and PM<sub>10</sub> have a similar spatial distribution throughout 200 Philadelphia (Figure 3). Similarities between the  $PM_{10}$  and the  $PM_{2.5}$  and  $PM_1$  distributions 201 202 indicate that finer PM constitutes a significant proportion of Philadelphia's PM and thus drives a 203 majority of the variation in PM<sub>10</sub> concentrations. The lowest concentrations of PM and BC in Philadelphia were generally found in Philadelphia's Lower North (LNO), West Park (WP), and 204 205 West (W) planning zones. The highest concentrations of PM across all size fractions were found 206 in Philadelphia's North Delaware (NDEL), River Wards (RW), and North (NOR) planning zones. RW contains a port and large public utility properties, as well as other industries, which may be 207 208 significant sources of PM in this area<sup>34</sup>. Interestingly, BC concentrations vary considerably in these planning zones, which suggests that traffic-related emissions do not contribute as much 209 210 to the ambient air pollution in these particular areas relative to other sources. BC concentrations are highest in the RW, Lower Far Northeast (LFNR), and Upper Far Northeast 211 (UFNE) planning zones. It is possible that increased traffic-related emissions resulting from the 212 213 proximity of these zones to Interstate 95 may be the cause of high BC concentrations in this 214 region. The Northeast Philadelphia Airport in RFNE, where BC concentrations are elevated, may 215 also contribute significantly to BC emissions; landings and takeoffs by aircrafts at airports, have been shown to increase local BC concentrations in the atmosphere<sup>35</sup>. 216

Statistically significant hotspots were found on all days across all measured size fractions of PM and BC throughout Philadelphia. The average concentrations of hotspots (Figure 4) within a given cell ranged from  $8.7 \pm 4.6 \ \mu g/m^3$  for BC;  $18.7 \pm 7.1 \ \mu g/m^3$  for PM<sub>1</sub>;  $28.0 \pm 8.8 \ \mu g/m^3$  for PM<sub>2.5</sub>; and  $46.0 \pm 17.3 \ \mu g/m^3$  for PM<sub>10</sub>. While there is slight variation in the location of hotspots among the different pollutants, the overall spatial distribution of hotspots throughout 222 Philadelphia is similar across all PM size fractions and BC (Figure 4). Relatively few hotspots were found in northern and western Philadelphia, which are located well outside of 223 224 Philadelphia's urban core. In these areas, traffic-related emissions are likely not as prominent as 225 in the urban core, and a greater abundance of vegetation may attenuate air pollution primarily by uptake via leaf stomata and particle deposition<sup>36,37</sup>. Each pollutant had hotspots that 226 exhibited a tendency to recur in the same locations across multiple days; hotspots for PM<sub>1</sub> 227 appeared in the same cell on as many as six separate days throughout the data collection 228 period, while hotspots for PM<sub>2.5</sub>, PM<sub>10</sub>, and BC appeared in the same cell on up to five different 229 days. The recurrence of hotspots in specific locations suggests that there are areas in 230 231 Philadelphia where pollutant concentrations are constantly elevated relative to the surrounding area. A notable cluster of cells in the NDEL, RW, and NOR planning zones contain high 232 concentration PM hotspots across multiple days. Other clusters of recurring hotspots are found 233 within the University Southwest (USW) and LFNE planning zones. 234



Figure 4. Maps displaying the locations and average concentrations of hotspots for (A) PM<sub>1</sub>, (B) PM<sub>2.5</sub>, (C) PM<sub>10</sub>, and (D) BC in Philadelphia.

## 240 Temporal Variation

241



Figure 4. Heatmap of log<sub>10</sub> PM mass values across 24 fine and coarse size fractions throughout
 each day of data collection. Time of day is denoted on the x-axis. Hotspots for PM<sub>2.5</sub> fraction
 covering > 2 minute periods are identified by vertical red boxes. July 15, 2019 was excluded
 from hotspot analysis; as such, no hotspots are identified.

