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Developing Multipollutant Exposure Indicators of Traffic Pollution: The Dorm Room Inhalation to Vehicle Emissions (DRIVE) Study

Jeremy A. Sarnat, Armistead (Ted) Russell, Donghai Liang, Jennifer L. Moutinho, Rachel Golan, Rodney J. Weber, Dong Gao, Stefanie Ebelt Sarnat, Howard H. Chang, Roby Greenwald, and Tianwei Yu

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with a Critique by the HEI Review Committee

Research Report 196

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ABOUT HEI

The Health Effects Institute is a nonprofit corporation chartered in 1980 as an independent research organization to provide high-quality, impartial, and relevant science on the effects of air pollution on health. To accomplish its mission, the institute

- Identifies the highest-priority areas for health effects research;
- Competitively funds and oversees research projects;
- Provides intensive independent review of HEI-supported studies and related research;
- Integrates HEI's research results with those of other institutions into broader evaluations; and
- Communicates the results of HEI's research and analyses to public and private decision makers.

HEI typically receives balanced funding from the U.S. Environmental Protection Agency and the worldwide motor vehicle industry. Frequently, other public and private organizations in the United States and around the world also support major projects or research programs. HEI has funded more than 340 research projects in North America, Europe, Asia, and Latin America, the results of which have informed decisions regarding carbon monoxide, air toxics, nitrogen oxides, diesel exhaust, ozone, particulate matter, and other pollutants. These results have appeared in more than 260 comprehensive reports published by HEI, as well as in more than 1,000 articles in the peer-reviewed literature.

HEI's independent Board of Directors consists of leaders in science and policy who are committed to fostering the public–private partnership that is central to the organization. The Research Committee solicits input from HEI sponsors and other stakeholders and works with scientific staff to develop a Five-Year Strategic Plan, select research projects for funding, and oversee their conduct. The Review Committee, which has no role in selecting or overseeing studies, works with staff to evaluate and interpret the results of funded studies and related research.

All project results and accompanying comments by the Review Committee are widely disseminated through HEI's website (*www.healtheffects.org*), printed reports, newsletters and other publications, annual conferences, and presentations to legislative bodies and public agencies.

ABOUT THIS REPORT

Research Report 196, *Developing Multipollutant Exposure Indicators of Traffic Pollution: The Dorm Room Inhalation to Vehicle Emissions (DRIVE) Study*, presents a research project funded by the Health Effects Institute and conducted by Dr. Jeremy A. Sarnat of Emory University, Atlanta, Georgia, and his colleagues. The report contains three main sections.

The HEI Statement, prepared by staff at HEI, is a brief, nontechnical summary of the study and its findings; it also briefly describes the Review Committee's comments on the study.

The Investigators' Report, prepared by Sarnat and colleagues, describes the scientific background, aims, methods, results, and conclusions of the study.

The Critique, prepared by members of the Review Committee with the assistance of HEI staff, places the study in a broader scientific context, points out its strengths and limitations, and discusses remaining uncertainties and implications of the study's findings for public health and future research.

This report has gone through HEI's rigorous review process. When an HEI-funded study is completed, the investigators submit a draft final report presenting the background and results of the study. This draft report is first examined by outside technical reviewers and a biostatistician. The report and the reviewers' comments are then evaluated by members of the Review Committee, an independent panel of distinguished scientists who have no involvement in selecting or overseeing HEI studies. During the review process, the investigators have an opportunity to exchange comments with the Review Committee and, as necessary, to revise their report. The Critique reflects the information provided in the final version of the report.

PREFACE

HEI's Research Program to Improve Assessment of Exposure to Traffic-Related Air Pollution

INTRODUCTION

Traffic emissions are an important source of urban air pollution. Emissions from motor vehicles and ambient concentrations of most monitored traffic-related pollutants have decreased steadily over the last several decades in most high-income countries as a result of air quality regulations and improvements in vehicular emission control technologies, and this trend is likely to continue. However, these positive developments have not been able to fully compensate for the rapid growth of the motor vehicle fleet due to growth in population and economic activity and increased vehicular congestion, as well as the presence of older or malfunctioning vehicles on the roads.

In 2010, HEI published Special Report Number 17, *Traffic-Related Air Pollution: A Critical Review of the Literature on Emissions, Exposure, and Health Effects*. This report, developed by the HEI Panel on the Health Effects of Traffic-Related Air Pollution ("Panel"), summarized and synthesized research related to the health effects from exposure to traffic emissions. Among its conclusions, the Panel "identified an exposure zone within a range of up to 300 to 500 m from a major road as the area most highly affected by traffic emissions (the range reflects the variable influence of background pollution concentrations, meteorologic conditions, and season)." The Panel estimated that 30% to 45% of people living in large North American cities reside within these zones. Based on a review of health studies, the Panel concluded that exposure to traffic-related air pollution was causally linked to worsening asthma symptoms. It also found "suggestive evidence of a causal relationship with onset of childhood asthma, nonasthma respiratory symptoms, impaired lung function, total and cardiovascular mortality, and cardiovascular morbidity" (HEI Panel on the Health Effects of Traffic-Related Air Pollution 2010).

The report also noted that exposure assessment of traffic-related air pollution is challenging; it is a complex mixture of particulate and many gaseous pollutants, many of which are also emitted by other sources, and is characterized by high spatial and temporal variability, with the highest traffic-related air pollution concentrations occurring at or close to major roads. Therefore, identifying an appropriate exposure metric that uniquely indicates traffic-related air pollution and modeling the distribution of exposure at a sufficiently high degree of spatial and temporal resolution have been difficult.

The most commonly used exposure metrics are measured or modeled concentrations of individual pollutants considered to be indicators of traffic-related air pollution (such as nitrogen dioxide [NO_2] or black carbon [BC]) and simple indicators of traffic (such as distance of the residence from busy roads or traffic density near the residence).

A range of models, such as dispersion, land-use regression, and hybrid models, have been developed to estimate exposure. Some attempts to account for indoor infiltration and time–activity patterns have been made to refine such estimates. Although many improvements in these exposure models have occurred over time (especially the use of geographical information system approaches and the application of more sophisticated statistical methods), their usefulness still depends on the model assumptions and data quality. Few studies have compared the performance of different models and evaluated exposure measurement error and possible bias in health estimations.

To start addressing these issues, HEI issued a Request for Applications in 2013. To inform the development of the RFA, the HEI Research Committee held a

workshop in April 2012 with experts in the areas of atmospheric chemistry, pollutant measurements, exposure models, epidemiology, and health assessment in order to discuss and identify the highest priority research questions.

OBJECTIVES OF RFA 13-1

RFA 13-1, *Improving Assessment of Near-Road Exposure to Traffic Related Pollution*, aimed to solicit studies to improve exposure assessment for use in future work on the health effects of traffic-related air pollution. The RFA had three major objectives:

- Demonstrate novel surrogates of near-road traffic-related pollution, taking advantage of new sensors and/or existing monitoring data.
- Determine the most important variables that explain spatial and temporal variance of near-road traffic-related pollutant concentrations at the personal, residential, and/or community levels, and explain the implications of these for future monitoring, modeling, exposure, and health effects studies.
- Improve inputs for exposure models for traffic-related health studies; evaluate and compare the performance of alternative models to existing models and actual measurements to quantify exposure measurement error.

DESCRIPTION OF THE PROGRAM

Five studies were funded under RFA 13-1 to represent a variety of geographical locations and cover the various RFA objectives. The study by Sarnat and colleagues described in this report (Research Report 196) is the second to be published. All five studies are summarized below.

"The Hong Kong D3D Study: A Dynamic Three Dimensional Exposure Model for Hong Kong," Benjamin Barratt, King's College London, United Kingdom (Principal Investigator) Barratt and colleagues estimated exposure to traffic-related air pollution using a dynamic three-dimensional land-use regression model for Hong Kong, which has many high-rise buildings, resulting in street canyons. Different exposure models were developed with increasing complexity (e.g., incorporating infiltration indoors, vertical gradients, and time–activity patterns) and applied in an epidemiological study to evaluate the potential impact of exposure measurement error in mortality estimates. The study has been published (Research Report 194).

"Enhancing Models and Measurements of Traffic-Related Air Pollutants for Health Studies Using Bayesian Melding," Stuart Batterman, University of Michigan, Ann Arbor, Michigan (Principal Investigator) Batterman and colleagues estimated exposure to traffic-related air pollution using a variety of methods and models, including air pollution dispersion models and novel data fusion methods that would be able to propagate uncertainty more fully into the exposure estimates. The study made extensive use of data collected in the Near-road EXposures and effects of Urban air pollutants Study (NEXUS), a cohort study designed to examine the relationship between near-roadway pollutant exposures and respiratory outcomes in children with asthma who live close to major roadways in Detroit. The study has been completed and is currently in review.

"Characterizing the Determinants of Vehicle Traffic Emissions Exposure: Measurement and Modeling of Land-Use, Traffic, Transformation, and Transport," Christopher Frey, North Carolina State University, Raleigh, North Carolina (Principal Investigator) Frey and colleagues investigated key factors that influence exposure to traffic-related air pollution: traffic and its composition; built environment including road characteristics and land use; and dispersion, transport, and transformation processes. The study collected extensive measurements of fine particulate matter ($PM_{2.5}$), ultrafine particles (UFPs), oxides of nitrogen (NO_x), and semi-volatile organic compounds (SVOCs) in various near-road locations in the Raleigh–Durham area. This study has been completed and is currently in review.

"Developing Multipollutant Exposure Indicators of Traffic Pollution: The Dorm Room Inhalation to Vehicle Emissions (DRIVE) Study," Jeremy Sarnat, Emory University, Atlanta, Georgia (Principal Investigator) In the study presented in this report, Sarnat and colleagues evaluated novel multipollutant traffic surrogates by collecting measurements in and around two student dormitories in Atlanta and explored the use of metabolomics to identify possible exposure-related metabolites. The

DRIVE study made use of a unique emission-exposure setting in Atlanta, on the Georgia Institute of Technology campus, with one dorm immediately adjacent to the busiest and most congested highway artery in the city (with more than 300,000 vehicles per day) and another dorm located farther away.

"Evaluation of Alternative Sensor-Based Exposure Assessment Methods," Edmund Seto, University of Washington, Seattle, Washington (Principal Investigator) Seto and colleagues performed an evaluation of novel low-cost air pollution sensors to characterize traffic-related air pollution in the San Francisco Bay area. They have deployed various low-cost air pollution sensors — including Shinyei particulate matter sensors and Alphasense electrochemical sensors — for an extended period of time. Sensors were colocated with reference monitors to evaluate sensor performance. This study has been completed and is currently in review.

NEXT STEPS

As these studies are nearing completion, valuable lessons learned may be integrated in new research. HEI is committed to continuing research on traffic-related air pollution and, in January 2017, issued RFA 17-1, *Assessing Adverse Health Effects of Exposure to Traffic-Related Air Pollution, Noise, and Their Interactions with Socioeconomic Status*, seeking studies to assess adverse health effects of short- and/or long-term exposure to traffic-related air pollution. The applicants were asked to consider spatially correlated factors that may either confound or modify the health effects of traffic-related air pollution, most notably, traffic noise, socioeconomic status, and factors related to the built environment, such as presence of green space.

At the time of publication of this report, three studies have been selected for funding and are expected to start in 2018. Payam Dadvand and Jordi Sunyer from the Barcelona Institute for Global Health will set up a new cohort of healthy pregnant women in Barcelona to examine the effects of traffic-related pollution and other factors on birth weight, fetal growth, and placental function. Ole Raaschou-Nielsen from the Danish Cancer Society Research Center, Copenhagen, Denmark, will make use of very large administrative databases to evaluate the effects of traffic-related air pollution and other factors on myocardial infarction, stroke, and diabetes in Denmark. Meredith Franklin from the University of Southern California–Los Angeles, will build on the Children's Health Study in Southern California to evaluate the adverse effects of nontailpipe emissions and of noise on children's respiratory health.

In addition, since the release of HEI's critical review of the traffic literature in 2010, many additional studies about traffic-related air pollution have been published, and regulations and vehicular technology have advanced significantly. Therefore, HEI is currently in the process of conducting a new literature review of the health effects of traffic-related air pollution. Further information on these activities can be obtained at the HEI website, *www.healtheffects.org/air-pollution/traffic-related-air-pollution.*

REFERENCES

HEI Panel on the Health Effects of Traffic-Related Air Pollution. 2010. Traffic-Related Air Pollution: A Critical Review of the Literature on Emissions, Exposure, and Health Effects. HEI Special Report 17. Boston, MA:Health Effects Institute.

HEI STATEMENT

Synopsis of Research Report 196

Exposure to Traffic-Related Air Pollution in a Panel of Students Living in Dormitories

INTRODUCTION

Exposure to traffic-related air pollution has been associated with various adverse health effects. However, exposure assessment is challenging because traffic-related air pollution is a complex mixture of many particulate and gaseous pollutants and is characterized by high variability by location and by time of day or season. Approaches to assess exposure to traffic-related air pollution have included measurements made at fixed sites at various distances from busy roads or with a mobile platform and via models such as land-use regression and dispersion models. Sarnat and colleagues proposed to evaluate two multipollutant air pollutant metrics that had not previously been considered as metrics of exposure to traffic-related air pollution, by collecting measurements in and around two student dormitories in Atlanta that were located at different distances from a major highway. They also proposed a small panel study to compare the indoor and outdoor pollutant concentrations to personal exposures experienced by students who lived in the two dormitories. They intended to explore the use of metabolomics to identify possible biological markers that varied with exposure to traffic-related air pollution in those students. With the encouragement of HEI, they also colocated several low-cost sensors with monitors using more established methods to test the reliability of the new sensors.

APPROACH

The investigators conducted their study on the campus of the Georgia Institute of Technology in Atlanta, Georgia, near the downtown Connector where I-75 and I-85 merge, one of the most heavily trafficked highway arteries in the United States (~300,000 vehicles per day with little diesel traffic). The investigators conducted an extensive field campaign to measure levels of a number of air pollutants — including fine particulate matter, nitrogen dioxide (NO_2), and black carbon — at six outdoor sites and

What This Study Adds

- This study evaluated exposure to traffic-related air pollutants measured outside, inside, and by personal monitors. It also evaluated metabolomics profiles in students living in dormitories close to or away from a traffic hotspot.
- It had many strong aspects — a large number of measurements at a site next to one of the busiest highways in the United States, a multipollutant approach, various exposure metrics, and concurrent evaluation of low-cost sensors.
- Lower-than-expected air pollutant concentrations and less-steep gradients reported in this and other recent studies provide evidence that the near-road environment is improving, likely a consequence of air quality regulations and related improvements in vehicle emission control technologies.
- The multipollutant exposure metrics explored in the study did not appear to be useful to capture traffic-related air pollution in the near-road environment, and the low-cost sensors tested did not perform well.
- Because of the low exposure contrast and other differences between the dormitory populations, a metabolomics study to explore differences among residents of two dormitories — one close to and the other farther from the roadway — was not informative.

This Statement, prepared by the Health Effects Institute, summarizes a research project funded by HEI and conducted by Dr. Jeremy Sarnat, Emory University Rollins School of Public Health, Atlanta, Georgia, and colleagues. Research Report 196 contains both the detailed Investigators' Report and a Critique of the study prepared by the Institute's Review Committee.

inside two student dormitories at different distances from the highway. They also measured personal air pollutant exposures to NO_2 and black carbon with monitors carried by a panel of student participants, as well as biological markers in the study participants.

The study focused on evaluating two multipollutant metrics of exposure to traffic-related air pollution. The first, the integrated mobile source indicator (IMSI), combines measurements of several different pollutants (elemental carbon, carbon monoxide, and nitrogen oxides) into a single metric of the mixture of traffic-related air pollutants. The second, the oxidative potential of fine particulate matter (FPMOP), was considered a potential metric of exposure to traffic-related air pollution because it was associated with both mobile source emissions and cytotoxicity and heart- and lung-related emergency department visits in some earlier studies. Although it is a single measurement, FPMOP is considered a multipollutant metric because it is affected (albeit unpredictably) by many particle properties, including size, surface properties, and chemical composition.

First, Sarnat and colleagues evaluated how pollutant levels changed with weather and time of day or week at an air pollutant monitor located 10 m from the highway. Second, they compared the air pollutant levels at their other monitors with the levels at the highway monitor. Third, they tested which outdoor and indoor measurements were most strongly correlated with measurements made by monitors carried by the participants. Fourth, they measured metabolomics profiles in the study participants' blood and saliva to see if there were differences among the participants that lived in the two dormitories.

MAIN RESULTS AND INTERPRETATION

In its independent review of the study, the HEI Review Committee noted that Sarnat and colleagues conducted a comprehensive study to evaluate single-pollutant and multipollutant metrics of exposure to traffic-related air pollution. The large number of detailed measurements — including outdoor and indoor air pollutant levels, personal exposure to air pollutants, and measurements in blood and saliva — the multipollutant approach, the low participant drop-out rate, and the concurrent evaluation of low-cost sensors were strengths of the study.

The results did not provide strong evidence of the utility of the IMSI or FPMOP as multipollutant metrics of exposure to traffic-related air pollution for use in health studies in the near-road environment. There was limited variation in these proposed metrics over the study area on average, although the different sites did have some differences in air pollutant levels when stratified by time of day. In addition, NO_2 did not seem to be a good metric of exposure to traffic-related air pollution in this study, because NO_2 levels were not substantially higher near the highway than they were farther away, and indoor sources contributed to the NO_2 levels inside the dormitories. The Committee noted that the investigators had not adopted an a priori criterion by which to assess the suitability of candidate metrics of exposure to traffic-related air pollution; when the Committee did apply its own criteria, they concurred with the conclusion that the multipollutant metrics were not useful for this application.

An interesting approach in this study was that it included a panel study with biological sampling for metabolomics analyses, rather than stopping at assessment of exposure to air pollution; however, the usefulness of the panel study results was limited. Despite their locations at different distances from the roadways, the personal exposures measured among residents of the two dormitories were very similar. The Committee thought the reported metabolomics differences were likely a consequence of factors other than exposure to traffic pollution. Though more extensive analyses are under way, an expanded study design that included more dormitories with carefully controlled building and population characteristics to allow separation of metabolomic differences related to the dormitory from those related to traffic-related air pollution would have been preferable.

The overall lower-than-expected air pollutant concentrations and less-steep gradients reported in this and other recent studies provide evidence that the near-road environment is improving. This result was likely a consequence of air quality regulations and related improvements in vehicle emission control technologies. The changing near-road environment has important consequences for the design of new research assessing the adverse health effects of traffic-related air pollution because larger study populations will be needed to measure potential effects of smaller exposure contrasts. In addition, past near-road air pollution and health studies may become less relevant to the current and future near-road environment given the fast-paced changes in engine and fuel technologies and electrification of the fleet.

INVESTIGATORS' REPORT

Developing Multipollutant Exposure Indicators of Traffic Pollution: The Dorm Room Inhalation to Vehicle Emissions (DRIVE) Study

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ABSTRACT

Introduction The Dorm Room Inhalation to Vehicle Emissions (DRIVE*) study was conducted to measure traditional single-pollutant and novel multipollutant traffic indicators along a complete emission-to-exposure pathway. The overarching goal of the study was to evaluate the suitability of these indicators for use as primary traffic exposure metrics in panel-based and small-cohort epidemiological studies.

Methods Intensive field sampling was conducted on the campus of the Georgia Institute of Technology (GIT) between September 2014 and January 2015 at 8 monitoring sites (2 indoors and 6 outdoors) ranging from 5 m to 2.3 km from the busiest and most congested highway artery in Atlanta. In addition, 54 GIT students living in one of two dormitories either near (20 m) or far (1.4 km) from the highway were recruited to conduct personal exposure sampling and weekly biomonitoring. The pollutants measured were selected to provide information about the heterogeneous particulate and gaseous composition of primary traffic emissions, including the traditional traffic-related species (e.g., carbon monoxide [CO], nitrogen dioxide [NO_2], nitric oxide [NO], fine particulate matter [$PM_{2.5}$], and black carbon [BC]), and of secondary species (e.g., ozone [O_3] and sulfate as well as organic carbon [OC], which is both primary and secondary) from traffic and other sources. Along with these pollutants, we also measured two multipollutant traffic indicators: integrated mobile source indicators (IMSIs) and fine particulate matter oxidative potential (FPMOP). IMSIs are derived from elemental carbon (EC), CO, and nitrogen oxide (NO_x) concentrations, along with the fractions of these species emitted by gasoline and diesel vehicles, to construct integrated estimates of gasoline and diesel vehicle impacts. Our FPMOP indicator was based on an acellular assay involving the depletion of dithiothreitol (DTT), considering both water-soluble and insoluble components (referred to as $FPMOP^{total\text{-}DTT}$). In addition, a limited assessment of 18 low-cost sensors was added to the study to supplement the four original aims.

Results Pollutant levels measured during the study showed a low impact by this highway hotspot source on its surrounding vicinity. These findings are broadly consistent with results from other studies throughout North America showing decreased relative contributions to

This Investigators' Report is one part of Health Effects Institute Research Report 196, which also includes a Critique by the Review Committee and an HEI Statement about the research project. Correspondence concerning the Investigators' Report may be addressed to Dr. Jeremy Sarnat, Emory University Rollins School of Public Health, 1518 Clifton Rd., Atlanta, GA 30322; e-mail: *jsarnat@emory.edu*.

Although this document was produced with partial funding by the United States Environmental Protection Agency under Assistance Award CR–83467701 to the Health Effects Institute, it has not been subjected to the Agency's peer and administrative review and therefore may not necessarily reflect the views of the Agency, and no official endorsement by it should be inferred. The contents of this document also have not been reviewed by private party institutions, including those that support the Health Effects Institute; therefore, it may not reflect the views or policies of these parties, and no endorsement by them should be inferred.

* A list of abbreviations and other terms appears at the end of this volume.

urban air pollution from primary traffic emissions. We view these reductions as an indication of a changing near-road environment, facilitated by the effectiveness of mobile source emission controls. Many of the primary pollutant species, including NO, CO, and BC, decreased to near background levels by 20 to 30 m from the highway source. Patterns of correlation among the sites also varied by pollutant and time of day. NO_2 exhibited spatial trends that differed from those of the other single-pollutant primary traffic indicators. We believe this was caused by kinetic limitations in the photochemical chemistry, associated with primary emission reductions, required to convert the NO-dominant primary NO_x, emitted from automobiles, to NO_2. This finding provides some indication of limitations in the use of NO_2 as a primary traffic exposure indicator in panel-based health effect studies. Roadside monitoring of NO, CO, and BC tended to be more strongly correlated with sites, both near and far from the road, during morning rush hour periods and often weakly to moderately correlated during other time periods of the day. This pattern was likely associated with diurnal changes in mixing and chemistry and their impact on spatial heterogeneity across the campus. Among our candidate multipollutant primary traffic indicators, we report several key findings related to the use of oxidative potential (OP)-based indicators. Although earlier studies have reported elevated levels of FPMOP in direct exhaust emissions, we found that atmospheric processing further enhanced $FPMOP^{total\text{-}DTT}$, likely associated with the oxidation of primary polycyclic aromatic hydrocarbons (PAHs) to quinones and hydroxyquinones and with the oxidization and water solubility of metals. This has important implications in terms both of the utility of $FPMOP^{total\text{-}DTT}$ as a marker for exhaust emissions and of the importance of atmospheric processing of particulate matter (PM) being tied to potential health outcomes. The results from the personal exposure monitoring also point to the complexity and diversity of the spatiotemporal variability patterns among the study monitoring sites and the importance of accounting for location and spatial mobility when estimating exposures in panel-based and small-cohort studies. This was most clearly demonstrated with the personal BC measurements, where ambient roadside monitoring was shown to be a poor surrogate for exposures to BC. Alternative surrogates, including ambient and indoor BC at the participants' respective dorms, were more strongly associated with personal BC, and knowledge of the participants' mean proximity to the highway was also shown to explain a substantial level of the variability in corresponding personal exposures to both BC and NO_2. In addition, untargeted metabolomic indicators measured in plasma and saliva, which represent emerging methods for measuring exposure, were used to extract approximately 20,000 and 30,000 features from plasma and saliva, respectively. Using hydrophilic interaction liquid chromatography (HILIC) in the positive ion mode, we identified 221 plasma features that differed significantly between the two dorm cohorts. The bimodal distribution of these features in the HILIC column was highly idiosyncratic; one peak consisted of features with elevated intensities for participants living in the near dorm; the other consisted of features with elevated intensities for participants in the far dorm. Both peaks were characterized by relatively short retention times, indicative of the hydrophobicity of the identified features. The results from the metabolomics analyses provide a strong basis for continuing this work toward specific chemical validation of putative biomarkers of traffic-related pollution. Finally, the study had a supplemental aim of examining the performance of 18 low-cost CO, NO, NO_2, O_3, and $PM_{2.5}$ pollutant sensors. These were colocated alongside the other study monitors and assessed for their ability to capture temporal trends observed by the reference monitoring instrumentation. Generally, we found the performance of the low-cost gas-phase sensors to be promising after extensive calibration; the uncalibrated measurements alone, however, would likely not have led to reliable results. The low-cost PM sensors we evaluated had poor accuracy, although PM sensor technology is evolving quickly and warrants future attention.

Conclusions An immediate implication of the changing near-road environment is that future studies aimed at characterizing hotspots related to mobile sources and their impacts on health will need to consider multiple approaches for characterizing spatial gradients and exposures. Specifically and most directly, the mobile source contributions to ambient concentrations of single-pollutant indicators of traffic exposure are not as distinguishable to the degree that they have been in the past. Collectively, the study suggests that characterizing exposures to traffic-related pollutants, which is already difficult, will become more difficult because of the reduction in traffic-related emissions. Additional multi-tiered approaches should be considered along with traditional measurements, including the use of alternative OP measures beyond those based on DTT assays, metabolomics, low-cost sensors, and air quality modeling.

INTRODUCTION

Recent interest in air pollution health effects and regulatory intervention has shifted toward adopting multipollutant perspectives (Greenbaum and Shaikh 2010; Johns et

al. 2012; Mauderly et al. 2010; Solomon et al. 2012; Vedal and Kaufman 2011). For highly heterogeneous sources, including primary traffic emissions, a multipollutant framework provides new opportunities to characterize biologically relevant exposures. Improving exposure assessment to traffic-related pollution is particularly critical given the abundance of studies reporting associations between traffic sources and numerous adverse health effects, as well as changing trends affecting the sources and impacts of mobile source emissions (HEI Panel on the Health Effects of Traffic-Related Air Pollution 2010).

Multipollutant approaches, including the use of source-apportioned measures of primary traffic pollution emissions, have been used to estimate health risks from combined exposures to traffic mixtures (Janssen et al. 2011; Ostro et al. 2007; Sarnat et al. 2008a). There is growing evidence, however, that the near-road environment is changing rapidly and that traditional source contributions, fate and transport properties, and exposure factors differ from those reported to exist in the literature (Blanchard et al. 2013a, 2013b, 2013c; Henneman et al. 2015; Vijayaraghavan et al. 2014). Substantial gaps exist in our understanding of how both traditional and nontraditional metrics vary in space and time in the changing near-road environment, how they compare with each other, and whether they offer accurate and biologically relevant means of assigning exposures to primary traffic emissions. Indeed, a leading factor facilitating the recent establishment of the U.S. Environmental Protection Agency (U.S. EPA)-supported near-road monitoring network (Batterman 2013) was an interest in improving exposure assessment for primary traffic emissions, for large urban populations, and for individuals living near roadways who may be especially vulnerable to this pollution source. Monitoring from the network's 75 near-road sites, which are largely located within 30 m of highly trafficked roadways, commenced on January 1, 2014.

To address these research gaps and more closely examine emerging trends related to characterizing traffic pollution exposures, we conducted DRIVE, a multi-tiered field study. A focus of the study centered on the near-road environment, understanding the impact of a roadway on its adjacent environment and the potential implications related to exposures for individuals living in this environment. We examined traditional traffic indicators as well as IMSIs and FPMOP, two less well-studied multipollutant traffic indicators. Briefly, IMSIs are integrated estimates of gasoline and diesel vehicle impacts constructed from EC, CO, and NO_x concentrations and the fractions of these species emitted by gasoline and diesel vehicles (Pachon et al. 2012). FPMOP is a biologically relevant, cumulative measure that characterizes pollutant exposures through their ability to elicit reactive oxygen species (ROS) (Ghio et al. 2012; Li et al. 2003; Squadrito et al. 2001; Tao et al. 2003). Here, FPMOP was derived using an acellular assay measuring the oxidative activity of water-soluble particles based on their ability to catalyze the transfer of electrons from DTT to oxygen, generating superoxide radical anions ($O_2\bullet^-$). Results using this assay are referred to as $FPMOP^{total\text{-}DTT}$. Our earlier studies found that ambient $FPMOP^{DTT}$ was associated with mobile source emissions (Bates et al. 2015; Verma et al. 2014); laboratory studies have also found that engine-emitted particles are DTT-active (McWhinney et al. 2011, 2013b). Thus, for near-road measurements, FPMOP, characterized using DTT depletion or some other assay, may also be a useful, biologically relevant method for characterizing primary traffic emissions.

A key component of the study was a panel of participants living near (within 50 m) or farther from (1.4 km) one of the busiest, most congested highways in Atlanta, Georgia. Although the study was designed as a monitoring and exposure assessment, we chose to use a panel-based design as a framework, with the potential to inform epidemiologic investigations involving those living near roadways. Panel-based and small-cohort designs have proven to be especially effective approaches to investigate traffic-related air pollution and adverse health, given the ability to measure both exposure and health parameters on an individual level (Delfino et al. 2006, 2008; McCreanor et al. 2007; Sarnat et al. 2012). Previous studies have examined the suitability of using broad proximity-based markers (Hoek et al. 2002) as well as single-species tracers of traffic pollution (Lewne et al. 2004; McCreanor et al. 2007; Oftedal et al. 2003; Rijnders et al. 2001), which have been useful in informing exposure assignment, particularly as it relates to measurement error, for epidemiologic analyses (Batterman et al. 2014a; Beckerman et al. 2008; Zhou and Levy 2008; Zhu et al. 2002b). For panel-based and small-cohort studies that aim to assess health effects of traffic pollution, improved exposure assignment may lead to reductions in the relevant components of measurement error for epidemiologic study designs examining short-term exposure and acute health response. Thus, a significant area of interest for our research group in conducting the current research was to assess relationships between outdoor primary traffic exposure measures in small panels of individuals living in close proximities to these sources, using exposure metrics based on near-road monitoring sites, and more proximal and biologically based metrics.

Collectively, the study comprehensively characterized a traffic pollution hotspot adjacent to residential areas, focusing on assessing the emission-to-exposure pathway for specific primary traffic pollutant components. This report summarizes the aims, design, and key findings of this effort.

SPECIFIC AIMS

The DRIVE study was designed around a near-road emission-to-exposure setting, based along the busiest and most congested highway artery in the geographic core of Atlanta. Its overarching goal was to evaluate the suitability of both traditional single-pollutant and novel multipollutant indicators for use as primary traffic exposure metrics in panel-based and small-cohort epidemiological studies. To achieve this goal, we set out and undertook the following four specific aims in our initial proposal:

- **Specific Aim 1.** To examine associations between traditional measured and modeled pollutants and novel traffic indicators at a dedicated near-road monitoring site. As part of this aim, we also proposed to assess factors affecting roadside concentrations of each pollutant and multipollutant indicator.
- **Specific Aim 2.** To assess the spatiotemporal variability of outdoor and indoor primary traffic pollutant components as well as multipollutant indicators along a near-road to mid-distance spatial gradient (5 m–2.3 km) from the traffic pollution source.
- **Specific Aim 3.** To evaluate how well traffic pollutant components and multipollutant traffic indicators measured at near-road and other fixed monitoring sites reflect corresponding personal exposures.
- **Specific Aim 4.** To explore the feasibility of using high-throughput metabolomics as an approach in panel-based or small-cohort studies for identifying environmental exposures, including primary traffic.

In addition to the four original aims, we added a fifth supplemental aim in response to a request from HEI. This aim was developed once the project was already under way; however, field sampling for the aim was still concurrent with that conducted for the other four aims above:

- **Supplemental Aim 5.** To examine the performance of multiple low-cost pollutant sensor sampling platforms in a near-road field setting.

METHODS AND STUDY DESIGN

STUDY LOCATION AND OVERALL DESIGN

The study was conducted to measure traditional and multipollutant traffic indicators along a full emission-to-exposure pathway. Intensive field sampling was conducted on the campus of GIT in Atlanta at outdoor and indoor monitoring sites adjacent to one of the most heavily trafficked highway arteries in the United States ("the Connector," a section of highway where Interstates 75 and 85 merge). Although numerous smaller roadways surround the GIT campus, the Connector is the dominant mobile emissions source, with an annual average daily traffic count at least 15 times that of the other roads in the area. Fifty-four GIT students living in one of two dormitories located at two different proximities to the highway were recruited to participate in personal exposure sampling and weekly biomonitoring. Sampling was conducted from September 2014 to January 2015.

Measurements included, but were not limited to, primary traffic pollutants using a variety of sampling platforms at several exposure tiers. Tier 1 included measurements conducted at six outdoor sites at various distances from the highway; Tier 2 included measurements conducted indoors at the two dorms; Tier 3 included personal exposure measurements of GIT students; and Tier 4 included metabolomics analyses of biological samples from the student participants. In addition to the field and personal exposure measurements, we conducted Research LINE-source (RLINE) dispersion modeling of several traffic pollutants with the goal of enhancing the characterization of the spatial distribution of pollution around the traffic hotspot and to provide additional information for personal exposure assessment.

OUTDOOR AND INDOOR MEASUREMENTS AND MODELING (TIERS 1 AND 2)

Sampling Locations

Sampling was conducted at eight dedicated monitoring sites (six outdoors and two indoors) ranging from 5 m to 2.3 km from the Connector (Figure 1). The main near-road sampling site (Roadside, or RDS) consisted of a highly instrumented trailer with an inlet 3 m from the ground and at a distance of 10 m from the closest lane of the 15-lane highway. The area surrounding this site was open, being in a campus parking lot. Near-road pollutant measurements were also made at a Georgia Department of Natural Resources' Environmental Protection Division (EPD) monitoring site with an inlet 3 m from the ground and 5 m from the closest highway lane. The site was established as part of the U.S. EPA near-road monitoring network, which commenced operation in June 2014 and was built in a more tree-lined portion of the Connector on the GIT campus about 500 m north of the RDS site. Outdoor monitoring data farther from the road were collected at an existing network site (Rooftop, or RFT) from a fourth-floor roof lab in the Ford Environmental Science and Technology Building located in the middle of the GIT campus, approximately 500 m west of the Connector, as well as at the Jefferson

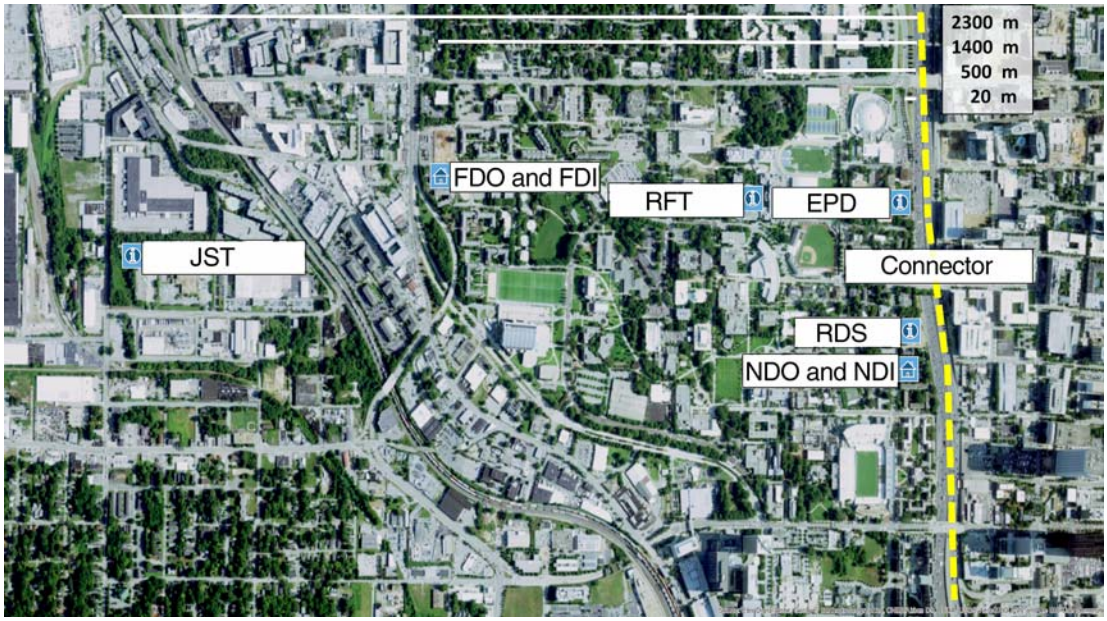

Figure 1. Map of sampling sites: Jefferson Street (JST), far dorm outdoor (FDO) and indoor (FDI), rooftop (RFT), Georgia EPD roadside (EPD), current study roadside (RDS), near-dorm outdoor (NDO) and indoor (NDI), and the I-75/I-85 highway Connector.

Street (JST) site of the Southeastern Aerosol Research and Characterization (SEARCH) network, located 2.3 km (Hansen et al. 2006) west of the Connector. Data from the JST site have been used for generating population exposure estimates examining longitudinal associations between air pollution and acute morbidity in many previous series of the Study of Particles and Health in Atlanta (SOPHIA) (Darrow et al. 2008, 2011; Metzger et al. 2003a, 2003b; Peel et al. 2003; Sarnat et al. 2008b, 2010; Strickland et al. 2010; Tolbert et al. 2000) and are generally considered to be representative of the pollutant concentrations and composition of Atlanta's urban background (Edgerton et al. 2005; Liu et al. 2005; Solomon et al. 2003). Two additional outdoor sites, along with two indoor sites, were located at the two student dormitories and were denoted as the near dorm outside (NDO) site, approximately 20 m west of the Connector, and the far dorm outside (FDO) site, approximately 1.4 km west of the Connector. The two indoor sites were inside the two dorms and were denoted as NDI and FDI for the near and far dorms, respectively.

Primary Instrumentation, Laboratory Analyses, and Multipollutant Methods

Pollutant measurements conducted included both commonly monitored species (e.g., CO and NO_x) as well as more novel measurements (e.g., OP), which included both primary species (e.g., EC) and secondary species related to traffic emissions (e.g., O_3), with the instrumentation varying among the sites (Table 1). The RDS site used the Southeastern Center for Air Pollution and Epidemiology (SCAPE) trailer equipped with a wide range of instruments to measure both gaseous and particulate pollutants as well as meteorological variables. A lab in the Ford Environmental Science and Technology Building provided reference instruments for calibration purposes. The JST site provided an ability to link DRIVE observations to long-term measurements.

Traditional primary traffic-related pollutants — BC, CO, NO_x, and NO_2 — were measured continuously at each sampling site. This information was used to characterize the heterogeneity of traffic pollutants across the sampling domain and to evaluate and calibrate the air quality model. At the RDS site, continuous instrumentation included BC (Aethalometer AE-31, Magee Scientific Company, Berkeley, CA, USA), CO (model 48i, Thermo Electron, Franklin, MA,

Table 1. Summary of Measurements Conducted at Each Monitoring Site[a]

Tier / Site / Measure	Instrument Model	Frequency (*n* = target sample number)
Tier 1: Outdoor		
1a — RDS		
CO	Thermo 48i	Continuous
NO-NO_2-NO_x	Teledyne 200A	Continuous
NO_2	Aerodyne CAPS	Continuous
O_3	Thermo 49C	Continuous
BC	Magee Scientific Aethalometer	Continuous
$PM_{2.5}$ Sulfate	ACSM	Continuous
$PM_{2.5}$ Mass and BC	Gravimetric and Reflectance	Integrated (48-hr)–2/wk
$PM_{2.5}$ OC and EC	Sunset Labs TOR with IMPROVE	Integrated (48-hr)–2/wk
Traffic count and composition	GEOcounts (GA DOT) with Automatic Traffic Recorders	Continuous
Meteorology (temp, RH, wind)	HOBO U30	Continuous
1b — EPD		
CO	Thermo 48i-TLE	Continuous
NO-NO_2-NO_x	Thermo 42i	Continuous
BC	Thermo MAAP 5012	Continuous
1c — NDO, FDO		
CO	Teledyne 300E	Continuous
NO-NO_2-NO_x	Thermo 42C Low Source	Continuous
NO_2	Ogawa badges	Integrated (48-hr)–2/wk * 2 dorms
$PM_{2.5}$ BC	microAeth AE51	Continuous
$PM_{2.5}$ CPC	TSI 3785 (Near); TSI 3022A (Far)	Continuous
$PM_{2.5}$ Mass	GRIMM	Continuous
$PM_{2.5}$ Mass and BC	Gravimetric and Reflectance	Integrated (48-hr)–2/wk * 2 dorms
$PM_{2.5}$ OC and EC	Sunset Labs Aerosol Analyzer with IMPROVE	Integrated (48-hr)–2/wk * 2 dorms
1d — RFT		
CO	Thermo 48C Trace Level	Continuous
NO-NO_2-NO_x	Thermo 42i Trace Level	Continuous
$PM_{2.5}$ BC	Thermo MAAP 5012	Continuous
$PM_{2.5}$ Mass	TEOM 1400a	Continuous
$PM_{2.5}$ Mass and BC	Gravimetric and Reflectance	Integrated (48-hr)–2/wk
$PM_{2.5}$ OC and EC	Sunset Labs Aerosol Analyzer with IMPROVE	Integrated (48-hr)–2/wk
Meteorology (temp, RH, wind)	Davis Vantage Pro2	Continuous
1e — JST		
Gases: CO, NO-NO_2-NO_x, NO_2, O_3	(See Hansen et al. 2012.)	Continuous
$PM_{2.5}$ (mass, ions, OC, EC)		Continuous
$PM_{2.5}$ (metals)		Integrated (24-hr)–1-in-3 day
Meteorology (temp, RH, wind)		Continuous

Table continues next page

[a] Details in Appendix A.

USA), NO_x (model 200A, Teledyne API, San Diego, CA, USA), O_3 (model 49C, Thermo Electron), particle composition, and directly measured NO_2 (Cavity Attenuated Phase Shift, Aerodyne Research, Billerica, MA, USA) (see Table 1 in this report and Appendix A, available on the HEI website). At the RFT site, continuous instrumentation included BC (multi-angle absorption photometer, model 5012, Thermo Electron), CO (48C Trace Level, Thermo Electron), and NO_x (42i Trace Level, Thermo Electron). In addition to continuous and semi-continuous measurements, we collected 48-hour quartz and Teflon filter–based samples for EC, OC, and particle mass analyses. The filters

Table 1 (*Continued*). Summary of Measurements Conducted at Each Monitoring Site[a]

Tier / Site / Measure	Instrument Model	Frequency (n = target sample number)
Tier 2: Indoor		
2 — NDI, FDI		
CO	Teledyne 300E	Continuous
$NO\text{-}NO_2\text{-}NO_x$	Thermo 42C Low Source	Continuous
NO_2	Ogawa badges	Integrated (48-hr)–2/wk* 2 dorms
$PM_{2.5}$ BC	microAeth AE51	Continuous
$PM_{2.5}$ CPC	TSI 3785 (Near); TSI 3022A (Far)	Continuous
$PM_{2.5}$ Mass	GRIMM	Continuous
$PM_{2.5}$ Mass and BC	Gravimetric and Reflectance	Integrated (48-hr)–2/wk * 2 dorms
$PM_{2.5}$ OC and EC	Sunset Labs Aerosol Analyzer with IMPROVE	Integrated (48-hr)–2/wk * 2 dorms
Tier 3–4: Personal Biomonitoring		
Participants		
NO_2	Ogawa badges	Integrated (48-hr)–2/wk/participant * 6 participants/wk
$PM_{2.5}$ Mass	MicroPEM Nephelometer	Continuous
$PM_{2.5}$ Mass and BC	Gravimetric and Reflectance	Integrated (48-hr)–2/wk/participant * 6 participants/wk
$PM_{2.5}$ OC and EC	Sunset Labs Aerosol Analyzer with IMPROVE	Integrated (48-hr)–2/wk/participant * 6 participants/wk
Location Tracking	GPS	Continuous
Time-activity diary (Tier 3)		4/wk/participant * 6 participants/wk (n = 288)
Recall survey (Tier 4)		1/wk/participant * 60 participants/wk (n = 720)
Saliva		1/wk/participant * 60 participants/wk (n = 720)
Bloods (plasma)		1/m/participant * 60 participants/month (n = 240)

[a] Details in Appendix A.

were also used for $FPMOP^{DTT}$ measurements to characterize the FPMOP multipollutant traffic indicator. A total of 55 research-grade instruments and 18 low-cost sensors were used, providing air pollutant concentration data at time scales from minutes to week-long averages.

The sampling instruments located inside the two dorms were identical (Table 1) and used a three-way valve to alternate sampling between indoor and outdoor air. In addition to the instrumentation in the two dorm rooms, integrated NO_2 passive sampling was conducted, using Ogawa badges, both inside and outside of the dedicated sampling rooms, as well as in a subset of the student dorm rooms to provide a means of assessing the representativeness of the sampling sites. The Ogawa badges contained cellulose filters coated with triethanolamine (Ogawa 1998), which were analyzed using spectrophotometric methods.

In the near dorm, instrumentation was placed in a first-floor dorm room that was being used as an administrative office. The room had a microwave, dorm-size refrigerator, and a small space heater running periodically. The dorm was five stories high, built in 1961, and most recently renovated in 2002; the first floor was used for administrative offices, and the top four floors were occupied student rooms (typically with two students per room). Each floor of the dorm was laid out similarly, with about 12 double rooms (11 ft × 15 ft) and a single shared bathroom. Each floor also had a single larger utility room (33 ft × 15 ft) used as a conference room on the first floor, as an exercise room on the second floor, and as kitchenettes on the third through fifth floors. The outdoor sampling inlet tube was located approximately 0.5 m off the ground. The indoor sampling inlet tube was raised approximately 0.25 m off the rug flooring for all continuous instrumentation operating on

the valve. The integrated measurements collected on filters were about 1.5 m off the floor, which was about the height of the window used for the outdoor sampling inlet.

In the far dorm, sampling was conducted in one-half of a two-bedroom suite on the first floor of the five-story building, which was built in 1984. There were seven suites per floor. Each suite had two bedrooms occupied by four students and a bathroom. On each floor there was one kitchen shared by the seven suites. The room in the suite used for sampling was not occupied during the study and was not adjacent to the kitchen.

The outdoor sampling inlet tube was 2 m off the ground, and the indoor sampling inlet tube was raised 0.25 m off the linoleum flooring for all continuous instrumentation operating on the valve. The filters on which the integrated measurements were collected were about 1.5 m off the floor, which was about the height of the window used for the outdoor sampling inlet. Both buildings used two-pipe heating and air conditioning systems that transitioned or started in response to ambient air temperatures. Less heated water was produced for heating when the outside air temperature was above 55° F, and less chilled water was produced for air conditioning when the temperature was below 65° F. The near dorm's system made this transition automatically; the far dorm's system required manual conversion between air conditioning and heating.

The study protocol called for two duplicate quartz filters to be used during both indoor and outdoor sampling at the dorms. Mass from one of the filters was originally intended for use in the characterization of particulate water-soluble organic carbon (WSOC) concentrations. Given the relatively low mass filter loadings, especially for the indoor samples, we chose to extract and composite mass from both duplicate quartz filters to increase analytical sensitivity for use with the DTT analyses (see below). Because WSOC was a pollutant we identified as being less associated with primary traffic emissions at this location (Weber et al. 2007), we felt the trade-off for enhanced confidence in the DTT quantitation was justifiable.

IMSIs The IMSIs were constructed using pollutant concentration measurements that are typically more readily available from routine monitors — namely EC, CO, and NO_x — along with the fraction of these species emitted by gasoline and diesel vehicles, as:

$$IMSI = \frac{\left(\frac{EC_{mob}}{EC_{tot}}\right)_{Emis}\frac{EC}{\sigma_{EC}} + \left(\frac{NO_{x_{mob}}}{NO_{x_{tot}}}\right)_{Emis}\frac{NO_x}{\sigma_{NO_x}} + \left(\frac{CO_{mob}}{CO_{tot}}\right)_{Emis}\frac{CO}{\sigma_{CO}}}{\left(\frac{EC_{mob}}{EC_{tot}}\right)_{Emis} + \left(\frac{NO_{x_{mob}}}{NO_{x_{tot}}}\right)_{Emis} + \left(\frac{CO_{mob}}{CO_{tot}}\right)_{Emis}}, \quad \text{(Eq. 1)}$$

where the scaled concentrations are normalized by the standard deviations σ of the pollutant concentrations and the terms in the parentheses are emission ratios calculated as the fraction of the specific species emissions from mobile sources and total species emissions. The emissions estimates were developed using the Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system and the EPA emissions modeling 2011v6.2 platform (*https://www.epa.gov/air-emissions-modeling/2011-version-62-platform*) with the 2011NEI version 2 emissions inventory (Houyoux and Vukovich 1999). The emissions inventories are from the 4-km grid cell that includes the GIT campus and the JST sampling site. Detailed information about the assumptions, mathematical derivation, and previous use of IMSIs is included in a study by Pachon and colleagues (2012).

FPMOP FPMOP is expressed from the generation of water-soluble ROS catalyzed by PM, also referred to as PM OP. OP has been hypothesized as one mechanism of PM toxicity that may elicit adverse PM-related health effects (Ayres et al. 2008). A variety of PM chemical constituents emitted by vehicles, such as BC, PAHs, oxygenated PAHs such as quinones and hydroxyquinones, and transition metal species, have been linked to ROS generation in previous studies (Cheung et al. 2010; de Kok et al. 2005; Surawski et al. 2010). OP has thus been hypothesized as a particulate property mediating traffic exposure and adverse health response. In the DRIVE study, we used the common DTT acellular assay to measure the OP of the water-soluble fraction ($OP^{WS\text{-}DTT}$) of $PM_{2.5}$ collected at various sampling sites. Briefly, in this assay electrons are transferred from antioxidant DTT to dissolved oxygen with the catalytic assistance of PM, leading to DTT depletion and ROS generation, mimicking a similar process in vivo with physiological antioxidants in place of DTT and resulting in oxidative stress. When this reaction is monitored under conditions of excess DTT, the DTT consumption rate is proportional to the concentrations of redox-active species in the PM extracts. The measured $OP^{WS\text{-}DTT}$ is normalized by volume of sampled air (expressed in units of pmol/min/m^3) to provide a measure of atmospheric concentration of aerosol OP. Alternatively, OP can be normalized by $PM_{2.5}$ mass to provide a measure of the

intrinsic OP of the overall $PM_{2.5}$ (i.e., an indication of the toxicity of the $PM_{2.5}$ in terms of OP). Intrinsic OP also provides insights into the contributions of specific sources to the overall aerosol OP.

Assessment of Data Quality

All field instruments used to measure continuous pollutant concentrations were evaluated, refurbished if needed, and calibrated before field sampling. In order to compare concurrent pollutant measurements across the sampling sites and ensure accurate concentrations during the sampling period, instruments measuring the same pollutant parameters were also colocated at times before, during, and after the 13-week intensive field sampling period. Calibration was done by varying the blend of pollutant gas from a cylinder of known concentration with a cylinder of zero air at given flow rates (Bios DryCal, Mesa Labs, Lakewood, CO, USA). Instrument colocations were conducted for continuous NO–NO_2–NO_x, CO, and integrated $PM_{2.5}$ mass and reflectance over a multiday period, both before and after field sampling, to assess method precision and potential instrument offset. Final concentration data reported were adjusted based on the time-weighted average of the calibration curves and the colocated measurements. Temporal corrections were also necessary for all the measurements conducted at the two dormitory sites because of a slight drift (approximately 10–15 seconds per day) in the timer used to control the value that regulated the change in indoor and outdoor sampling. A full discussion of the data processing and quality assurance/quality control (QA/QC) protocol is presented in Appendix A, available on the HEI website.

All CO instruments were modified by adding a CO scrubber (Parrish et al. 1994) to provide zero air to the instrument for 50% of the sampling period for accurate CO measurements. Because measured CO concentrations are affected by changes in ambient temperature and the sampling locations were not kept at a constant temperature throughout the day, the zero CO concentration measurement varied throughout the day. By measuring the zero air concentration each 15-minute sampling period, the true CO concentrations were calculated by taking the difference in the measured CO concentrations and the zero concentration reading.

For the continuously measured pollutant parameters, the primary unit of temporal aggregation used to calculate completeness was 15 minutes (e.g., 95% completeness indicates that for a specified sampling period, typically 48 hours, 95% of the 15-minute temporal blocks were processed and valid for use in data analysis).

Low-Cost Multisensory Platforms

Interest in the use of low-cost sensors has recently increased because of their convenience and portability as well as their small size, light weight, and low power consumption. Although sensor technology has improved rapidly, there are still concerns about the accuracy of low-cost sensors. Evaluating various low-cost sensors for specific applications and conditions is important to better understand their current capabilities and limitations (Johnson et al. 2016). Low-cost sensors are currently being used in a wide variety of applications: to increase the spatial coverage of measurement data (e.g., Gao et al. 2015), to collect personal exposure data (Nieuwenhuijsen et al. 2015; Steinle et al. 2015), and to make mobile monitoring measurements (Van den Bossche et al. 2015).

After the study commenced and shortly before the intensive portion of the field study began, discussions between our study team and HEI staff led to the study being extended to include the deployment of low-cost sensors to assess their accuracy and potential application in assessing exposures in a near-road environment, building on earlier work at GIT and the availability of such sensors. The study provided an opportunity to deploy a set of low-cost sensors that could run simultaneously alongside reference instrumentation at several different locations. However, earlier work at GIT identified limitations to the various PM sensors that were to be used during the study, mainly their accuracy across a dynamic range of concentration levels, with the result that the addition of the sensors was viewed as an opportunity to assess issues, not to provide more spatial coverage. With reference instrumentation located at varying distances from vehicle emission sources, the sensors could be moved throughout the sampling period to assess how well each sensor captured the variability at various concentration levels. In addition to the PM sensors, gas-phase pollutant sensors were tested as part of the study. The use of the low-cost sensors required additional electronic components that needed to be weather protected. Deployment in the study provided a platform to test improvements made to previously designed low-cost sensor packages (Johnson et al. 2016). Although this part of the project was added at the start of the sampling period, the availability of previously designed and tested low-cost sensor units enabled rapid deployment with only minor modifications.

Three different low-cost PM sensors were tested for accuracy compared with reference instruments and for consistency between the various sensor models. Because the sensors used different measurement techniques and had different reported limits of detection (LODs), it was important to determine which sensor would work best in the study environment. Gaseous pollutants (CO, NO, NO_2,

and O_3) were also measured by low-cost sensors to assess their accuracy. Additional details about the various sensor models used and an explanation of how they work can be found in Appendix C, available on the HEI website.

The sensors were configured in four multisensory units so that they could run simultaneously at the various sampling locations. In each multisensory unit, an Arduino Mega microcontroller (model 2560, Arduino, *www.arduino.cc*) was used with a data-logging shield and real-time clock (product ID 1141, Adafruit, New York, NY, USA) to log the sensor's analog signal at one-minute averages as comma-separated values on a secure digital memory (SD) card. The sampling boxes were 11.81 in × 7.87 in × 6.39 in opaque plastic junction boxes. Inlet and outlet holes were drilled into each box, and a 25-mm fan was placed at the exhaust to draw air through the box. As designed, the multisensory units had various configurations of sensors to achieve the specific objectives of the low-cost sensor deployment and to maximize the utility of the available equipment. Each box also contained temperature and relative humidity (RH) sensors (Sensirion SHT15, Staefa, Zurich, Switzerland) to measure conditions inside the box.

The first multisensory unit contained three different low-cost PM sensors that were tested for accuracy compared with the reference instrument and for consistency among the sensor models. This unit was placed at the NDI site for 3 weeks, at the FDO site for 2 weeks, and at the RDS site for 1 week. The second unit contained one PM sensor and four gas-phase electrochemical sensors measuring CO, NO_x, NO_2, and O_3. It was placed outside at the NDO site for approximately 1 week (November 15–25), at the RDS site for 2 weeks, and at the FDO site for 1 week. The last two multisensory units contained a single PM sensor. One unit was placed outside at the FDO site for 5 days and outside at the NDO site for 1 week. The other unit was placed at the RDS site for 5 days, indoors at the NDI site for 2 weeks, and outside at the NDO site for 1 week. (The complete rotation schedule is shown in Table C.3 in Appendix C, available on the HEI website.)

Given the late addition of this component of the sampling, much of the sensor calibration was conducted during and after the intensive sampling period. Equations for converting recorded electrical signals to particle mass concentration were not provided by the manufacturer (Shinyei 2002; Shinyei Technology Co. 2010, 2013). We used an empirical calibration, via Deming regression modeling (Linnet 1993), based on the 1-hour average concentrations recorded by the sensors and reference instruments. The calibration was developed between the sensor total PM concentrations and the reference $PM_{2.5}$ measurements in order to understand how well the sensor could capture the $PM_{2.5}$ trend. It is important to note that the particle sensors measure using light scattering. The particles are detected in the light source, and the output is the ratio between the length of time when the pulses occur and the total time. This fraction can be influenced by the particles' properties, mainly their chemical composition and size distribution, which vary with location. Laboratory studies have found that such outputs can vary by a factor of 10 depending on composition (Wang Y et al. 2015) and by a factor of 12 depending on size (Austin et al. 2015). Therefore, assessing and calibrating to the local ambient aerosols is important and adds to the uncertainly of the measurements.

The gas sensors are electrochemical cells that generate a current that is linearly proportional to the concentration of the pollutant. Each gas sensor was provided with a sensor-specific calibration provided by the manufacturer. The CO, NO, and NO_2 sensors were calibrated using only the provided equation. The O_3 sensor required further calibration incorporating the reference instrument measurements as well as the temperature and RH. See Appendix C, available on the HEI website, for calibration details.

PANEL STUDY (TIERS 3 AND 4)

Participant Recruitment

To address specific aim 3, we conducted a nested panel study (from September 8, 2014, to January 5, 2015) as a component in the indoor and outdoor monitoring campaign. We recruited a cohort of students living in the same dormitories that housed the NDI and FDI sampling instrumentation. Exclusion criteria included smokers or those currently living with smokers. Individuals were compensated up to $300 for their participation throughout the study. Recruitment was conducted during a 3-week period to accommodate dorm move-in dates and the start of the fall school semester. Recruitment occurred on-site at the dorms by researchers in accordance with pre-established protocols and the Institutional Review Board at GIT. Recruitment flyers were posted in each of the dorms, and a series of informational sessions was held at each of the dorms in early August. Of the 28 and 38 students who signed consent forms to participate from the near dorm and far dorm, respectively, 26 from the near dorm and 31 from the far dorm were enrolled based on an assessment of their availability during the semester and likely compliance with the study protocol. During the 12-week personal sampling period, two participants from the near dorm and one from the far dorm dropped out of the study. No specific reasons were given for this attrition. In total, 54 students participated in the study.

Once enrolled, participants were given a baseline questionnaire detailing sociodemographic information,

preliminary health, and typical time–activity patterns. The participants were then informed of the weekly personal exposure monitoring and were given the opportunity to volunteer through an electronically distributed scheduling system. Of the 54 participants, 51 chose to participate in the personal exposure sampling, 23 from the near dorm and 28 from the far dorm.

Personal Exposure Monitoring

As part of the panel study, we conducted personal exposure monitoring during the entire 12-week-long data collection period to coincide with times when students were on campus (i.e., no monitoring was conducted during the Thanksgiving holiday break). Each week approximately six students participated in two consecutive 48-hour personal exposure sampling sessions. On Mondays, the participants were given personal sampling packs to measure 48-hour integrated personal $PM_{2.5}$, EC, and NO_2 exposures (Monday A.M. to Wednesday A.M.). On Wednesdays of the same week, field staff met with the participants to replace filters and batteries for the second 48-hour sampling period (Wednesday A.M. to Friday A.M.).

The personal sampling packs weighed approximately 3 lbs and were easily attachable to the strap of a backpack or bag in order to be near the participants' breathing zone with minimal discomfort or alteration to their daily activity. Continuous $PM_{2.5}$ concentrations were measured via nephelometry by a personal sampling device (MicroPEM V3.2A, RTI International, Research Triangle Park, NC, USA). The MicroPEMs also contained 37-mm Teflon filters (Gelman Sciences, Ann Arbor, MI, USA) that collected particles through the sampling inlets at a 0.5-L/min flow rate. Particle mass was determined via gravimetric analysis on the 48-hour integrated filters in a temperature- (18°–24° C) and RH- (40 ± 5%) controlled weighing room by a trained staff member. Personal $PM_{2.5}$ concentrations were calculated using the mass measurements and the total air volumes recorded by the personal sampling devices. The gravimetric filter mass was then processed and used to correct the continuous nephelometer output. The BC components of $PM_{2.5}$ collected on the MicroPEM filters were measured via reflectance (model M43D, EEL Smokestain Reflectometer, Diffusion Systems, London, UK) and following protocols established for the European Study of Cohorts for Air Pollution Effects (ESCAPE Reflectance Standard Operating Procedure, RUPIOH 4.0, 2002); personal BC concentrations were expressed as absorption coefficients (unitless). NO_2 was collected using Ogawa passive samplers (Ogawa & Company, Pompano Beach, FL, USA) containing cellulose filters coated with triethanolamine (Ogawa & Company 1998), which were analyzed using spectrophoto-metric methods.

Time–activity pattern data were collected through portable global positioning system (GPS) trackers that were attached to the side of the pack and recorded participant locations continuously over the two consecutive 48-hour cycles. Locations within a radius of 20 m where the participant stayed for more than 5 minutes were collected as waypoints (i.e., longitude and latitude). To eliminate recall bias that may have occurred, we chose to use GPS trackers instead of diaries or questionnaires to analyze time–activity patterns. Distances from the waypoints to the Connector were calculated using a geographic information system (ArcMAP version 10.4.1, Esri, Redlands, CA, USA) and were calculated based on the shortest distance from the Connector, because it was the main source of traffic pollutants during the sampling period. The logged GPS data were then used to aid in quantifying time spent in various microenvironments and proximity to traffic sources as potential modifiers of personal exposures. Specifically, each GPS location was categorized as an indoor or outdoor environment and by campus area (near, center, far, or other).

During the study, we also modeled traffic-related CO, NO_x, and $PM_{2.5}$ concentrations hourly throughout the sampling domain at a 25-m × 25-m grid resolution using the fine-scale dispersion model RLINE (Batterman et al. 2014b; Snyder et al. 2013). To estimate traffic-related personal exposures, we integrated participant GPS locations with corresponding on-road dispersion modeling output. (Full details on the RLINE models, assumptions, limitations, and modeled pollutant dispersion results are in Appendix D, available on the HEI website.)

For the personal exposure samples, which included filters with low mass loadings, we conducted DTT analyses using a time-intensive manual dilution and analysis method that we had hoped would provide greater analytical sensitivity to measure concentrations in this mass loading range. Unfortunately, results from the method were not successful, and we were ultimately unable to quantify personal DTT exposure.

Biomonitoring and Metabolomics Analyses

All 54 students participated in the environmental metabolomics analysis by contributing up to four (monthly) venous blood and twelve (weekly) saliva samples. In total, 175 plasma and cell samples (average of 3.2 repeated samples per participant) and 621 2-ml vials of saliva (average of 11.5 repeated samples per participant) were collected. Metabolomics analyses were conducted on the samples, using liquid chromatography–mass spectrometry techniques (Q Exactive HF hybrid quadrupole-Orbitrap, Thermo Fisher Scientific, Waltham, MA, USA). Two technical columns, one in hydrophilic positive ion mode and one in C18 hydrophobic negative ion mode, were used to

detect metabolites in each sample, with triplicate runs of each sample conducted in both columns. Only the features exhibiting a median coefficient of variation among the triplicate measures of less than 30% and a pairwise Pearson correlation within the triplicate greater than 0.7 were included in the subsequent analyses. Following quality assessment, the triplicate measures of each sample were averaged, and two R packages, apLCMS and xMSanalyzer (created internally at Emory), were used to extract metabolite features. Finally, only the metabolites present in at least 15% of all samples (by biological media) were included in the final statistical analyses; this criterion was established in order to enhance generalizability and to reduce the possibility that comparisons of metabolic profiles between dorm cohorts (the main comparison of interest; see below) were unduly influenced by individual samples or participants. A log2 transformation, followed by quantile normalization, was performed to reduce the effects of technical errors on downstream statistical analysis and biological interpretation. (Further details on the technical aspects of metabolomics analyses can be found in Appendix E, available on the HEI website.)

DATA ANALYSIS METHODS

Our analyses consisted of comparisons of both single- and multipollutant traffic indicators. The single-pollutant indicators included CO, NO, NO_2, EC, and $PM_{2.5}$. The multipollutant indicators included IMSI and FPMOP. Additional analyses were conducted as part of aim 5, the low-cost sensor technology assessment.

Aim 1 Analysis: Associations among Indicators at a Near-Road Site

Aim 1 focused on characterizing pollutant levels, pollutant composition, and temporal variability of pollutant concentrations at the RDS monitoring site, which served as the near-road anchor site. To address this aim, we conducted descriptive statistics, pairwise pollutant correlations, and regression analyses.

Specifically, to evaluate associations more closely among several key physical and atmospheric factors and the corresponding pollutant concentrations or IMSI values at the RDS site, we conducted linear mixed modeling. Multivariate linear mixed regression modeling was conducted to assess factors that affected temporal variability in the concentrations of each single pollutant or IMSI values measured at the site. The basic form of the model was as follows:

$$RDS_t = \beta Z_t + \theta_t + \varepsilon_t, \quad \text{(Eq. 2)}$$

where RDS_t denotes the concentration of BC, CO, NO, NO_2, NO_x, O_3, or IMSI values at the site during hour t, and β is the coefficient of interest that describes the influence of factor Z_t on the hourly RDS pollutant level. Factors of interest included time period of the day (categorical), temperature, wind speed, RH, wind direction (categorical), day of the week (categorical), and traffic counts; the factors were included simultaneously in models predicting each pollutant. Finally, θ_t represents time-specific random intercepts used to capture potential variations not explained by Z_t, and ε_t represents residual random normal error. For these and all of the subsequent mixed effects models, covariance was modeled using a lag-one autoregressive matrix.

Aim 2 Analysis: Spatiotemporal Variability of Indicators at Multiple Near-Road Sites

For Aim 2, we assessed the spatiotemporal variability of outdoor pollutant levels and IMSIs along the near-road to mid-distance spatial gradient from the Connector. As part of this aim, we evaluated factors affecting the observed strengths of associations, including traffic patterns and meteorology. To address this aim, much as for Aim 1, we also conducted descriptive statistics, inter-site pollutant correlations, and multivariate linear mixed effect modeling. Regression analyses enabled formal assessment of spatiotemporal variability in pollutant levels among the outdoor monitoring sites, particularly as a function of proximity to the Connector. We considered the following regression model:

$$Ratio_{st} = \beta Z_{st} + \theta_t + \varepsilon_{st}, \quad \text{(Eq. 3)}$$

where $Ratio_{st}$ denotes the ratio of the pollutant concentration measured at site s during hour t to the pollutant concentration at the RDS reference site during hour t. Here, s indexed the five additional outdoor monitoring sites: the EPD near-road site, the NDO site, the RFT rooftop site, the FDO site, and the SEARCH network's JST site. β is the coefficient of interest that describes the influence of factor Z_{st} on spatial gradients of the pollutant or IMSI measures. Factors of interest included proximity from the monitoring site s to the RDS site (distance), time period of the day (categorical), temperature, wind speed, RH, wind direction (categorical), day of the week (categorical), and traffic counts. The factors were included in separate models predicting the ratios for each pollutant; interactions between proximity of the monitoring site to the RDS site and each factor were also considered. Finally, component θ_t is the time-specific random intercept used to capture potential variations not explained by Z_{st}, and ε_{st} represents residual random normal error.

Aim 3 Analysis: Associations Between Traffic Indicators, Indoor Pollutants, and Personal Exposures

For Aim 3, we evaluated how well several different indicators reflected personal exposures to traffic pollution, as follows:

$$Personal_{i(s)t} = \beta Traffic_t + \theta_t + \varepsilon_{i(s)t}, \quad \text{(Eq. 4)}$$

where $Personal_{i(s)t}$ denotes the personal exposure to BC, NO_2, or $PM_{2.5}$ for participant i from dorm s (i.e., near dorm or far dorm) during 48-hour period t, and β is the coefficient describing the influence of *Traffic*, the indicator of traffic of interest, on personal exposure levels. The various traffic indicators were assessed in separate models and included (1) the directly measured BC and NO_2 concentrations at the RDS site during 48-hour period t, (2) the directly measured BC and NO_2 concentrations at the participants' respective outdoor dorm monitoring site (NDO or FDO) during 48-hour period t, (3) the directly measured BC and NO_2 concentrations at the participants' respective indoor dorm monitoring site (NDI or FDI) during 48-hour period t, (4) time-weighted 48-hour mean proximity in meters of a participant to the highway source during personal sampling, (5) the participant's dorm residence (categorical), and (6) RDS IMSI levels during 48-hour period t. A product term between *Traffic* and the participants' dorm was also examined to evaluate potential effect modification. R^2 was calculated fitting the predicted value against the observed value as a criterion in evaluating the performance of each of the six indicators in predicting personal exposures to BC and NO_2.

Aim 4 Analysis: Differences in Exposures and Metabolomic Changes in Study Participants

For Aim 4, we were interested in examining whether differences in indicators of primary traffic pollution were associated with corresponding metabolomic changes in the study participants. To do so, we followed an untargeted metabolomics workflow, where differences in metabolic profiles were analyzed without prior knowledge of their chemical identity. As an initial step toward this aim, we conducted random effect linear models to assess associations between metabolite feature intensity (i.e., relative concentration) and the participant's dormitory location (near dorm versus far dorm) as the primary exposure indicator of interest:

$$\begin{aligned} Y_{hi(s)t} = {} & \mu + \theta_{i(s)} + \beta_1 Dorm_s + \beta_2 Age_{i(s)} \\ & + \beta_3 Sex_{i(s)} + \beta_4 BMI_{i(s)} + \beta_5 Race_{i(s)} \\ & + \beta_6 Collegeyr_{i(s)} + \beta_7 Movingdays_{i(s)t} \\ & + \beta_8 Timepoints_{i(s)t} + \varepsilon_{hi(s)t}, \end{aligned} \quad \text{(Eq. 5)}$$

where $Y_{hi(s)t}$ refers to intensity (i.e., relative concentration) of metabolite h for participant i from dorm s on biosampling date t. Separate models were conducted for each metabolite, from each of the four biomatrix columns (plasma positive ions, plasma negative ions, saliva positive ions, and saliva negative ions). μ is the fixed-effect intercept and a random effect. $\theta_{i(s)}$ was included to control for potential between-participant variation. $Dorm_s$ refers to the dorm location for participant i and was our primary variable of interest. Other covariates were included to control for potential confounding by age, sex (categorical), body mass index (continuous), race (categorical), and college year (categorical). We also controlled for $Movingdays_{i(s)t}$, the total number of days between the biosample collection date and the date that participant i moved into dorm s, and $Timepoints_{i(s)t}$, the month number for plasma or week number for saliva when the biosample was collected from participant i in dorm s. $\varepsilon_{hi(s)t}$ represents residual random normal error. Hypothesis tests to identify differentially expressed features between the near dorm and far dorm comparison groups (by biomatrix column) were adjusted for multiple comparisons using the Benjamini–Hochberg false discovery rate procedure. Results for equation 5 are presented using Manhattan plots, which plot the retention time of each metabolite feature on the x-axis against the negative logarithm of its P value on the y-axis.

Future analyses, currently beyond the scope of this report, will include assessment of traffic indicators beyond the dorm locations (e.g., outdoor pollutant concentration levels at the RDS or other sites) as predictors of metabolite intensity in equation 5. In addition, pathway and functional analysis using an untargeted network analysis approach will be conducted (Li et al. 2013), along with validation of the chemical identity of specific features via tandem mass spectrometry analyses.

Aim 5 Analysis: Performance of Low-Cost Sensors as Indicators of Pollutant Exposure

For Aim 5, we compared the concentration measurements from the low-cost sensors with those from the relevant reference instrument using least square linear fitting and evaluated mean differences in the absolute concentrations and the 95% confidence intervals between the reference instrument and the sensor values. The comparisons were conducted at the individual monitoring sites to assess how well the sensors captured concentrations across a range of pollutant levels. For the PM sensors, a continuous monitor (model 1.109 optical particle counter, Grimm Aerosol, Ainring, Germany) located at the NDO site was used as the reference instrument, because it was the only reference PM instrument available. For the gas sensors, the CO and NO_x

instruments at each site were used to check the manufacturer calibration curves in each rotation location. Because further calibration was required for the O_3 sensor, correlation with the reference O_3 instrument was assessed at each calibration step to verify that the correlation was improving with each additional calibration. The equation and correlation for each sensor at each location are shown in Appendix C.

RESULTS

EXECUTION OF STUDY DESIGN

The centerpiece of the study was an intensive multimonth field sampling campaign, consisting of eight outdoor and indoor monitoring sites concurrently measuring numerous traffic- and non-traffic-related air pollutants, dispersion modeling, and a nested panel study of 54 participants that included both personal exposure measurements and biomonitoring for markers of traffic exposure. Participant recruitment, sampler deployment, staff field training, and site synchronization were conducted under unusually accelerated conditions, given the study design and its linkage to the GIT academic semester. Student recruitment and designation of indoor sampling sites, for example, could not be undertaken until after the students arrived on campus for the start of the school year. Despite this, we were able to complete the targeted number of sampling weeks successfully and largely adhere to the original research objectives and study scope.

Overall, we were satisfied with the data quality for the measured and modeled pollutants as well as the biological samples collected. A considerable number of the existing air monitoring resources belonged to the GIT and Emory researchers; the study also used other existing state monitoring resources. Additional monitors were loaned by non-GIT and Emory researchers, requiring additional calibration. Our assessments of completeness indicated acceptable levels of data loss for the targeted data parameters during the study (see Appendix Table A.1, available on the HEI website). Data losses throughout the sampling period were primarily caused by technical problems (power outages and instrumentation malfunction) and field technician error. In particular, we experienced data loss for indoor and outdoor particle count concentrations at both dorm sites because of sampler malfunctions. Finally, because of a malfunction of the indoor–outdoor valve switching mechanism at the far dorm between September 19 and October 3, only indoor levels for continuously measured pollutants were measured on these dates. Completeness for the continuously measured BC concentrations was variable and was similarly associated with suboptimal performance of the personal aethalometers (microAeth model AE51, AethLabs, San Francisco, CA, USA) used in the dorm sampling locations. (Complete information on data processing and quality are presented in Appendix A, available on the HEI website.)

Results for each of the five aims are presented below.

AIM 1: CHARACTERIZING ROADSIDE PATTERNS OF SINGLE-POLLUTANT AND MULTIPOLLUTANT TRAFFIC INDICATORS

The RDS near-road site provided key measurements to assess the temporal variability of the traditional and multipollutant primary traffic pollutants (Appendix B, available on the HEI website, contains complete descriptive statistics for the pollutant and meteorological parameters). Easterly winds (i.e., from the Connector toward the campus monitoring sites) predominated for more than 70% of the time, enhancing direct influence from the highway emissions (see wind rose in Appendix B). Mean pollutant concentrations measured at the RDS site were typically higher than the urban background level; however, they were not as high as those reported in previous near-road field studies (Beckerman et al. 2008; Fruin et al. 2008; Fujita et al. 2011; Kozawa et al. 2009; MacNaughton et al. 2014; Zhu et al. 2008). The key pollutant species measured at the RDS site were NO, NO_2, CO, BC, and O_3, with mean concentrations (standard deviation) of 20.9 (23.5) ppb, 29.1 (15.6) ppb, 424.9 (209.8) ppb, 1.6 (1.3) $\mu g/m^3$, and 18.2 (11.7) ppb, respectively. Direct NO_2 measurements were also made at the RDS site, with a mean concentration of 22.0 (11.7) ppb. The near-road concentrations recorded at the EPD site (e.g., mean NO_2 levels of 19.5 [8.6] ppb during the study), which is part of the U.S. EPA's national near-road monitoring network, were also low compared with historical near-road pollutant concentrations. The levels at the RDS and EPD sites were, however, entirely consistent with other current near-road monitoring results throughout the United States. For example, initial monitoring results from approximately 50 near-road monitoring network sites in 2014 showed annual NO_2 means ranging from 9 to 24 ppb (U.S. EPA 2016b). These relatively low near-road concentrations are reflective of broad emissions reductions throughout the U.S. traffic fleet, and the findings are indicative of a changing near-road environment.

The temporal variability of pollutant concentrations, IMSIs, and traffic counts at the RDS site was characterized using time-series and diurnal profile plots (Appendix B). In order to compare the diurnal profiles of the species, traffic conditions, and meteorological conditions, a single diurnal

profile was generated by normalizing the diurnal profiles to the overall mean of each pollutant (Figure 2). The diurnal profiles for BC, CO, and NO_2 concentrations were similar, and because the IMSI values were developed from the concentrations of these species, the IMSIs also showed a similar profile: a morning peak in concentrations at 10 A.M., an evening peak at 10 P.M., and minimum concentrations at 3 A.M. and 4 P.M. Diurnal O_3 profiles showed a pronounced concentration peak at 3–4 P.M. and a minimum at 6–7 A.M.

Interestingly, the diurnal profile of traffic counts also differed from that observed for the traffic-related pollutants (Figure 2), with traffic counts rising quickly from 5 A.M. to 7 A.M., rising more slowly until the maximum at 3 P.M., dropping slowly until about 7 P.M., and then dropping off more quickly until the minimum at 3 A.M. Indeed, traffic counts alone were shown to be poorly associated with the corresponding roadside pollutant measurements (Table 2).

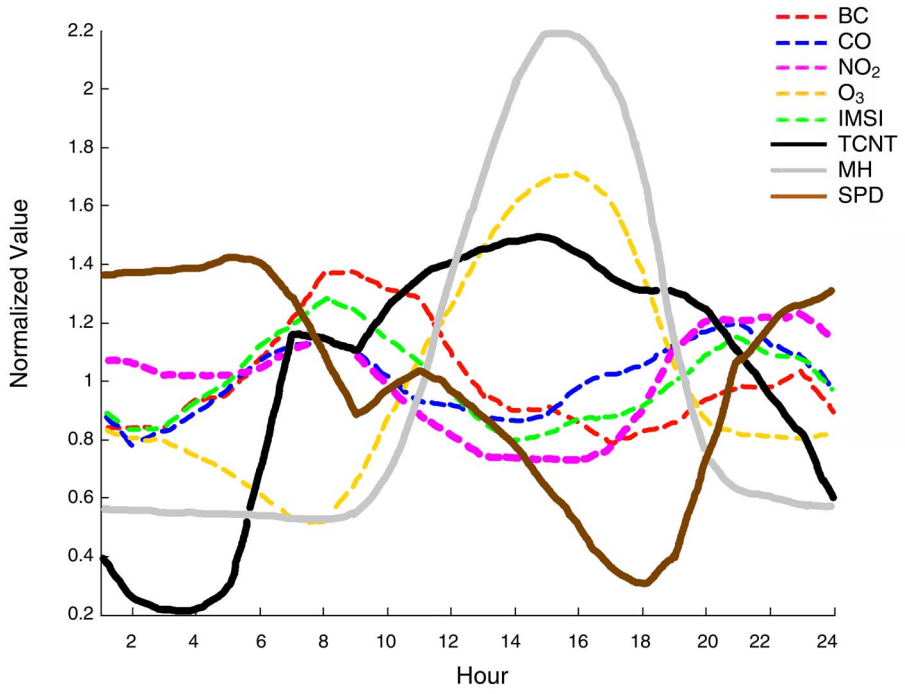

Figure 2. Normalized diurnal concentration profiles for BC, CO, NO_2, O_3, and the IMSI as well as traffic count (TCNT), mixing height (MH), and traffic speed (SPD) for the RDS site from September 1 to December 31, 2014.