247 PM emissions were not uniform across all size fractions measured. All observations 248 indicate the presence of particulate matter of 5  $\mu$ m in diameter or smaller. PM exceeding 5  $\mu$ m 249 in diameter is not as ubiquitous throughout the data collection period, with larger particles not 250 being detected at times throughout each day. Mass values observed for particles 1.6  $\mu$ m 251 diameter and larger generally demonstrated the greatest variation throughout each day, with 252 particles with a diameter 0.5  $\mu$ m and smaller also showing less within-day variation (Figure 4). 253 Trends emerged despite significant temporal variation in the concentration and distribution of  $PM_{1}$ ,  $PM_{2.5}$ , and  $PM_{10}$  (p < 0.05, Tables SI-1 – SI-3). The  $PM_{2.5}$  size fraction (Figure 2D) clustered 254 255 into two distinct time periods separated at approximately 11:08 AM, which complements our 256 finding of BC clusters at approximately 10:56 AM (Figure 2C). While this relationship is expected due to BC largely contributing to  $PM_{2.5}$  composition in urban areas from vehicles<sup>38</sup>, larger ( $PM_{10}$ ) 257 and smaller size (PM<sub>4</sub>) fractions varied in their separation of peaks by time. PM<sub>4</sub> displayed less 258 259 discrete temporal clustering (Figure 2E), with a break in clustering at approximately 10:08 AM. A cutoff was not found for PM<sub>10</sub>. The lack of temporal clustering for PM<sub>10</sub>, as seen in a previous 260 261 mobile monitoring study<sup>39</sup>, affirms that larger particulate matter emission is stochastic across the urban landscape, and may be attributed to crustal sources (e.g. dust resuspension). These 262 263 results complement findings in other mobile monitoring studies where PM size fractions exhibit different concentrations in the morning and afternoon<sup>40</sup>. 264

The number, duration, and timing of  $PM_{25}$  hotspots (Figure 4) varied from day to day; 265 however, they were most consistently seen from 8:00 – 9:00 AM (30.19% of all hotspots) in 266 complement with other studies<sup>41,42</sup>. These hotspots are likely attributed to primary particles 267 268 emitted from morning rush-hour traffic, where the number and density of vehicles on the road is high relative to the rest of the day. The presence of hotspots outside the morning trend may 269 270 be attributed to areas closer to industrial sites, such as those located along the I-676 and I-95 corridors<sup>43</sup>. While a substantial fraction of the PM<sub>2.5</sub> in urban areas originates from combustion 271 272 engines, increased solar radiation during the summer months likely enhances the contribution of secondary particles formed from photooxidation of precursor molecules<sup>44</sup>. 273

274 In this study, we demonstrate the potential for a mobile monitoring approach to 275 examine fine-scale spatiotemporal distribution of air pollutants in a major city. Our findings 276 demonstrate the variability of PM and BC concentrations in space and time and indicate that 277 trends in variation are dependent on the size and type of pollutant. Our analysis is limited by 278 relatively few repetitions of routes and variability in the accuracy of geolocation data. As our 279 sampling occurs entirely on Philadelphia roadways, it should be noted that our measurements may be slightly different relative to ambient air further from roads. While urban air pollution 280 near roadways tends to be higher due to the influence of traffic-related emissions<sup>45-47</sup>, a 2017 281 282 study of air pollution along pedestrian walkways in Philadelphia observed higher average 283 pollutant concentrations than those observed in this study<sup>6</sup>. More extensive sampling would allow for additional confidence in observed trends and provide opportunities to observe air 284 285 pollution patterns at other temporal scales; sampling during late afternoon and evening hours would provide additional insight into air pollution trends throughout the day, while increased 286 287 repetition of measurements both within and across seasons would allow for a seasonal analysis of air pollution trends<sup>48</sup>. Hot spot analysis reveals regions in Philadelphia that merit further 288 289 study, especially in the context of vulnerable and socioeconomically disadvantaged populations 290 that may be disproportionately impacted by air pollution.

Though our analysis reveals spatiotemporal variation in PM and BC and possible causes of this variation, it stops short of estimating the contributions of specific sources to variability; as our air pollution measurements covary in space and time, it is difficult to quantify the extent of variation resulting from spatial influences (locations of point sources, movement of nonpoint sources) and temporal influences (temporally-sensitive atmospheric processes, random

events) separately. Future analyses should focus on the influence of urban structure on air pollution. Cities can be quite different from one another compositionally and structurally, and the roles of urban structure<sup>49</sup> and land use<sup>6,50</sup> may be important drivers of variation in urban air pollution. This analysis can be used to help identify general times and places across urban environments where air pollution may have the greatest adverse impacts on human and environmental health, which is paramount to effective air pollution mitigation and reduction of negative health impacts associated with air pollution.

303

## 304 Acknowledgements:

305 We would like to thank Meghan Conway and Alexander Saad for their assistance in data 306 collection. Financial support for this study was provided through National Science Foundation 307 (NSF) grant #1832407.

308

#### 309 Conflicts of Interest:

310 The authors declare no conflicts of interest.

311

#### 312 Supporting Information:

- 313 Supporting Information includes:
- Maps showing variation in the route travels (Figure SI-1)
- Spearman correlation tests of BC and PM<sub>2.5</sub> (Figure SI-2)
- Pairwise Mann-Whitney U-tests comparing pollutant observations between days (Tables

317 SI-1 - 4)

## 318 References:

319 (1) Pulmonary Health Effects of Air Pollution 320 https://www.ncbi.nlm.nih.gov/pmc/articles/PMC4776742/ (accessed Jun 29, 2020). 321 (2) Strosnider, H.; Kennedy, C.; Monti, M.; Yip, F. Rural and Urban Differences in Air Quality, 2008-322 2012, and Community Drinking Water Quality, 2010-2015 - United States. MMWR Surveillance 323 Summaries 2017, 66 (13), 2010–2015. https://doi.org/10.15585/mmwr.ss6613a1. 324 (3) World Population Projected to Reach 9.7 Billion by 2050 | UN DESA | United Nations Department 325 of Economic and Social Affairs. 326 (4) Tsapakis, M.; Lagoudaki, E.; Stephanou, E. G.; Kavouras, I. G.; Koutrakis, P.; Oyola, P.; von Baer, D. 327 The Composition and Sources of PM2.5 Organic Aerosol in Two Urban Areas of Chile. 328 Atmospheric Environment 2002, 36 (23), 3851–3863. https://doi.org/10.1016/S1352-329 2310(02)00269-8. 330 (5) Kelly, F. J.; Fussell, J. C. Size, Source and Chemical Composition as Determinants of Toxicity 331 Attributable to Ambient Particulate Matter. Atmospheric Environment 2012, 60, 504-526. https:// 332 doi.org/10.1016/j.atmosenv.2012.06.039. 333 (6) Shakya, K. M.; Kremer, P.; Henderson, K.; McMahon, M.; Peltier, R. E.; Bromberg, S.; Stewart, J. 334 Mobile Monitoring of Air and Noise Pollution in Philadelphia Neighborhoods during Summer 335 2017. Environ. Pollut. 2019, 255 (Pt 1), 113195-113195. 336 https://doi.org/10.1016/j.envpol.2019.113195. 337 (7) US EPA, O. Report on the Environment (ROE); Collections and Lists; 2015. (8) 338 Hotspots of black carbon and PM2.5 in an urban area and relationships to traffic characteristics -339 ScienceDirect https://www.sciencedirect.com/science/article/pii/S0269749116305978 (accessed 340 Jun 29, 2020). (9) 341 Dominici, F.; Peng, R. D.; Bell, M. L.; Pham, L.; McDermott, A.; Zeger, S. L.; Samet, J. M. Fine 342 Particulate Air Pollution and Hospital Admission for Cardiovascular and Respiratory Diseases. 343 JAMA 2006, 295 (10), 1127-1134. https://doi.org/10.1001/jama.295.10.1127. 344 (10) Dockery, D. W.; Pope, C. A.; Xu, X.; Spengler, J. D.; Ware, J. H.; Fay, M. E.; Ferris, B. G.; Speizer, F. 345 E. An Association between Air Pollution and Mortality in Six U.S. Cities. New England Journal of 346 Medicine 1993, 329 (24), 1753-1759. https://doi.org/10.1056/NEJM199312093292401. 347 (11)Miller, F. J.; Gardner, D. E.; Graham, J. A.; Jr, R. E. L.; Wilson, W. E.; Bachmann, J. D. Size 348 Considerations for Establishing a Standard for Inhalable Particles. Journal of the Air Pollution 349 Control Association 1979, 29 (6), 610-615. https://doi.org/10.1080/00022470.1979.10470831. 350 Shakya, K. M.; Rupakheti, M.; Aryal, K.; Peltier, R. E. Respiratory Effects of High Levels of (12) 351 Particulate Exposure in a Cohort of Traffic Police in Kathmandu, Nepal: Journal of Occupational 352 and Environmental Medicine **2016**, 58 (6), e218-e225. 353 https://doi.org/10.1097/JOM.000000000000753. 354 (13) Rabinovitch, N.; Strand, M.; Gelfand, E. W. Particulate Levels Are Associated with Early Asthma 355 Worsening in Children with Persistent Disease. Am. J. Respir. Crit. Care Med. 2006, 173 (10), 356 1098-1105. https://doi.org/10.1164/rccm.200509-1393OC. 357 (14) Paul, G.; Nolen, J. E.; Alexander, L.; Bender, L. K.; Vleet, V.; Barrett, W.; Jump, Z.; Rappaport, S.; 358 Samet, J. M.; Ballentine, N.; Nimirowski, T.; Innocenzi, L.; Wojs, V.; Lavelle, L.; Clark, C.; Fitzgerald, 359 J.; Eyer, A.; Lacina, K.; Macmunn, A.; Tubbs, G.; Meyer, E.; Albiero, M.; Montague, S.; Finstad, C.; 360 Boucher, L.; Martin, V.; Consulting, B. R.; Designs, O. State of the Air 2019. 2019. 361 (15) Cao, C.; Jiang, W.; Wang, B.; Fang, J.; Lang, J.; Tian, G.; Jiang, J.; Zhu, T. F. Inhalable 362 Microorganisms in Beijing's PM 2.5 and PM 10 Pollutants during a Severe Smog Event. Appl. 363 Environ. Microbiol. 2014. https://doi.org/10.1021/es4048472.