Table 2. Pairwise Correlation Coefficients for Roadside Hourly Pollutant Concentrations and Selected Environmental Factors, During Periods When Wind Originated from the East

Species / Factor	Temp (°F)	RH (%)	1/ Mixing Height	Traffic Count	Wind Speed	Traffic Count/ Mixing Height
NO (ppb)	0.00	0.22	0.28	0.00	0.39	0.00
NO_2 (ppb)	0.10	0.00	0.41	0.00	0.69	0.10
CO (ppb)	0.10	0.00	0.22	0.00	0.49	0.10
O_3 (ppb)	0.53	0.57	0.57	0.30	0.45	0.53
BC ($\mu g/m^3$)	0.10	0.22	0.30	0.00	0.49	0.10
IMSI	0.00	0.30	0.40	0.00	0.58	0.00

Mean diurnal traffic speeds, an indicator of congestion patterns, similarly, did not correspond with diurnal trends observed for the RDS primary pollutant levels. Specifically, we observed traffic counts rising at 6 A.M., with corresponding reductions in mean traffic speeds from approximately 70 mph to 40 mph by 8 A.M. Traffic count remains high throughout the day, with mean traffic speeds increasing to 45 mph at 10 A.M. before dropping to 15 mph at 5 P.M. Because estimated emission rates remain fairly constant above 20 mph (Appendix B, Figure B.5, available on the HEI website) and traffic counts remain elevated throughout the daytime, congestion variability throughout the day cannot explain the variability in the observed diurnal pollutant concentrations. Moreover, diurnal emissions trends are highest when the observed traffic-related primary species levels are low.

To better understand the role of traffic count and congestion as predictors and potential surrogates of exposure to primary traffic pollution, we conducted pairwise correlation analyses between RDS pollutant concentrations and key physical and meteorological indicators, including vehicle traffic count and mixing height. In order to control for the influence of wind direction on measured concentrations at the study monitoring sites, we limited analyses to periods when winds originated from the east (Table 2). Mixing height data were generated using the weather research and forecasting (WRF) model (version 3.6) to generate the meteorological fields. The modeling system nests its 4-km grid in a 12-km mother grid covering Georgia and portions of neighboring states and uses a 36-km outer grid over the continental United States to provide boundary conditions. The WRF was initialized and constrained at the boundaries using analysis products from the North American Mesoscale model (*nomads.ncdc.noaa.gov*). Mixing heights remained low until about 10 A.M., increased during the day until reaching a maximum at about 3 P.M., then decreased until approximately 10 P.M. The results show that increased mixing height (using the inverse of mixing height in the analysis) was correlated with decreased concentrations for all of the pollutants except O_3. Increased mixing height is typically associated with more rapid, convectively driven turbulence (along with mechanical turbulence generated by vehicles and the wind), likely dispersing primary pollutants more rapidly than during the night and early morning hours, when convection is lower (Seinfeld and Pandis 2006).

Increasing temperature (related to increased photolytic activity) and decreasing RH (associated in part with decreasing cloudiness) were noticeably correlated with increasing O_3 concentrations but not strongly correlated with any of the other pollutant concentrations. Photochemical activity drives both O_3 formation and mixing height, resulting in peaks for both parameters between 1 P.M. and 6 P.M. In addition, a larger mixed layer will contribute O_3 from aloft from prior-day formation, further increasing ground-level concentrations. Traffic count, as noted above, was not strongly correlated with pollutant concentrations; however, for most primary pollutants we did observe a slightly higher correlation between traffic count divided by mixing height than the inverse of the mixing height alone, indicating a potential, though minor, impact. Increasing wind speed had the largest effect on the concentration measurements, leading to negative correlations for all species except O_3. Overall, at the RDS site, the observed trends in primary traffic-related pollutants were explained more by rapid pollutant dispersion, particularly associated with the increase in convective mixing and the resulting increase in mixing height along with increased wind speeds.

As shown in Table 3, multiple factors were significantly associated with the levels of the traffic-related single pollutants and IMSI at the RDS site. Different time periods over the course of the day had different RDS concentrations; however, the magnitude and statistical significance of the effects varied by pollutant. RDS BC and NO concentrations, for example, were highest during the morning periods, when mixing height was low and traffic counts were increasing, than during other periods of the day — a result that was not the case for NO_2 and CO. The afternoon period, when mixing height and traffic counts were both high (Figure 2), had lower primary pollutant concentrations than did most other times of day. The effect of the weekend was another important predictive factor for all pollutants (except CO), with significantly lower concentrations of BC, NO, NO_2, and NO_x on weekends compared with weekdays, indicative of the decreased presence of diesel trucks.

As shown in the earlier analysis (Table 2), temperature was positively associated with CO and O_3 levels, indicative of sunlight-induced photochemical formation for both pollutants. Increasing wind speed and wind direction, specifically easterly or southerly with respect to the RDS site, was found to enhance dispersion and reduce roadside level of all the pollutants except O_3. IMSI levels were associated with the same factors. This finding was not altogether unexpected, given the associations between these factors and the constituent IMSI components: CO, NO_x, and BC.

AIM 2: CHARACTERIZING SPATIAL AND TEMPORAL VARIABILITY PATTERNS OF SINGLE-POLLUTANT AND MULTIPOLLUTANT TRAFFIC INDICATORS

When assessing overall spatial gradients in the traditional single-pollutant indicators, all (except NO_2) showed steep gradients of decreasing concentrations with

Table 3. Regression Coefficients from Multivariate Models Examining Associations Between Multiple Factors and Hourly Pollutant Concentrations at the RDS Site[a]

	Estimate of Coefficient (95% CI)	Estimate of Coefficient (95% CI)	Estimate of Coefficient (95% CI)
	BC (μg/m³)	**CO (ppb)**	
Late evening (9 PM–12 AM)	0.27 (0.08 to 0.46)	11.56 (−21.26 to 44.38)	
Midnight & early morning (1–5 AM)	0.26 (0.06 to 0.47)	−4.66 (−40.85 to 31.53)	
Morning rush hour (6–9 AM)	0.43 (0.18 to 0.68)	−6.20 (−49.67 to 37.27)	
Midday (10 AM–3 PM)	0.10 (−0.07 to 0.28)	−31.86 (−62.41 to −1.32)	
Temperature	0.01 (0.00 to 0.02)	2.17 (0.56 to 3.77)	
Relative humidity	0.00 (0.00 to 0.01)	−0.16 (−1.08 to 0.76)	
Wind speed	−0.22 (−0.27 to −0.16)	−42.32 (−51.51 to −33.12)	
Winds from the north	−0.28 (−0.47 to −0.08)	−37.45 (−71.33 to −3.56)	
Winds from the east (from the highway)	−0.74 (−0.96 to −0.52)	−60.84 (−98.68 to −23.01)	
Winds from the south	−0.33 (−0.79 to 0.14)	−10.69 (−87.48 to 66.11)	
Weekend	−0.61 (−0.96 to −0.25)	−10.73 (−73.56 to 52.09)	
Traffic counts (per 1,000)	0.05 (0.04 to 0.07)	8.60 (6.56 to 10.64)	
	NO (ppb)	**NO_2 (ppb)**	**NO_x (ppb)**
Late evening (9 PM–12 AM)	5.27 (1.89 to 8.65)	1.16 (−0.73 to 3.05)	6.46 (1.86 to 11.05)
Midnight & early morning (1–5 AM)	5.80 (2.13 to 9.47)	−1.60 (−3.65 to 0.46)	4.19 (−0.80 to 9.18)
Morning rush hour (6–9 AM)	8.08 (3.69 to 12.46)	−0.92 (−3.39 to 1.54)	7.13 (1.17 to 13.10)
Midday (10 AM–3 PM)	2.84 (−0.30 to 5.97)	−2.68 (−4.43 to −0.93)	0.11 (−4.14 to 4.36)
Temperature	−0.12 (−0.28 to 0.04)	0.01 (−0.08 to 0.10)	−0.11 (−0.33 to 0.11)
Relative humidity	0.23 (0.13 to 0.32)	−0.11 (−0.16 to −0.05)	0.12 (−0.01 to 0.25)
Wind speed	−1.97 (−2.93 to −1.02)	−5.30 (−5.83 to −4.77)	−7.25 (−8.55 to −5.95)
Winds from the north	−5.71 (−9.29 to −2.14)	−4.51 (−6.49 to −2.52)	−10.25 (−15.09 to −5.40)
Winds from the east (from the highway)	−19.17 (−23.16 to −15.18)	−10.48 (−12.70 to −8.27)	−29.72 (−35.13 to −24.31)
Winds from the south	−21.02 (−29.36 to −12.68)	−3.91 (−8.55 to 0.73)	−24.98 (−36.29 to −13.66)
Weekend	−8.97 (−14.80 to −3.15)	−4.81 (−8.05 to −1.57)	−13.78 (−21.82 to −5.73)
Traffic counts (per 1,000)	1.08 (0.87 to 1.29)	0.48 (0.36 to 0.60)	1.56 (1.28 to 1.84)
	O_3 (ppb)	**IMSI**	
Late evening (9 PM–12 AM)	0.81 (−0.26 to 1.89)	0.15 (0.03 to 0.27)	
Midnight & early morning (1–5 AM)	1.73 (0.57 to 2.90)	0.12 (−0.02 to 0.25)	
Morning rush hour (6–9 AM)	1.33 (−0.12 to 2.78)	0.15 (−0.02 to 0.31)	
Midday (10 AM–3 PM)	2.05 (1.06 to 3.03)	−0.03 (−0.14 to 0.09)	
Temperature	0.42 (0.37 to 0.48)	0.00 (−0.01 to 0.01)	
Relative humidity	−0.25 (−0.28 to −0.22)	0.00 (0.00 to 0.01)	
Wind speed	2.69 (2.39 to 2.99)	−0.22 (−0.26 to −0.18)	
Winds from the north	1.91 (0.80 to 3.02)	−0.21 (−0.33 to −0.10)	
Winds from the east (from the highway)	5.24 (4.00 to 6.48)	−0.47 (−0.60 to −0.34)	
Winds from the south	3.80 (1.17 to 6.42)	−0.29 (−0.56 to −0.02)	
Weekend	2.18 (0.14 to 4.23)	−0.28 (−0.56 to 0.01)	
Traffic counts (per 1,000)	−0.30 (−0.36 to −0.23)	0.04 (0.04 to 0.05)	

[a] All covariates were included simultaneously in the model for each pollutant of interest.

increasing distance from the highway, with the steepest part of the gradient within the first 20 m (Figure 3 and Figure 4; and Appendix Table B.1, available on the HEI website). The mean CO concentration at the RDS site, for example, was approximately 400 ppb and decreased to roughly 200 ppb at the far dorm site, pointing to the impacts of primary vehicle emissions (Figure 3, Appendix Table B.1). Outdoor and indoor CO concentrations at the

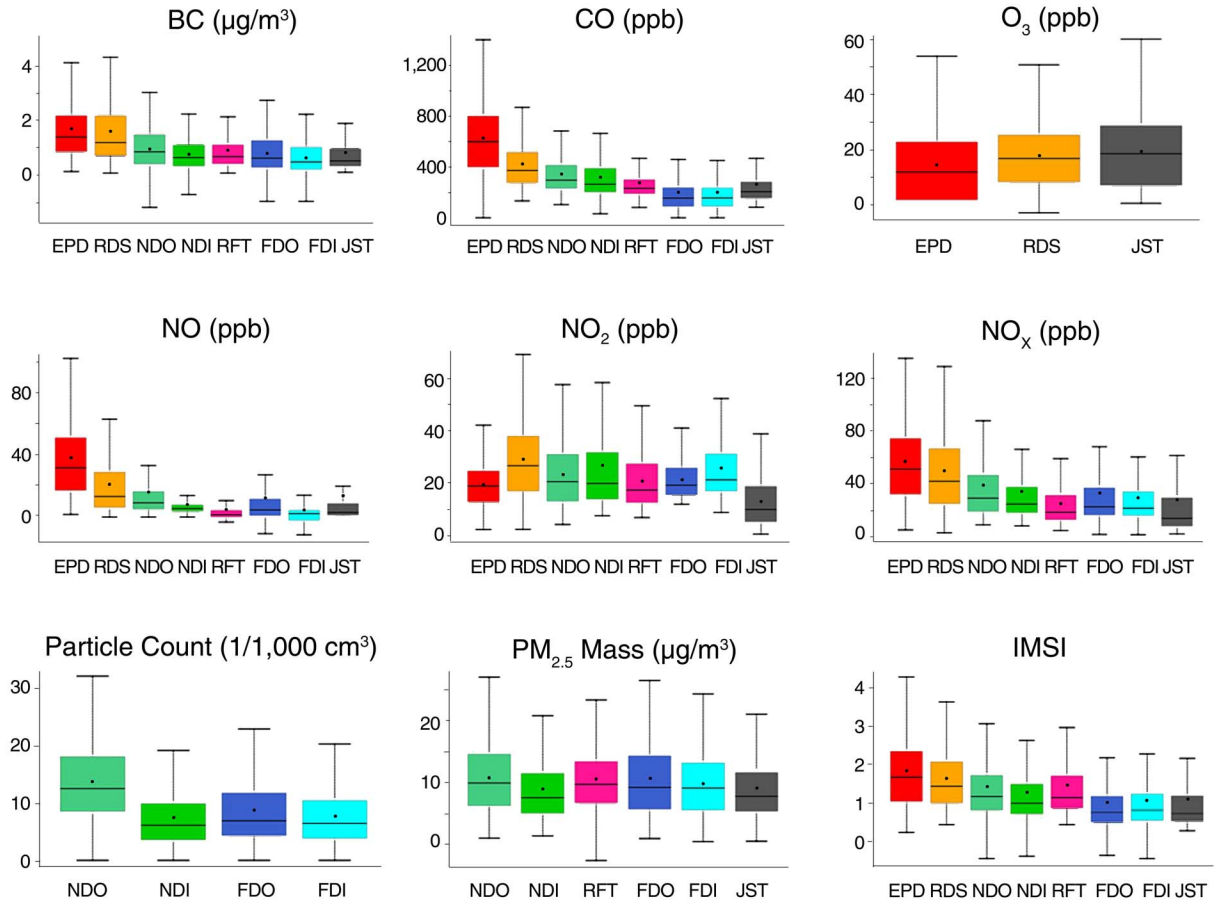

Figure 3. Boxplots showing the distribution of hourly concentrations of BC, CO, O_3, NO, NO_2, NO_x, condensation particle count, $PM_{2.5}$, and the IMSI between September 8, 2014, and January 5, 2015, at the study's outdoor and indoor sampling sites by increasing distance from the Connector: EPD (5 m), RDS (10 m), NDO (20 m), NDI (20 m), RFT (500 m), FDO (1.4 km), FDI (1.4 km), and (8) JST (2.3 km). The horizontal lines represent medians, the dots represent means, the boxes enclose the 25th to 75th percentiles, and the whiskers enclose the 5th to 95th percentiles.

near dorm were also consistently higher than those measured at the far dorm, reflecting the impact the highway has on local CO levels. A similar trend was observed for NO and BC concentrations (mean concentrations measured at the RDS site of about 20 ppb and 1.6 µg/m³, decreasing to 10 ppb and 0.8 µg/m³, respectively, at the FDO site). CO, NO, and BC levels at the EPD site were generally comparable to, if not higher than, those at the RDS site (Figure 3, Appendix Table B.1). This may have been caused by sampling location differences (the RDS site was more open, being at the edge of a parking area; the EPD site was located in a break in trees used to buffer the campus from the Connector in that area) (Amorim et al. 2013; Batterman et al. 2014b; Brantley et al. 2014; Dadvand et al. 2014; Jeanjean et al. 2015; Jin et al. 2016; Morakinyo and Lam 2016; Tong et al. 2015, 2016) as well as by instrument errors among the samplers used at both sites. In contrast to the spatial gradient patterns for CO, NO, and BC, the spatial gradient pattern for NO_2 was more muted in the 1.3-km GIT sampling domain, with mean NO_2 levels approximately 30 ppb at the RDS site and 21 ppb at the FDO site. There was some indication of lower mean NO_2 concentrations at the JST (urban background) site, with levels averaging 13 ppb during sampling for the current study. NO_2 at the EPD site was also lower than at the RDS site, possibly because of reduced oxidation from reduced mixing. (A summary of the mean and standard deviation of measurements made at each site for the September 8 to January 5 sampling period is in Appendix B.)

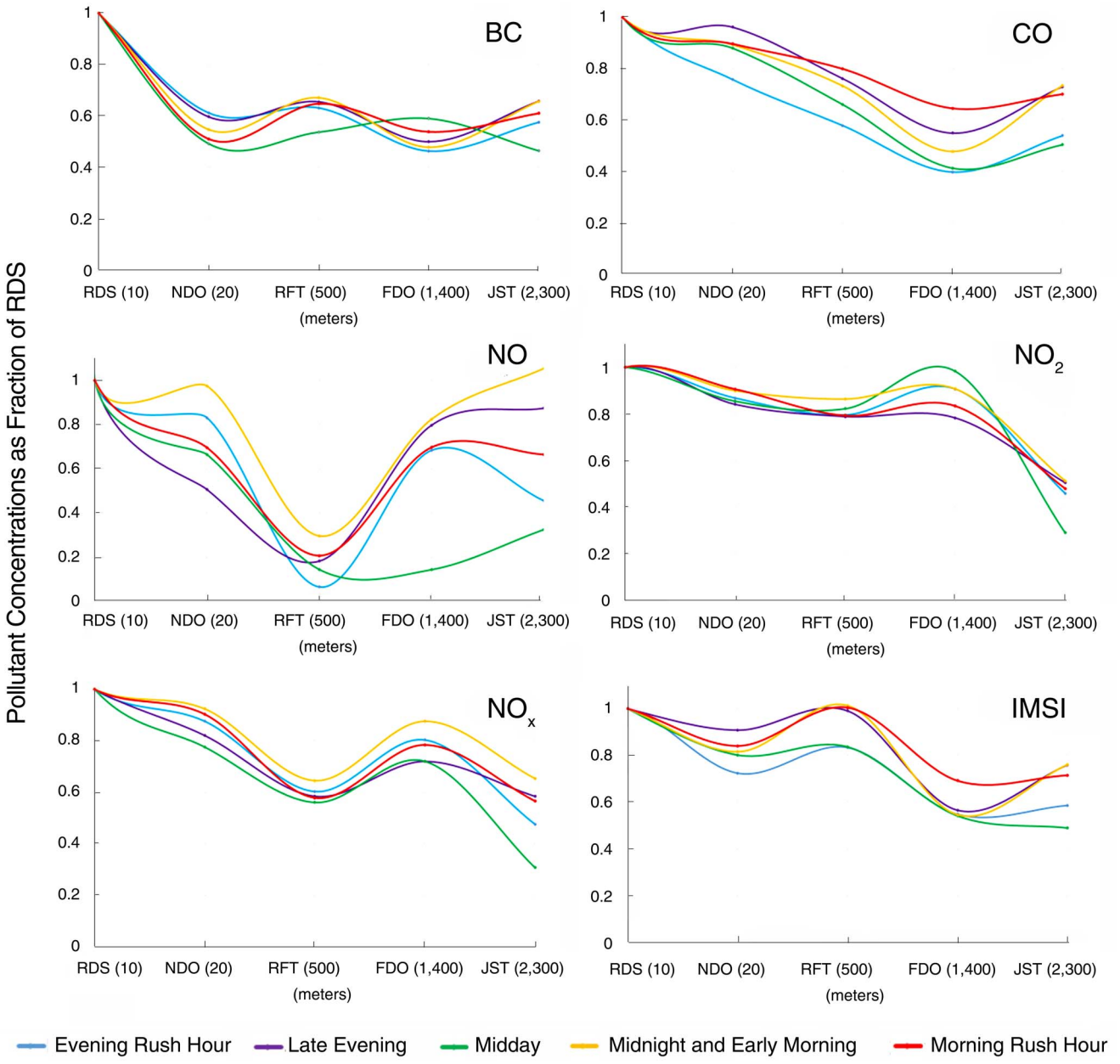

Figure 4. Spatial gradients of outdoor BC, CO, NO, NO_2, NO_x, and IMSI concentrations shown by increasing distance from the Connector and by time period of the day.

We examined diurnal profiles for each pollutant at the eight sampling locations (Figure 5). The two near-road sites (RDS and EPD) showed initial peak CO concentrations at 7 A.M., with peaks occurring slightly later in the morning between 8 and 9 A.M. at the farther locations, highlighting dispersion and transport processes near the highway as well as potentially different timing in traffic intensities on campus. Similarly, morning peak NO concentrations at the RDS site occurred mainly around 7 A.M., reaching a maximum for the day of about 35 ppb. Steep near-road (within 30 m) gradients were again observed in BC concentrations for the two near-road sites, with concentrations averaging about 0.5 μg/m^3 higher than those of all the other sites. For BC, little noticeable spatial gradient was evident among the non-near-road sites; however, BC levels at all sites followed a similar diurnal profile, with a main concentration

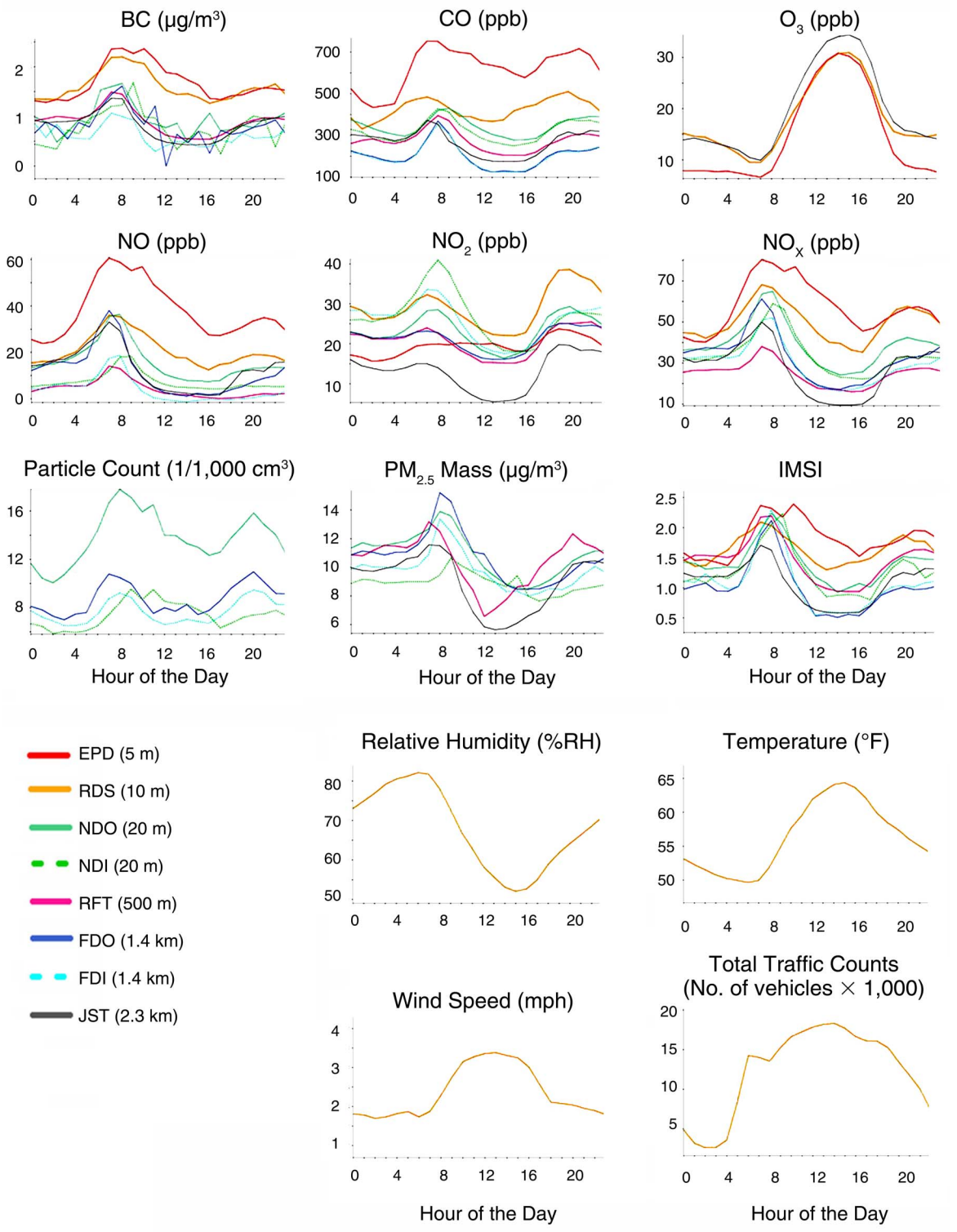

Figure 5. Diurnal profile plots showing mean hourly concentrations of BC, CO, O_3, NO, NO_2, NO_X, particle number, $PM_{2.5}$, and the IMSI, as well as relative humidity, temperature, wind speed, and total traffic counts at the study's outdoor and indoor sites between September 8, 2014, and January 5, 2015.

peak between 7 and 10 A.M. and another, more gradual peak in the evening. This might suggest that, although the highway leads to an increase in BC close to the road, it does not affect the overall microenvironment as much as expected because the regional levels of BC from traffic and other sources (e.g., biomass burning) lead to a background sufficiently high to mask the gradient beyond that distance on the campus. The dorm measurements had a more variable diurnal profile, though this may be an artifact of the personal aethalometers used. At the other ambient sites, either standard aethalometers or multi-angle absorption photometers were used to conduct measurements.

Near-road chemistry provides insight into the diurnal patterns and spatial gradients of both O_3 and NO_2. Nitrogen oxides are mainly emitted as NO. In a high-O_3 environment, NO_2 forms relatively quickly from the reaction of NO and O_3. This relationship can be observed when assessing the three near-road diurnal profiles in Figure 5. When NO concentrations peak in the morning, the O_3 is consumed and is thus at a minimum, and because NO_2 is the reaction product, a small NO_2 morning peak is seen as well (though it is less pronounced than those of species that are dominantly primary). During the middle of the day (10 A.M. to 6 P.M.), the NO concentration drops because of an increase in the boundary layer mixing height, and the O_3 concentration increases because of increased photochemical reactions and fumigation of O_3 from aloft. The NO_2 concentration drops with NO concentrations. From 4 P.M. to 8 P.M., the NO and NO_2 concentrations increased; however, the NO_2 rate of increase was much faster. The NO concentration increases as the boundary layer collapses and as O_3 production slows and is consumed to form NO_2 and a decrease in O_3.

Associations Between Pollutant Concentrations at RDS and Other Monitoring Sites

Inter-site Spearman correlations were used to examine how well temporal variability patterns at the RDS site reflected corresponding temporal variability of single pollutants and the IMSI at various distances from the highway source (Table 4). Correlations among 24-hour average pollutant concentrations at the various sites ranged from 0.13 to 0.97. Concentrations of NO_2 measured at the RDS site had observed Spearman correlations greater than 0.7 with the other ambient sites and were more temporally correlated across the domain than those of the three other primary traffic species (BC, CO, and NO). For BC, CO, and NO, stronger correlations with the RDS site were generally found for sites closer to the RDS site compared with those farther away.

We conducted additional inter-site correlation analyses examining correlations between 1-hour average pollutant concentrations. These results showed that the strength of the correlations between the RDS site and the other outdoor sites varied considerably both during the course of a day and across pollutants (Figure 6). In the early morning hours, the range of correlations was 0.35 to 0.7 among all pollutants, with the exception of NO_2. After 7 A.M., correlations dropped steadily until about 5 P.M. (with correlations ranging from 0.05 to 0.7 among all pollutants). These results are consistent with the idea that concentrations at each site are affected by the highway as a main source, as traffic increases for the morning rush hour, chemical reactions are less impactful, and there is less vertical diffusion. Conversely, correlation trends after morning rush hours are driven in part by enhanced photochemistry during the afternoon and increased vertical mixing.

To further quantify the potential spatial gradient from the traffic source observed in the descriptive analysis and the factors that accounted for the degree of the gradient, regression analyses were conducted to model the log ratio of the pollutant concentration measured at the other sites to the concentration measured at the RDS site (Appendix Table B.3). As expected based on the descriptive plots of the pollutant distributions, results from this analysis indicate that distance from the highway served as a significant factor influencing the traffic spatial gradient, with negative associations observed for all pollutants. In addition, results showed significant modification in the gradient by time of day, with the strongest decay occurring during midday hours for all traffic pollutants. For all of the measured pollutants, the effects of distance at midday were significantly different from those during the evening rush hour, which served as the reference level in the time-of-day analysis (Appendix Table B.3). In addition, in all cases, the midnight and early morning periods had the weakest decay (i.e., with largest distance estimates). These results suggest that strength of decay is likely dependent on the magnitude of source emissions; lower emissions from the highway at midnight compared with midday allow for more homogeneity of pollutant concentrations across space.

FPMOP Results

To assess the effects of roadway emissions on the near-road environment, we first compared and contrasted the spatial distribution of $FPMOP^{WS\text{-}DTT}$ with that of other species that were also measured with the integrated filters. The original study focused on the water-soluble component of FPMOP; however, after the main data collection period, we expanded our analytical capability to FPMOP $^{total\text{-}DTT}$. To test the method, a smaller study was

Table 4a. Inter-site Spearman Correlations of Daily Means for BC, Based on Measurements Made from September 8, 2014, to January 5, 2015[a]

	BC_RDS	BC_EPD	BC_NDI	BC_NDO	BC_RFT	BC_FDI	BC_FDO	BC_JST
BC_RDS		0.80	0.52	0.59	0.73	0.34	0.39	0.64
BC_EPD			0.75	0.78	0.80	0.47	0.55	0.69
BC_NDI				0.53	0.54	0.34	0.35	0.50
BC_NDO					0.66	0.42	0.46	0.59
BC_RFT						0.53	0.53	0.88
BC_FDI							0.51	0.51
BC_FDO								0.50

[a] Intensity of shading indicates relative strength of linear association.

Table 4b. Inter-site Spearman Correlations of Daily Means for CO, Based on Measurements Made from September 8, 2014, to January 5, 2015[a]

	CO_RDS	CO_EPD	CO_NDI	CO_NDO	CO_RFT	CO_FDI	CO_FDO	CO_JST
CO_RDS		0.44	0.67	0.66	0.51	0.52	0.50	0.48
CO_EPD			0.53	0.52	0.5	0.49	0.48	0.34
CO_NDI				0.90	0.69	0.70	0.68	0.58
CO_NDO					0.69	0.72	0.70	0.55
CO_RFT						0.81	0.79	0.79
CO_FDI							0.97	0.77
CO_FDO								0.74

[a] Intensity of shading indicates relative strength of linear association.

Table 4c. Inter-site Spearman Correlations of Daily Means for CPC, Based on Measurements Made from September 8, 2014, to January 5, 2015[a]

	CPC_NDI	CPC_NDO	CPC_FDI	CPC_FDO
CPC_NDI		0.73	0.56	0.53
CPC_NDO			0.35	0.36
CPC_FDI				0.96

[a] Intensity of shading indicates relative strength of linear association.

Table 4d. Inter-site Spearman Correlations of Daily Means for NO, Based on Measurements Made from September 8, 2014, to January 5, 2015[a]

	NO_RDS	NO_EPD	NO_NDI	NO_NDO	NO_RFT	NO_FDI	NO_FDO	NO_JST
NO_RDS		0.67	0.53	0.75	0.45	0.34	0.41	0.55
NO_EPD			0.37	0.58	0.5	0.46	0.51	0.47
NO_NDI				0.75	0.13	0.14	0.22	0.36
NO_NDO					0.37	0.34	0.41	0.45
NO_RFT						0.42	0.48	0.48
NO_FDI							0.89	0.51
NO_FDO								0.62

[a] Intensity of shading indicates relative strength of linear association.

Table 4e. Inter-site Spearman Correlations of Daily Means for NO_2, Based on Measurements Made from September 8, 2014, to January 5, 2015[a]

	NO_2_RDS	NO_2_EPD	NO_2_NDI	NO_2_NDO	NO_2_RFT	NO_2_FDI	NO_2_FDO	NO_2_JST
NO_2_RDS		0.84	0.70	0.79	0.74	0.79	0.81	0.68
NO_2_EPD			0.66	0.73	0.68	0.69	0.73	0.54
NO_2_NDI				0.86	0.70	0.68	0.61	0.57
NO_2_NDO					0.75	0.69	0.73	0.64
NO_2_RFT						0.89	0.91	0.84
NO_2_FDI							0.92	0.78
NO_2_FDO								0.85

[a] Intensity of shading indicates relative strength of linear association.

Table 4f. Inter-site Spearman Correlations of Daily Means for NO_x, Based on Measurements Made from September 8, 2014, to January 5, 2015[a]

	NO_x_RDS	NO_x_EPD	NO_x_NDI	NO_x_NDO	NO_x_RFT	NO_x_FDI	NO_x_FDO	NO_x_JST
NO_x_RDS		0.72	0.8	0.85	0.66	0.62	0.61	0.64
NO_x_EPD			0.70	0.70	0.6	0.58	0.58	0.49
NO_x_NDI				0.93	0.69	0.68	0.64	0.62
NO_x_NDO					0.71	0.67	0.67	0.64
NO_x_RFT						0.8	0.81	0.79
NO_x_FDI							0.96	0.78
NO_x_FDO								0.78

[a] Intensity of shading indicates relative strength of linear association.

Table 4g. Inter-site Spearman Correlations of Daily Means for O_3, Based on Measurements Made from September 8, 2014, to January 5, 2015[a]

	O_3_RDS	O_3_EPD	O_3_JST
O_3_RDS		0.77	0.87
O_3_EPD			0.85

[a] Intensity of shading indicates relative strength of linear association.

Table 4h. Inter-site Spearman Correlations of Daily Means for $PM_{2.5}$, Based on Measurements Made from September 8, 2014, to January 5, 2015[a]

	$PM_{2.5}$_NDI	$PM_{2.5}$_NDO	$PM_{2.5}$_RFT	$PM_{2.5}$_FDI	$PM_{2.5}$_FDO	$PM_{2.5}$_JST
$PM_{2.5}$_NDI		0.76	0.61	0.76	0.76	0.66
$PM_{2.5}$_NDO			0.76	0.95	0.94	0.80
$PM_{2.5}$_RFT				0.66	0.70	0.85
$PM_{2.5}$_FDI					0.93	0.78
$PM_{2.5}$_FDO						0.79

[a] Intensity of shading indicates relative strength of linear association.

Table 4i. Inter-site Spearman Correlations of Daily Means for IMSI, Based on Measurements Made from September 8, 2014, to January 5, 2015[a]

	IMSI_RDS	IMSI_EPD	IMSI_NDI	IMSI_NDO	IMSI_RFT	IMSI_FDI	IMSI_FDO	IMSI_JST
IMSI_RDS		0.78	0.69	0.74	0.72	0.63	0.68	0.65
IMSI_EPD			0.81	0.81	0.75	0.65	0.72	0.67
IMSI_NDI				0.80	0.75	0.66	0.68	0.62
IMSI_NDO					0.78	0.70	0.72	0.65
IMSI_RFT						0.80	0.83	0.88
IMSI_FDI							0.87	0.76
IMSI_FDO								0.78

[a] Intensity of shading indicates relative strength of linear association.

conducted in which filters were collected at two of the study sites and analyzed for both $FPMOP^{WS\text{-}DTT}$ and $FPMOP^{total\text{-}DTT}$, along with measurements of OC and EC. As a further means of assessing FPMOP, we also expanded the smaller study to include transition metals. The main $FPMOP^{WS\text{-}DTT}$ results are discussed first, followed by the results from the smaller study that included $FPMOP^{total\text{-}DTT}$ and metals.

The spatial distributions of $PM_{2.5}$ mass, OC, EC, $FPMOP^{WS\text{-}DTT}$ normalized by volume of sampled air ($FPMOP^{WS\text{-}DTT}/m^3$; unless explicitly stated otherwise, $FPMOP^{WS\text{-}DTT}$ means $FPMOP^{WS\text{-}DTT}/m^3$), and $FPMOP^{WS\text{-}DTT}$ normalized by $PM_{2.5}$ mass ($FPMOP^{WS\text{-}DTT}/\mu g$) are shown in Figure 7. Spatial distributions based on the integrated data were consistent with the continuous measurements discussed above. Both BC (Figure 3) and EC (Figure 7) showed some evidence of a decreasing trend with distance from the roadway, and $PM_{2.5}$ mass, determined both from optical particle number size distributions (Figure 3) and gravimetrically using filter samples (Figure 7), showed no clear spatial trend. Of the integrated filter measurements in Figure 7, only EC showed a discernible decreasing concentration with increasing distance from the roadway (similar to other primary gaseous species, such as CO and NO_x, discussed above). $PM_{2.5}$ mass and OC showed no trends, likely because they are largely composed of secondary species, and primary roadway emissions only make up a small fraction of $PM_{2.5}$ mass. Below we show that a similar explanation applies to the FPMOP.

$FPMOP^{WS\text{-}DTT}$ normalized to $PM_{2.5}$ mass ($FPMOP^{WS\text{-}DTT}/\mu g$) is a measure of the intrinsic OP of the overall $PM_{2.5}$. It is an indication of the toxicity of the $PM_{2.5}$ in terms of OP. Intrinsic FPMOP also provides insight into the contributions of specific sources to overall aerosol OP. Based on a source-apportionment analysis, Bates and colleagues (2015) found that light-duty gasoline vehicle emissions had the highest intrinsic FPMOP, followed by biomass burning and heavy-duty diesel vehicle emissions. Similar to $PM_{2.5}$ mass, OC, and $FPMOP^{WS\text{-}DTT}/m^3$, $FPMOP^{WS\text{-}DTT}/\mu g$ was also spatially uniform on average (Figure 7), which confirms that the contribution of primary vehicle emissions to FPMOP was not substantial compared with that of the other primary sources or secondary particle formation.

Correlations of FPMOP with other air quality parameters can also provide insights into sources, although the correlations for the 48-hour integrated filter-based data provided less insight than those for the continuous measurements discussed above. Pearson product-moment correlation coefficients for the linear regression between $FPMOP^{WS\text{-}DTT}$ and various measured chemical components were calculated for each measurement site (Appendix Table B.5). $FPMOP^{WS\text{-}DTT}/m^3$ was highly correlated with OC at all sites. It also correlated well with EC, CO, NO_x, and the IMSI measures (online data were averaged over the filter sampling times). The lowest correlations were generally found at the RDS site. Correlations between $FPMOP^{WS\text{-}DTT}/\mu g$ and these other components produced similar findings (Appendix Table B.7). Overall, the results were consistent with the spatial uniformity of $FPMOP^{WS\text{-}DTT}$; FPMOP was most strongly correlated with species that were also spatially uniform and less correlated with those that were more spatially heterogeneous. However, generally good correlations were seen with all the species, because no species had a consistently high degree of spatial heterogeneity and the data were highly averaged.

The indoor dorm data for FPMOP (Figure 7) were similar to the outdoor data in that there were no large systematic differences between indoor and outdoor levels for either $FPMOP^{WS\text{-}DTT}/m^3$ or $FPMOP^{WS\text{-}DTT}/\mu g$. However, at the near dorm, $PM_{2.5}$ mass, BC, and $FPMOP^{WS\text{-}DTT}/m^3$ were higher outside the dorm compared with inside it (OC was opposite). In contrast, indoor and outdoor $FPMOP^{WS\text{-}DTT}/\mu g$ were similar at both the near and far dorms. $FPMOP^{WS\text{-}DTT}/m^3$ (Appendix Table B.5) was more highly correlated with the other measured parameters at the FDO site than at the NDO and NDI sites, where r (Pearson product-moment correlation) was generally greater than 0.6. A comparison of indoor versus outdoor correlations for individual species provides insights into infiltration of outdoor pollutants or contributions from indoor sources. Table 5 shows the indoor–outdoor r for each species at the two dorm sites. At the near dorm, the gas species (CO, NO_x, and the IMSI values) were highly correlated ($r > 0.97$), whereas the aerosol components (FPMOP, PM, OC, EC, and BC) were not well correlated. In contrast, at the far dorm, except for PM mass, there was a good indoor and outdoor correlation among all species. Overall, these data suggest that for FPMOP, there were no significant unique sources inside the dorms, given the similar $FPMOP^{WS\text{-}DTT}/\mu g$ between indoor and outdoor sites. Slightly higher outdoor levels of filter-measured $PM_{2.5}$ roadway emissions at the RDS site compared with indoor levels and higher correlations between indoor and outdoor gases versus particles suggest that the indoor environment was somewhat protective against nearby roadway particulate emissions and less so for gases. At the far dorm, there was more uniformity between indoor and outdoor levels for both gases and particles.

These results are consistent with those from our previous studies, showing that secondary processing is a significant source of $FPMOP^{WS\text{-}DTT}$. This includes, but is not

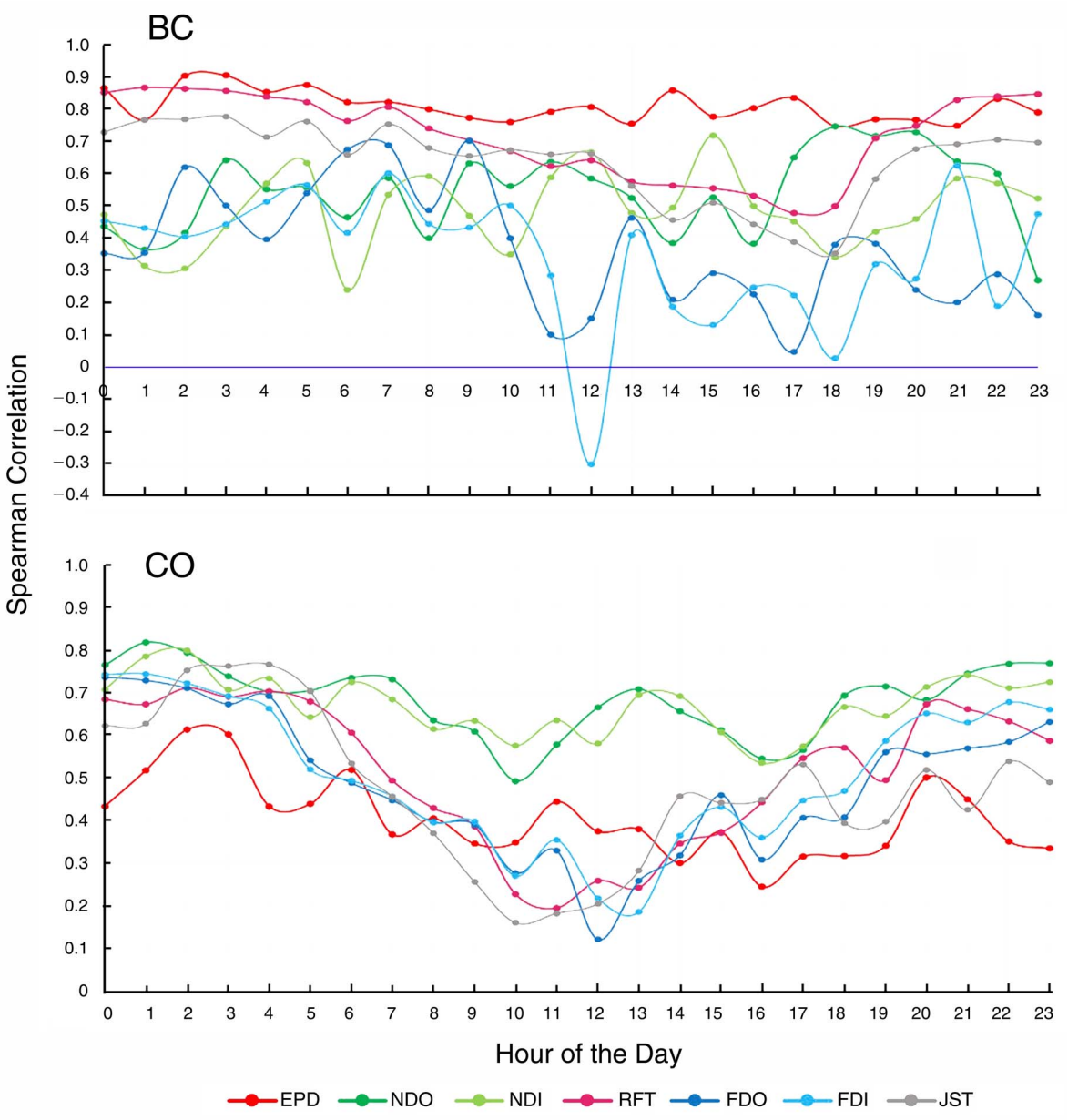

Figure 6. Spearman correlations between BC, CO, NO, and NO_2 measurements at the RDS site and all other sites by hour of the day. *(Figure continues next page.)*

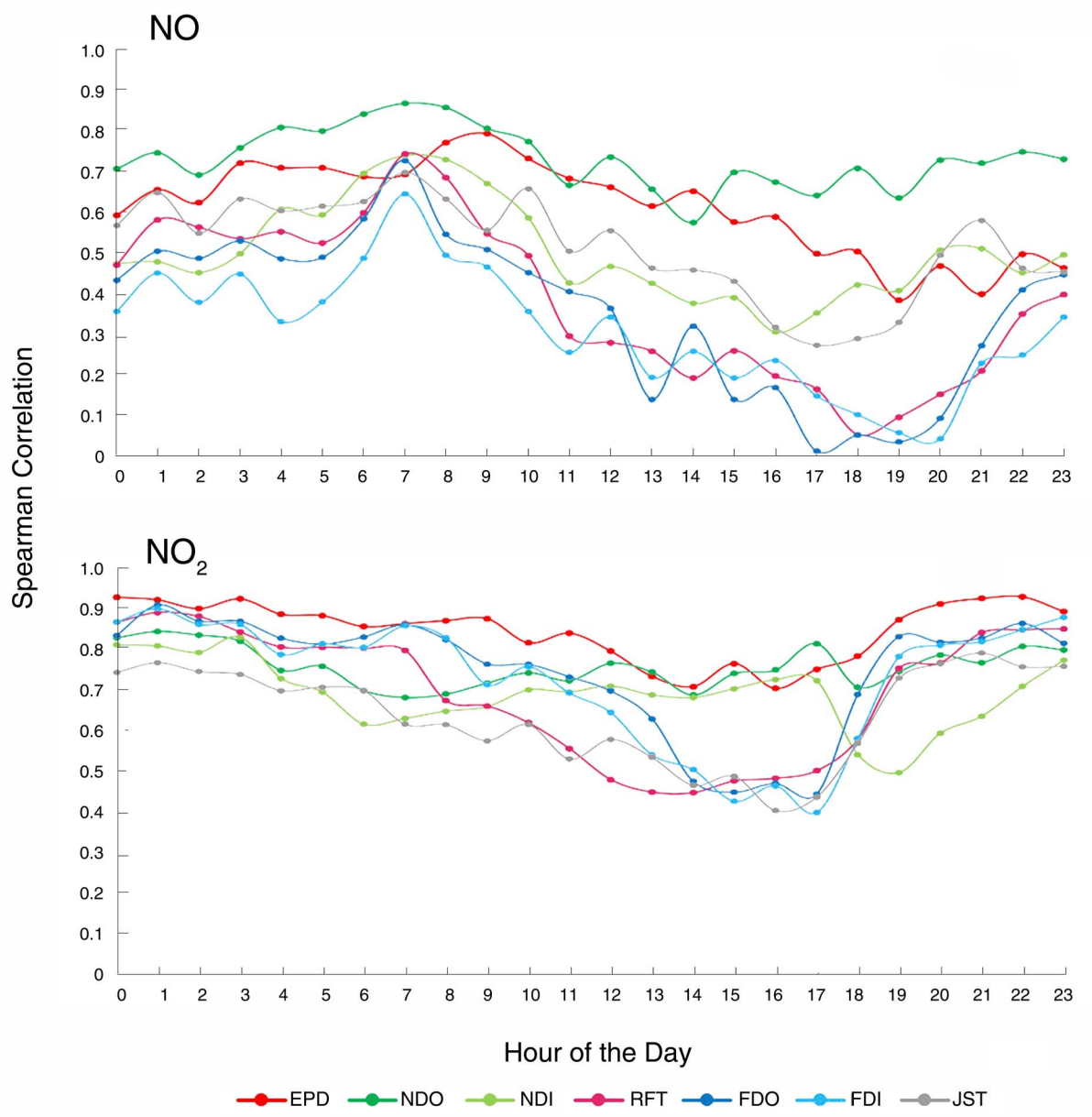

Figure 6 ***(Continued).***

limited to, processing of primary traffic emissions. For example, studies have also shown that $FPMOP^{WS\text{-}DTT}/m^3$ is related mainly to oxidized aromatic species (PAHs converted to quinones and hydroxyquinones) and water-soluble transition metals (e.g., copper [Cu] and manganese [Mn]) (Verma et al. 2014, 2015a, 2015b). Vehicles emit precursors for both: tailpipe emissions from incomplete combustion are sources of PAHs, the precursor of quinones, and brake or tire wear and resuspended road dust are precursors for water-soluble transition metals. Although vehicles are a source of the precursors, oxidation is first required for the emitted PAHs, and acid dissolution for emitted metals, to be converted to water-soluble species that become DTT-active (Cho et al. 2005; Fang et al. 2015; Meskhidze et al. 2003; Nenes et al. 2011; Rattanavaraha et al. 2011). (Note that metal dissolution by the formation of metal-organic complexes does not seem to play a significant role.) Thus, although a distinct signal from primary emissions exists, $FPMOP^{WS\text{-}DTT}$ does not appear to be a good mobile source–specific indicator, particularly in near-road environments.

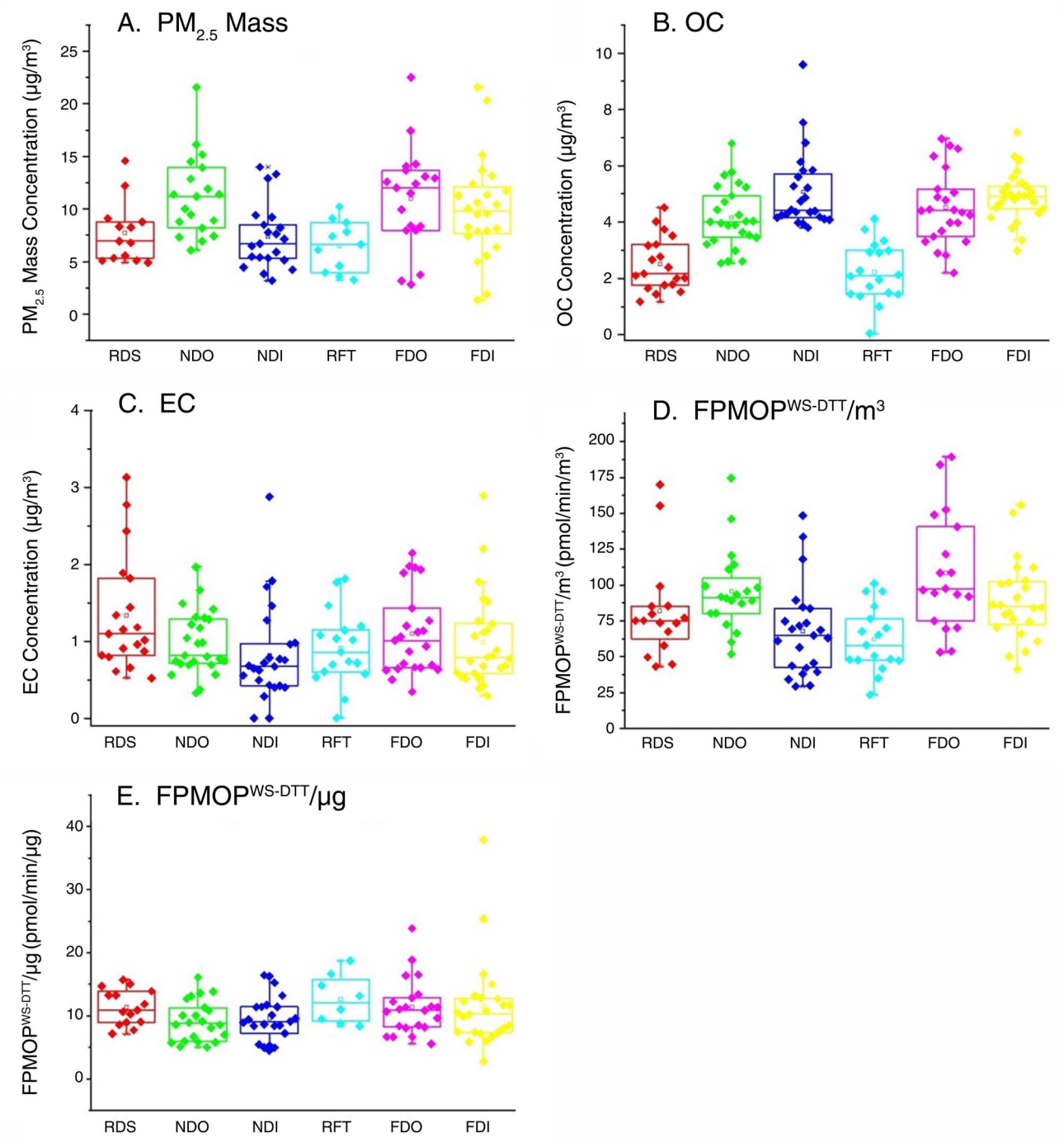

Figure 7. Spatial distribution of concentrations of $PM_{2.5}$ mass, OC, EC, $FPMOP^{WS\text{-}DTT}/m^3$, and $FPMOP^{WS\text{-}DTT}/\mu g$ from 48-hour integrated filter measurements at the sampling sites by increasing distance from the Connector. The horizontal lines represent medians, the empty dots represent means, the boxes enclose the 25th to 75th percentiles, and the whiskers enclose the 5th to 95th percentiles.

Table 5. Pearson Correlation (*r*) Between Indoor and Outdoor Concentrations at the Near and Far Dorms for Each Species Measured

	Near Dorm	Far Dorm
$FPMOP^{WS\text{-}DTT}$	0.60 ($N = 22$)	0.88 ($N = 22$)
PM	0.62 ($N = 23$)	0.60 ($N = 24$)
OC	0.42 ($N = 24$)	0.76 ($N = 23$)
EC	0.17 ($N = 24$)	0.88 ($N = 3$)
CO	0.98 ($N = 24$)	0.76 ($N = 20$)
NO_x	0.99 ($N = 24$)	0.99 ($N = 19$)
BC	0.17 ($N = 24$)	0.88 ($N = 3$)
IMSI	0.97 ($N = 20$)	0.98 ($N = 17$)

Measurement of $FPMOP^{WS\text{-}DTT}$ did not include possible contributions from insoluble species. Previous studies have found that DTT-active PM components emitted by vehicles may have low solubility and be a significant component of vehicle (specifically diesel) tail-pipe emissions (McWhinney et al. 2013b). Measurements of $FPMOP^{WS\text{-}DTT}$ may then underestimate primary traffic emissions as a source for FPMOP, and measurements of insoluble species to OP may provide evidence of a stronger roadway signal than does $FPMOP^{WS\text{-}DTT}$. A method for quantifying $FPMOP^{total\text{-}DTT}$ was developed in the final year of the current study. In this method, water extracts from the ambient air filter samples are not filtered (i.e., via liquid filtration), so that the insoluble species and particles remain in the reaction aliquot and can participate in the DTT consumption reaction. (The method and verification of the approach have been detailed by Gao and colleagues [2017].)

The method was applied to sets of filters collected at the EPD and RFT sites from April 2016 to May 2016, subsequent to the main measurement campaign. A total of 64 quartz high-volume filters were collected (32 at each site). In addition to $FPMOP^{total\text{-}DTT}$ analysis, these filters were also analyzed using the $FPMOP^{WS\text{-}DTT}$ method. OC and EC were measured by thermal optical transmission (same as for the filters discussed above). The water-soluble and total elements were also analyzed via inductively couple plasma mass spectrometry. (A brief description of the metals analysis method is provided in Appendix B, which is available on the HEI website.) In the following section we discuss the results from these filters. No ancillary gas-phase data or other aerosol composition data similar to that of the main study were available. However, the paired filter sampling at the EPD and RFT sites contrasting $FPMOP^{WS\text{-}DTT}$, $FPMOP^{total\text{-}DTT}$, and selected water-soluble and total elements allowed further analysis of roadways as a source for FPMOP.

Panels A through F of Figure 8 compare the average concentrations of potassium (K), Mn, iron (Fe), Cu, zinc (Zn), and $FPMOP^{total\text{-}DTT}$ at the EPD and RFT sites. (A summary of the data and statistics is shown in Appendix Table B.8.) First, we compared $FPMOP^{total\text{-}DTT}$ with $FPMOP^{WS\text{-}DTT}$. As expected, $FPMOP^{total\text{-}DTT}/m^3$ was on average higher than $FPMOP^{WS\text{-}DTT}/m^3$; the ratio of $FPMOP^{WS\text{-}DTT}$ to $FPMOP^{total\text{-}DTT}$ was on average 65% at the RFT site and 62% at the EPD site. This indicates for these data that $FPMOP^{total\text{-}DTT}$ contained on average 35% to 38% insoluble species. An example could be quinones that are DTT-active yet remain attached to the surface of soot (Antinolo et al. 2015).

Various degrees of spatial uniformity were seen for the various metals (Figure 8A–C). The largest difference was for Mn and Fe, for which both total and water-soluble Mn and Fe were higher at the EPD site. As seen before from the main study, $FPMOP^{WS\text{-}DTT}$ was fairly uniform; on average it was about 2% higher at the EPD site. Curiously, a similar result was found for $FPMOP^{total\text{-}DTT}$; it was 6% higher. Most of the Cu, Fe, and Zn measured at both sites were water-insoluble, whereas for K and FPMOP, the water-soluble fractions were the larger percentages of the totals.

We used daily concentration ratios for a more direct day-to-day comparison of average concentrations (Figure 8). The daily ratio summary results are similar to those of Figure 8. In this analysis, it can also be seen that both $FPMOP^{WS\text{-}DTT}$ and $FPMOP^{total\text{-}DTT}$ were at similar levels at the two sites. As noted, the uniformity of $FPMOP^{WS\text{-}DTT}$ for this study period was consistent with that of the earlier study period discussed above. The new noteworthy feature was that $FPMOP^{total\text{-}DTT}$ was just as uniform as

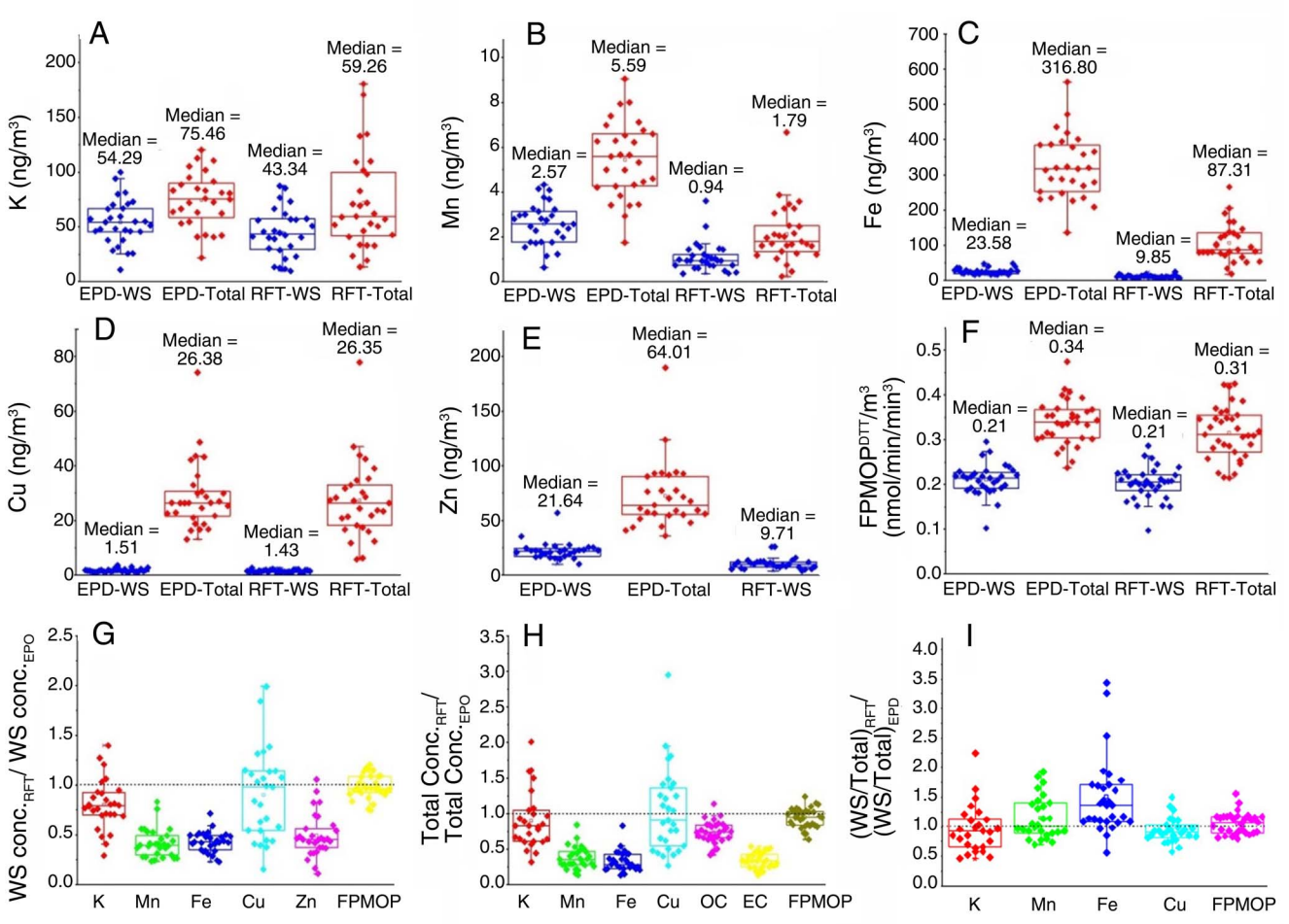

Figure 8. (A through F) Average ambient water-soluble and total metal (i.e., K, Mn, Fe, Cu, and Zn) and FPMOP^{DTT} concentrations at the EPD and RFT sites. Blue boxes denote water-soluble (WS) concentrations; red boxes denote total concentrations (total Zn data for the RFT site are missing because of experimental issues). The data are also summarized in Appendix Table B.8. Concentration ratios of simultaneous 24-hour integrated filter measurements ($N = 32$) made at the EPD and RFT sites of (**G**) water-soluble metals and FPMOP and (**H**) total metals, OC, EC, and FPMOP (total Zn data for the RFT site are missing). The data are also summarized in Appendix Table B.9. (**I**) Ratios of water-soluble metals and FPMOP at the RFT and EPD sites. A ratio of one means that the water-soluble fraction was the same at both sites; a ratio larger than one means that the water-soluble fraction was larger farther away from the Connector. The data are also summarized in Appendix Table B.9. The horizontal lines represent medians, the empty dots represent means, the boxes enclose the 25th to 75th percentiles, and the whiskers enclose the 5th to 95th percentiles.

FPMOP$^{WS\text{-}DTT}$, which was not expected. For the metals, water-soluble and total K were only slightly higher at the EPD site than at the RFT site, whereas Mn, Fe, and Zn were clearly higher at the EPD site. A significant source of K is biomass burning, of which a large fraction is soluble (Zhang et al. 2010), with minor contributions from roadway emissions, consistent with these observations. Mn, Fe, and Zn compounds are known to be associated with brake wear or resuspended traffic road dust (Fang et al. 2015; Gietl et al. 2010; Heal et al. 2005; Johansson et al. 2009). Cu is also emitted from roadways (Fang et al. 2015, 2016); however, in this small-sample-size study, it was highly variable and no specific conclusions about Cu can be reached from these measurements. OC and EC data were consistent with those of the main study; EC was much higher at the EPD roadside site than at the RFT site, and OC was only slightly higher (Figure 8). For reference, a correlation matrix for the various water-soluble and total metals and FPMOP is provided in Appendix Table B.10, available on the HEI website.

The ratio of the fraction of the water-soluble components at the RFT site to the fraction of water-soluble components at the EPD site was also compared more quantitatively (Figure 8G–I). A ratio of one means the

water-soluble fraction was the same at both sites. A ratio larger than one means the fraction of water-soluble species was higher farther away from the road (which would be expected if there were some form of processing leading to an increase in solubility after emission). Water-soluble fractions for Mn and Fe were generally higher away from the road; however, for K, Cu (Cu data were suspicious because of their highly scattered distribution), and FPMOP, there was not a large difference. Thus, although Figure 8G–I shows that $FPMOP^{WS\text{-}DTT}$ was less than $FPMOP^{total\text{-}DTT}$ at both sites (the $FPMOP^{WS\text{-}DTT}/FPMOP^{total\text{-}DTT}$ ratio was 0.62 to 0.65), there were no significant differences in the water-soluble fractions between the two sites. This suggests that even the water-insoluble components of FPMOP must undergo some form of processing. Similarly, metals emitted by roadway emission (Mn and Fe, ignoring Cu) showed only a slightly larger soluble fraction away from the road. Overall, this analysis of fractions of soluble species showed that no significant degree of processing of the aerosol, in terms of degree of solubility, happened in the time the aerosol was emitted from the roadway and dispersed to the RFT site.

General Temporal and Spatial Variability Results

By monitoring the concentrations of traditional primary traffic-related pollutants at six ambient sites within 2 km of a major interstate highway, this study has shown the spatial variability as well as the effects on temporal variability of distance from the main vehicle emissions source. First, evaluation of the near-road measurements highlighted the decrease in vehicle emissions as a major source, resulting in low ambient air pollutant concentrations relative to historic near road levels in Atlanta. As a consequence, meteorological conditions have become increasingly important in determining concentration levels in near-road environments, and different exposure assessment strategies should be evaluated. Second, assessing spatial variability patterns confirmed that levels of most of the traffic indicators decreased with distance from the highway, with the exception of NO_2. Because NO_2 is often presented as a surrogate of traffic pollution, it is important to recognize that the lack of gradient affects the way exposures should be accounted for. Third, the spatial gradients also exhibited substantial diurnal variability, with peak concentrations reached at different times depending on the pollutant and the distance from the highway. The diurnal variability led to poor spatial correlations across the sites and also has significant implications for how exposure is assigned in epidemiology studies when a single near-road site is used as a proxy for exposure. Finally, for a given pollutant and building, infiltration and indoor pollutant sources can become a large part of exposure and should not be ignored. By characterizing the spatiotemporal variability that exists in a microenvironment surrounding a major traffic emission source, we have highlighted the limitations of using a single sampling location and single traffic-emission indicators as surrogates of exposure for participants living within a few kilometers of the traffic hotspot.

In addition to assessing single pollutant exposure indicators, two multipollutant indicators were evaluated as alternative surrogates of exposure. FPMOP exhibited a more uniform gradient, linked more closely with secondary components than with primary emissions. Although OP is potentially a good measure for overall exposure, it does not appear to be a good measure of exposure for primary vehicle emissions. IMSIs, in contrast, are constructed from the mobile source contributions to the concentrations of BC, CO, and NO_x to better reflect the spatial gradients of mobile source impacts. The spatial gradient and diurnal profile patterns of the IMSIs were similar to those of BC, CO, and NO_x (all three were similar). The IMSIs had few significant terms, however, when assessing the impact of factors on the spatial gradients, suggesting that IMSIs would be a more stable way to assess exposures compared with single traffic-related indicators.

AIM 3: CHARACTERIZING SPATIAL AND TEMPORAL VARIABILITY PATTERNS OF PERSONAL EXPOSURE

In total, we collected 116 48-hour personal sampling sessions from 51 participants. Baseline information collected from each participant at the commencement of sampling showed generally similar demographic characteristics among participants in both dorms, with a greater relative number of sophomore (second-year) students living in the far dorm (42.9%) compared with the near dorm (8.7%) (Table 6).

Time–Activity Summary

Among the 51 personal exposure participants, continuous GPS data were collected from 43 participants (21 from the near dorm and 22 from the far dorm) over the course of 11 sampling weeks. GPS device malfunction resulted in the loss of data during several collection periods, and there were consequently fewer GPS 48-hour sample cycles collected compared with the $PM_{2.5}$ and NO_2 samples. GPS cycles averaged 2,873 minutes (47.9 hours) and were collected during every week of the intensive sampling period, except for fall break (October 13 to 17, 2014) and Thanksgiving (November 26 to 28, 2014).

A total of 1,273 stationary waypoints were collected from the 43 participants over the data collection period.

On average, GPS data from each participant registered approximately 13 waypoints per 48-hour sampling period, with an average of 203 minutes (3.4 hours) spent at each waypoint. There were 554 waypoints for the 22 far dorm participants, resulting in approximately 25.2 waypoints per participant, with an average of 233 minutes spent at each waypoint. In contrast, the 21 near dorm participants registered 719 waypoints, resulting in 34.2 waypoints per participant, with an average of 180 minutes spent at each waypoint. As expected, in addition to GPS clusters at both of the dorms, we also observed substantial clustering in the center of the GIT campus (250 to 1000 m from the Connector), where most of the academic classes are held (Figure 9 and Figure 10). The central campus region was the area with the most recorded waypoints during sampling, with 178 (35% of total waypoints).

There was a clear bimodal distribution in the locations of participants from the two dorms, with students spending much of their time inside or near their respective dorms (Figure 9). Participants living in the far dorm, for example, spent 37% of their time within or near the far dorm (1.4 km from the Connector); the near dorm participants spent a roughly equivalent amount of time (39%) within or near the near dorm (20 m from the Connector). Overall, roughly 10% of the participants' time was spent outside the campus perimeter (farther than 1.6 km from the Connector).

Personal Pollutant Exposure Summary

Direct comparisons of the personal exposure distributions by dorm should be viewed cautiously, given the unbalanced sampling design, in which students from one of the dorms participated in greater numbers than those from the other dorm. With this caveat, results from the personal exposure measurements show that the participants from both dorms had similar measured personal exposures to NO_2, BC (expressed as absorption coefficients), and

Table 6. Baseline Demographic Information on Participants in the Personal Exposure Sampling

Variable	Overall (n = 51)	Near Dorm (n = 23)	Far Dorm (n = 28)
Age, mean (SD)	19.3 (0.85)	19.2 (0.9)	19.4 (0.8)
BMI (SD)	23.3 (3.0)	22.7 (3.1)	23.9 (2.9)
Sex, n (%)			
Female	24 (47.1)	11 (47.8)	13 (46.4)
Male	27 (52.9)	12 (52.2)	15 (53.6)
Academic year, n (%)			
Freshman	29 (56.9)	16 (69.6)	13 (46.4)
Sophomore	14 (27.5)	2 (8.7)	12 (42.9)
Junior	7 (13.7)	4 (17.4)	3 (10.7)
Senior	1 (2.0)	1 (4.3)	0 (0.0)
Time in dorm prior to first plasma sample collection	69 (119)	86 (161)	55 (67)
Time spent outdoors,[a] n (%)			
Less than 1 hr	4 (8.0)	2 (8.7)	2 (7.4)
1–2 hr	21 (42.0)	11 (47.8)	10 (37.0)
3–4 hr	18 (36.0)	8 (34.8)	10 (37.0)
5 hr or more	7 (14.0)	2 (8.7)	5 (18.5)
Time spent in vehicle,[a] n (%)			
Less than 1 hr	20 (40.0)	8 (34.8)	12 (44.4)
1–2 hr	27 (54.0)	14 (60.9)	13 (48.1)
3–4 hr	2 (4.0)	1 (4.3)	1 (3.7)
5 hr or more	1 (2.0)	0 (0.0)	1 (3.7)

[a] Daily estimated average of time–activity patterns prior to the study, obtained from participant baseline questionnaires.

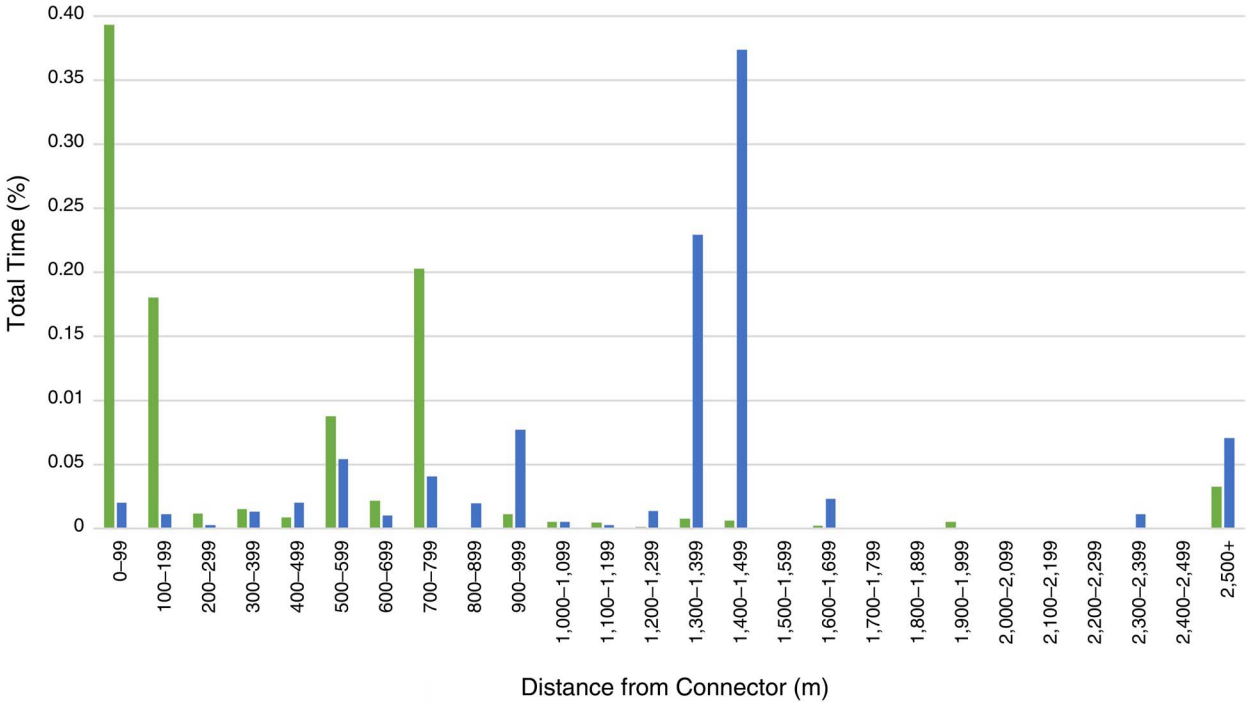

Figure 9. Percentage of 48-hour personal sampling period spent at various distances from the Connector by participants living in the near dorm (green bars) or far dorm (blue bars).

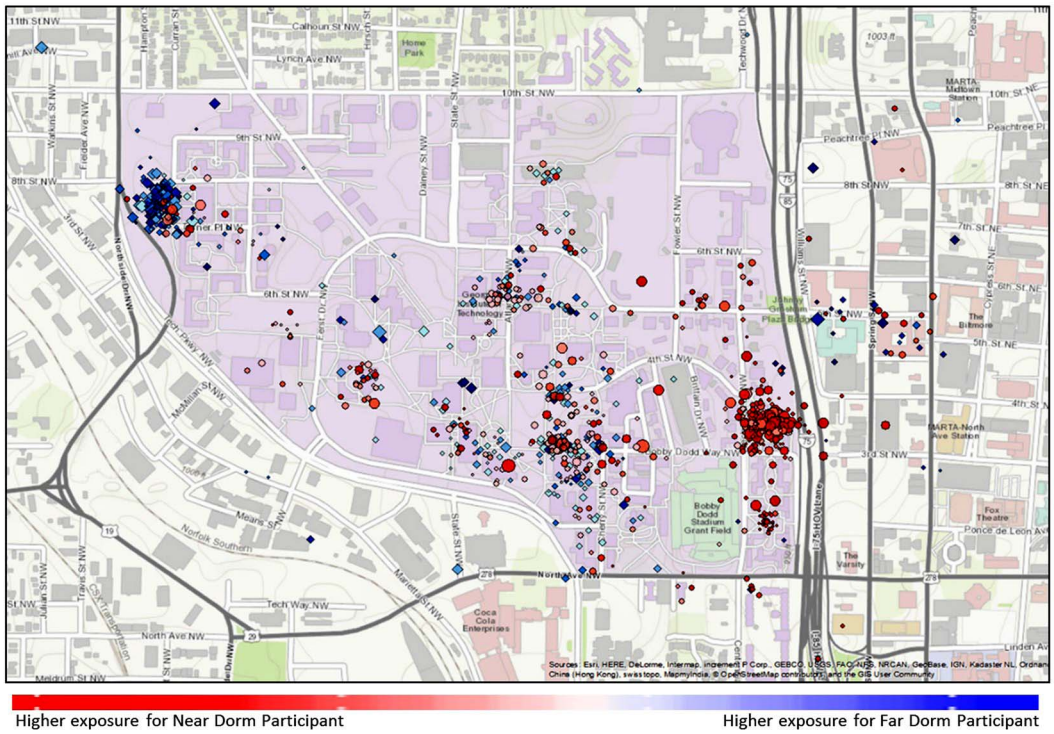

Figure 10. Personal IMSI exposure to primary traffic emissions among participants living in the near dorm (red circles) or far dorm (blue diamonds). Color gradients reflect relative exposures (darker = higher exposure), and sizes reflect the amount of time spent in each location.

$PM_{2.5}$ (Table 7). Integrated participant-specific 48-hour personal NO_2 exposures ranged from 6.9 to 28.8 ppb in the near dorm and 2.2 to 43.5 ppb in the far dorm, which were also broadly comparable with corresponding outdoor concentrations measured at the other study sites during this time. Notably, consistent with the aggregated exposure results, matched comparisons of the unbalanced personal exposures by week and participant dorm location also provided some indication of roughly comparable exposures to each of the measured pollutants between dorms (Appendix E, available on the HEI website).

Along with the directly measured personal pollutant exposures, we integrated each participant's GPS information with temporally resolved RLINE modeled concentrations to generate modeled personal exposures to NO_x, BC, and CO. Unlike the integrated direct measurements, all three modeled pollutant exposures were based solely on outdoor estimated spatial gradients and indicated higher overall levels in the near dorm participants compared with the far dorm participants. These modeled estimates do not include any terms to predict indoor or nonvehicle source contributions to personal exposure. We used the RLINE modeled pollutant personal exposure output to create spatiotemporally resolved estimates of personal IMSI exposures as a multipollutant integrated measure of personal exposure to traffic emissions (Appendix D).

Apart from the clear spatial clustering for the two dormitories, the exposure map indicates that for the near dorm participants the highest IMSI exposures occurred at locations closest to the highway source. IMSI exposures were also elevated for the cluster of waypoints near the far dorm, which may reflect contributions from the in-dorm cafeteria in the far dorm and a fairly sizable surface road along the dorm's northern boundary. IMSIs for waypoints located in the central campus area were typically lower than those modeled for areas near the two dorms.

Using mixed effects regression modeling, we examined specific indicators of 48-hour integrated personal exposures in the participants. The indicators we examined were designed to assess differences among a range of exposure surrogates that varied by proximity to student dorm residence as well as other factors that accounted for individual spatiotemporal mobility patterns and residence. The independent terms were (1) pollutant levels measured at the RDS site, (2) pollutant levels measured outside a participant's dorm, (3) pollutant levels measured inside a participant's dorm, (4) mean proximity (in meters) of the participant to the highway during the 48-hour sampling period, (5) whether the participant lived at the near or far dorm (categorical), and (6) IMSI levels measured at the RDS site.

As shown in Table 8, neither of the near-road predictors of personal BC, measured concentrations and IMSI values at the RDS site, was associated with corresponding personal BC; and variability in BC for the RDS metrics explained less variability in personal BC than the other

Table 7. Summary Statistics for Personal Pollutant Exposures

Dorm Location		RLINE Estimate				Direct Measurement			
		RLINE CO (ppb)	RLINE NO_x (ppb)	RLINE $PM_{2.5}$ (μg/m3)	IMSI	$PM_{2.5}$ BC (using Reflectance) (μg/m3)	$PM_{2.5}$ (using Gravimetry) (μg/m3)	Personal NO_2 (ppb)	Dorm Room NO_2 (ppb)
Near dorm	*N*	47	47	47	625	46	47	45	16
	Mean	57.4	9.5	1.4	1.3	1.0	8.2	14.7	10.7
	Minimum	3.9	0.6	0.1	0.02	0.2	0.7	6.9	5.4
	Maximum	164.8	26.8	4.0	42.5	2.6	17.6	28.8	16.9
Far dorm	*N*	58	58	58	463	58	58	58	22
	Mean	28.8	4.7	0.7	0.7	1.1	9.0	17.5	15.2
	Minimum	3.2	0.5	0.1	0.02	0.3	1.0	2.2	5.6
	Maximum	93.9	15.3	2.2	29.5	2.6	17.4	43.5	24.1

Table 8. Evaluation of Performance of Various Parameters in Predicting Pollutant Concentrations at the Personal Level Using Generalized Linear Mixed Models

	BC (Reflectance)				NO_2			
Predictor	*N*	Estimate	95% CI	R^2	*N*	Estimate	95% CI	R^2
RDS site concentrations	76	1.16	(−0.28 to 2.61)	0.46	103	0.03	(−0.15 to 0.20)	0.57
Dorm concentrations								
Outdoor	104	0.76[a]	(0.59 to 0.92)	0.56	103	0.31[a]	(0.10 to 0.51)	0.43
Indoor	104	0.77[a]	(0.64 to 0.89)	0.63	103	0.36[a]	(0.19 to 0.53)	0.48
Proximity (per 50 m)	82	0.08	(−0.02 to 0.17)	0.69	81	0.09	(−0.02 to 0.20)	0.57
Dorm residence (far)	104	1.84[a]	(0.12 to 3.55)	0.67	103	2.58[a]	(0.59 to 4.57)	0.56
IMSI at RDS site	76	2.40	(0.19 to 4.60)	0.44	75	2.42	(−0.85 to 0.46)	0.44

[a] P value < 0.05

indicator variables examined. In contrast, personal BC was moderately and significantly associated with measurements both outside and inside of the dorms, with observed R^2 values of 0.56 and 0.63, respectively. Based on these results showing stronger associations between personal BC and monitors located closer to the participants' residences, it makes sense that the use of a simple dorm location term to predict personal exposure was also a significant predictor of personal BC exposure, explaining a roughly similar degree of variability (R^2 = 0.67).