- 364 (16) Zhang, Y.-L.; Cao, F. Fine Particulate Matter (PM 2.5) in China at a City Level. Scientific Reports
   365 2015, 5 (1), 14884. https://doi.org/10.1038/srep14884.
- 366 (17) Vallius, M.; Janssen, N. A. H.; Heinrich, J.; Hoek, G.; Ruuskanen, J.; Cyrys, J.; Van Grieken, R.; de
  367 Hartog, J. J.; Kreyling, W. G.; Pekkanen, J. Sources and Elemental Composition of Ambient PM2.5
  368 in Three European Cities. *Science of The Total Environment* 2005, *337* (1), 147–162.
  369 https://doi.org/10.1016/j.scitotenv.2004.06.018.
- 370 (18) Zhang, A.; Qi, Q.; Jiang, L.; Zhou, F.; Wang, J. Population Exposure to PM2.5 in the Urban Area of
  371 Beijing. *PLOS ONE* 2013, 8 (5), e63486. https://doi.org/10.1371/journal.pone.0063486.
- 372 (19) A population exposure model for particulate matter: case study results for PM 2.5 in Philadelphia,
   373 PA | Journal of Exposure Science & Environmental Epidemiology
- https://www.nature.com/articles/7500188 (accessed Jul 24, 2020).
- (20) Van Poppel, M.; Peters, J.; Bleux, N. Methodology for Setup and Data Processing of Mobile Air
   Quality Measurements to Assess the Spatial Variability of Concentrations in Urban Environments.
   Environmental Pollution 2013, 183, 224–233. https://doi.org/10.1016/j.envpol.2013.02.020.
- 378 (21) Deville Cavellin, L.; Weichenthal, S.; Tack, R.; Ragettli, M. S.; Smargiassi, A.; Hatzopoulou, M.
  379 Investigating the Use Of Portable Air Pollution Sensors to Capture the Spatial Variability Of Traffic380 Related Air Pollution. *Environ. Sci. Technol.* 2016, 50 (1), 313–320.
- 381 https://doi.org/10.1021/acs.est.5b04235.
- Targino, A. C.; Rodrigues, M. V. C.; Krecl, P.; Cipoli, Y. A.; Ribeiro, J. P. M. Commuter Exposure to
  Black Carbon Particles on Diesel Buses, on Bicycles and on Foot: A Case Study in a Brazilian City.
  Environmental Science and Pollution Research 2018, 25 (2), 1132–1146.
  https://doi.org/10.1007/s11356-017-0517-x.
- 386 (23) Van den Bossche, J.; Peters, J.; Verwaeren, J.; Botteldooren, D.; Theunis, J.; De Baets, B. Mobile
  387 Monitoring for Mapping Spatial Variation in Urban Air Quality: Development and Validation of a
  388 Methodology Based on an Extensive Dataset. Atmospheric Environment 2015, 105, 148–161.
  389 https://doi.org/10.1016/j.atmosenv.2015.01.017.
- 390 (24) Gozzi, F.; Della Ventura, G.; Marcelli, A. Mobile Monitoring of Particulate Matter: State of Art and
  391 Perspectives. Atmospheric Pollution Research 2016, 7 (2), 228–234.
  392 https://doi.org/10.1016/j.apr.2015.09.007.
- 393 (25) Hamstead, Z. A.; Kremer, P.; Larondelle, N.; McPhearson, T.; Haase, D. Classification of the
  394 Heterogeneous Structure of Urban Landscapes (STURLA) as an Indicator of Landscape Function
  395 Applied to Surface Temperature in New York City. *Ecological Indicators* 2016, 70, 574–585.
  396 https://doi.org/10.1016/j.ecolind.2015.10.014.
- Stewart, J. D.; Kremer, P.; Shakya, K. M.; Conway, M.; Saad, A. Outdoor Atmospheric Microbial
   Diversity Is Associated with Three-Dimensional Urban Landscape Structure and Differs from
   Indoor-Transit Systems. *bioRxiv* 2020, 2020.06.17.157651.
- 400 https://doi.org/10.1101/2020.06.17.157651.
- 401 (27) Getis, A.; Ord, J. K. The Analysis of Spatial Association by Use of Distance Statistics. *Geographical* 402 *Analysis* 1992, 24 (3), 189–206. https://doi.org/10.1111/j.1538-4632.1992.tb00261.x.
- 403 (28) City of Philadelphia, Department of Public Health, Air Management Services. *Philadelphia's Air* 404 *Quality Report 2018*; 2019.
- 405 (29) Burton, R. M.; Suh, H. H.; Koutrakis, P. Spatial Variation in Particulate Concentrations within
  406 Metropolitan Philadelphia. *Environmental Science and Technology* **1996**, 30 (2), 400-407. https://
  407 doi.org/10.1021/es950030f.
- 408 (30) Yu, N.; Zhu, Y.; Xie, X.; Yan, C.; Zhu, T.; Zheng, M. Characterization of Ultrafine Particles and Other
  409 Traffic Related Pollutants near Roadways in Beijing. *Aerosol and Air Quality Research* 2015, 15.
  410 https://doi.org/10.4209/aaqr.2014.11.0295.