Results from the personal NO_2 exposure models were more similar across metrics in terms of strengths of association and model fit than those for BC. R^2 values among the six metrics ranged from 0.43 to 0.57. Notably, for both of the directly measured pollutants, the mean distance from the highway of the participant during the 48-hour sampling period explained the greatest variability in corresponding personal exposure. This term, in both pollutant models, was not itself significant, however, indicating a greater likelihood that the observed results were caused by chance. Similarly, neither of the IMSI values measured at the RDS site for BC or NO_2 were shown to be associated with personal BC or NO_2.

AIM 4: METABOLOMICS ANALYSIS

A central exploratory aim of the study's was to examine whether exposures to traffic pollution can be detected using highly novel environmental metabolomics methods. Specifically, we assessed whether observable changes in specific metabolic profiles can be detected in plasma and saliva samples collected repeatedly throughout the sampling period. To our knowledge, the work conducted as part of Aim 4 constitutes the largest prospective assessment of traffic pollution exposure and metabolic response biomarkers to date.

Blood sampling was conducted four times during the 12-week intensive sampling period, with one of the four collection periods occurring at the beginning of the study (September 19, 2014). In total, we collected 175 plasma and cell samples from the 54 participants, averaging 3.2 samples per participant. Saliva collection was conducted every sampling week during the 12-week study period. In total, we collected 621 2-ml vials of saliva, averaging 11.5 samples per participant.

Among the samples, we extracted 20,766 metabolic features from the plasma samples and 29,013 from the saliva samples. QA/QC results indicated excellent overall data quality, with a median coefficient of variation across the triplicate analyses for all metabolites of <30%. A moderate fraction of features (39.2%) were identified and shared in both plasma and saliva samples (Appendix Figure F.1, available on the HEI website). Over 45% of the ions extracted were found to have exact mass-to-charge (m/z) matches (m/z difference less than 10^{-5}) with metabolites identified in either the Human Metabolome Database (*http://www.hmdb.ca/*) or the U.S. EPA's Mobile Air Toxics database (*www.epa.gov/otaq/toxics.htm*).

We developed mixed effects linear models to examine associations between feature intensity (i.e., relative concentration) and dormitory location (i.e., near dorm versus far

dorm) in each of the four technical columns, controlling for random intercepts and potential confounding factors (age, sex, race, dorm move-in day, and repeated sampling over time). In total, we identified 221 features that significantly differed between the dorms in intensity ($P < 0.05$, using the Benjamini–Hochberg false discovery rate correction procedure). Moreover, 220 of the 221 significant features were from the plasma samples, providing an initial indication that saliva might not be a sensitive-enough matrix for detecting relevant metabolic changes at the scale of a small panel-based environmental exposure study (Figure 11).

We note that these analyses reflect metabolomic differences associated with a broad indicator of exposure (i.e., residential location) and thus may not be associated with any environmental exposures relevant to the study aims. We are currently conducting comprehensive bioinformatics analyses to elucidate potential metabolites as biomarkers related to either traffic exposure or its relevant health endpoints. Future work will include linear modeling that includes more targeted indicators of primary traffic as well as formal validation and identification of individual metabolomics features using tandem mass spectrometry.

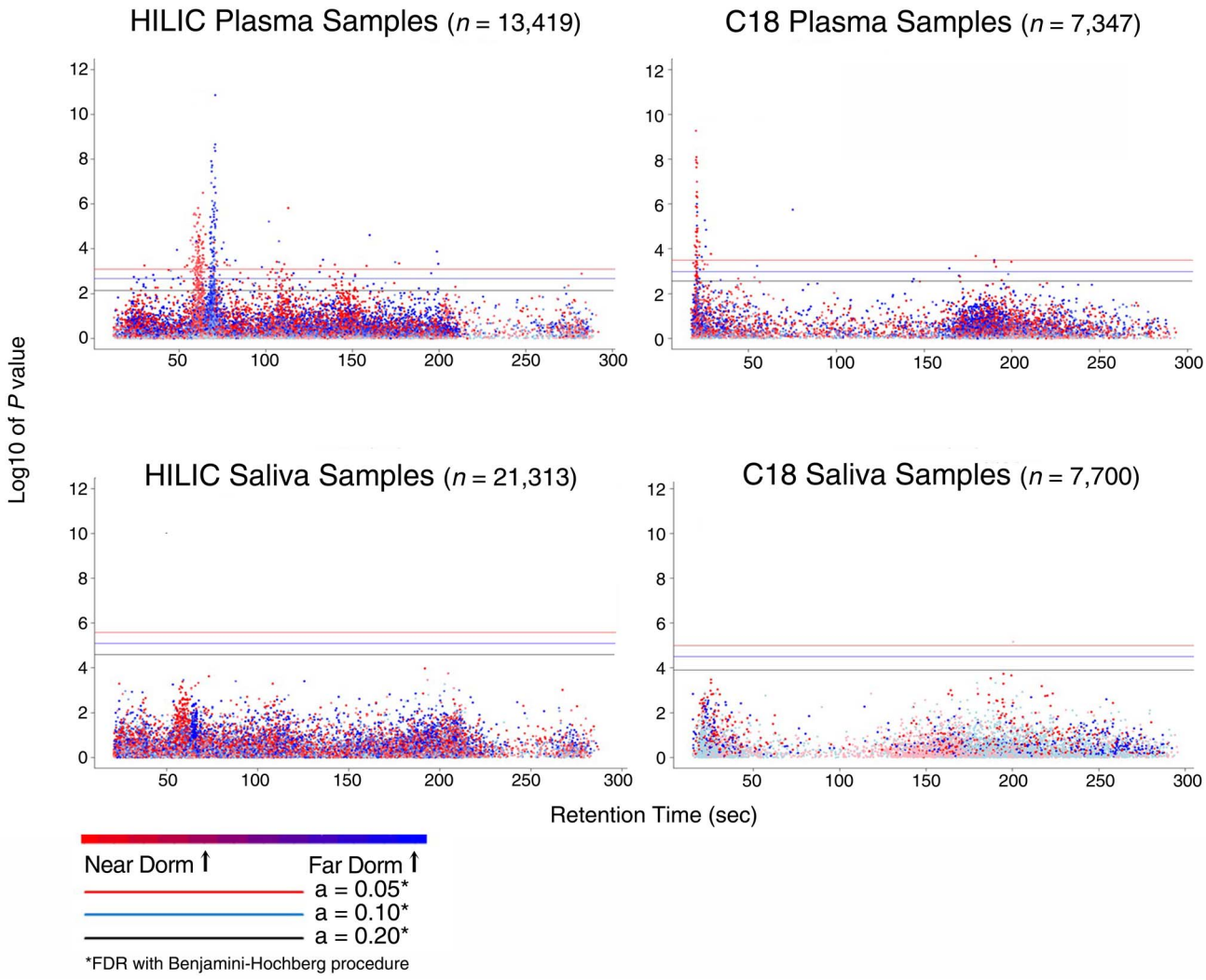

Figure 11. Manhattan plots of metabolites by dorm, using mixed effects linear models. False discovery rate (FDR) correction was made using the Benjamini–Hochberg procedure. At $a = 0.05$, 175 metabolites were found in the HILIC (hydrophilic positive ion mode) plasma matrix to have statistically significant differences in relative concentrations between the near dorm participants' metabolic profiles and those of the far dorm participants. Forty-five significant metabolites were identified in the C18 (hydrophobic negative ion mode) plasma matrix. Zero and only one significant metabolite were identified in the HILIC saliva and C18 saliva matrices, respectively.

AIM 5: LOW-COST SENSOR MEASUREMENTS

Low-cost sensors have the potential to be easier to use than traditional measurement instrumentation because of their small size and low power consumption. This allows a greater number of sensors to be deployed, increasing the density of spatial measurements over that of reference instruments. Our objective for implementation during the study was to assess how well the sensors captured the temporal trends at various locations and distances from the main traffic emissions source. For this reason, the gas and particulate sensors were assembled into four multisensory units and rotated around the sampling locations. The gas-phase sensors were deployed at the NDO and FDO sites as well as at the RDS site. At least one particulate sensor was in each multisensory unit, sampling inside and outside the near dorm, outside the far dorm, and at the RDS site. (See Appendix C, available on the HEI website, for a more detailed table of the rotation schedule.) By rotating the particulate sensors, the various models and configurations could be tested at locations with different concentration ranges and in comparison with the reference instruments. Further, sensors of the same model type could be compared with each other to assess their consistency. Although easy to use and rotate, the sensors required calibration checks similar to those of the reference instruments.

Our assessment of the use of the low-cost sensors was based on examining how well the sensor results compared with those of the reference instruments both after the manufacturers' recommended calibration and then after our further calibration using the in-field observations and in-lab calibrations after the intensive monitoring sessions. Application of the manufacturer-supplied calibration curves to convert sensor voltage outputs to CO and NO concentrations (Appendix C) led to good correlations with the reference instruments at the RDS site (R^2 = 0.90 and 0.92, respectively). The correlations between sensors and reference monitors were slightly stronger at the NDO site (R^2 = 0.85 and 0.86) and weakest at the FDO site (R2 = 0.51 and 0.79). Drifts in the linear regression slopes were also observed as the sensors were rotated between the sites, with the CO sensor calibration slope decreasing from 0.76 to 0.62 and the NO sensor calibration slope increasing from 0.69 to 0.87, either because of a change in the accuracy of the sensor or changes in the concentration ranges at the various sampling locations (Appendix C). NO_2 sensor measurements using the manufacturer calibration did not agree with the concentrations measured by the reference instrument as well as the other gas-phase sensors at the RDS, NDO, and FDO sites (R^2 = 0.54, 0.78, and 0.78, respectively). The sensor appeared to follow a similar trend capturing the peaks; however, the sensor peaks were biased high compared with the reference monitor peaks (Figure 12). The bias was also captured in the change of the linear regression slope between the NO_2 sensor and the various reference monitors from 2.11 at the NDO site to 3.89 at the FDO site. The change in slope was possibly because of an incorrect calibration curve supplied by the manufacturer. (See Appendix C for more detailed results.)

NO_2 sensor measurements using the manufacturer calibration did not agree with the concentrations measured by the reference instrument as well the other gas-phase sensors at the RDS, NDO, and FDO sites (R^2 = 0.54, 0.78, and 0.78, respectively). The sensor appeared to follow a similar trend capturing the peaks; however, the sensor peaks were biased high compared with the reference monitor peaks (Figure 12). The bias was also captured in the change of the linear regression slope between the NO_2 sensor and the reference monitor from 2.11 to 3.89, possibly because of an incorrect calibration curve supplied by the manufacture. (See Appendix C for more detailed results.)

The O_3 sensor (Figure 12) also came with a calibration curve from the manufacturer; however, it was not adequate, and further calibration was necessary using concurrent temperature and RH measurements. After application of the supplied calibration curve, the sensor concentrations ranged from about 250 ppb to 2000 ppb, with a majority of the concentrations being negative. Although the application of a log-regression improved the R^2 of the O_3 sensor calibration relative to reference instruments only from 0.40 to 0.44, the data no longer contained negative concentrations and captured the reference instrument's observed peaks. To further increase the correlation (R^2 = 0.67), a multivariable calibration approach including the temperature and RH measurements was used. Appendix C includes further details about the calibration process used to correct the O_3 sensor. Of interest, because the O_3 calibration curve included the NO_2 concentration and the NO_2 sensor overestimated concentrations compared with the reference analyzer, a separate correction to the O_3 sensor data was to use the NO_2 reference measurement concentrations. This helped to assess how dependent the O_3 concentrations were on the accuracy of the NO_2 measured concentrations. Overall, the O_3 concentration range decreased (100 ppb to 150 ppb), and the sensor was able to capture the peaks in the reference instrument measurements, although about half the values were still negative. (A complete description of how the O_3 sensor measurements were calibrated can be found in Appendix C.)

The PM sensors were not supplied with $PM_{2.5}$ calibration curves from the manufacturer and therefore required the development of a calibration curve comparing the sensor with a reference instrument. All the sensors were

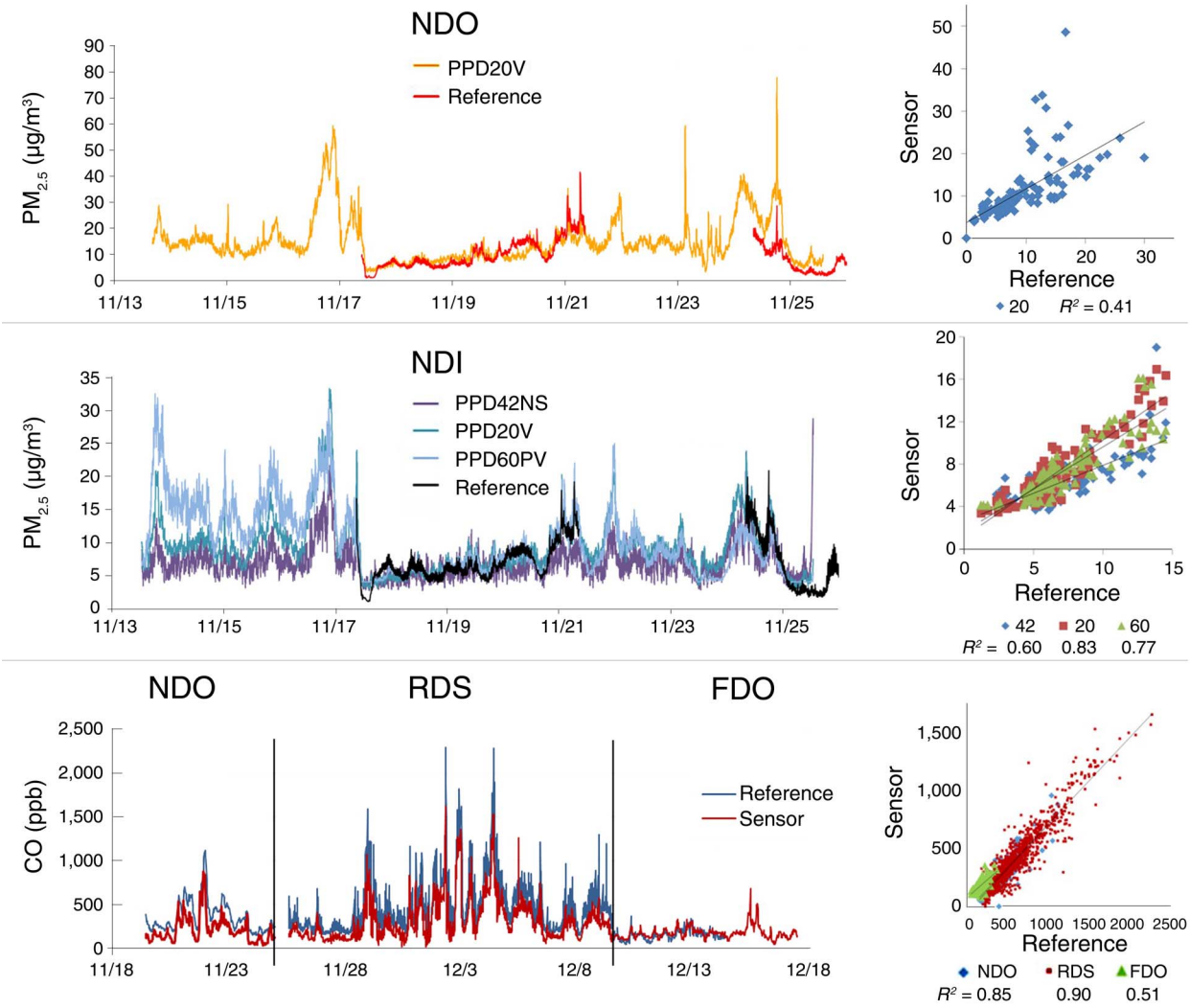

Figure 12. Time series (hourly resolution) of the comparison of particle and gas sensor measurements compared with reference instrument measurements. (*Figure continues next page.*)

located at the RFT site in their multisensory units, and a Deming regression calibration curve was developed based on 3 days of tapered element oscillating balance (TEOM) data. The PM sensors were placed at the NDI and NDO sampling sites and compared with $PM_{2.5}$ measurements from a Grimm monitor (Figure 12). The various sensor models did not perform equally, but overall their measurements compared reasonably with the Grimm measurements; however, there might also have been inaccuracies in the Grimm measurements and the method by which the $PM_{2.5}$ concentrations were calculated. Overall, further analysis and comparisons would be necessary before the particulate sensors could be used reliably. However, based on these initial results and the fact that sensor technology has greatly improved since these measurements were collected, this study has shown the potential of low-cost sensors and how it is important to evaluate various models in a study's various locations to determine which models most accurately measure the local ambient conditions.

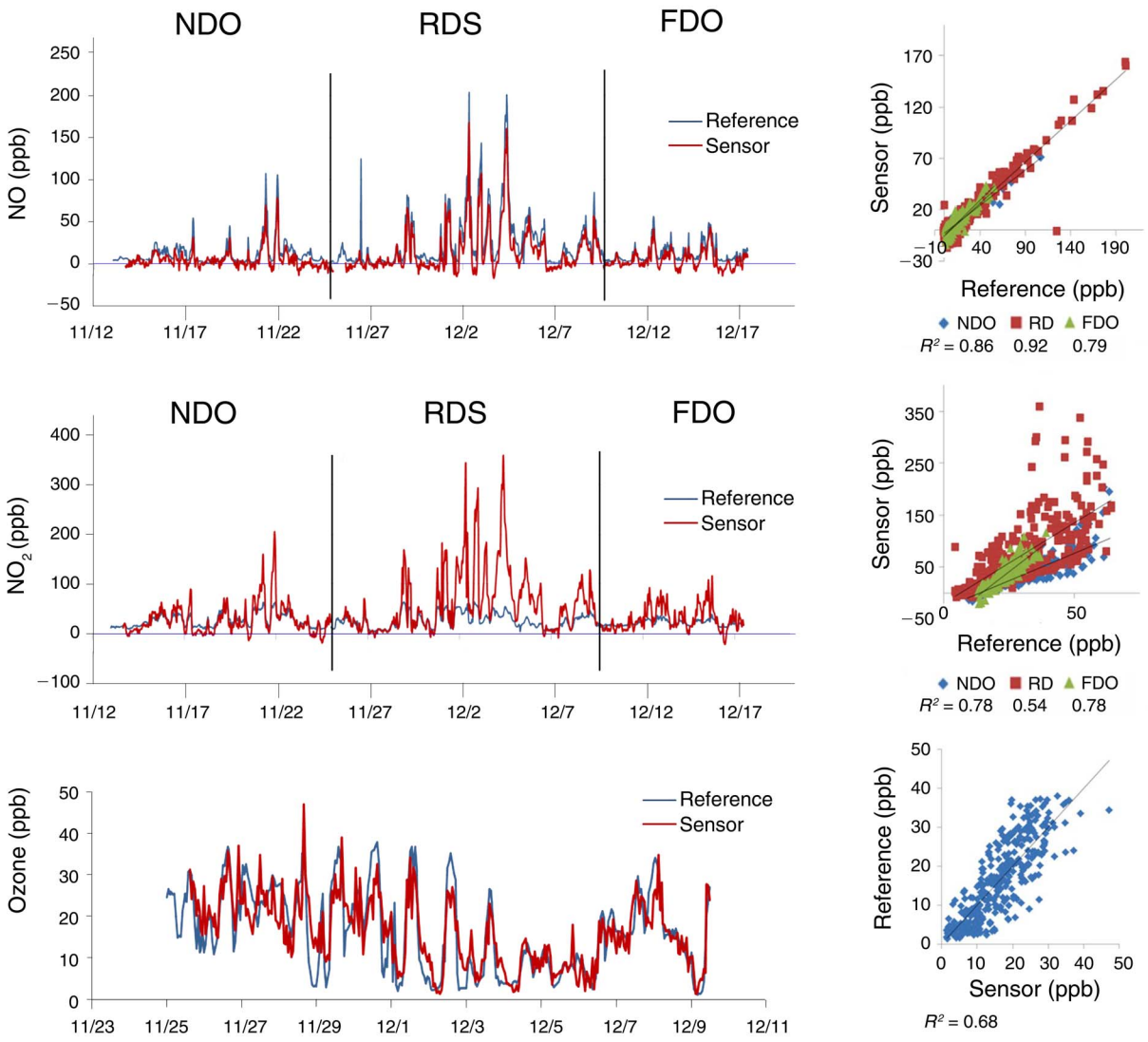

Figure 12 (*Continued*).

DISCUSSION AND CONCLUSIONS

The overarching aim of the study was to obtain a better understanding of primary traffic pollutant spatiotemporal patterns and how they relate to exposure characterization in panel-based and small-cohort health effect studies. The study location, adjacent to a major highway intersecting metropolitan Atlanta, with an annual average daily traffic count of 305,365 vehicles (Wiegand and Smith 2013), provided a unique setting for examining emission-to-exposure pathways. In total, the study team was successful in collecting extensive pollutant measurements at six sampling locations, using 26 continuous instruments, 28 weekly integrated filter measurements, and 18 low-cost sensors. A panel of 54 participants living in the study domain also contributed valuable information related to personal exposures to these pollutants and the potential for using novel metabolomics as indicators of biologically relevant exposures.

The results from over 2,800 hours of speciated measurements and modeling point to complex and dynamic conditions within and adjacent to this major near-road microenvironment. It follows that the current findings did not identify a specific pollutant indicator or model that was able to independently reflect the full nature of these observed patterns of variability in space and time. Differences between results for the RDS and EPD monitoring

sites provide an illustrative example of how variability differed between roadside monitors located within 5 m of the highway source and approximately 500 m from each other. At these two sites CO concentrations, for example, were only weakly correlated over time ($r = 0.44$) during the 4 months of sampling. Local geophysical conditions may explain some of the weak linear association; the monitor at the EPD site, for example, was sited in an area with greater foliage buffering than that of the RDS site. The apparent complexity of spatiotemporal variability increased when comparing measurements at outdoor and indoor sites with personal exposures. Within the near-road complexity captured in the study, however, we were able to identify several key findings that inform our understanding of this microenvironment and ultimately our ability to better assign exposures to primary pollution in panel-based epidemiology studies of traffic exposure.

THE CHANGING NEAR-ROAD ENVIRONMENT

Study measurements showed that today's near-highway pollutant concentrations are less elevated above background levels than those found in earlier investigations at this site and other near-road settings. Indeed, accurate and sensitive quantitation of CO at the RDS site, for example, necessitated use of a stringent zero-air calibration protocol every 15 minutes to measure concentrations that were mostly less than 1 ppm. We view these levels as reflections of general reductions in primary automotive emissions and indicative of broader trends occurring throughout North America. These reductions can also be observed in the long-term sampling data from another EPD-run site cited to measure maximum neighborhood impact of air pollution in Atlanta's South Dekalb (SDK) nonattainment site, 600 m from the principal highway bypass (Interstate 285) southeast of the GIT campus (Appendix B, available on the HEI website). NO_2 and CO concentrations at the SDK site have both decreased by about 50% since 2000 and 2003, respectively, highlighting the drastic decrease in mobile emissions in the Atlanta area. Initial results from the U.S. EPA's near-road monitoring network, pollutant trends analysis, and emissions estimates support this supposition. For example, using data from measurements around two Los Angeles area interstates in 2001–2002, Zhu and colleagues (2002a,b) found CO levels about 1.5 ppm higher near the highway versus 300 m away and BC levels that were elevated by about 4 $\mu g/m^3$ over the same distance. In spite of the high traffic density, increases of this magnitude are no longer found in Atlanta along the highway (Vijayaraghavan et al. 2014; Yan et al. 2009), which had an annual average daily traffic count of more than 300,000 vehicles in 2012 (Wiegand and Smith 2013). In contrast, we observed average increments of only 200 ppb CO, 0.8 $\mu g/m^3$ BC, and 17 ppb for NO_x between the RDS and FDO sites. The study roadside levels, while not consistent with those of earlier studies, were, however, very consistent with other current near-road monitoring results throughout the United States, reflective of broad emissions reductions throughout the U.S. traffic fleet. The mean NO_2 level measured at the EPD site during sampling, for example, was 19.5 (standard deviation 8.6) ppb. Initial monitoring results from approximately 50 near-road monitoring network sites in 2014 showed annual NO_2 means ranging from 9 to 24 ppb (U.S. EPA 2015).

Nationally, estimates of on-road mobile source CO emissions show that 2014 levels were about 49% of those in 2004 and 25% of those in 1994 (U.S. EPA, Air Pollutant Emissions Trends Data; accessed 15 August 2016a). Similarly, for NO_x, the decrease was about 50% since 2004, with some evidence that mobile source NO_x emission estimates may be biased high and that automobile emission reductions have been greater than estimated (Anderson et al. 2014). For $PM_{2.5}$, similarly, the U.S. EPA has estimated on-road mobile-source emissions decreases of approximately 43% since 2004. In the Atlanta metro area specifically, long-term analysis of data from the SEARCH network's JST site shows that CO, NO_x, and BC levels decreased by 350 ppb, 35 ppb, and 1.25 $\mu g/m^3$, respectively, from 1999 to 2011, with source apportionment analysis indicating that mobile source–related $PM_{2.5}$ decreased by about half over the same period (Blanchard et al. 2013b) (Appendix B). Zhai and colleagues (2016) estimated that mobile source PM impacts in Georgia decreased by about 30% between 2002 and 2013. These declining trends are likely to persist in the future, as new mobile source emission controls are introduced and additional policy interventions are implemented.

An immediate implication of the changed near-road environment is that future studies aimed at characterizing mobile source–related hotspots and their impact on health will need to consider using multiple approaches for characterizing spatial gradients and exposures. Specifically and most directly, ambient concentrations of single-pollutant indicators of exposure are no longer measurable to the degree they were just 10 years ago, with CO perhaps providing the clearest example of this trend. Near-road levels of key pollutants such as NO_x and $PM_{2.5}$ are also already only a fraction above urbanwide levels. Similarly, annual mean EC levels (often used as a tracer for diesel engine emissions) measured at the JST site declined from 1.6 to 0.8 $\mu g/m^3$ between 2000 and 2012. Of equal importance to the reductions in primary traffic pollutant emissions are the substantial changes in secondary atmospheric

processes associated with these reductions. Pollutants, once considered to be reliable source-indicative traffic markers, are now more predominantly associated with other sources and formation processes. The JST annual average CO, produced by atmospheric reactions of biogenic organic gases, was down to 199 ppb in 2010, while Yorkville, a rural site near Atlanta, averaged 163 ppb in the same year. Indeed, urban CO levels are approaching background levels. Again, this traditional indicator of primary traffic emissions is no longer predominantly affected by traffic. OC, also used to indicate mobile source impacts, exhibits a similar pattern; from 2000 to 2012, OC levels at the JST site have dropped from 4.1 to 2.7 μg/m^3, while Yorkville levels dropped from 3.3 to 2.4 μg/m^3). Future studies should not rely on measuring the "traditional" markers alone when assessing the impact of traffic-related emissions on a microenvironment. An added consideration is that traffic composition may change dramatically with the increased use of electric vehicles, which contribute no primary exhaust emissions but contribute to emissions from brake and tire wear and resuspended road dust.

SPATIOTEMPORAL PATTERNS OF PRIMARY TRAFFIC INDICATORS

Extensive observations and modeling were used to characterize the spatiotemporal dynamics of ambient traffic pollutants across the study domain, and both single- and more novel multipollutant indicators were examined. As discussed above, concentrations of the traditional primary traffic indicators measured (e.g., CO, NO_x, and EC) were low compared with historical levels but still elevated in the near-road environment.

Pollutant Spatial Variability

Although difficult to ascertain with precision, given location-specific idiosyncrasies that are common when conducting ambient pollutant measurements, the distances at which the traffic pollutants reached background levels, or levels common at non-near-road sites in the city, were similar to, if not slightly lower than, than those reported previously in the literature (Beckerman et al. 2008; Fruin et al. 2008; Fujita et al. 2011; Kozawa et al. 2009; MacNaughton et al. 2014; Zhu et al. 2008). Our assessment of spatial variability showed that pollutant concentrations decreased rapidly with increasing distance from the highway, with noticeable reductions between the roadside sampling sites (at approximately 5 m from the highway) and the near dorm (at approximately 20 m to 30 m from the highway). This is consistent with the results from RLINE dispersion modeling, which also showed pronounced reductions over this distance. Further reductions in CO, NO_x, and EC were measured at the more distant study monitoring locations. These findings are also broadly consistent with overall trends in reduced primary emissions. Again, although part of the decrease might have been caused by sampling location characteristics, the results from the RLINE dispersion modeling are qualitatively consistent with the ambient monitoring, showing sharp pollutant reductions over relatively short distances from the highway.

Pollutant Temporal Variability

The pollutant diurnal patterns found during sampling were consistent with those found in earlier studies showing elevated concentrations of the primary traffic pollutant occurring during the morning rush hours, when mixing is weak and emissions are high, then decreasing as the boundary layer increases. During the evening, the concentrations of primary traffic pollutants increased and remained high throughout the night in spite of the greatly reduced emissions (as estimated using the U.S. EPA's Motor Vehicle Emissions Simulator and link-based traffic estimates). Notably, there was a sharp difference between these typical, bimodal diurnal peaks for many of the pollutants and the corresponding hourly patterns for traffic count, which is itself used as a proxy for exposure to traffic pollutants in health effects studies (Gilbert et al. 2005; Janssen et al. 2003; Wichmann et al. 2005). Traffic counts on the Connector during sampling exhibited distinctive patterns of rising sharply in the early morning, concordant with the beginning of the morning rush hour, reaching peak vehicle-per-hour counts of approximately 20,000, and remaining elevated from 10 A.M. to 4 P.M. The differences in diurnal patterns between traffic counts and traffic pollutant levels were striking and highlighted the predominant role meteorology, and specifically its influence on vertical dispersion, have on the impact of a traffic hotspot on adjacent areas. The regression model results in Table 3 highlight the diminished predictive power of traffic count on roadside pollutant levels. In this model, which examined how various periods of the day explained corresponding levels at the RDS site, the midday period (from 10 A.M. TO 3 P.M.) — during which traffic counts were highest — was found to be the only period that did not have statistically different concentrations of the traditional primary traffic pollutants compared with the evening rush hour period.

Correlations between pollutants measured at the RDS site and the other sites in the study sampling area varied by pollutant and time of day. Generally, temporal correlations were moderate to strong between measurements at the highway source (i.e., the RDS and EPD sites) and at the

other sites throughout the campus area, including the more distant sites from the highway (e.g., the FDO site). Hourly resolved correlation analysis showed, however, that the strengths of the linear associations varied markedly throughout the day. This was most apparent for the primary traffic pollutant NO, for which correlations between the roadside sites were strong throughout the sampling domain during rush hour periods (6 A.M.–10 A.M.). The correlations decreased during the later morning and afternoon, even over the small domain studied here; these are periods of higher thermally driven turbulence and dispersion. Further, regression analysis showed that distance from the highway was a dominant explanatory variable. Although neither of these findings was wholly unexpected, the degree of reduced correlation in the afternoon is important in terms of interpreting how well near-road observations reflect temporal variability patterns in other locations. Our results suggest that the spatial environment over which a near-road monitor can be used to assess temporal variability patterns is limited. For epidemiological studies that are specifically looking at highly resolved exposures and responses, it may not be sufficient to rely on near-road monitoring as a surrogate of exposure. The afternoon correlation for EC was about 0.5 between the near-road RDS site and the NDO site, which was only 20 m from the highway. CO dropped from a correlation of 0.8 in the morning to approximately 0.6 in the afternoon. Again, the decreased correlation may be due, in part, to specific sampling location issues, but the trend toward weaker linear associations during the afternoon for all the single-pollutant primary traffic indicators at the more distant monitors suggests that additional observations or modeling tools should be used to characterize pollutant dynamics over such scales when a study is relying on capturing temporal exposure trends.

A key finding was related to differences in the spatiotemporal behavior of NO_2 and that of CO and BC, two other single-pollutant primary traffic indicators. During sampling, NO_2 levels did not exhibit as strong a spatial gradient as CO and BC did. Absolute NO_2 concentrations were moderately homogeneous from the RDS site to the FDO site. This is caused, in part, by kinetic limitations in the photochemistry required to convert the NO-dominant primary NO_x, emitted from automobiles, to NO_2. It is possible that some of the stoichiometric dynamics involved are more broadly related to the lower primary traffic emissions noted above. Related to these processes were the observed O_3 levels, which were on average low. Peak O_3 levels during the day averaged about 30 ppb, well below the National Ambient Air Quality Standard of 70 ppb. Mean O_3 levels at night were between 15 ppb at the JST site and 5 ppb at the EPD site, showing more NO scavenging when diffusion of O_3 from aloft was inhibited. O_3 observations during the study showed typical diurnal trends, with low levels at night and higher levels during the day, and were relatively homogeneous spatially. The daytime O_3 levels near the road were slightly lower than those of locations farther away, which was expected because of the titration of O_3 by fresh NO emissions. However, the decrease in O_3 was much smaller than would have occurred if all of the NO reacted with O_3, showing that titration was kinetically (e.g., due to turbulent diffusion and advection from the road) limited over the time scales at work. This has implications related to how models should capture NO-to-NO_2 conversion when being used to assess exposure in near- and on-road environments.

Finally, study measurements clearly pointed to NO_2 levels that were generally higher inside both dorms compared with the outdoor environment, highlighting non-outdoor source contributions to indoor concentrations. Collectively, the current results suggest that NO_2 might not capture the true patterns of variability associated with primary traffic emissions at this site and thus might serve as a less useful exposure indicator than the other single pollutant indicators NO, CO, or BC, particularly when considering personal exposure monitoring, given both the elevated levels of NO_2 and the amounts of time spent indoors.

The study was designed to inform questions related to the potential impact of spatial and temporal variability patterns of primary traffic pollutant indicators on epidemiologic analyses of traffic-related health effects. A means of formalizing these observations would include simulated estimates of the impact of uncertainty within a measurement error framework, using the study's empirical spatiotemporal variability measurements. In the absence of this formal analytical exercise, which is being conducted by our group, the current results do indicate that for panel-based and small-cohort studies, the use of roadside measurements as surrogates of exposure to primary traffic pollution is likely to result in exposure assignments that differ substantially from direct measurements of primary traffic emissions. We observed this to be true even for those participants living in residences within 20 m of the roadway source, with greater differences for those living farther from the roadway.

INDICATORS OF PERSONAL EXPOSURE TO PRIMARY TRAFFIC POLLUTION

Nested within the intensive study's ambient and indoor monitoring was a personal exposure sampling campaign, which included 51 48-hour sampling sessions that included participants from both dorms. As with many panel-based

personal exposure designs (Ebelt et al. 2000; Janssen et al. 2000; Sarnat et al. 2005), these results have the potential to inform discussions about the suitability of the use of personal exposure surrogates. For these analyses, we used the results from the personal exposure study to address issues related to participant mobility patterns and whether roadside temporal variability reflected corresponding personal exposures to specific traffic indicators. We were limited in the pollutant targets we could measure using our personal monitoring platform. For example, current methods for characterizing personal exposure to CO do not have sufficient sensitivities to reliably measure levels similar to those observed in this study, and thus we were not able to directly measure exposure to this primary traffic pollutant. Personal IMSI exposures were also generated using modeled, spatially resolved RLINE concentrations linked to concurrent GPS location. Given this limitation, analyses assessing personal exposure surrogates focused on results for BC and NO_2 exclusively, because these constituted the only traffic-indicator pollutant species that were directly measured.

Our geospatial mapping and GPS waypoints histogram clearly indicated that the study participants tended to stay in or near their places of residence during much of the time they participated in their personal exposure assessments. Thus, the participants who lived in the dorm nearest the highway spent more time close to the highway compared with the participants living in the far dorm. This finding was not unexpected for a college cohort, for whom daily weekday mobility patterns may be more limited than those for a working adult population. This result, combined with the pronounced pollutant gradients from the highway, led us to hypothesize that personal exposures to both BC and NO_2 would be higher among participants living in the near dorm. Instead, personal pollutant concentrations were comparable among the participants from both dorms. This result was likely caused by several factors related to the potential true differences in source contributions to personal pollutant exposures as well as to artifacts of the study's personal sampling design. Indoor NO_2 and other non-ambient source contributions may have contributed to total personal NO_2 exposures in ways that may have led to an overall attenuation of the differences in exposure between residents of the two dorms. Alternatively, as mentioned above (Aim 3 Results), weekly participation in the personal exposure monitoring was not balanced by dorm because of logistical considerations, with a greater number of participants living in the far dorm than in the near dorm. It is possible, therefore, that weekly ambient pollutant levels were higher in weeks when more participants in the far dorm participated in personal exposure monitoring compared with other weeks.

Although the direct personal exposure measurements were limited to NO_2 and BC, the results from the personal exposure monitoring also point to the complexity and diversity of the patterns of variability in space and time among the monitoring sites and the importance of accounting for location and spatial mobility when estimating exposure in panel-based and small-cohort studies. This was most clearly demonstrated with the personal BC measurements, where ambient roadside monitoring was shown to be a poor surrogate of corresponding exposures to this pollutant. Alternative surrogates, including ambient and indoor BC at the participants' respective dorms, were more strongly associated with personal BC, for which spatial and temporal variability patterns differed, than with those measured at the highway. Moreover, although not statistically significant, the participants' mean proximity to the highway also explained a substantial level of the variability in corresponding personal exposures to both BC and NO_2, again highlighting the importance of capturing individual mobility for panel-based exposure studies.

Collectively, the personal exposure results point to the need to develop more sensitive indictors of exposure, which could include microsensors and biologically based indicators of exposure, such as metabolomics. The combination of the RLINE dispersion model with the geographic information system (GIS) data showed promise in improving the personal exposure prediction performance and for informing future personal exposure assessments, because it does not require extensive monitoring data measured inside or outside participants' residential locations and it accounts for variances in personal activity via precise GIS data. An advanced, fully specified model incorporating ambient pollutant measurement, residential proximity and location, and data on personal activity and meteorology might also improve the prediction performance; such a model is currently being developed as an extension of the RLINE model but is beyond the scope of the current report. Similarly, future work using data from the personal exposure monitoring will link exposures with corresponding metabolomic response.

PERFORMANCE OF THE MULTIPOLLUTANT INDICATORS OF PRIMARY TRAFFIC EMISSIONS

A key aim of the study was to assess the potential of two multipollutant indicators to characterize exposure to traffic-related pollutants. FPMOP levels were quantified by the depletion of DTT in the presence of ambient aerosol in aqueous extracts. Laboratory studies have shown mobile source emissions to be DTT-active (Li et al. 2009;

McWhinney et al. 2011, 2013a, 2013b; Rattanavaraha et al. 2011). Field studies further support the link between DTT activity and mobile sources (Bates et al. 2015; Biswas et al. 2009; Boogaard et al. 2012; Janssen et al. 2014; Kelly et al. 2011; Verma et al. 2011, 2012; Yang et al. 2015a). Studies have also found an association between measures of FPMOP and various health outcomes (Bates et al. 2015, 2016; Bilenko et al. 2015; Boogaard et al. 2012; Dadvand et al. 2014; Steenhof et al. 2011, 2013; Yang et al. 2015b, 2016). Yang and colleagues (2016) found that $FPMOP^{total\text{-}DTT}$ was more strongly associated with respiratory outcomes than was $PM_{2.5}$, while another measure of FPMOP, using electron spin resonance, was not found to be associated with these same outcomes. Similarly, we previously found that $FPMOP^{WS\text{-}DTT}$ was associated with respiratory and cardiovascular health outcomes in a long-term analysis, while FPMOP characterized using an ascorbic assay was not, and that the DTT assay is more sensitive to organic compounds (Fang et al. 2016). Collectively, these findings led us to hypothesize that $FPMOP^{WS\text{-}DTT}$ may constitute an alternate indicator for characterizing mobile source hotspot environments.

Similar to overall study results highlighting the complexity of the near-road environment, the current finding also supports the conclusion that the relationship between $FPMOP^{WS\text{-}DTT}$ and direct mobile source emissions is highly complex. The complexity of this relationship, however, is important. First, $FPMOP^{WS\text{-}DTT}$ was shown to be moderately correlated with the traditional and more widely measured markers of mobile source emissions (e.g., EC/BC, NO_x, and CO). Pearson correlations between $FPMOP^{WS\text{-}DTT}$ and the species measured at the RDS and NDO sites ranged from about 0.4 to 0.8. However, the correlations dropped off with distance from the roadside, and the levels did not decrease as much as for the single-species primary traffic species. As discussed, atmospheric processing appears to play an important role in these spatial trends. Atmospheric processing would lead to a continued correlation with other co-emitted species but alter the relationship (both the degree of correlation and the relative magnitude), which is what we found during sampling.

Earlier research suggested that mobile source–derived pollutants would be enriched in ROS-active species, leading to expectations of higher FPMOP at near-road sites (Bates et al. 2015; Cho et al. 2005; Verma et al. 2014). An important finding from the study observations suggests that, although roadway emissions are a source of components that contribute to FPMOP, secondary processes are required to convert the roadway emissions to species with measurable OP for $FPMOP^{WS\text{-}DTT}$. A major contribution of this work was extending this finding to $FPMOP^{total\text{-}DTT}$, which was found to behave similarly to $FPMOP^{WS\text{-}DTT}$. Thus, neither $FPMOP^{WS\text{-}DTT}$ nor $FPMOP^{total\text{-}DTT}$ is largely directly emitted by traffic, and so these are not appropriate as multipollutant indicators of primary traffic emissions. This can be explained by the fact that FPMOP is associated with primary traffic emissions that have undergone a chemical transformation. For example, quinones and hydroxyquinones are highly DTT-active (Verma et al. 2015b) and are derived from the oxidation of combustion-emitted PAHs, whereas PAHs themselves do not contribute to $FPMOP^{total\text{-}DTT}$ (Cho et al. 2005). Water-insoluble species that contribute to $FPMOP^{total\text{-}DTT}$ must also undergo some degree of processing (acid dissolution, in this case).

Although measurements of $FPMOP^{total\text{-}DTT}$ were made subsequent to the intensive 4-month sampling period, comparisons between the EPD roadside site and the RDS near-road site resulted in similar findings as those for $FPMOP^{WS\text{-}DTT}$. Both forms of FPMOP, as measured by DTT, had important and unexpected spatial patterns, revealing a complex relationship between mobile source emissions and FPMOP. Specifically, the results here underscored the importance of the influence of atmospheric processing on FPMOP levels. Our findings further suggest that directly emitted traffic pollutants (e.g., PAHs and metals) are oxidized (the PAHs) or converted to soluble forms (the metals, by acid dissociation), leading to other species and an increase in both $FPMOP^{WS\text{-}DTT}/m^3$ and $FPMOP^{total\text{-}DTT}/m^3$ levels. If, as recent studies have suggested (Bates et al. 2015; Fang et al. 2016; Weichenthal et al. 2016; Yang et al. 2016), OP measured using DTT assays is associated with various health outcomes, then there is increased importance in understanding the atmospheric transformation processes that increase the OP activity of pollutant emissions.

It is somewhat surprising that atmospheric processing also affects spatial distributions of $FPMOP^{total\text{-}DTT}$. It is, for example, generally believed that the oxidation of primary organic aerosol leads to water-soluble components; thus if oxidation is needed to make vehicle-emission components of FPMOP, one would expect $FPMOP^{total\text{-}DTT}$ to be nearly equal to $FPMOP^{WS\text{-}DTT}$. The data presented here show there is a measurable fraction of $FPMOP^{total\text{-}DTT}$ (~35%) that is not water soluble yet still requires atmospheric processing to be DTT-active (i.e., to contribute to FPMOP). An example, as noted above, is that of PAHs on the surface of insoluble soot emitted by incomplete combustion. These only become a measurable contribution to $FPMOP^{total\text{-}DTT}$ when oxidized by O_3, resulting in the PAHs' conversion to quinones. The particle remains insoluble, but it is now DTT-active. Other types of processes may also occur. Water-soluble transition metals, for example, contribute significantly to FPMOP (Charrier and Anastasio 2012; Cho

et al. 2005; Schoonen et al. 2006; Vejerano et al. 2015). In the current study, we have also shown that for a selected group of transition metals, the metals are largely not emitted from traffic in a water-soluble form and that at the spatial scales of the study, they are not transformed to water-soluble components when measured at a near-road site compared with a roadside site.

The implications of our work, along with that of other research, are that FPMOP, as characterized by the DTT assay, is a valuable component to measure as part of studies aimed at characterizing mobile source impacts on both air quality and health, though the relationship is complex. FPMOP is a more integrative measure and includes aspects of atmospheric transformation of mobile source–related pollutants; it is also sensitive to non-tailpipe emissions (brake and dust). Background and secondary pollutants, such as widely dispersed emissions from biomass burning, are also a main driver of FPMOP. For a changing near-road environment, where the influence of primary traffic is becoming less pronounced, this is important.

The other novel indicator investigated in this study was the IMSI. IMSI measurements are a combination of EC, CO, and NO_x observations, so it is not surprising that IMSI values are very highly correlated with each of these species individually. It is not apparent from the current findings that the IMSIs measurements captured aspects of variability, either in space or time, that differed much from the traditional, single-pollutant tracers. Based on these results, we did not feel this multipollutant metric represented the optimal approach for characterizing primary traffic emissions over the limited temporal and spatial scale of the study. Despite this, IMSIs are not as sensitive as individual species to non-mobile sources emissions (e.g., EC and BC or CO from biomass burning) and are formulated to account for emissions from other sources that contribute to an urban background. Roadside measurements show that IMSIs are also similarly correlated with $FPMOP^{total\text{-}DTT}$ and individual components (Appendix B, available on the HEI website). This, along with an earlier study showing IMSIs to be slightly more strongly associated with acute cardiovascular outcomes in population time-series studies in Atlanta than with individual traffic components (Pachon et al. 2012) and a study finding IMSIs to be more strongly associated across monitoring sites than with single pollutants or positive matrix factorization factors (Oakes et al. 2014), suggests that IMSIs may still be useful in future work examining multipollutant mobile source impacts on health.

In our study, multiple ambient sampling sites provided direct measurements of traffic-related pollutant concentrations near the highway. RLINE modeling conducted at a very fine 25-m resolution provided additional spatial information and linkage of air pollutant levels to not only the highway, but also to the surface streets on and around the GIT campus. The RLINE modeling specifically targeted the fraction of emissions originating from the highway and proved valuable in characterizing the spatial and temporal gradients found in our study domain. The resulting spatial fields show the concentration trends that were not captured by the measurements. The resolution was chosen to capture concentrations at a resolution that could be used in our personal exposure assessment, providing simulated ambient exposures in areas where observations were not available and at time scales not captured by personal monitoring. RLINE modeling can also directly link concentrations to sources. The RLINE results indicated that, although the highway certainly led to the highest levels of ambient traffic-related pollutant levels, local surface streets around campus could lead to locally elevated levels very near the streets that decreased rapidly with increased distance from those streets. However, on-campus traffic led to relatively small contributions in the core study area. This is important with respect to assessing personal exposures, because participants walked to various activities. For the most part, the interstate and the main roads around, not on, campus were the primary contributors.

Together, the study results highlight the difficulty of adequately capturing the desired spatiotemporal pollutant dynamics using traditional sampling approaches. Even with the extensive sampling conducted here, measurements alone could not provide detailed spatiotemporal fields for exposure assessments that account for the complex traffic networks in the area. Some level of modeling, be it dispersion modeling as conducted here or other approaches (e.g., land-use regression) (de Hoogh et al. 2014; Hoek et al. 2008; Yang et al. 2015b, 2016), is suggested. Dispersion modeling has the benefit of providing direct source–receptor relationships, but as found here, the models at present require some level of calibration.

USE OF ENVIRONMENTAL METABOLOMICS IN PANEL STUDIES

Untargeted metabolomics-based indicators represent emerging, yet still uncertain, means for measuring exposures and responses to potentially thousands of xenobiotic agents (Ellis et al. 2012; Park et al. 2012; Walker et al. 2016). To date, most examples of using environmental metabolomics have either been in cohorts of several thousand (Ganna et al. 2016; Pallister et al. 2016) or in smaller panels of individuals exposed to extremely elevated concentrations of specific chemicals in occupational settings (Dudka et al. 2014; Romano et al. 2012; Wang Z et al. 2015; Wei et al. 2013). In our study, as part of a central exploratory aim, we

specifically assessed whether changes in specific metabolic profiles associated with environmental exposures were discernible in repeated plasma and saliva samples from the student participants. To our knowledge, the work conducted as part of this aim constitutes the single largest prospective assessment so far of traffic pollution exposures and potential metabolic responses.

Generally, the results from the metabolomics analyses provided a strong basis for continuing this work toward chemical validation of putative biomarkers of traffic-related pollution. Work conducted as part of this aim informs questions related to the analytical sensitivity of high-resolution platforms to generate high-quality data, the future feasibility of using plasma and saliva metabolite concentrations as exposure indicators in relatively small environmental exposure assessments, and optimal biological matrices for use in research applications. Broadly, we were satisfied with the overall metabolomics data quality when assessed using accepted data quality standards related to the coefficient of variation among the triplicate measures and to ion abundances in the panel (Go et al. 2015; Uppal et al. 2013). The current results demonstrate that it is possible to reliably extract tens of thousands of features using our high-resolution mass spectrometry platform. The number of features, a unique aspect of high-resolution mass spectrometry techniques compared with other methods (e.g., nuclear magnetic resonance spectroscopy), is critical in that it enhances the potential discovery of new markers both of exposure and of biological responses associated with exposures of interest.

In total, approximately 20,000 and 30,000 features were extracted from plasma and saliva, respectively. Of these, roughly 20% were matched by mass and identified in both biomatrices. Similar to other high-dimensional methods, the features in the data were not identified. Annotation and formal validation of the ions in the study samples, via tandem mass spectrometry, were beyond the scope of work for the current study. Despite this, our plasma metabolomics results pointed to the presence of distinct metabolite fingerprints differentiating participants living in the dorm closer to highway from those living in the dorm farther from the highway. Using mixed effects modeling while controlling strictly both for multiple comparisons and for other factors likely to affect metabolite expression, we identified 221 features that differed significantly by dorm. The bimodal distribution of these features in the HILIC column was highly idiosyncratic, with one peak consisting of features with elevated intensities for participants in the near dorm and the other consisting of features with elevated intensities for participants in the far dorm. Both peaks were characterized by relatively short retention times, indicative of the hydrophobicity of the identified features. Moreover, the features that differed significantly were abundant in many of the participants, providing some indication that the differences were not driven by extreme responses in relatively few participants. It should be emphasized that it is not possible, at this stage in the metabolomics analysis workflow, to infer that any difference was associated with differences among the participants in terms of their exposure to primary traffic pollution.

It is worth emphasizing that the current metabolomics analyses included only a rough indicator of exposure to primary traffic pollution, namely residential dorm location. It is possible, and perhaps likely, that the observed differences in the expressed features, by dorm location, reflected other factors and exposures unrelated to traffic sources. Confounding by cleaning agents used at the two dorms, cafeteria-related dietary differences, or other unknown and unspecified factors may be the true source of the differences in these profiles. Although we designed the study to recruit panels of participants from both dorms that were demographically balanced, discrepancies existed. Participants in the near dorm had lived in their dorm rooms a month longer, on average, than participants in the far dorm (Table 6), potentially contributing to differences in exposure. Similarly, the far dorm panel included a larger fraction of sophomores, comprising approximately 43% of the total panel from this dorm compared with the near dorm panel (approximately 9% sophomores). Despite this, we believe that these findings do reflect some measures of environmental difference in exposures in the two dorm panels, which may include diet or other non-traffic pollutant sources, providing support for future annotation of the features as well as external validation of their identity.

In contrast with the plasma metabolomics results, findings for the saliva samples were more equivocal, with some indication, albeit nonsignificant, that between-dorm differences in feature expression were similar to those found in the plasma metabolomics results. Overall, only one saliva feature differed significantly by dorm in the mixed effects modeling results (in contrast with the hundreds found in the plasma samples). These biological matrix differences may have been caused by several factors, including differential sampling and analytical errors involved in the collection of saliva compared with blood. Although we adopted consistent methods for the collection of both sets of biological samples, there was inherently more variability in the saliva sample collection because it was done by the participants themselves using a passive drool protocol. The blood samples, in contrast, were drawn by four trained phlebotomists. It is also possible that the differences in the feature profiles might have been

related to the biologically relevant metabolites of exposure existing only in plasma and not in saliva. Future validation of the features that differed by dorm may elucidate the nature of the differences in the results by biological matrix. Until these further analytical steps are completed, our findings support the use plasma over saliva as a preferred matrix for reflecting differences in metabolomic profiling.

Collectively, the metabolomics results demonstrate initial proof of concept for this approach in being able to resolve statistically robust metabolic differences in a small panel setting. Future steps in our work will include additional analyses examining associations between specific pollutant metrics and corresponding metabolite feature expression and the validation of specific metabolites with the goal of identifying metabolic products of exposure or responses to traffic pollutants.

LOW-COST POLLUTANT SENSOR SAMPLING PLATFORMS IN A FIELD SETTING

Intensive field sampling using traditional monitoring methods is both very demanding in terms of personnel time involved in siting, deploying, and maintaining equipment and assessing and analyzing the data and very costly in terms of expenditures on equipment, calibration gases, and maintenance. Further, siting limitations can prohibit placing monitors where desired. Therefore, large reference instruments alone could not capture pollutant dynamics or detailed inputs for exposure assessment.

Low-cost sensors, although still in the development stage, are also seen as being potentially attractive for studies such as ours. Because of their ease of use, small size, and low power consumption, low-cost sensors can be deployed in larger numbers compared with traditional instruments, providing more spatially resolved measurements. The attractiveness and potential utility of such sensors was borne out in a supplemental component of the current study. In particular, after calibration the gas-phase sensors, but not the PM sensors, proved reliable and much easier to deploy and operate than traditional instrumentation. In spite of findings from other studies that have provided support for the potential utility of low-cost PM sensors (Austin et al. 2015; Holstius et al. 2014; Piedrahita et al. 2014; Reis et al. 2015), their performance in this study would likely not have led to reliable results. However, it is important to note that PM sensor technologies are evolving rapidly, and future studies of this kind should strongly consider using both gas-phase and PM pollutant sensors. Such studies should stress calibration of the low-cost sensors against reliable and well-calibrated instruments.

LIMITATIONS

Care should be taken in interpreting the study results, and we wanted to present what is surely an incomplete list of some the most prominent limitations. First, the location, while well suited for our purpose, was still a single location with its own unique characteristics. The section of highway running next to GIT has a higher proportion of light-duty gasoline versus heavy-duty diesel vehicles than other area highways. Atlanta is effectively in the middle of a forest with a substantial canopy of urban foliage. Biogenic emissions are thus substantial contributors to atmospheric emissions, particularly VOCs, that oxidize to form an important fraction of the PM and CO in the region. The acellular $FPMOP^{WS\text{-}DTT}$ assay is one of a variety of measures of PM OP. Although there is a growing body of literature using this assay, the literature is still limited. Further evaluation of $FPMOP^{WS\text{-}DTT}$ and non-water-soluble DTT is important. The use of integrated sampling on filters instead of online techniques for the OP analysis limited the interpretation of OP results in this study in a number of ways: correlations analysis of integrated samples is less effective than time-resolved data, no diurnal trends data are available, and artifacts, especially semivolatile species known to be associated with fresh traffic emissions, can lead to either positive or negative artifacts. The other multipollutant metric studied here, the IMSI, has seen limited use, and although it has some attractive characteristics, it is not apparent from our results that it was the best approach for deriving such an indicator for use over the limited temporal and spatial scale of the study. A near-road air pollution model had significant spatial and temporal biases, and our calibration efforts did not remove all the errors. Use of air pollution models, therefore, should include recognition that such errors exist. The intensive sampling was conducted over a period of roughly 4 months. Meteorological factors, including photochemically driven pollutant formation and other transport processes were, accordingly, limited to conditions and seasonal patterns similar to those during field sampling. It is worth noting, however, that the field campaign start and end dates were intentionally designed to maximize variability in meteorological conditions.

Finally, although we viewed as promising the results from the largely exploratory aim involving the use of environmental metabolomics as potential biologically based traffic indicators, we also wish to stress that these analyses reflected metabolomic differences associated with a broad indicator of exposure (i.e., residential location) and thus may not be associated with any environmental exposures relevant to the study aims.

DISCUSSION AND IMPLICATIONS OF FINDINGS

The overarching goal of the study was to evaluate the suitability of single and multipollutant traffic indicators for use in small-cohort and panel-based epidemiological studies. Toward this end, several salient findings emerged from our analyses. The first finding documented the reduced impact of the highway source on its surrounding vicinity. The Atlanta Connector is one of the busiest, most congested highway arteries in the United States. Pollutant levels measured during the study were much lower than previous measurements at these sites and were broadly consistent with results from other studies throughout North America that have pointed to a reduced relative contribution to urban air pollution from primary traffic emissions. We view these reductions as an indication of the effectiveness of mobile source controls (e.g., Georgia EPD 1999, 2013; U.S. EPA 1999, 2000, 2005, 2011).

The reductions have implications in terms of the types and composition of human exposures to traffic pollution on and near roadways and of the pollutants that we can reliably use as indicators of exposure to primary traffic sources. The study was designed to examine various aspects of this issue through five specific aims.

Specific Aim 1 focused on associations between traditional measured and modeled pollutants and novel traffic indicators at our dedicated near-road monitoring site. Our findings clearly showed that NO_2, among the pollutants frequently used as source-indicative tracers of traffic pollutant, exhibited patterns in space and time that differed from those of the other single-species tracers and of our multipollutant candidate indicators, FPMOP and the IMSI. Throughout our sampling domain, NO_2 levels were more homogeneous compared with those of the other pollutants. We believe this difference is also related to reductions in primary emissions and corresponding changes in near-road NO_x chemistry. An additional, unexpected example of the varying role of traffic indicators involved traffic counts on the highway. Although vehicle numbers certainly drive emissions and measured pollutant levels, the impact of traffic count was modified by time of day and by differential meteorological processes that affect local dispersion patterns. Put differently, the times of the day with the most vehicles on the highway were not the same times when traffic pollutant levels were highest, and traffic count alone was not a suitable predictor of pollutant concentrations. For panel-based and cohort studies, the choice of a traffic indicator must therefore be carefully considered and not assumed to be valid based on historical pollutant levels and study designs.

The results also identified specific patterns of pollutant spatial distribution with implications for how we assign exposure to traffic emissions. Although absolute roadside pollutant levels were lower, the monitoring did show elevated levels of vehicle-derived pollutants such as BC, CO, and NO in the near-road environments (both the roadside and near-highway dorm sampling locations) compared with those of other sites. For exposure characterization in panel-based and cohort designs, this also has implications, particularly for designs that use proximity as a surrogate of exposure, as is common in many epidemiological studies of traffic pollution (Hoek et al. 2002). Our findings suggest that earlier ordinal proximity–based surrogates of exposure may no longer reflect current spatial distributions of primary traffic pollution.

Specific Aim 2 assessed the spatiotemporal variability of outdoor and indoor primary traffic pollutant components as well as multipollutant indicators along a near-road to mid-distance spatial gradient (5 m to 2.3 km) from the traffic pollution source. As with spatial variability, we observed several patterns of pollutant temporal variability that we believe may have meaningful implications for traffic pollution exposure assessment. These results are especially pertinent, given the recent efforts in the United States to establish an extensive network of near-roadway monitoring. The degree to which near-roadway monitoring reflects corresponding temporal patterns in pollution at adjacent sites was therefore a primary interest of ours. With respect to this question, our findings showed varying degrees of temporal linear association between our RDS site and the other sites in the sampling area. Broadly, and not unexpectedly, correlations tended to be strongest between the RDS site and the sites located most closely to it. When assessed as mean concentrations over 24-hr periods, correlations were typically moderate to strong for CO, NO, BC, NO_2, and the multipollutant indicators we considered. A more complex picture emerged when examining the diurnal pattern of correlation, or correlations resolved at each hour of the day. For these analyses, our findings showed that roadside monitoring tended to be more strongly correlated with sites both near and far from the road and during morning rush hour periods and often to be weakly to moderately correlated during other periods of the day. This pattern is likely associated with diurnal changes in mixing and chemistry and their impact on spatial heterogeneity across the campus.

We examined two very different types of multipollutant indicators in the study, one based on observations of more traditionally monitored species, the other based on an acellular assay of ROS as a component of PM. As discussed above, earlier research has suggested that mobile source–derived pollutants would be enriched in ROS-active species. An important finding from the observations conducted

as part of the current study, along with recent measurements from the SCAPE study, suggests that, although fresh exhaust emissions do have elevated ROS activity, the ROS activity increases with atmospheric processing. This can be explained in part by the formation of quinones and hydroxyquinones from the oxidation of PAHs and further by the oxidation of transition metals. Thus, although ROS activity as measured by DTT can be used as an indicator of mobile source emissions and potential health impacts, these results suggest that atmospheric processing is also potentially important to the resulting health effects. Further, the DTT assay is not a good indicator of fresh automotive emissions. Although a limitation of this assay is that it is acellular, past studies did find that $OP^{WS\text{-}DTT}$ activity was more strongly associated with emergency department admissions for asthma and cardiovascular disease than were other traffic-related pollutants such as NO_2, BC, CO, and $PM_{2.5}$.

The ROS assay indicator led to a very different, and very important, conclusion with respect to the potential importance of multipollutant indicators of traffic-related pollutants. Specifically, traditional single-pollutant indicators (e.g., OC, EC, CO, NO_X, or individual VOCs) do not capture the increase in per mass ROS, and some of these pollutants are even photodegraded rather than components of ROS. Given recent findings from our earlier studies and those by other groups, increased ROS activity may be a more powerful indicator of potential health impact than any single pollutant indicator. Future studies of the impact of traffic-related pollutants on health should strongly consider going beyond measuring only the traditional pollutants associated with traffic-related emissions and using ROS-related measurements, given their potential linkage to health. IMSIs or many other approaches using regularly observed pollutants to develop multipollutant indicators of traffic impacts (e.g., positive matrix factorization) would not capture the changes in ROS activity. Although not studied here, more advanced photochemical modeling can theoretically capture atmospheric transformations, but this is not a capability of current models. The current results suggest that alternative measures of traffic-related ROS activity may also be useful, beyond the DTT-based assays we used.

In earlier studies in Atlanta (Pachon et al. 2012) and elsewhere (Oakes et al. 2014), the IMSI multipollutant approach tended to have stronger correlations between sites than the single pollutants did and also slightly stronger associations with observed health endpoints. From the DRIVE results, however, it was not apparent that IMSIs captured aspects of variability, either in space or time, that differed much from those captured by the traditional single-pollutant tracers. The IMSI did not provide much added insight into exposures to traffic-related pollutants over the spatial domains considered, and we collectively did not feel that this multipollutant metric represented the optimal approach to characterizing primary traffic emissions over the limited temporal and spatial scale of the study. This may have been caused by a variety of factors, including the limited sampling duration (5 months), the closeness of the monitors to each other or the fact that the approach was not as useful as had previously been found.

Nevertheless, our results can provide insights into the use of multipollutant indicators in future exposure and health studies. IMSIs in particular can take advantage of observations coming from near-road network monitoring. Earlier studies using this approach considered larger spatial domains. We believe our current findings support further analysis to determine spatial and temporal scales appropriate for this approach and whether a similar alternative multipollutant indicator may provide additional insights.

Specific Aim 3 dealt specifically with evaluating how well traffic pollutant components and multipollutant traffic indicators measured at near-road and other fixed monitoring sites reflect corresponding personal exposures. We generally found weak associations between the roadside monitors and the corresponding personal exposures. Neither of the near-road predictors of personal BC, concentrations measured at the RDS site and the RDS IMSI, was associated with corresponding personal BC. In contrast, personal BC was moderately and significantly associated with measurements both outside and inside of the participants' respective dorms. Based on these results showing stronger associations between personal BC and monitors located closer to the participants' residences, it followed that a simple dorm location term to predict personal exposure was also a significant predictor of personal BC exposure, explaining a roughly similar degree of variability.

Notably, for both of the personally measured pollutants (i.e., BC and NO_2), using the mean distance from the highway of a participant during the 48-hour sampling period was shown to explain more variability in corresponding personal exposures than did other factors tested. The distance from highway term was not itself significant in either pollutant model, however, indicating a greater likelihood that the observed results were caused by chance. Similarly, neither IMSI value measured at the RDS site was shown to be associated with personal BC or NO_2.

Fine-scale air quality modeling may provide additional detail for use in panel-based and small-cohort assessments of exposure to primary traffic pollution. In our case, we

used the RLINE dispersion model along with a high-resolution link-based emissions inventory. Although the application of RLINE did provide concentration fields at a very fine-scale 25-m resolution for each of the pollutants simulated, biases were found in the hourly concentration data. In particular, very high concentrations were found directly over and near the major roadways. Although part of the high bias may have been caused by a bias in the emissions, the reduced bias farther away from the major roadways would suggest that the near-source bias was caused by model parameterizations. Model calibration was used to make use of the model results in the exposure assessment, though the fit remained low. Land use regression models could provide additional spatial detail, similar to the dispersion model, but would require further field measurements and would not tie levels directly to the source or explicitly account for meteorological impacts.

Specific Aim 4 explored the feasibility of using environmental metabolomics as an approach in panel-based or small-cohort studies for identifying environmental exposures, including primary traffic. We were encouraged by the results from our novel biomonitoring analyses. Although these findings have yet to be validated, we view the initial results as promising. The metabolomics approach has the potential of generating new hypotheses and guiding traffic pollution exposure research in the development of new, biologically relevant exposure indicators for panel and cohort studies. It is important to emphasize that the current metabolomics analyses and findings used a broad, nonspecific indicator of traffic pollution exposure, namely participant dorm location. These results thus say little about actual exposure to pollutants associated with primary traffic. Future studies will include analyses examining associations between specific pollutant metrics and corresponding metabolite expression as well as validation of specific metabolites for identifying metabolic products of exposure or responses to traffic pollution. Despite this, we believe, the metabolomics results demonstrated initial proof of concept in resolving statistically robust metabolic differences in a small panel setting — an incremental yet critical step leading to the potential development of a specific biological indicator of exposure to traffic pollution.

Finally, as part of **Supplemental Aim 5**, we examined the performance of multiple low-cost pollutant sensor sampling platforms in a near-road field setting. Low-cost sensor technologies appear to be poised to radically change the approach to conducting observational field studies of exposure by providing increased spatial and temporal coverage of pollutant concentrations. We believe the study findings support the future development and use of these direct sensing technologies. The gas-phase sensors for NO_2, CO, and O_3, in particular, provided what appear to be sufficiently reliable results for future health-related studies of this type (though extensive evaluation and calibration of the sensors were necessary). The results from the PM sensors indicate that the specific models used in the study would be less useful for such studies. The PM sensors agreed less well, not only with more traditional monitoring instrumentation, but also with each other. Given the low-cost and relatively easy deployment of such sensors, future field studies should strongly consider employing low-cost sensors widely, though calibration and testing will still be critical.

Although we were successful at addressing our overarching aim and meeting the specific aims, there are several modifications to the designs we would pursue were we to conduct a similar study in the future. First, our use of low-cost sensors leads us to believe that our gas-phase measurements could largely be replaced with low-cost sensors, along with one or two high-grade reference monitors. Care would have to be taken to calibrate the sensors adequately. The case for PM monitoring is not as clear, but it should not be ruled out if the technology continues to progress. (We would still recommend using various PM sensors for comparison.) Multisensor units could be placed at much greater density than was possible given the study constraints when using traditional instrumentation. The modeling aspect of this study was also successful in that it provided a conceptual picture of the spatial patterns of pollutant concentrations not available from the observations alone. On the other hand, it was also apparent that modeled estimates of traffic-related air pollution levels can be highly biased. It was not apparent what else might be done (beyond the calibrations done here), outside of a specialized study focusing on model accuracy in similar settings. Thus, adding another approach for fine-scale spatial exposure field development, such as land use regression, that could utilize the spatial density of monitoring allowed by low-cost sensors, could be of interest. Future studies should consider using multiple measures of OP (e.g., cellular and other non-DTT assays) and greater use of metabolomics analyses.

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REFERENCES

Amorim JH, Rodrigues V, Tavares R, Valente J, Borrego C. 2013. CFD modelling of the aerodynamic effect of trees on urban air pollution dispersion. Sci Tot Environ 461:541–551.

Anderson DC, Loughner CP, Diskin G, Weinheimer A, Canty TP, Salawitch RJ, et al. 2014. Measured and modeled CO and NOy in DISCOVER-AQ: An evaluation of emissions and chemistry over the eastern US. Atmos Environ 96:78–87.

Antinolo M, Willis MD, Zhou S, Abbatt JPD. 2015. Connecting the oxidation of soot to its redox cycling abilities, Nature Comm, 6:6812. doi:10.1038/ncomms7812.

Austin E, Novosselov I, Seto E, Yost MG. 2015a. Laboratory evaluation of the Shinyei PPD42NS low-cost particulate matter sensor. Plos One 10:e0137789. Available: *https://doi.org/10.1371/journal.pone.0137789*.

Ayres JG, Borm P, Cassee FR, Castranova V, Donaldson K, Ghio A, et al. 2008. Evaluating the toxicity of airborne particulate matter and nanoparticles by measuring oxidative stress potential — A workshop report and consensus statement. Inhal Toxicol 20:75–99.

Bates JT, Weber R, Abrams J, Verma V, Fang T, Ivey C, et al. 2018. Source impacts on and cardiorespiratory effects of reactive oxygen species generated by water-soluble $PM_{2.5}$ across the eastern United States. In: Mensink C., Kallos G. (eds) Air Pollution Modeling and Its Application XXV. ITM 2016. Springer Proceedings in Complexity. Springer, Cham. *https://doi.org/10.1007/978-3-319-57645-9_79*.

Bates JT, Weber RJ, Abrams J, Verma V, Fang T, Klein M, et al. 2015. Reactive oxygen species in atmospheric particulate matter suggest a link to cardiorespiratory effects. Environ Sci Technol; doi:10.1021/acs.est.5b02967 [Online 27 Oct 2015].

Batterman S. 2013. The near-road ambient monitoring network and exposure estimates for health studies. EM (Pittsburgh, Pa) 2013(7):24–30.

Batterman S, Burke J, Isakov V, Lewis T, Mukherjee B, Robins T. 2014a. A comparison of exposure metrics for traffic-related air pollutants: Application to epidemiology studies in Detroit, Michigan. Int J Environ Res Public Health 11:9553–9577.

Batterman S, Ganguly R, Isakov V, Burke J, Arunachalam S, Snyder M, et al. 2014b. Dispersion modeling of traffic-related air pollutant exposures and health effects among children with asthma in Detroit, Michigan. Transport Res Rec 2452:105–113.

Beckerman B, Jerrett M, Brook JR, Verma DK, Arain MA, Finkelstein MM. 2008. Correlation of nitrogen dioxide with other traffic pollutants near a major expressway. Atmos Environ 42:275–290.

Bilenko N, Brunekreef B, Beelen R, Eeftens M, de Hoogh K, Hoek G, et al. 2015. Associations between particulate matter composition and childhood blood pressure - the PIAMA study. Environ Int 84:1–6.

Biswas S, Verma V, Schauer JJ, Cassee FR, Cho AK, Sioutas C. 2009. Oxidative potential of semi-volatile and non volatile particulate matter (PM) from heavy-duty vehicles retrofitted with emission control technologies. Environ Sci Tech 43:3905–3912.

Blanchard CL, Hidy GM, Tanenbaum S, Edgerton ES, Hartsell BE. 2013a. The Southeastern Aerosol Research and Characterization (SEARCH) Study: Spatial variations and chemical climatology, 1999-2010. J Air Waste Manage Assoc 63:260–275.

Blanchard CL, Hidy GM, Tanenbaum S, Edgerton ES, Hartsell BE. 2013b. The Southeastern Aerosol Research and Characterization (SEARCH) Study: Temporal trends in gas and PM concentrations and composition, 1999-2010. J Air Waste Manage Assoc 63:247–259.

Blanchard CL, Tanenbaum S, Hidy GM. 2013c. Source attribution of air pollutant concentrations and trends in the Southeastern Aerosol Research and Characterization (SEARCH) network. Environ Sci Technol 47:13536–13545.

Boogaard H, Janssen NAH, Fischer PH, Kos GPA, Weijers EP, Cassee FR, et al. 2012. Contrasts in oxidative potential and other particulate matter characteristics collected near major streets and background locations. Environ Health Perspect 120:185–191.

Brantley HL, Hagler GSW, Deshmukh PJ, Baldauf RW. 2014. Field assessment of the effects of roadside vegetation on near-road black carbon and particulate matter. Sci Tot Environ 468:120–129.

Charrier JG, Anastasio C. 2012. On dithiothreitol (DTT) as a measure of oxidative potential for ambient particles: Evidence for the importance of soluble transition metals. Atmos Chemis Phys 12:9321–9333.

Cheung KL, Ntziachristos L, Tzamkiozis T, Schauer JJ, Samaras Z, Moore KF, et al. 2010. Emissions of particulate trace elements, metals and organic species from gasoline, diesel, and biodiesel passenger vehicles and their relation to oxidative potential. Aerosol Sci Tech 44:500–513.

Cho AK, Sioutas C, Miguel AH, Kumagai Y, Schmitz DA, Singh M, et al. 2005. Redox activity of airborne particulate matter at different sites in the Los Angeles basin. Environ Res 99:40–47.

Dadvand P, Ostro B, Figueras F, Foraster M, Basagana X, Valentin A, et al. 2014. Residential proximity to major roads and term low birth weight the roles of air pollution, heat, noise, and road-adjacent trees. Epidemiology 25:518–525.

Darrow LA, Klein M, Correa A, Flanders WD, Waller L, Marcus M, et al. 2008. Ambient air pollution and preterm birth in Atlanta, 1994-2004: A time-series analysis. Birth Defects Research Part A. Clin Molec Teratology 82:367.

Darrow LA, Klein M, Strickland MJ, Mulholland JA, Tolbert PE. 2011. Ambient air pollution and birth weight in full-term infants in Atlanta, 1994-2004. Environ Health Perspect 119:731–737.

de Hoogh K, Korek M, Vienneau D, Keuken M, Kukkonen J, Nieuwenhuijsen MJ, et al. 2014. Comparing land use regression and dispersion modelling to assess residential exposure to ambient air pollution for epidemiological studies. Environ Int 73:382–392.

de Kok TM, Hogervorst JG, Briede JJ, van Herwijnen MH, Maas LM, Moonen EJ, et al. 2005. Genotoxicity and physicochemical characteristics of traffic-related ambient particulate matter. Environ Molecular Mutagenesis 46:71–80.

Delfino RJ, Staimer N, Gillen D, Tjoa T, Sioutas C, Fung K, et al. 2006. Personal and ambient air pollution is associated with increased exhaled nitric oxide in children with asthma. Environ Health Perspect 114:1736–1743.

Delfino RJ, Staimer N, Tjoa T, Gillen D, Kleinman MT, Sioutas C, et al. 2008. Personal and ambient air pollution exposures and lung function decrements in children with asthma. Environ Health Perspect 116:550–558.

Dudka I, Kossowska B, Senhadri H, Latajka R, Hajek J, Andrzejak R, et al. 2014. Metabonomic analysis of serum of workers occupationally exposed to arsenic, cadmium and lead for biomarker research: A preliminary study. Environ Int 68:71–81.

Ebelt ST, Petkau AJ, Vedal S, Fisher TV, Brauer M. 2000. Exposure of chronic obstructive pulmonary disease patients to particulate matter: Relationships between personal and ambient air concentrations. J Air Waste Manage Assoc 50:1081–1094.

Edgerton ES, Hartsell BE, Saylor RD, Jansen JJ, Hansen DA, Hidy GM. 2005. The Southeastern Aerosol Research and Characterization Study: Part II. Filter-based measurements of fine and coarse particulate matter mass and composition. J Air Waste Manage Assoc 55:1527–1542.

Ellis JK, Athersuch TJ, Thomas LD, Teichert F, Perez-Trujillo M, Svendsen C, et al. 2012. Metabolic profiling detects early effects of environmental and lifestyle exposure to cadmium in a human population. BMC Med 10:61.

Fang T, Guo H, Verma V. Peltier RE, Weber RJ. 2015. $PM_{2.5}$ water-soluble elements in the southeastern United States: Automated analytical method development, spatiotemporal distributions, source apportionment, and implications for heath studies. Atmos Chem Phys 15(20):11667–11682.

Fang T, Verma V, Bates JT, Abrams J, Klein M, Strickland MJ, et al. 2016. Oxidative potential of ambient water-soluble $PM_{2.5}$ in the southeastern United States: Contrasts in sources and health associations between ascorbic acid (AA) and dithiothreitol (DTT) assays. Atmos Chem Phys 16:3865–3879.

Fruin S, Westerdahl D, Sax T, Sioutas C, Fine P. 2008. Measurements and predictors of on-road ultrafine particle concentrations and associated pollutants in Los Angeles. Atmos Environ 42:207–219.

Fujita EM, Campbell DE, Zielinska B, Arnott WP, Chow JC. 2011. Concentrations of air toxics in motor vehicle-dominated environments. Research Report 156. Boston, MA:Health Effects Institute.

Ganna A, Fall T, Salihovic S, Lee W, Broeckling CD, Kumar J, et al. 2016. Large-scale non-targeted metabolomic profiling in three human population-based studies. Metabolomics 12:13.

Gao D, Fang T, Verma V, Zeng L, Weber RJ. 2017. A method for measuring total aerosol oxidative potential (OP) with the dithiothreitol assay and comparisons between an urban and roadside site of water-soluble and total OP. Atmos Meas Tech 10:2821–2835.

Gao ML, Cao JJ, Seto E. 2015. A distributed network of low-cost continuous reading sensors to measure spatiotemporal variations of PM2.5 in Xi'an, China. Environ Pollut 199:56–65.

Georgia EPD (Georgia Department of Natural Resources Environmental Protection Division Air Protection Branch). 1999. Georgia Gasoline Marketing Rule.

Georgia EPD (Georgia Department of Natural Resources Environmental Protection Division Air Protection Branch). 2013. Rules for Air Quality Control.

Ghio AJ, Carraway MS, Madden MC. 2012. Composition of air pollution particles and oxidative stress in cells, tissues, and living systems. J Toxicol Environ Health B Crit Rev 15:1–21.

Gietl JK, Lawrence R, Thorpe AJ, Harrison RM. 2010. Identification of brake wear particles and derivation of a quantitative tracer for brake dust at a major road. Atmos Environ 44:141–146.

Gilbert NL, Goldberg MS, Beckerman B, Brook JR, Jerrett M. 2005. Assessing spatial variability of ambient nitrogen dioxide in Montreal, Canada, with a land-use regression model. J Air Waste Manage Assoc 55:1059–1063.

Go Y-M, Walker DI, Liang Y, Uppal K, Soltow QA, Tran V, et al. 2015. Reference standardization for mass spectrometry and high-resolution metabolomics applications to exposome research. Toxicol Sci 148:531–543.

Greenbaum D, Shaikh R. 2010. First steps toward multipollutant science for air quality decisions. Epidemiology 21:195–197.

Hansen DA, Edgerton E, Hartsell B, Jansen J, Burge H, Koutrakis P, et al. 2006. Air quality measurements for the aerosol research and inhalation epidemiology study. J Air Waste Manage Assoc 56:1445–1458.

Hansen DA, Edgerton ES, Hartsell BE, Jansen JJ, Kandasamy N, Hidy GM, et al. 2012. The Southeastern Aerosol Research and Characterization Study: Part 1 — overview. J Air Waste Manag Assoc 53:14601471. Available: *https://doi.org/10.1080/10473289.2003.10466318* [accessed 23 February 2018].