- 411 (31) Spatial and Temporal Variability of the PM2.5/PM10 Ratio in Wuhan, Central China - Aerosol and 412 Air Quality Research https://aaqr.org/articles/aaqr-16-09-oa-0406 (accessed Jun 29, 2020). 413 (32) Ni, M.; Huang, J.; Lu, S.; Li, X.; Yan, J.; Cen, K. A Review on Black Carbon Emissions, Worldwide and 414 in China. Chemosphere 2014, 107, 83-93. https://doi.org/10.1016/j.chemosphere.2014.02.052. 415 (33) Kim, S.; Yu, S.; Yun, D. Spatiotemporal Association of Real-Time Concentrations of Black Carbon 416 (BC) with Fine Particulate Matters (PM2.5) in Urban Hotspots of South Korea. International 417 Journal of Environmental Research and Public Health 2017, 14 (11), 1350. 418 https://doi.org/10.3390/ijerph14111350. 419 (34) Philadelphia City Planning Commission. Philadelphia2035. 420 (35) Agarwal, A.; Speth, R. L.; Fritz, T. M.; Jacob, S. D.; Rindlisbacher, T.; Iovinelli, R.; Owen, B.; Miake-421 Lye, R. C.; Sabnis, J. S.; Barrett, S. R. H. SCOPE11 Method for Estimating Aircraft Black Carbon 422 Mass and Particle Number Emissions. Environ. Sci. Technol. 2019, 53 (3), 1364-1373. 423 https://doi.org/10.1021/acs.est.8b04060. Beckett, K. P.; Freer-Smith, P. H.; Taylor, G. The Capture of Particulate Pollution by Trees at Five 424 (36) 425 Contrasting Urban Sites. Arboricultural Journal 2000, 24 (2-3), 209-230. https://doi.org/10.1080/03071375.2000.9747273. 426 427 (37) Nowak, D. J.; Crane, D. E.; Stevens, J. C. Air Pollution Removal by Urban Trees and Shrubs in the 428 United States. Urban Forestry & Urban Greening **2006**, 4 (3), 115–123. 429 https://doi.org/10.1016/j.ufug.2006.01.007. 430 (38) Ježek, I.; Katrašnik, T.; Westerdahl, D.; Močnik, G. Black Carbon, Particle Number Concentration 431 and Nitrogen Oxide Emission Factors of Random in-Use Vehicles Measured with the on-Road 432 Chasing Method. Atmospheric Chemistry and Physics 2015, 15 (19), 11011–11026. 433 https://doi.org/10.5194/acp-15-11011-2015. 434 (39) Peters, J.; Theunis, J.; Poppel, M. V.; Berghmans, P. Monitoring PM10 and Ultrafine Particles in 435 Urban Environments Using Mobile Measurements. Aerosol Air Qual. Res. 2013, 13 (2), 509–522. 436 https://doi.org/10.4209/aaqr.2012.06.0152. 437 (40) Hankey, S.; Marshall, J. D. On-Bicycle Exposure to Particulate Air Pollution: Particle Number, Black 438 Carbon, PM2.5, and Particle Size. Atmospheric Environment 2015, 122, 65-73. 439 https://doi.org/10.1016/j.atmosenv.2015.09.025. 440 (41) Tunno, B. J.; Shields, K. N.; Lioy, P.; Chu, N.; Kadane, J. B.; Parmanto, B.; Pramana, G.; Zora, J.; 441 Davidson, C.; Holguin, F.; Clougherty, J. E. Understanding Intra-Neighborhood Patterns in PM2.5 442 and PM10 Using Mobile Monitoring in Braddock, PA. Environ Health 2012, 11 (1), 76. 443 https://doi.org/10.1186/1476-069X-11-76. 444 (42) Zhao, X.; Zhang, X.; Xu, X.; Xu, J.; Meng, W.; Pu, W. Seasonal and Diurnal Variations of Ambient 445 PM2.5 Concentration in Urban and Rural Environments in Beijing. Atmospheric Environment 446 2009, 43 (18), 2893–2900. https://doi.org/10.1016/j.atmosenv.2009.03.009. 447 (43) Chow, J. C.; Watson, J. G.; Fujita, E. M.; Lu, Z.; Lawson, D. R.; Ashbaugh, L. L. Temporal and Spatial 448 Variations of PM2.5 and PM10 Aerosol in the Southern California Air Quality Study. Atmospheric 449 Environment 1994, 28 (12), 2061-2080. https://doi.org/10.1016/1352-2310(94)90474-X. 450 (44) Claeys, M.; Graham, B.; Vas, G.; Wang, W.; Vermeylen, R.; Pashynska, V.; Cafmeyer, J.; Guyon, P.; 451 Andreae, M. O.; Artaxo, P.; Maenhaut, W. Formation of Secondary Organic Aerosols Through 452 Photooxidation of Isoprene. Science 2004, 303 (5661), 1173-1176. 453 https://doi.org/10.1126/science.1092805. 454 (45) Barzyk, T. M.; George, B. J.; Vette, A. F.; Williams, R. W.; Croghan, C. W.; Stevens, C. D. 455 Development of a Distance-to-Roadway Proximity Metric to Compare near-Road Pollutant Levels 456 to a Central Site Monitor. Atmospheric Environment 2009, 43 (4), 787-797.
- 457 https://doi.org/10.1016/j.atmosenv.2008.11.002.