Heal MR, Hibbs LR, Agius RM, Beverland LJ. 2005. Total and water-soluble trace metal content of urban background PM_{10}, $PM_{2.5}$ and black smoke in Edinburgh, UK. Atmos Environ 39:1417–1430.

HEI Panel on the Health Effects of Traffic-Related Air Pollution. 2010. Traffic-Related Air Pollution: A Critical Review of the Literature on Emissions, Exposure, and Health Effects. Special Report 17. Boston, MA:Health Effects Institute.

Henneman LRF, Holmes HA, Mulholland JA, Russell AG. 2015. Meteorological detrending of primary and secondary pollutant concentrations: Method application and evaluation using long-term (2000-2012) data in Atlanta. Atmos Environ 119:201–210.

Hoek G, Beelen R, de Hoogh K, Vienneau D, Gulliver J, Fischer P, et al. 2008. A review of land-use regression models to assess spatial variation of outdoor air pollution. Atmos Environ 42:7561–7578.

Hoek G, Brunekreef B, Goldbohm S, Fischer P, van den Brandt PA. 2002. Association between mortality and indicators of traffic-related air pollution in the Netherlands: A cohort study. Lancet 360:1203–1209.

Holstius DM, Pillarisetti A, Smith KR, Seto E. 2014. Field calibrations of a low-cost aerosol sensor at a regulatory monitoring site in California. Atmos Measure Tech 7:1121–1131.

Houyoux MR, Vukovich JM. 1999. Updates to the Sparse Matrix Operator Kernel Emissions (SMOKE) Modeling System and Integration with Models-3, paper presented at The Emission Inventory: Regional Strategies for the Future, Raleigh, NC, 26–28 October.

Janssen NAH, Brunekreef B, van Vliet P, Aarts F, Meliefste K, Harssema H, et al. 2003. The relationship between air pollution from heavy traffic and allergic sensitization, bronchial hyperresponsiveness, and respiratory symptoms in Dutch schoolchildren. Environ Health Perspect 111: 1512–1518.

Janssen NA, de Hartog JJ, Hoek G, Brunekreef B, Lanki T, Timonen KL, et al. 2000. Personal exposure to fine particulate matter in elderly subjects: Relation between personal, indoor, and outdoor concentrations. J Air Waste Manage Assoc 50:1133–1143.

Janssen NA, Hoek G, Simic-Lawson M, Fischer P, van Bree L, ten Brink H, et al. 2011. Black carbon as an additional indicator of the adverse health effects of airborne particles compared with PM_{10} and $PM_{2.5}$. Environ Health Perspect 119:1691–1699.

Janssen NAH, Yang AL, Strak M, Steenhof M, Hellack B, Gerlofs-Nijland ME, et al. 2014. Oxidative potential of particulate matter collected at sites with different source characteristics. Sci Total Environ 472:572–581.

Jeanjean APR, Hinchliffe G, McMullan WA, Monks PS, Leigh RJ. 2015. A CFD study on the effectiveness of trees to disperse road traffic emissions at a city scale. Atmos Environ 120:1–14.

Jin XM, Yang LJ, Du XZ, Yang YP. 2016. Particle transport characteristics in the micro-environment near the roadway. Build Environ 102:138–158.

Johansson C, Norman M, Burman L. 2009. Road traffic emission factors for heavy metals. Atmos Environ 43:4681–4688.

Johns DO, Stanek LW, Walker K, Benromdhane S, Hubbell B, Ross M, et al. 2012. Practical advancement of multipollutant scientific and risk assessment approaches for ambient air pollution. Environ Health Perspect 120:1238–1242.

Johnson KK, Bergin MH, Russell AG, Hagler GSW. 2016. Using low cost sensors to measure ambient particulate matter concentrations and on-road emissions factors Atmos Meas Tech Discuss; *https://doi.org/10.5194/amt-2015-331*.

Kelly F, Anderson HR, Armstrong B, Atkinson R, Barratt B, Beevers S, et al. 2011. Part 2. Analysis of the oxidative potential of particulate matter. In: The Impact of the Congestion Charging Scheme on Air Quality in London. Research Report 155. Boston, MA:Health Effects Institute.

Kozawa KH, Fruin SA, Winer AM. 2009. Near-road air pollution impacts of goods movement in communities adjacent to the ports of Los Angeles and Long Beach. Atmos Environ 43:2960–2970.

Lewne M, Cyrys J, Meliefste K, Hoek G, Brauer M, Fischer P, et al. 2004. Spatial variation in nitrogen dioxide in three European areas. Sci Total Environ 332:217–230.

Li N, Sioutas C, Cho A, Schmitz D, Misra C, Sempf J, et al. 2003. Ultrafine particulate pollutants induce oxidative stress and mitochondrial damage. Environ Health Perspect 111:455–460.

Li QF, Wyatt A, Kamens RM. 2009. Oxidant generation and toxicity enhancement of aged-diesel exhaust. Atmos Environ 43:1037–1042.

Li S, Park Y, Duraisingham S, Strobel FH, Khan N, Soltow QA, et al. 2013. Predicting network activity from high throughput metabolomics. PLoS Comput Biol 9:e1003123.

Linnet K. 1993. Evaluation of regression procedures for methods comparison studies. Clin Chem 39:424–432.

Liu W, Wang YH, Russell A, Edgerton ES. 2005. Atmospheric aerosol over two urban-rural pairs in the southeastern United States: Chemical composition and possible sources. Atmos Environ 39:4453–4470.

MacNaughton P, Melly S, Vallarino J, Adamkiewicz G, Spengler JD. 2014. Impact of bicycle route type on exposure to traffic-related air pollution. Sci Total Environ 490:37–43.

Mauderly JL, Burnett RT, Castillejos M, Ozkaynak H, Samet JM, Stieb DM, et al. 2010. Is the air pollution health research community prepared to support a multipollutant air quality management framework? Inhal Toxicol 22:1–19.

McCreanor J, Cullinan P, Nieuwenhuijsen MJ, Stewart-Evans J, Malliarou E, Jarup L, et al. 2007. Respiratory

effects of exposure to diesel traffic in persons with asthma. N Eng J Med 357:2348–2358.

McWhinney RD, Badali K, Liggio J, Li SM, Abbatt JPD. 2013a. Filterable redox cycling activity: A comparison between diesel exhaust particles and secondary organic aerosol constituents. Environ Sci Tech 47:3362–3369.

McWhinney RD, Gao SS, Zhou SM, Abbatt JPD. 2011. Evaluation of the effects of ozone oxidation on redox-cycling activity of two-stroke engine exhaust particles. Environ Sci Tech 45:2131–2136.

McWhinney RD, Zhou S, Abbatt JPD. 2013b. Naphthalene SOA: Redox activity and naphthoquinone gas-particle partitioning. Atmos Chem Phys 13:9731–9744.

Meskhidze N, Chameides WL, Nenes A, Chen G. 2003. Iron mobilization in mineral dust: Can anthropogenic SO_2 emissions affect ocean productivity? Geophys Res Lett 30.

Metzger KB, Tolbert PE, Klein M, Peel JL, Flanders WD. 2003a. Case-crossover analyses of cardiovascular emergency department visits and ambient air quality, Atlanta, Georgia, 1993-2000. Epidemiology 14:S66.

Metzger KB, Tolbert PE, Klein M, Peel JL, Flanders WD, Todd KH, et al. 2003b. Ambient air pollution and emergency department visits for specific cardiovascular conditions, Atlanta, Georgia, 1993-2000. Amer J Epidemiol 157:S27.

Morakinyo TE, Lam YF. 2016. Simulation study of dispersion and removal of particulate matter from traffic by roadside vegetation barrier. Environ Sci Poll Res 23:6709–6722.

Nenes A, Krom MD, Mihalopoulos N, Van Cappellen P, Shi Z, Bougiatioti A, et al. 2011. Atmospheric acidification of mineral aerosols: A source of bioavailable phosphorus for the oceans. Atmos Chem Phys 11:6265–6272.

Nieuwenhuijsen MJ, Donaire-Gonzalez D, Rivas I, de Castro M, Cirach M, Hoek G, et al. 2015. Variability in and agreement between modeled and personal continuously measured black carbon levels using novel smartphone and sensor technologies. Environ Sci Tech 49:2977–2982.

Oakes MM, Baxter LK, Duvall RM, Madden M, Xie MJ, Hannigan MP, et al. 2014. Comparing multipollutant emissions-based mobile source indicators to other single pollutant and multipollutant indicators in different urban areas. Int J Environ Res Pub Health 11:11727–11752.

Oftedal B, Nafstad P, Magnus P, Bjorkly S, Skrondal A. 2003. Traffic related air pollution and acute hospital admission for respiratory diseases in Drammen, Norway, 1995-2000. Eur J Epidemiol 18:671–675.

Ogawa & Company. 1998. NO, NO_2, NO_x and SO_2 Sampling Protocol Using the Ogawa Sampler. Ogawa Company USA.

Ostro B, Feng WY, Broadwin R, Green S, Lipsett M. 2007. The effects of components of fine particulate air pollution on mortality in California: Results from CALFINE. Environ Health Perspect 115:13–19.

Pachon JE, Balachandran S, Hu YT, Mulholland JA, Darrow LA, Sarnat JA, et al. 2012. Development of outcome-based, multipollutant mobile source indicators. J Air Waste Manage Assoc 62:431–442.

Pallister T, Jennings A, Mohney RP, Yarand D, Mangino M, Cassidy A, et al. 2016. Characterizing blood metabolomics profiles associated with self-reported food intakes in female twins. Plos One 11:16.

Park YH, Lee K, Soltow QA, Strobel FH, Brigham KL, Parker RE, et al. 2012. High-performance metabolic profiling of plasma from seven mammalian species for simultaneous environmental chemical surveillance and bioeffect monitoring. Toxicology 295:47–55.

Parrish DD, Holloway JS, Fehsenfeld FC. 1994. Routine, continuous measurement of carbon-monoxide with parts-per-billion precision. Environ Sci Tech 28:1615–1618.

Peel JL, Tolbert PE, Klein M, Metzger KB, Flanders WD, Todd K, et al. 2003. Examining the relationship between ambient air pollution and asthma emergency department visits in Atlanta, 1993-2000. Epidemiology 14:S70.

Piedrahita R, Xiang Y, Masson N, Ortega J, Collier A, Jiang Y, et al. 2014. The next generation of low-cost personal air quality sensors for quantitative exposure monitoring. Atmos Measure Tech 7:3325–3336.

Rattanavaraha W, Rosen E, Zhang HF, Li QF, Pantong K, Kamens RM. 2011. The reactive oxidant potential of different types of aged atmospheric particles: An outdoor chamber study. Atmos Environ 45:3848–3855.

Reis S, Seto E, Northcross A, Quinn NWT, Convertino M, Jones RL, et al. 2015. Integrating modelling and smart sensors for environmental and human health. Environ Modell Softw 74:238–246.

Rijnders E, Janssen NAH, van Vliet PHN, Brunekreef B. 2001. Personal and outdoor nitrogen dioxide concentrations in relation to degree of urbanization and traffic density. Environ Health Perspect 109:411–417.

Romano A, Aiani MR, Brambilla M, Brambilla P, Carcano M, Galbiati E, et al. 2012. [Metabolomic profiles of exhaled

breath condensate of 39 nickel exposed workers]. G Ital Med Lav Ergon 34:682–686.

Sarnat JA, Brown KW, Schwartz J, Coull BA, Koutrakis P. 2005. Ambient gas concentrations and personal particulate matter exposures: Implications for studying the health effects of particles. Epidemiology 16:385–395.

Sarnat SE, Klein M, Sarnat JA, Flanders WD, Waller LA, Mulholland JA, et al. 2010. An examination of exposure measurement error from air pollutant spatial variability in time-series studies. J Expo Sci Environ Epidemiol 20:135–146.

Sarnat JA, Marmur A, Klein M, Kim E, Russell AG, Sarnat SE, et al. 2008a. Fine particle sources and cardiorespiratory morbidity: An application of chemical mass balance and factor analytical source-apportionment methods. Environ Health Perspect 116:459–466.

Sarnat SE, Raysoni AU, Li WW, Holguin F, Johnson BA, Luevano SF, et al. 2012. Air pollution and acute respiratory response in a panel of asthmatic children along the U.S.-Mexico border. Environ Health Perspect 120:437–444.

Sarnat SE, Sarnat JA, Klein M, Goldman G, Mulholland J, Russell AG, et al. 2008b. Applying alternative approaches to characterizing air pollution exposure in an epidemiologic study in Atlanta. Epidemiology 19:S38.

Schoonen MAA, Cohn CA, Roemer E, Laffers R, Simon SR, O'Riordan T. 2006. Mineral-induced formation of reactive oxygen species. Med Mineral Geochem 64:179–221.

Seinfeld JH, Pandis S. 2006. Atmospheric Chemistry and Physics. 2nd ed. Hoboken, NJ:John Wiley.

Shinyei K. 2002. Product specifications PPD20V. In: SP-30-E-99007, 1-6.

Shinyei Technology Co. L. 2010. Specification sheet of PPD42NS.

Shinyei Technology Co. L. 2013. Product specifications PPD60PV-T2. In: SP-30-E-08003(V01).

Snyder MG, Venkatram A, Heist DK, Perry SG, Petersen WB, Isakov V. 2013. RLINE: A line source dispersion model for near-surface releases. Atmos Environ 77:748–756.

Solomon PA, Chameides W, Weber R, Middlebrook A, Kiang CS, Russell AG, et al. 2003. Overview of the 1999 Atlanta supersites project. J Geophys Res 108; doi:10.1029/2001JD001458 [Online 15 April 2003].

Solomon PA, Costantini M, Grahame TJ, Gerlofs-Nijland ME, Cassee FR, Russell AG, et al. 2012. Air pollution and health: Bridging the gap from sources to health outcomes: Conference summary. Air Qual Atmos Health 5:9–62.

Squadrito GL, Cueto R, Dellinger B, Pryor WA. 2001. Quinoid redox cycling as a mechanism for sustained free radical generation by inhaled airborne particulate matter. Free Radic Biol Med 31:1132–1138.

Steenhof M, Gosens I, Strak M, Godri KJ, Hoek G, Cassee FR, et al. 2011. In vitro toxicity of particulate matter (PM) collected at different sites in the Netherlands is associated with PM composition, size fraction and oxidative potential — the RAPTES project. Part Fibre Toxicol 8:26.

Steenhof M, Mudway IS, Gosens I, Hoek G, Godri KJ, Kelly FJ, et al. 2013. Acute nasal pro-inflammatory response to air pollution depends on characteristics other than particle mass concentration or oxidative potential: The RAPTES project. Occup Environ Med 70:341–348.

Steinle S, Reis S, Sabel CE, Semple S, Twigg MM, Braban CF, et al. 2015. Personal exposure monitoring of $PM_{2.5}$ in indoor and outdoor microenvironments. Sci Total Environ 508:383–394.

Strickland MJ, Darrow LA, Klein M, Flanders WD, Sarnat JA, Waller LA, et al. 2010. Short-term associations between ambient air pollutants and pediatric asthma emergency department visits. Amer J Respir Criti Care Med 182:307–316.

Surawski NC, Miljevic B, Roberts BA, Modini RL, Situ R, Brown RJ, et al. 2010. Particle emissions, volatility, and toxicity from an ethanol fumigated compression ignition engine. Environ Sci Tech 44:229–235.

Tao F, Gonzalez-Flecha B, Kobzik L. 2003. Reactive oxygen species in pulmonary inflammation by ambient particulates. Free Rad Biol Med 35:327–340.

Tolbert PE, Klein M, Metzger KB, Peel J, Flanders WD, Todd K, et al. 2000. Interim results of the study of particulates and health in Atlanta (SOPHIA). J Expo Anal Environ Epidemiol 10:446–460.

Tong ZM, Baldauf RW, Isakov V, Deshmukh P, Zhang KM. 2016. Roadside vegetation barrier designs to mitigate near-road air pollution impacts. Sci Total Environ 541:920–927.

Tong ZM, Whitlow TH, MacRae PF, Landers AJ, Harada Y. 2015. Quantifying the effect of vegetation on near-road air quality using brief campaigns. Environ Pollut 201:141–149.

Uppal K, Soltow QA, Strobel FH, Pittard WS, Gernert KM, Yu T, et al. 2013. xMSanalyzer: Automated pipeline for improved feature detection and downstream analysis of large-scale, non-targeted metabolomics data. BMC Bioinformatics 14:15.

U.S. EPA (U.S. Environmental Protection Agency). 2015. Aerminute User's Guide. Available: *https://www3.epa.gov/ttn/scram/7thconf/aermod/aerminute_userguide.pdf*.

U.S. EPA (U.S. Environmental Protection Agency). 2016a. Air Pollutant Emissions Trends Data. Available *https://www.epa.gov/air-emissions-inventories/air-pollutant-emissions-trends-data* [accessed 15 August 2016].

U.S. EPA (U.S. Environmental Protection Agency). 2005. Clean Air Interstate Rule.

U.S. EPA (U.S. Environmental Protection Agency). 1999. Impact Analysis Control of Air Pollution from New Motor Vehicles: Tier 2 Motor Vehicle Emissions Standards and Gasoline Sulfur Control Requirements.

U.S. EPA (U.S. Environmental Protection Agency). 2016b. Integrated Science Assessment for Oxides of Nitrogen — Health Criteria (Final, 2016). EPA/600/R-15/068. Available: *http://ofmpub.epa.gov/eims/eimscomm.getfile?p_download_id=526855*.

U.S. EPA (U.S. Environmental Protection Agency). 2011. Mobile Source Air Toxins Rule.

U.S. EPA (U.S. Environmental Protection Agency). 2000. Regulatory Impact Analysis: Heavy-duty Engine and Vehicle Standards and Highway Diesel Fuel Sulfur Control Requirements. Chapter 3.

Van den Bossche J, Peters J, Verwaeren J, Botteldooren D, Theunis J, De Baets B. 2015. Mobile monitoring for mapping spatial variation in urban air quality: Development and validation of a methodology based on an extensive dataset. Atmos Environ 105:148–161.

Vedal S, Kaufman JD. 2011. What does multi-pollutant air pollution research mean? Amer J Respir Crit Care Med 183:4–6.

Vejerano EP, Ma YJ, Holder AL, Pruden A, Elankumaran S, Marr LC. 2015. Toxicity of particulate matter from incineration of nanowaste. Environ Sci: Nano 2:143–154.

Verma V, Fang T, Guo H, King L, Bates JT, Peltier RE, et al. 2014. Reactive oxygen species associated with water-soluble $PM_{2.5}$ in the southeastern United States: Spatiotemporal trends and source apportionment. Atmos Chem Phys 14:12915–12930.

Verma V, Fang T, Xu L, Peltier RE, Russell AG, Ng NL, et al. 2015a. Organic aerosols associated with the generation of reactive oxygen species (ROS) by water-soluble $PM_{2.5}$. Environ Sci Tech 49:4646–4656.

Verma V, Pakbin P, Cheung KL, Cho AK, Schauer JJ, Shafer MM, et al. 2011. Physicochemical and oxidative characteristics of semi-volatile components of quasi-ultrafine particles in an urban atmosphere. Atmos Environ 45:1025–1033.

Verma V, Rico-Martinez R, Kotra N, King L, Liu J, Snell TW, et al. 2012. Contribution of water-soluble and insoluble components and their hydrophobic/hydrophilic subfractions to the reactive oxygen species-generating potential of fine ambient aerosols. Environl Sci Tech 46:11384–11392.

Verma V, Wang Y, El-Afifi R, Fang T, Rowland J, Russell AG, et al. 2015b. Fractionating ambient humic-like substances (HULIS) for their reactive oxygen species activity — assessing the importance of quinones and atmospheric aging. Atmos Environ 120:351–359.

Vijayaraghavan K, DenBleyker A, Ma L, Lindhjem C, Yarwood G. 2014. Trends in on-road vehicle emissions and ambient air quality in Atlanta, Georgia, USA, from the late 1990s through 2009. J Air Waste Manage Assoc 64(7):808–816.

Walker DI, Go Y-M, Liu K, Pennell KD, Jones DP. 2016. Population screening for biological and environmental properties of the human metabolic phenotype: Implications for personalized medicine. In: Metabolic Phenotyping in Personalized and Public Healthcare. Boston, MA:Academic Press, 167–211.

Wang Y, Li JY, Jing H, Zhang Q, Jiang JK, Biswas P. 2015. Laboratory evaluation and calibration of three low-cost particle sensors for particulate matter measurement. Aerosol Sci Tech 49:1063–1077.

Wang Z, Zheng Y, Zhao B, Zhang Y, Liu Z, Xu J, et al. 2015. Human metabolic responses to chronic environmental polycyclic aromatic hydrocarbon exposure by a metabolomic approach. J Proteome Res 14:2583–2593.

Weber RJ, Sullivan AP, Peltier RE, Russell A, Yan B, Zheng M, et al. 2007. A study of secondary organic aerosol formation in the anthropogenic-influenced southeastern United States. J Geophys Res-Atmos 112.

Wei Y, Wang Z, Chang CY, Fan T, Su L, Chen F, et al. 2013. Global metabolomic profiling reveals an association of metal fume exposure and plasma unsaturated fatty acids. PLoS One 8:e77413.

Weichenthal S, Crouse DL, Pinault L, Godri-Pollitt K, Lavigne E, Evans G, et al. 2016. Oxidative burden of fine particulate air pollution and risk of cause-specific mortality in the Canadian Census Health And Environment Cohort (CANCHEC). Environ Res 146:92–99.

Wichmann J, Janssen NAH, van der Zee S, Brunekreef B. 2005. Traffic-related differences in indoor and personal absorption coefficient measurements in Amsterdam, the Netherlands. Atmos Environ 39:7384–7392.

Wiegand K, Smith J. 2013. Traffic monitoring program. Georgia Department of Transportation Office of Transportation Data. Available: *http://www.dot.ga.gov/DriveSmart/Data/Documents/2013 Georgia Traffic Monitoring Program 4.pdf.*

Yan B, Zheng M, Hu YT, Ding X, Sullivan AP, Weber RJ, Baek J, Edgerton ES, Russell AG. 2009. Roadside, urban, and rural comparison of primary and secondary organic molecular markers in ambient $PM_{2.5}$. Environ Sci Technol 43(12): 4287–4293.

Yang AL, Hellack B, Leseman D, Brunekreef B, Kuhlbusch TAJ, Cassee FR, et al. 2015a. Temporal and spatial variation of the metal-related oxidative potential of $PM_{2.5}$ and its relation to $PM_{2.5}$ mass and elemental composition. Atmos Environ 102:62–69.

Yang A, Janssen NAH, Brunekreef B, Cassee FR, Hoek G, Gehring U. 2016. Children's respiratory health and oxidative potential of PM2.5: The PIAMA birth cohort study. Occup Environ Med 73:154–160.

Yang A, Wang M, Eeftens M, Beelen R, Dons E, Leseman D, et al. 2015b. Spatial variation and land use regression modeling of the oxidative potential of fine particles. Environ Health Perspect 123:1187–1192.

Zhai X, Russell A, Sampath P, Mulholland J, Kim B-U, Kim Y, et al. 2016. Calibrating R-LINE model results with observational data to develop annual mobile source air pollutant fields at fine spatial resolution: Application in Atlanta. Atmos Environ 147:446–457.

Zhang X, Hecobian A, Zheng M, Frank NH, Weber RJ. 2010. Biomass burning impact on $PM_{2.5}$ over the southeastern US during 2007: Integrating chemically speciated FRM filter measurements, MODIS fire counts and PMF analysis. Atmos Chem Phys 10:6839–6853.

Zhou Y, Levy JI. 2008. The impact of urban street canyons on population exposure to traffic-related primary pollutants. Atmos Environ 42:3087–3098.

Zhu Y, Fung DC, Kennedy N, Hinds WC, Eiguren-Fernandez A. 2008. Measurements of ultrafine particles and other vehicular pollutants inside a mobile exposure system on Los Angeles freeways. J Air Waste Manage Assoc 58:424–434.

Zhu YF, Hinds WC, Kim S, Shen S, Sioutas C. 2002a. Study of ultrafine particles near a major highway with heavy-duty diesel traffic. Atmos Environ 36:4323–4335.

Zhu YF, Hinds WC, Kim S, Sioutas C. 2002b. Concentration and size distribution of ultrafine particles near a major highway. J Air Waste Manage Assoc 52:1032–1042.

MATERIALS AVAILABLE ON THE HEI WEBSITE

Appendices A through F contain supplemental material not included in the printed report. They are available on the HEI website at *www.healtheffects.org/publications.*

Appendix A. Sampling Instrumentation and QA/QC Protocol

Appendix B. Measurement Details

Appendix C. Low-Cost Sensors

Appendix D. RLINE Calibration

Appendix E. Personal Exposures: Matching by Sampling Week

Appendix F. Metabolomics Analysis

ABOUT THE AUTHORS

Jeremy A. Sarnat, Ph.D., is an associate professor of environmental health at the Rollins School of Public Health at Emory University in Atlanta, Georgia. He served as the principal investigator for the DRIVE study. His research focuses primarily on measuring exposures and acute health responses to urban air pollution in various populations. He earned his B.A. in anthropology from the University of Michigan in Ann Arbor, Michigan, and his MPH and Sc.D. from the Harvard School of Public Health in Boston, Massachusetts. He serves on the Science Advisory Boards for Nitrogen Oxides and Particulate Matter in the U.S. EPA's Clean Air Scientific Advisory Committee.

Armistead (Ted) Russell, Ph.D., a co-investigator on the DRIVE study, is a professor of civil and environmental engineering at GIT in Atlanta, Georgia, where his research is aimed at better understanding the dynamics of air pollutants at urban and regional scales and at assessing their impacts on health and the environment. He earned his B.S. in mechanical engineering at Washington State University

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Donghai Liang, M.P.H., is a doctoral candidate in the Department of Environmental Health at Emory University. He earned his B.S. in biology at Peking University in Beijing, China, and his M.P.H. in environmental health sciences at Yale University in New Haven, Connecticut. His current research interests include exposure science, environmental epidemiology, and metabolomics in the research area of air pollution. For the DRIVE study, he served as a lead researcher, contributing to field sampling, data management, and data analysis.

Jennifer L. Moutinho, M.S., is a graduate student in the School of Civil and Environmental Engineering at GIT in Atlanta, Georgia. She earned her B.S. in chemical engineering at Worcester Polytechnic Institute in Worcester, Massachusetts, and her M.S. in environmental engineering at GIT. For the DRIVE study, she was responsible for managing and maintaining the instrumentation at the monitoring sites on the GIT campus and processing instrumentation data.

Rachel Golan, Ph.D., earned her B.Sc. in nutrition at the Hebrew University of Jerusalem in Jerusalem, Israel, and her M.P.H. and Ph.D. in epidemiology at Ben-Gurion University of the Negev in Beer-Sheeva, Israel. She recently began a tenure track position at Ben-Gurion University of the Negev after a 2-year postdoctoral fellowship in the Department of Environmental Health at Emory University. As a postdoctoral researcher, she led the field campaign of the DRIVE study. Her research today focuses on the effect of air pollution on human health and combines both clinical research and basic science.

Rodney J. Weber, Ph.D., is a professor in the School of Earth and Atmospheric Sciences at GIT in Atlanta, Georgia. He earned his Ph.D. in mechanical engineering at the University of Minnesota in Minneapolis and Saint Paul, Minnesota. His research interests focus on atmospheric aerosols, outdoor air quality, and environmental impacts, including the sources and fates of air pollutants, aerosol dynamics, particle radiative properties, development of particle measurement systems, regional air quality, and health effects. As a co-investigator on the DRIVE study, he contributed to the field-study aspects of the project, focusing on measurements of OP.

Dong Gao, B.E., is currently a Ph.D. student in the School of Civil and Environmental Engineering at GIT in Atlanta, Georgia. She earned her B.E. in environmental engineering at Sun Yat-sen University in Guangzhou City, Guangdong, China. In the DRIVE project, she was responsible for the measurements of FPMOP using the DTT assay. Her research interests include atmospheric chemistry, sources, and processing.

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CRITIQUE

Review Committee

HEI

Research Report 196, *Developing Multipollutant Exposure Indicators of Traffic Pollution: The Dorm Room Inhalation to Vehicle Emissions (DRIVE) Study*, J.A. Sarnat et al.

INTRODUCTION

Traffic emissions are an important source of urban air pollution, and exposure to traffic-related air pollution has been associated with various adverse health effects. In 2010, HEI published Special Report 17, *Traffic-Related Air Pollution: A Critical Review of the Literature on Emissions, Exposure, and Health Effects.* That report, developed by the HEI Panel on the Health Effects of Traffic-Related Air Pollution, summarized and synthesized research related to the health effects from exposure to traffic emissions. The Panel concluded that exposure to traffic-related air pollution was causally linked to worsening asthma symptoms. It also found suggestive evidence of a causal relationship with onset of childhood asthma, nonasthma respiratory symptoms, impaired lung function, total and cardiovascular mortality, and cardiovascular morbidity (HEI Panel on the Health Effects of Traffic-Related Air Pollution 2010). In a review by the World Health Organization (WHO*), these adverse effects on health were observed after adjusting for socioeconomic status and noise (WHO 2013).

Because traffic-related air pollution is of public health interest, it is important to understand where and at what level people are exposed to traffic emissions. However, exposure assessment is challenging because traffic-related air pollution is a complex mixture of many particulate and gaseous pollutants and is characterized by high spatial and temporal variability (HEI Panel on the Health Effects of Traffic-Related Air Pollution 2010). The highest levels of traffic-related air pollution occur within a few hundred meters of major roads, with the impact zone depending on the pollutant, geographical and land-use characteristics, and meteorological conditions (Karner et al. 2010; Zhou and Levy 2006).

Identifying an appropriate metric of exposure to traffic-related air pollution has been difficult, because many of the pollutants are also emitted from other combustion sources. Development and evaluation of metrics of exposure to traffic-related air pollution rely both on measurement campaigns with adequate design and instrumentation and on tools to infer traffic-related air pollution exposure from these measurements. Approaches to assess exposure to traffic-related air pollution have included measurements made at various distances from busy roads using fixed sites or mobile platforms as well as models such as land-use regression and dispersion models. In some cases, infiltration and time–activity patterns have been included for more accurate estimates of personal exposure to air pollution from traffic and other outdoor sources. Each of these exposure estimation approaches has limitations that have been discussed before (HEI Panel on the Health Effects of Traffic-Related Air Pollution 2010). In 2013, following the recommendation of the HEI Traffic Review Panel to improve exposure assessment of traffic-related air pollution for use in health studies, HEI issued Request for Applications (RFA) 13-1, *Improving Assessment of Near-Road Exposure to Traffic Related Pollution*. Since then, HEI has funded five studies under RFA 13-1 (see Preface).

In response to RFA 13-1, Dr. Jeremy Sarnat of Emory University, Atlanta, Georgia, and colleagues proposed a 2-year study, "Developing Multipollutant Exposure Indicators of Traffic Pollution: The Dorm Room Inhalation to Vehicle Emissions (DRIVE) Study." Sarnat and colleagues proposed to evaluate two multipollutant air pollutant metrics that had not previously been evaluated as metrics of exposure to traffic-related air pollution by conducting measurements in and around two student dormitories in Atlanta that were located at different distances from a major highway. The first multipollutant metric would combine levels of multiple traffic-related air pollutants with their relative emissions to achieve a single metric of exposure. The second metric would be an acellular assay of the oxidative potential of fine particulate matter. They also proposed a small panel study to explore the use of metabolomics to identify possible biological markers (metabolites) that varied with exposure to traffic-related air pollution in students who lived in the two dormitories. The HEI

Dr. Jeremy A. Sarnat's 2-year study, "Developing Multipollutant Exposure Indicators of Traffic Pollution: The Dorm Room Inhalation to Vehicle Emissions (DRIVE) Study," began in March 2014. Total expenditures were $694,800. The draft Investigators' Report from Sarnat and colleagues was received for review in September 2016. A revised report, received in April 2017, was accepted for publication in June 2017. During the review process, the HEI Review Committee and the investigators had the opportunity to exchange comments and to clarify issues in both the Investigators' Report and the Review Committee's Critique.

* A list of abbreviations and other terms appears at the end of this volume.

Research Committee recommended Dr. Sarnat's application for funding because they thought the application was comprehensive and included a novel combination of approaches, such as the multipollutant metrics of exposure to traffic-related air pollution, the unique design of two dormitories located different distances from a very busy highway, and analysis of metabolites as potential biomarkers of exposure.

This Critique provides the HEI Review Committee's evaluation of the study. It is intended to aid the sponsors of HEI and the public by highlighting both the strengths and limitations of the study and by placing the Investigators' Report in scientific and regulatory context.

SUMMARY OF THE STUDY

SPECIFIC AIMS

The study by Sarnat and colleagues had four original specific aims and one supplemental aim. The original aims of the study were:

- To examine associations between traditional and two novel metrics of exposure to traffic-related air pollution — an integrated mobile source indicator (IMSI) and fine particulate matter oxidative potential (FPMOP) — at a roadside monitoring site;
- To assess the spatiotemporal variability of outdoor and indoor levels of traffic-related air pollutant components, IMSI, and FPMOP at monitoring sites 5 m to 2.3 km from the roadside monitoring site compared with concentrations at the roadside monitoring site;
- To evaluate how well traffic pollutant components and the multipollutant metrics of exposure measured at the roadside and other fixed monitoring sites reflect corresponding personal exposures of students living in dormitories at different distances from a highway; and
- To explore the feasibility of using high-throughput metabolomics in a panel of dormitory residents for identifying environmental exposures, including exposure to traffic-related air pollution.

In addition, at the request of HEI, a supplemental aim was added to the study to examine the performance of several low-cost sensors at a few locations already selected for the main study.

STUDY DESIGN AND APPROACH

The DRIVE study took place on the campus of the Georgia Institute of Technology (GIT) in Atlanta, Georgia, near the downtown Connector where I-75 and I-85 merge, one of the busiest highway arteries in the United States (~300,000 vehicles per day). This is a unique section of road because it is highly congested but has very little diesel traffic (Georgia DOT Office of Planning 2015). The diesel traffic volume is particularly low because of a ban on through trucks on I-75 and I-85 in Atlanta. Between September 2014 and January 2015, the investigators conducted four tiers of measurements, including air pollutant measurements on the GIT campus between 5 m and 2.3 km from the highway at (1) six outdoor sites and (2) inside two student dormitories, as well as (3) measurements of personal exposure to air pollutants and (4) biological measurements in a panel of student participants (Critique Table 1). The two dormitories were selected because one was adjacent to the Connector and the other was 1.4 km away in a section of the GIT campus that was farther from roads with high traffic volumes. The investigators measured a number of pollutants at the outdoor and indoor sites, including fine particulate matter ($PM_{2.5}$), nitrogen dioxide (NO_2), and black carbon (BC). They also measured personal exposure to NO_2 and BC with monitors carried by the study participants. Measurements were conducted using the same instrumentation at all sites when possible, but often it was necessary to use different equipment across the sites (see Table 1 in the Investigators' Report). The investigators conducted detailed quality assurance and quality control tests, including time drift and alignment adjustments and instrument colocations before and after monitoring periods. In total, 54 students participated in the personal exposure and metabolomics measurements.

The study focused on evaluating two multipollutant metrics of exposure to traffic-related air pollution. The IMSI was of interest because it incorporated measurements of several different pollutants into a single metric of the mixture of traffic-related air pollutants (Pachon et al. 2012). The IMSI was defined to be the average concentrations of elemental carbon (EC), carbon monoxide (CO), and nitrogen oxides (NO_x), weighted by modeled estimates of the fraction of these concentrations contributed by emissions from gasoline and diesel motor vehicles. It was calculated from measured values at the outdoor and indoor sites and from a combination of measurements (CO and NO_2) and modeled values (EC) for personal exposures (see Equation 1 in the Investigators' Report).

FPMOP was considered a second potential multipollutant metric of exposure to traffic-related air pollution because it was associated with both mobile source emissions (McWhinney et al. 2011; Verma et al. 2014) and cytotoxicity and heart- and lung-related emergency department visits in some earlier studies (Abrams et al. 2017; Ayres et al.

Critique Table 1. Details of the DRIVE Measurement Campaign Including Measurement Type, Site, Measurement Frequency[a,b]

	Site (distance from highway)									
	Outdoor Air						Indoor Air			
Measurement Type	EPD (5 m)	RDS (10 m)	NDO (20 m)	RFT (500 m)	FDO (1.4 km)	JST (2.3 km)	NDI (20 m)	FDI (1.4 km)	Personal Exposure	Biological Measure-ments
CO	1-hr	1-hr	1-hr	1-hr	1-hr	1-hr	1-hr	1-hr		
$NO\text{-}NO_2\text{-}NO_x$	1-hr	1-hr	1-hr	1-hr	1-hr	1-hr	1-hr	1-hr		
NO_2		1-hr	48-hr		48-hr	1-hr	48-hr	48-hr	48-hr	
O_3	1-hr	1-hr				1-hr				
$PM_{2.5}$ mass	1-hr		1-hr	1-hr	1-hr	1-hr	1-hr	1-hr	1-hr	
$PM_{2.5}$ mass and BC		48-hr	48-hr	48-hr	48-hr		1-hr	1-hr	48-hr	
BC	1-hr	1-hr	1-hr	1-hr	1-hr		1-hr	1-hr		
$PM_{2.5}$, OC, and EC		48-hr	48-hr	48-hr	48-hr	1-hr	48-hr	48-hr	48-hr	
$PM_{2.5}$, sulfate		1-hr				1-hr				
Other $PM_{2.5}$ ions						1-hr				
$PM_{2.5}$ metals						E3D				
PNC[c]			1-hr		1-hr		1-hr	1-hr		
IMSI	1-hr	1-hr	1-hr	1-hr	1-hr	1-hr	1-hr	1-hr	1-hr (modeled)	
FPMOP		48-hr	48-hr	48-hr	48-hr		48-hr	48-hr		
Traffic count and composition		1-hr								
Temperature, RH, wind speed and direction		1-hr		1-hr		1-hr				
GPS location tracking									1-hr (4 days)	
Time–activity diary									Daily (4 days)	
Saliva										Weekly
Blood samples (plasma)										Monthly

CO = carbon monoxide; NO_2 = nitrogen dioxide; $NO\text{-}NO_2\text{-}NO_x$ = oxides of nitrogen; O_3 = ozone; $PM_{2.5}$ = fine particulate matter and its various components including EC = elemental carbon and OC = organic carbon, and PNC = particle number concentration; FPMOP = fine particulate matter oxidative potential; IMSI = integrated mobile source indicator; and RH = relative humidity.

[a] Measurements were made in 4 tiers from September 8, 2014, to January 5, 2015: (1) 6 outdoor sites at various distances from the highway (17 weeks); (2) 2 indoor sites at GIT dormitories at different distances from the highway (17 weeks); (3) personal exposures of study participants (4 days total per participant); and (4) biological samples from study participants (12 saliva samples and 4 blood samples per participant).

The sites were EPD = US EPA's NO_2 near-road monitoring site for Atlanta, RDS = roadside monitoring site, NDO = outdoor monitoring site at the dormitory near the highway, RFT = rooftop monitoring site, FDO = outdoor monitoring site at the dormitory far from the highway, JST = Jefferson Street monitoring site, NDI = indoor monitoring site at the dormitory near the highway, and FDI = indoor monitoring site at the dormitory far from the highway.

[b] Pollutants were measured continuously at 1-hour resolution (1-hr), as integrated 48-hour measurements twice per week [48-hr], or as integrated 24-hour measurements every third day [E3D]. For the indoor and outdoor measurements, air pollutant level records were about 80% complete overall, with completeness for most individual pollutants ranging from 61% to 100% (see IR Table A.1 for details).

[c] PNC was measured only outside of the dorms and had 21% complete data at FDO and 35% complete data at NDO.

2008; Bates et al. 2015). Some other studies have reported poor correlations between FPMOP and traffic-related air pollutant levels, suggesting that FPMOP may capture the properties of only a subset of the more common metrics of exposure to traffic-related air pollution (Hu et al. 2008; Kelly et al. 2011). However, even if uncorrelated with most metrics of exposure to traffic-related air pollution, FPMOP could still be useful if the health effects of traffic-related air pollution are related to oxidative potential rather than to overall levels of fine particles. Although a single measurement, FPMOP is considered a multipollutant metric because it is believed to be affected by many particle properties, including size, surface properties, and chemical composition. It was measured in an acellular assay on 48-hour $PM_{2.5}$ filter samples based on the ability to catalyze the transfer of electrons from dithiothreitol (DTT) to oxygen.

To address aim 1, Sarnat and colleagues measured air pollutant levels at a roadside monitoring site (called RDS) 10 m from the highway. Pollutants measured for each aim are listed in Critique Table 1. The investigators used linear mixed models to assess the relationship between hourly outdoor concentrations at the RDS monitoring site and weather and traffic conditions. The following variables were included in a combined model: time of day, temperature, relative humidity, wind speed, wind direction, day of week (i.e., weekday vs. weekend), and traffic counts.

To address aim 2, the investigators measured air pollutant levels at five other outdoor monitoring sites, up to 2.3 km from the highway edge (see Critique Table 1). They compared hourly pollutant and IMSI levels and their correlations at each of these additional sites to levels at the RDS site used in aim 1. Air pollutant concentrations and correlations of the indoor and outdoor sites relative to those of the RDS site were reported as spatial gradients for five periods of the day, including morning and evening rush hours.

To address aim 3, the investigators asked study participants to carry portable monitors measuring $PM_{2.5}$, NO_2, BC, OC, and EC for two 48-hour periods. The participants also carried GPS tracking devices and completed diaries of their locations for the periods during which they carried the monitors. After the study period, the investigators ran generalized linear mixed models of personal exposure to BC and NO_2 on potential metrics of exposure to traffic-related air pollution. The traffic-related air pollution exposure metrics they considered were (1) concentrations at the RDS roadside monitor, (2) outdoor and (3) indoor concentrations at the participants' dormitories, (4) participant proximity to the highway during personal sampling, (5) an indicator variable for the participant's dormitory residence (i.e., dormitory near to or far from the highway), and (6) IMSI levels at the RDS monitor. They evaluated the performance of the metrics using the R^2 of the predicted values from the models on the personal exposure measurements of BC and NO_2.

To address aim 4, the investigators collected monthly blood samples and weekly saliva samples from the study participants. Then they assessed metabolite features in the blood and saliva samples. To determine whether dormitory was associated with these features, the investigators developed random effect linear models to assess associations between the metabolite features and an indicator variable of the participant's dormitory location. They developed separate models for each metabolite measured in the plasma and saliva samples. Models included potential confounder variables such as age, sex, body mass index, race, college year, and the amount of time since a participant had moved into the dormitory. Associations of biological markers (metabolites) with dormitory location were considered significant using $P < 0.05$, after correction for multiple comparisons among the 20,766 plasma and 29,013 saliva metabolite features.

To address supplemental aim 5, the investigators compared hourly concentrations of PM, CO, NO_x, and ozone (O_3) measured by low-cost sensors with colocated hourly reference instrument measurements using linear regression. The PM sensors used a light-scattering detection method (Shinyei Technology Co. 2002, 2010, 2013), and the gaseous sensors for CO, nitric oxide (NO), NO_2, and O_3 used electrochemical detection. Four units with multiple sensors were rotated for one to three weeks at a time among sites inside and outside of the dormitory near the highway, outside the dormitory far from the highway, and at the RDS site. The sensor evaluation was performed between November 4 and December 17, 2014. The sensors were calibrated first with manufacturer-supplied calibration curves where available and then by comparison with in-field observations for temperature and humidity corrections.

SUMMARY OF RESULTS

- The investigators reported relatively low levels of roadside air pollution concentrations. Still, they reported higher levels of most of the metrics of exposure to traffic-related air pollution closer to the roadway than farther away, with the exception of NO_2 and FPMOP. Mixing height and wind speed were more important determinants of hourly roadside pollutant levels than were traffic variables at a single near-road site. Correlations among levels of most pollutants at the various outdoor sites were relatively moderate and varied substantially by time of day.

- Based on findings from aims 1 and 2, the investigators concluded that the current study did not provide strong evidence of the utility of the IMSI and FPMOP as metrics of exposure to traffic-related air pollution. Additionally, NO_2 was less useful as a metric of exposure to traffic-related air pollution than were NO, CO, or BC, because NO_2 was higher indoors than outdoors and had shallower gradients near the roadside than the other pollutants measured.
- In the aim 3 analyses, measured personal exposures were similar for occupants of the two dormitories, although models predicted higher exposures at the dormitory near the highway. Pollutant levels measured at the roadside monitor predicted less of the variation in personal exposure to BC and NO_2 than ambient and indoor concentrations at the students' dormitories did.
- The investigators identified 221 metabolite features that differed significantly among the students from the two dormitories. However, the differences in metabolite features between the dormitory populations were not necessarily related to differences in exposure to traffic-related air pollution because there were also differences in year of college and access to kitchens or a cafeteria.
- Finally, the results of the add-on low-cost sensor validation study showed that the low-cost sensors tested did not perform well. Performance (linear regression slope, bias, and R^2) of the sensors was generally better for the gaseous sensors after extensive calibration than it was for $PM_{2.5}$.

HEI REVIEW COMMITTEE'S EVALUATION

In its independent review of the study, the HEI Review Committee noted that the report represents an impressive amount of effort and compiled data from a project examining an important topic. The Committee commended the investigators for conducting detailed measurements for six outdoor sites, two indoor residential (dormitory) sites, 54 study participants who carried personal exposure monitors, and 51 study participants who gave biological samples. The investigators used these data to compare and contrast various metrics of exposure to traffic-related air pollution. They also attempted to evaluate the suitability of these metrics, although the ability to do so was limited by the use of different monitors at different sites and the small number of pollutants measured in the panel study. The application of metabolomics to a near-road environmental panel study was an interesting aspect of the study.

The Committee considered the multipollutant approach to be a major strength of the study. In addition to measuring many pollutants, Sarnat and colleagues also assessed two multipollutant metrics of exposure to traffic-related air pollution. The investigators included many quality assurance and quality control tests to make sure the data were of good quality. Another strength of the study was the high retention of study participants, with 51 of 54 recruited students completing all study activities. The Committee also appreciated the low-cost sensor evaluation, because the development and use of such sensors is spreading at a fast pace, and they are increasingly being considered for use in air pollution research. Below, we discuss the Committee's detailed observations about the contributions and limitations of the study.

USEFULNESS OF THE TWO MULTIPOLLUTANT METRICS

The investigators' stated overarching goal for the current study was to evaluate the suitability of single and multipollutant metrics of exposure to traffic-related air pollution for use in small cohort and panel studies. The Review Committee thought that evaluating the suitability of various metrics of exposure to traffic-related air pollution was an important goal because the results of such a study could enhance exposure assessment for some types of future health studies, although the IMSI and FPMOP techniques tested in this study may be limited in their application for such studies given that they provided ambient concentrations and not personal measures of exposure. Their approach allowed the investigators to explore the concordance among the various metrics of exposure to traffic-related air pollution at different locations, although the investigators did not take advantage of the opportunity to directly compare levels and correlations among the air pollutants, IMSI, and FPMOP at the RDS monitoring site.

The Committee agreed in general with the investigators' conclusions that neither the IMSI nor FPMOP provided much benefit for use in health studies related to multipollutant exposure to traffic-related air pollutants in the near-road environment. In the absence of a description of clear criteria in the report, the Committee identified three main criteria to determine the suitability of a metric of exposure to traffic-related air pollution for use in panel studies: the metric should generally have (1) a value that is inversely related to the distance from the roadway, (2) reasonable agreement with personal exposure measurements of BC and NO_2 or other measured traffic-related air pollutants, and (3) relevance to health or other appropriate biological outcomes. In addition, a simpler metric would be preferable to

a complicated or difficult-to-measure metric if both had similar performance.

The Committee concluded that, based on these criteria, there was little evidence that the multipollutant metrics tested in the current study were useful for exposure assessment in the near-road environment because neither of the multipollutant metrics improved much upon single-pollutant measurements such as EC, NO_x, or CO.

The investigators' rationale for exploring the usefulness of IMSI was based on an earlier report that IMSI values were highly correlated at two ambient monitoring sites (including the Jefferson Street site) (Panchon et al 2012). However, the Committee noted that the two studies had methodological differences, particularly because IMSI was compared at two urban sites in the earlier study but at six sites — most of which were highly influenced by nearby traffic — in the current study, and because the weights of mobile to total emissions were based on different geographical areas in the two studies.

Regarding the other metric, the Committee noted that Sarnat and colleagues found little variability in either total or water-soluble FPMOP across the sites. Also, atmospheric processing appeared to increase rather than decrease the levels of FPMOP, in agreement with several earlier studies on the influence of metal and volatile particle components on particle evolution and oxidative potential (Kelly et al. 2011; Verma et al. 2011). These findings did not provide strong evidence for the utility of FPMOP as a metric of exposure to traffic-related air pollutants in the near-road environment. The investigators indicated that they plan to continue to improve the FPMOP assay and to further explore its utility in future analyses; the Committee thought this would be a worthwhile endeavor.

Regardless of the utility of the particular multipollutant metrics assessed in the study, the investigators performed detailed near-road measurements that could be used in the future to test alternative combinations of pollutants to develop additional multipollutant traffic metrics. They also reported that personal exposure was more closely related to measurements near (either indoors or outdoors) where the participants spent their time than to measurements made close to the freeway. They concluded that where the monitors are located may be just as important as which pollutants are measured if the goal is to identify spatial and temporal trends in air pollution levels for residents of near-highway buildings. The Committee agreed with the investigators that different metrics from those used in the past may need to be used or developed in the future so that future metrics will be more strongly associated with health effects and reflect the changing near-road environment.

THE CHANGING NEAR-ROAD ENVIRONMENT

Lower-than-expected air pollutant concentrations near roadways and shallower gradients in measured concentrations between roadside and more distant sites, as reported in this and other recent studies, provide evidence that the near-road environment is improving, at least in some locations. Levels of near-road traffic-related air pollutants were lower in the current study than in the earlier studies of air pollution near North American highways with mainly gasoline-fueled vehicles (e.g., Gilbert et al. 2003; Zhu et al. 2002; see also Critique Table 2). For example, near-road PNC was greater than 40,000 particles/cm^3 in some of the earlier studies (Kuhn et al. 2005a, 2005b; Zhu et al. 2002) and less than 20,000 particles/cm^3 in the current study. Similarly, BC was 4 to 5 μg/m^3 in earlier studies (Massoli et al. 2012; Zhu et al. 2002) and only about 2 μg/m^3 in the current study. Levels of both PNC and BC in the current study were similar to levels of these two pollutants in recent studies in Montreal and Boston (Levy et al. 2014; Patton et al. 2014). However, a more detailed comparison among these and other studies in Critique Table 2 is limited by the variability of the locations of near-road sites from 0 to 123 m from the edges of the highways in different studies.

Some early studies of traffic-related air pollution have relied on NO_2 level as a metric for exposure, although the HEI Panel on the Health Effects of Traffic-Related Air Pollution (2010) found NO_2 not to be a good surrogate for exposure to traffic-related air pollution because of its many other outdoor and indoor sources. Low NO_2 concentrations, similar to those reported in the current study, were also observed throughout the U.S. EPA near-road monitoring network, which included the EPD site and began operation in densely populated and high-traffic areas in 2014 (near the start of the current study) (Batterman 2013; U.S. EPA 2016), again highlighting that the roadside environment is changing.

Sarnat and colleagues suggested that the lower-than-expected air pollutant concentrations in both the current study and the U.S. EPA's near-road monitoring network may be related to lower emission rates from motor vehicles and a downward trend in ambient pollutant concentrations. The Committee agreed with the investigators that lower emissions and overall air pollutant concentrations with less spatial variation near roads are likely a result of air quality regulations and related improvements in vehicle emission control technologies.

Another feature of prior roadside studies has been a strong pattern of diurnal peaks in air pollution, related to peaks in traffic volumes. In the current study, peak morning concentrations of different traffic pollutants measured at the roadside site did not occur at the same time, and the

timing of peak concentrations did not coincide with the timing of peak traffic volume or congestion. The Committee thought that diurnal patterns that vary by pollutant and proximity to the highway have important implications because they suggest that models of near-road air pollutant exposure may require more than a single temporal adjustment factor (as obtained by a fixed site) to capture spatial–temporal variation in air pollution levels.

If decreasing near-road air pollutant concentrations are a continuing trend and traffic volume is a less important predictor of near-road pollutant levels than it was in the past, then historical near-road air pollution and health studies may become less relevant to the current near-road environment, and new studies may be needed to assess the health effects of traffic-related air pollution. The investigators suggested that in the future it will be even more important to consider multiple metrics of exposure to traffic-related air pollution and to study the near-road environment in a multipollutant framework. The Committee agreed with this suggestion and thought that multipollutant research would contribute to key questions related to which air pollutants (or exposure proxies) are most strongly associated with health effects. The Committee also thought that if traffic-related air pollution concentrations continue to decline, both exposure contrast and magnitude of adverse health effects may decrease. However, traffic-related air pollution remains a complex mixture, and there may be unmeasured or rarely measured pollutants, as well as interactions between pollutants, that are relevant to health. Moreover, there are many other factors associated with traffic exposure, most notably traffic noise, socioeconomic status, and factors related to the built environment that may either confound or modify the health effects of traffic-related air pollution. Future studies should find ways to exploit exposure contrasts and settings where correlated factors can be quantified and separated.

Though the results of this well-conducted and extensive study are useful, their generalizability should be viewed with caution. As noted, measured concentrations were typical of near-road concentrations measured in other recent studies; however, the study was conducted for part of one year, and whether these results are representative of the full year or long-term trends is not known. Additionally, the study was conducted in an O_3 nonattainment area and adjacent to one of the busiest highway arteries in the United States (>300,000 vehicles per day in 2010) (Wiegand and Smith 2013). Still, this was a very a useful study of the effects of large numbers of gasoline vehicles largely in the absence of diesel vehicles (Georgia DOT Office of Planning 2015), and the overall results may be applicable to the impacts of inner-urban major roads that also have limits on diesel traffic.

PANEL STUDY

Personal Exposure Contrast Between the Dormitories

The assumption underlying the panel study — that there would be sufficient personal air pollution exposure contrasts between participants living in the two dormitories, one closer to and the other farther from the highway — was not upheld by observations. Air pollutant concentrations near the roadside were lower than expected (see section above), contributing to limited exposure contrasts. In addition, students spent only about 8 hours per 24-hour period at their dormitories and spent the rest of their time approximately evenly split between the center of campus and off-campus locations. Also, the sampling of participants from the dorms was not balanced, and personal sampling was conducted only for 4 days per participant as opposed to for the full period as it was at the fixed site monitors. In addition, some of the metrics of exposure to traffic-related air pollution (e.g., IMSI) were lowest at the center of the campus, suggesting that sources of traffic-related air pollutants other than the downtown Connector may have contributed some traffic-related air pollution exposure at the dormitory farther from the highway. The Committee agreed with the investigators that the limited exposure contrasts were an unexpected and unfortunate limitation of the panel study and that there were many possible explanations for the similarities of exposure for participants living in the two dormitories.

Metabolomics

The Committee agreed with the investigators' conclusion that the metabolomics profiles of participants living in the two dormitories were different, despite the similarities in air pollution exposure levels. The Committee appreciated that the investigators identified several differences — other than exposure to traffic-related air pollution — between the dormitory populations that could explain some of the differences observed in metabolomics profiles. These included differences in diet (there was a cafeteria in one of the dormitories and not in the other), age and college year, and cleaning agents used. Also, students self-selected into dormitories using a ranked preference system that prioritized more senior students (Georgia Tech Department of Housing 2018). Thus, the student populations in the dormitories differed in several ways that could have influenced the metabolomics profiles, independent of traffic exposures.

In principle, a basic design with two dormitories that have different exposure levels but similar occupant and building characteristics would be a reasonable approach; however, because of an unexpected lack of contrast in air

Critique Table 2. Studies Comparing Traffic-Related Air Pollutant Levels at Various Distances from North American Highways That Have a Low Percentage (<12%) of Diesel Vehicles[a]

Reference	Location	Period	Traffic Volume, (vehicles/day)	Monitoring Type[b]	Pollutants	Main Relevant Findings	Roadside Concentration
Current study	Atlanta, Georgia	Sept. 2014–Jan. 2015	300,000	Fixed sites: 5 m–2.3 km	CO, NO-NO_2-NO_x, NO_2, O_3, BC, $PM_{2.5}$ (sulfate, mass, BC, OC, EC, ions, metals), PNC	Concentrations twice as high at highway edge as at 20–500 m away, except for NO_2, which decreased by only ~20% in this distance (see Investigators' Report Figure 4)	1.7 µg/m^3 BC 624 ppb CO 15 ppb O_3 38 ppb NO 19 ppb NO_2 57 ppb NO_x 10 µg/m^3 $PM_{2.5}$ (see Investigators' Report Table B.1)
Patton et al. 2014, Padró-Martínez et al. 2012	Boston, Massachusetts	Sept. 2009–July 2012	150,000	Mobile: 0–400 m and >1,000 m	CO, NO-NO_2-NO_x, pPAHs, PNC (0.04–3,000 nm), $PM_{2.5}$, BC	Concentrations twice as high <400 m from the road than >1000 m away. Up to 44% decrease in the 0–200 m near-highway zone, depending on neighborhood, pollutant, and side of the highway	400–600 ppb CO 15–30 ppb NO 30–70 ppb NO_x 5–8 fA pPAHs (not converted to concentration) 30,000 #/cm^3 14 µg/m^3 $PM_{2.5}$ 0.4–0.8 µg/m^3 BC
Levy et al. 2014	Montreal, Quebec, Canada	Winter, summer, and autumn 2009	Unspecified	Mobile: city-wide	NO, NO_2, NO_y, NO_x, NO_z, SO_2, CO, O_3, PM_{10}, $PM_{2.5}$, $PM_{1.0}$, PNC, BC, OM, sulfate, NO_3^-, HOA, MZ57 benzene, C3-benzene, toluene, xylenes	Compared with a nearby urban back-ground site, concentrations by the main highway were higher by 210–265% NO_2, 180–200% for BC, and 300% for PNC. Elevated concentrations were detected as far as 600–700 m from the main highway	50 ppb NO_2 125 ppb NO 100,000 #/cm^3 4 µg/m^3 BC
Kimbrough et al. 2013a, Kimbrough et al. 2013b	Las Vegas, Nevada	Mid-Dec. 2008–mid-Dec. 2009	>200,000	Transect: Upwind: 100 m, Downwind: 20 m, 100 m, 300 m	NO_2, NO_x, CO, BC, $PM_{2.5}$, PM_{10}, PM_{coarse}	Highest concentrations (NO_2, NO_x, CO) closest to the road relative to farther sites	20 ppb NO_2 40 ppb NO_x 500 ppb CO
Massoli et al. 2012	New York, New York	July 13–Aug. 1, 2009	Unspecified	Mobile: 0–500 m	CO_2, NO, NO_2, PNC, BC, organics	Increasing pollutant levels during early morning hours; levels declined quickly within 150 m downwind of the highway	470 ppmv CO_2 170 ppbv NO 30 ppbv NO_2 60,000 #/cm^3 5 µg/m^3 BC
McAdam et al. 2011	Burlington, Ontario, Canada	June 17–Aug. 5, 2009	34,000	Transect: 10 m, 30 m, and 60 m	O_3, CO, SO_2, $PM_{2.5}$, NO, NO_2, NO_x	No decline in concentrations with distance for O_3, CO, SO_2, or $PM_{2.5}$; statistically significant decline in concentrations with distance for NO, NO_2, or NO_x	3.2 µg/m^3 $PM_{2.5}$ 2.9 ppb NO 5.4 ppb NO_2 8.7 ppb NO_x 0.2 ppb SO_2 26.9 ppb O_3 0.2 ppm CO

Table continues next page

Critique Table 2 (*Continued*). Studies Comparing Traffic-Related Air Pollutant Levels at Various Distances from North American Highways That Have a Low Percentage (<12%) of Diesel Vehicles[a]

Reference	Location	Period	Traffic Volume, (vehicles/day)	Monitoring Type[b]	Pollutants	Main Relevant Findings	Roadside Concentration
Hagler et al. 2009	Raleigh, North Carolina	July 27–Aug. 22, 2006	~125,000	Mobile: 0–300 m	PNC, NO, NO_2, CO, BC, $PM_{2.5}$, PM_{10}, benzene, toluene	Steep decline with distance of PNC in first 100 m, moderate to strong correlations at 20 m with air toxics (benzene, toluene), BC, NO, and CO	25,000 #/cm^3
Hu et al. 2009	Santa Monica, California	March–June 2008 (5 mornings total)	60 to 900 vehicles/5-minute period	Transect: 0–3,600 m	PNC, ultrafine particle size distribution, $PM_{2.5}$, BC, pPAHs, CO, CO_2, NO_x, NO, NO_2	Visible decline with distance out to ~1,200 m downwind of highway	~60,000 #/cm^3 ~150 ppb NO ~55 ng/m^3 pPAHs
Beckerman et al. 2008	Toronto, Ontario, Canada	Aug. 3–10, 2004	400,000	Transect: 4–986 m	NO_2, NO_x, NO, O_3, VOCs, $PM_{2.5}$, PM_{10}, BC, CO, SO_2	NO_2 levels declined to background levels by 300 m, and other pollutants had high correlations with NO_2	~20 ppb NO_2 ~30 ppb NO_x ~100,000 #/cm^3 ~100 µg/m^3 $PM_{2.5}$
Kuhn et al. 2005b	Pasadena, California	Weekdays Jan. 12–25, 2005	~80,000	Fixed sites: <2.5 m and ~50 m	PM_{10} (mass, NO_3^-, sulfate, OC, EC, Na, Mg, Al, Si, S, Cl) in coarse, accumulation, and ultrafine modes	Concentrations of elements in ultrafine (UF) and accumulation modes up to 36% higher near road than farther away	20 µg/m^3 PM_{UF+acc} 80,000 #/cm^3
Kuhn et al. 2005a	Pasadena, California	May 17–June 4, 2004	~90,000	Fixed sites: <2.5 m and ~50 m	PM_{10} (mass, NO_3^-, OC, EC, Na, Mg, Al, Si, S, Cl) in coarse and accumulation modes, and particle size distribution	Mixed results with PNC at 150 m only 30% of PNC at road edge	~46,000 #/cm^3
Reponen et al. 2003	Cincinnati, Ohio	Unspecified	135,000–150,000	Transect: 80–1600 m	PNC (0.02–1 µm, 0.3–20 µm), $PM_{2.5}$ (0.02–2.5 µm), Al, Si, S, K, Fe, EC, OC	UFP decreased to half ~50–150 m downwind; concentrations of elements indicative of road dust and diesel were elevated up to 400 m	~20,000 #/cm^3 ~20 µg/m^3 $PM_{2.5}$
Gilbert et al. 2003	Montreal, Quebec, Canada	Sept. 18–25, 2001	185,000	Transect: 0–1400 m	NO_2	Decreased almost to background (to half of peak) within 200 m of highway	~35 ppb NO_2
Zhu et al. 2002	Los Angeles, California	May 15–July 18, 2001	~ 300,000 (California Dept. of Transportation 2002) (13,900 vehicles/hr during study period)	Transect: 30–300 m	PNC and size distribution (6–220 nm), CO, BC, PM	Concentrations decreased by half within 50 m for CO and by half within 100 m for BC and PNC. The gradient in mass concentration was negligible.	~2 ppm CO ~5 µg/m^3 BC ~150,000 #/cm^3 ~50 µg/m^3 PM
Rodes and Holland 1981	Los Angeles, California	Aug. 1978	200,000	Transect: 1 upwind, 2 downwind, 123–388 m	NO, NO_2, and O_3	Sharp gradient of NO and NO_2 within 150 m; still higher than background at 400 m	400 ppb NO 90 ppb NO_2 480 ppb NO_x

[a] Abbreviations not defined in main text are C3-benzene = benzene measured by proton transfer reaction–mass spectrometry (PTR-MS); fA = electrical current measured in femtoamps; HOA = hydrocarbon-like organic aerosol; MZ57 benzene = benzene with a mass-to-charge ratio of 57; PM_{10} = particulate matter ≤ 10 µm in aerodynamic diameter; PM_{coarse} = particulate matter between 2.5 and 10 µm in aerodynamic diameter; PM_{UF+acc} = particulate matter in the ultrafine and accumulation size modes; PNC = particle number concentration measured in particles per volume (#/cm^3); pPAHs = particle-bound polycyclic aromatic hydrocarbons.

[b] Distances in meters from edge of road.

pollutant levels at the two dormitories as well as differences between the occupants and characteristics of the dormitories, it was difficult to definitively attribute metabolomic differences in participants living in the two dorms to traffic-related air pollution exposures from the analyses presented in the Investigators' Report. Though more extensive analyses are under way, an expanded study design that included more dormitories with carefully controlled building and population characteristics to allow separation of metabolomic differences related to the dormitory from those related to traffic-related air pollution would have been preferable.

SUMMARY AND CONCLUSIONS

Sarnat and colleagues conducted a comprehensive study to evaluate single- and multipollutant metrics of exposure to traffic-related air pollution. The large number of detailed measurements — including outdoor, indoor, personal exposure, and biological measurements — and the multipollutant approach were strengths of the study.

An important finding of this study — consistent with other recent studies — was that traffic-related air pollution levels, even next to one of the most highly traveled highways in America, have decreased significantly. Also, as in previous studies, the levels of pollutants decreased with increasing distance from roadways. Dr. Sarnat and his colleagues assessed the usefulness of two metrics of multipollutant exposures to traffic-related air pollution, namely, IMSI and FPMOP, but did not find strong evidence of the utility of either for the near-road environment. Though the investigators did not posit an a priori criterion to assess the suitability of these candidate metrics, the Committee applied its own criteria and came to largely the same conclusion that the metrics were not useful in this application.

An interesting approach in this study was that it included a panel study with biological sampling for metabolomics analyses, rather than stopping at assessment of exposure to air pollution; however, the usefulness of the panel study results was limited. Despite the locations of the dormitories of student participants at different distances from the roadways, the Committee thought the personal exposures measured among residents of the two dormitories were very similar; the reported metabolomics differences were likely a consequence of factors other than exposure to traffic pollution. An expanded study design that included more dormitories with carefully controlled building and population characteristics would have been preferable.

The overall lower-than-expected air pollutant concentrations and smaller contrasts in air pollutant levels between near-road and more distant sites reported in this and other recent studies provide evidence that the near-road environment is changing for the better, likely a consequence of air quality regulations and related improvements in vehicle emission control technologies. In particular, NO_2 did not seem to be a particularly good metric of traffic-related air pollution in the current study because NO_2 levels were not substantially higher near the highway than farther away, and indoor sources contributed to the NO_2 levels inside the dormitories. The changing near-road environment has important consequences for the design of new research assessing the adverse health effects of traffic-related air pollution. In addition, past near-road air pollution and health studies may become less relevant to the current and future near-road environment given the fast-paced changes in engine and fuel technologies and electrification of the fleet (HEI Special Committee on Emerging Technologies 2011).

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REFERENCES

Abrams JY, Weber RJ, Klein M, Samat SE, Chang HH, Strickland MJ, et al. 2017. Associations between ambient fine particulate oxidative potential and cardiorespiratory emergency department visits. Environ Health Perspect 125:107008; doi:10.1289/EHP1545.

Ayres JG, Borm P, Cassee FR, Castranova V, Donaldson K, Ghio A, et al. 2008. Evaluating the toxicity of airborne particulate matter and nanoparticles by measuring oxidative stress potential — A workshop report and consensus statement. Inhal Toxicol 20:75–99; doi:10.1080/08958370701665517.

Bates JT, Weber RJ, Abrams J, Verma V, Fang T, Klein M, et al. 2015. Reactive oxygen species generation linked to sources of atmospheric particulate matter and cardiorespiratory effects. Environ Sci Technol 49:13605–13612; doi:10.1021/acs.est.5b02967.

Batterman S. 2013. The near-road ambient monitoring network and exposure estimates for health studies. EM (Pittsburgh, Pa) 2013:24–30. Available: *http://pubs.awma.org/gsearch/em/2013/7/batterman.pdf* [accessed 3 March 2018].

Beckerman B, Jerrett M, Brook JR, Verma DK, Arain MA, Finkelstein MM. 2008. Correlation of nitrogen dioxide with other traffic pollutants near a major expressway. Atmos Environ 42:275–290; doi:10.1016/j.atmosenv.2007.09.042.

California Department of Transportation. 2002. 2001 Traffic Volumes on California State Highways. Sacramento, CA:State of California Department of Transportation. Available: www.dot.ca.gov/trafficops/census/docs/2001_aadt_volumes.pdf [accessed 8 December 2017.

Georgia DOT Office of Planning. 2015. Georgia Statewide Freight and Logistics Plan, 2010–2050: Truck Modal Profile. Georgia Department of Transportation. Available: *http://www.dot.ga.gov/InvestSmart/Freight/Documents/Plan/Task%203_Georgia%20Truck%20Freight%20Modal%20Profile.pdf* accessed 7 March 2018].

Georgia Environmental Protection Division. 2015. 2015 Five-Year Assessment of Ambient Air Monitoring Program. Atlanta, Georgia:Georgia Department of Natural Resources. Available: *https://www3.epa.gov/ttnamti1/files/networkplans/GAassess2015.pdf* [accessed 7 March 2018].

Georgia Tech Department of Housing. 2018. Georgia Tech Housing: A Better Plan. Available: *https://housing.gatech.edu/sites/default/files/documents/brochures/gstudentversionv18-web.pdf* [accessed 30 January 2018].

Gilbert NL, Woodhouse S, Stieb DM, Brook JR. 2003. Ambient nitrogen dioxide and distance from a major highway. Sci Total Environ 312:43–46; doi:10.1016/s0048-9697(03)00228-6.

Hagler GSW, Baldauf RW, Thoma ED, Long TR, Snow RF, Kinsey JS, et al. 2009. Ultrafine particles near a major roadway in Raleigh, North Carolina: Downwind attenuation and correlation with traffic-related pollutants. Atmos Environ 43:1229–1234; doi:10.1016/j.atmosenv.2008.11.024.

HEI Panel on the Health Effects of Traffic-Related Air Pollution. 2010. Traffic-Related Air Pollution: A Critical Review of the Literature on Emissions, Exposure, and Health Effects. Special Report 17. Boston, MA:Health Effects Institute.

HEI Special Committee on Emerging Technologies. 2011. The Future of Vehicle Fuels and Technologies: Anticipating Health Benefits and Challenges. Communication 16. Boston, MA:Health Effects Institute.

Hu S, Fruin S, Kozawa K, Mara S, Paulson SE, Winer AM. 2009. A wide area of air pollutant impact downwind of a freeway during pre-sunrise hours. Atmos Environ 43:2541–2549; doi:10.1016/j.atmosenv.2009.02.033.

Hu S, Polidori A, Arhami M, Shafer MM, Schauer JJ, Cho A, et al. 2008. Redox activity and chemical speciation of size fractioned PM in the communities of the Los Angeles-Long Beach harbor. Atmos Chem Phys 8:6439–6451; doi:10.5194/acp-8-6439-2008.

Karner AA, Eisinger DS, Niemeier DA. 2010. Near-roadway air quality: Synthesizing the findings from real-world data. Environ Sci Technol 44:5334–5344; doi:10.1021/es100008x.

Kelly F, Anderson H, Armstrong B, Atkinson R, Barratt B, Beevers S, et al. 2011. Part 2. Analysis of the oxidative potential of particulate matter. In: The Impact of the Congestion Charging Scheme on Air Quality in London. Research Report 155. Boston, MA:Health Effects Institute.

Kimbrough ES, Baldauf RW, Watkins N. 2013a. Seasonal and diurnal analysis of NO_2 concentrations from a long-duration study conducted in Las Vegas, Nevada. J Air Waste Man Assoc 63:934–942; doi:10.1080/10962247.2013.795919.

Kimbrough S, Baldauf RW, Hagler GSW, Shores RC, Mitchell W, Whitaker DA, et al. 2013b. Long-term continuous measurement of near-road air pollution in Las Vegas: Seasonal variability in traffic emissions impact on local air quality. Air Qual Atmos Health 6:295–305; doi:10.1007/s11869-012-0171-x.

Kuhn T, Biswas S, Fine PM, Geller M, Sioutas C. 2005a. Physical and chemical characteristics and volatility of PM in the proximity of a light-duty vehicle freeway. Aerosol Sci Technol 39:347–357; doi:10.1080/027868290930024.

Kuhn T, Biswas S, Sioutas C. 2005b. Diurnal and seasonal characteristics of particle volatility and chemical composition in the vicinity of a light-duty vehicle freeway. Atmos Environ 39:7154–7166; doi:10.1016/j.atmosenv.2005.08.025.

Levy I, Mihele C, Lu G, Narayan J, Hilker N, Brook JR. 2014. Elucidating multipollutant exposure across a complex metropolitan area by systematic deployment of a mobile laboratory. Atmos Chem and Phys 14:7173–7193; doi:10.5194/acp-14-7173-2014.

Massoli P, Fortner EC, Canagaratna MR, Williams LR, Zhang Q, Sun Y, et al. 2012. Pollution gradients and chemical characterization of particulate matter from vehicular traffic near major roadways: Results from the 2009 Queens College Air Quality Study in NYC. Aerosol Sci Technol 46:1201–1218; doi:10.1080/02786826.2012.701784.

McAdam K, Steer P, Perrotta K. 2011. Using continuous sampling to examine the distribution of traffic related air pollution in proximity to a major road. Atmos Environ 45:2080–2086; doi:10.1016/j.atmosenv.2011.01.050.

McWhinney RD, Gao SS, Zhou S, Abbatt JP. 2011. Evaluation of the effects of ozone oxidation on redox-cycling activity of two-stroke engine exhaust particles. Environ Sci Technol 45:2131–2136; doi:10.1021/es102874d.

Pachon JE, Balachandran S, Hu Y, Mulholland JA, Darrow LA, Sarnat JA, et al. 2012. Development of outcome-based, multipollutant mobile source indicators. Journal of the Air & Waste Management Association 62:431–442; doi:10.1080/10473289.2012.656218.

Padró-Martínez LT, Patton AP, Trull JB, Zamore W, Brugge D, Durant JL. 2012. Mobile monitoring of particle number concentration and other traffic-related air pollutants in a near-highway neighborhood over the course of a year. Atmos Environ 61:253–264; https://doi.org/10.1016/j.atmosenv.2012.06.088.

Patton AP, Perkins J, Zamore W, Levy JI, Brugge D, Durant JL. 2014. Spatial and temporal differences in traffic-related air pollution in three urban neighborhoods near an interstate highway. Atmos Environ 99:309–321; doi:10.1016/j.atmosenv.2014.09.072.

Reponen T, Grinshpun SA, Trakumas S, Martuzevicius D, Wang Z-M, LeMasters G, et al. 2003. Concentration gradient patterns of aerosol particles near interstate highways in the Greater Cincinnati airshed. J Environ Monit 5:557–562; doi:10.1039/b303557c.

Rodes CE, Holland DM. 1981. Variations of NO, NO2 and O_3 concentrations downwind of a Los Angeles freeway. Atmos Environ 15:243–250; doi:10.1016/0004-6981(81)90024-X.

Shinyei Technology Co., Ltd. 2002. PPD20V_DS Specification Sheet. Available: *www.gvzcomp.it/index.php/en/fis?format=raw&task=download&fid=459* [accessed 3 April 2018].

Shinyei Technology Co. 2010. Specification Sheet of PPD42NS. Available: *http://wiki.timelab.org/images/f/f9/PPD42NS.pdf* [accessed 3 March 2018].

Shinyei Technology Co., Ltd. 2013. PPD60PV-T2_DS Specification Sheet. Available: *https://github.com/vitiral/Airbeam/blob/master/BOM/Sensor%20Dust%20--%20PPD60PV-T2%20Spec%20Sheet.pdf* [accessed 7 March 2018].

U.S. EPA (U.S. Environmental Protection Agency). 2016. Revision to the Near-road NO_2 Minimum Monitoring Requirements (Final Rule). Fed Reg 81 FR 96381. Available: *https://www.gpo.gov/fdsys/pkg/FR-2016-12-30/html/2016-31645.htm* [accessed 11 December 2017].

Verma V, Fang T, Guo H, King L, Bates JT, Peltier RE, et al. 2014. Reactive oxygen species associated with water-soluble PM2.5 in the southeastern United States: Spatiotemporal trends and source apportionment. Atmos Chem Phys 14:12915–12930; doi:10.5194/acp-14-12915-2014.

Verma V, Pakbin P, Cheung KL, Cho AK, Schauer JJ, Shafer MM, et al. 2011. Physicochemical and oxidative characteristics of semi-volatile components of quasi-ultrafine particles in an urban atmosphere. Atmos Environ 45:1025–1033; *https://doi.org/10.1016/j.atmosenv.2010.10.044*.

Wiegand K, Smith J. 2013. Traffic Monitoring Program. Atlanta, GA:Georgia Department of Transportation, Office of Transportation Data. Available: www.dot.ga.gov/DriveSmart/Data/Documents/2013_Georgia_Traffic_Monitoring_Program_4.pdf [accessed 2 March 2018].

WHO (World Health Organization). 2013. Review of evidence on health aspects of air pollution — REVIHAAP Project: Technical Report. Available: *www.euro.who.int/__data/assets/pdf_file/0004/193108/REVIHAAP-Final-technical-report-final-version.pdf?ua=1* [accessed 14 December 2017].

Zhou Y, Levy JI. 2007. Factors influencing the spatial extent of mobile source air pollution impacts: A meta-analysis. BMC Public Health 7:89. Available: www.ncbi.nlm.nih.gov/pubmed/17519039 [accessed 3 April 2018].

Zhu Y, Hinds WC, Kim S, Sioutas C. 2002. Concentration and size distribution of ultrafine particles near a major highway. J Air Waste Manag Assoc 52:1032–1042.

ABBREVIATIONS AND OTHER TERMS

ARC	Atlanta Regional Commission
ACSM	aerosol chemical speciation monitor
BC	black carbon
CO	carbon monoxide
Cu	copper
DRIVE	Dorm Room Inhalation to Vehicle Emissions (study)
DTT	dithiothreitol
EC	elemental carbon
EPD	Environmental Protection Division (monitoring site)
FDI	far dorm indoor (monitoring site)
FDO	far dorm outdoor (monitoring site)
FDR	false discovery rate
Fe	iron
FPMOP	fine particulate matter OP
$FPMOP^{WS\text{-}DTT}$	FPMOP derived via depletion of water-soluble DTT in an acellular essay
$FPMOP^{total\text{-}DTT}$	FPMOP derived via depletion of water-soluble and insoluble DTT in an acellular assay
$FPMOP^{WS\text{-}DTT}/m^3$	volume-normalized FPMOP derived via depletion of water-soluble DTT in an acellular assay
$FPMOP^{WS\text{-}DTT}/\mu g$	mass-normalized FPMOP derived via depletion of water-soluble DTT in an acellular assay
GIS	geographic information system
GIT	Georgia Institute of Technology
GPS	global positioning system
HILIC	hydrophilic interaction liquid chromatography
IMSI	integrated mobile source indicator
JST	Jefferson Street (monitoring site)
K	potassium
LOD	limit of detection
MH	mixing height
Mn	manganese
MOVES	Motor Vehicle Emission Simulator (modeling system)
NDI	near dorm indoor (monitoring site)
NDO	near dorm outdoor (monitoring site)
NO_2	nitrogen dioxide
NO	nitric oxide
NO_x	nitrogen oxides
O_3	ozone
OC	organic carbon
OP	oxidative potential
OPC	optical particle counter
OP^{DTT}	OP derived via water-soluble and insoluble DTT depletion
PAH	polycyclic aromatic hydrocarbon
PM	particulate matter
$PM_{2.5}$	particulate matter $\leq$ 2.5 µm in aerodynamic diameter
QA/QC	quality assurance/quality control
R^2	coefficient of determination
r	correlation coefficient
RDS	roadside (monitoring site)
RFT	rooftop (monitoring site)
RH	relative humidity
RLINE	Research LINE source dispersion model
ROS	reactive oxygen species
SCAPE	Southeastern Center for Air Pollution and Epidemiology
SD	secure digital memory
SDK	South Dekalb
SEARCH	Southeastern Aerosol Research and Characterization
SMOKE	Sparse Matrix Operator Kernel Emissions
SOPHIA	Study of Particles and Health in Atlanta
SPD	traffic speed
TCNT	traffic count
TEOM	tapered element oscillating microbalance
U.S. EPA	U.S. Environmental Protection Agency
WRF	weather research and forecasting
WS	water soluble
WSOC	water-soluble organic carbon
Zn	zinc

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