- (46) Yu, C. H.; Fan, Z.; Lioy, P. J.; Baptista, A.; Greenberg, M.; Laumbach, R. J. A Novel Mobile
  Monitoring Approach to Characterize Spatial and Temporal Variation in Traffic-Related Air
  Pollutants in an Urban Community. *Atmospheric Environment* 2016, 141, 161–173.
  https://doi.org/10.1016/j.atmosenv.2016.06.044.
- 462 (47) Karner, A. A.; Eisinger, D. S.; Niemeier, D. A. Near-Roadway Air Quality: Synthesizing the Findings
  463 from Real-World Data. *Environ. Sci. Technol.* **2010**, 44 (14), 5334–5344.
- 464 https://doi.org/10.1021/es100008x.
  465 (48) Liu, Y.; Wu, J.; Yu, D.; Ma, Q. The Relationship between Urba
- 465 (48) Liu, Y.; Wu, J.; Yu, D.; Ma, Q. The Relationship between Urban Form and Air Pollution Depends on
  466 Seasonality and City Size. *Environ Sci Pollut Res* 2018, 25 (16), 15554–15567.
  467 https://doi.org/10.1007/s11356-018-1743-6.
- 468 (49) Cárdenas Rodríguez, M.; Dupont-Courtade, L.; Oueslati, W. Air Pollution and Urban Structure
- Linkages: Evidence from European Cities. *Renewable and Sustainable Energy Reviews* 2016, 53,
  1–9. https://doi.org/10.1016/j.rser.2015.07.190.
- 471 (50) Weng, Q.; Yang, S. Urban Air Pollution Patterns, Land Use, and Thermal Landscape: An
  472 Examination of the Linkage Using GIS. *Environ Monit Assess* **2006**, 117 (1), 463–489.
- 473 https://doi.org/10.1007/s10661-006-0888-9.
- 474

## 475 Supplemental



Figure SI-1. Maps showing all PM<sub>2.5</sub> observations on each day of data collection. As a result of
 intermittent road closures, the routes traveled on each day vary slightly.



Figure SI-2. Spearman correlations of BC and PM<sub>2.5</sub> concentrations on each day, with correlation
 coefficient (ρ) and p-value at the top left of each subplot.

Table SI-1: Pairwise Mann-Whitney U tests with Bonferroni correction for concentrations of
 PM<sub>1</sub>.

Date	6/27/1	7/1/	7/3/1	7/5/1	7/9/1	7/10/	7/12/	7/15/	7/16/	7/24/	7/26/
	9	19	9	9	9	19	19	19	19	19	19
6/27	0	0	0	0	0	0	0	0	0	0	0
/19											
7/1/	2.20E-	0	0	0	0	0	0	0	0	0	0
19	16										
7/3/	2.20E-	2.20	0	0	0	0	0	0	0	0	0
19	16	E-16									
7/5/	2.20E-	2.20	2.20E	0	0	0	0	0	0	0	0
19	16	E-16	-16								
7/9/	2.20E-	2.20	2.20E	2.20E	0	0	0	0	0	0	0
19	16	E-16	-16	-16							
7/10	2.20E-	2.20	2.20E	2.20E	2.20E	0	0	0	0	0	0
/19	16	E-16	-16	-16	-16						
7/12	2.20E-	2.20	2.20E	2.20E	2.20E	2.20E	0	0	0	0	0
/19	16	E-16	-16	-16	-16	-16					
7/15	2.20E-	2.20	2.20E	2.20E	2.20E	2.20E	2.20E	0	0	0	0
/19	16	E-16	-16	-16	-16	-16	-16				
7/16	2.20E-	2.20	2.20E	2.20E	2.20E	2.20E	6.00E	2.20E	0	0	0
/19	16	E-16	-16	-16	-16	-16	-14	-16			
7/24	2.20E-	2.20	2.20E	0	0						
/19	16	E-16	-16	-16	-16	-16	-16	-16	-16		
7/26	2.20E-	2.20	2.20E	0							
/19	16	E-16	-16	-16	-16	-16	-16	-16	-16	-16	
7/29	2.20E-	2.20	2.20E								
/19	16	E-16	-16	-16	-16	-16	-16	-16	-16	-16	-16

Table SI-2: Pairwise Mann-Whitney U tests with Bonferroni correction for concentrations of PM<sub>2.5</sub>.

Date	6/27/	7/1/1	7/10/	7/12/	7/15/	7/16/	7/24/	7/26/	7/29/	7/3/1	7/5/1
Date	10	0	10	10	10	10	10	10	10	0	0
( (07 (	17	7	17	17	17	17	17	17	17	7	7
6/2//	0	0	0	0	0	0	0	0	0	0	0
19											
7/1/1	6.60E	0	0	0	0	0	0	0	0	0	0
9	-270										
7/10/	2.20E	2.20E	0	0	0	0	0	0	0	0	0
19	-16	-16									
7/12/	2.20E	2.20E	2.20E	0	0	0	0	0	0	0	0
19	-16	-16	-16								
7/15/	2.20E	1.98E	2.20E	2.20E	0	0	0	0	0	0	0
19	-16	-10	-16	-16							
7/16/	2.20E	2.20E	2.20E	1.41E	2.20E	0	0	0	0	0	0
19	-16	-16	-16	-10	-16						
7/24/	1.90E	2.20E	2.20E	2.20E	2.20E	2.20E	0	0	0	0	0
19	-120	-16	-16	-16	-16	-16					
7/26/	2.20E	2.20E	2.20E	1.91E	2.20E	1.12E	2.20E	0	0	0	0
19	-16	-16	-16	-23	-16	-87	-16				
7/29/	2.20E	0	0	0							
19	-16	-16	-16	-16	-16	-16	-16	-16			
7/3/1	2.20E	2.20E	2.20E	1.45E	2.20E	6.32E	2.20E	3.82E	2.20E	0	0
9	-16	-16	-16	-71	-16	-157	-16	-20	-16		
7/5/1	2.20E	1.07E	2.20E	0							
9	-16	-16	-16	-16	-16	-16	-16	-16	-72	-16	
7/9/1	2.20E	2.20E	6.19E	2.20E							
9	-16	-16	-14	-16	-16	-16	-16	-16	-16	-16	-16



PM<sub>10</sub>.

Date	6/27/	7/1/1	7/10/	7/12/	7/15/	7/16/	7/24/	7/26/	7/29/	7/3/1	7/5/1
	19	9	19	19	19	19	19	19	19	9	9
6/27/	0	0	0	0	0	0	0	0	0	0	0
19											
7/1/1	2.20E	0	0	0	0	0	0	0	0	0	0
9	-16										
7/10/	2.20E	2.20E	0	0	0	0	0	0	0	0	0
19	-16	-16									
7/12/	2.20E	2.20E	2.20E	0	0	0	0	0	0	0	0
19	-16	-16	-16								
7/15/	9.43E	1.54E	2.20E	2.20E	0	0	0	0	0	0	0
19	-167	-12	-16	-16							
7/16/	2.20E	2.20E	6.07E	3.26E	2.20E	0	0	0	0	0	0
19	-16	-16	-211	-18	-16						
7/24/	2.64E	2.20E	2.20E	2.20E	2.20E	2.20E	0	0	0	0	0
19	-62	-16	-16	-16	-16	-16					
7/26/	2.20E	2.20E	2.20E	2.13E	2.20E	1.43E	2.20E	0	0	0	0
19	-16	-16	-16	-17	-16	-79	-16				
7/29/	2.20E	0	0	0							
19	-16	-16	-16	-16	-16	-16	-16	-16			
7/3/1	2.20E	2.20E	2.20E	8.51E	2.20E	4.71E	2.20E	9.48E	2.20E	0	0
9	-16	-16	-16	-103	-16	-217	-16	-46	-16		
7/5/1	2.20E	1.17E	2.20E	0							
9	-16	-16	-16	-16	-16	-16	-16	-16	-54	-16	
7/9/1	2.20E	2.20E	2.40E	9.23E	2.20E	4.87E	2.20E	2.20E	2.20E	2.20E	2.20E
9	-16	-16	-10	-242	-16	-150	-16	-16	-16	-16	-16

522 Table SI-4: Pairwise Mann-Whitney U tests with Bonferroni correction for concentrations of BC.

Date 6/27/ 7/1/1 7/10/ 7/12/ 7/15/ 7/16/ 7/24/ 7/26/ 7/29/	7/3/1	7/5/1
--	-------	-------

	19	9	19	19	19	19	19	19	19	9	9
6/27/	0	0	0	0	0	0	0	0	0	0	0
19											
7/1/1	35.32	0	0	0	0	0	0	0	0	0	0
9	32										
7/10/	0.042	0.148	0	0	0	0	0	0	0	0	0
19	016	038									
7/12/	25.67	40.92	2.470	0	0	0	0	0	0	0	0
19	4	66	38								
7/15/	7.840	1.436	2.09E	0.891	0	0	0	0	0	0	0
19	8	16	-05								
7/16/	0.274	1.382	51.58	3.749	0.001	0	0	0	0	0	0
19	692	7	56	46	339						
7/24/	1.794	4.491	20.35	15.43	0.004	29.33	0	0	0	0	0
19	54	3	44	08	103	04					
7/26/	1.45E	1.45E-	4.8E-	1E-11	1.45E	9.07E	4.27E-	0	0	0	0
19	-14	14	09		-14	-07	10				
7/29/	7.6E-	0.000	1.820	0.017	4.26E	2.889	0.280	0.002	0	0	0
19	05	101	28	714	-09	48	566	312			
7/3/1	8.78E	7.22E-	0.678	0.010	1.15E	0.863	0.150	0.000	60.90	0	0
9	-06	06	48	501	-10	28	282	626	48		
7/5/1	24.70	4.370	1.51E	2.991	29.85	0.006	0.042	1.45E	1.67E	2.12E-	0
9	38	52	-05	78	84	712	88	-14	-09	12	
7/9/1	0.003	0.004	20.31	0.528	1.64E	17.94	5.938	1.47E	9.57	8.995	3.81E
9	51	811	48	726	-07	54	68	-07		8	-08