

HHS Public Access

Author manuscript Environ Res. Author manuscript; available in PMC 2021 May 01.

Published in final edited form as: Environ Res. 2020 May ; 184: 109389. doi:10.1016/j.envres.2020.109389.

Evaluating a Multipollutant Metric for Use in Characterizing Traffic-Related Air Pollution Exposures within Near-Road Environments

Jennifer L. Moutinho1,¶ , **Donghai Liang**2,#,¶ , **Rachel Golan**3, **Stefanie T. Ebelt**2, **Rodney Weber**1, **Jeremy A. Sarnat**2, **Armistead G. Russell**¹

¹School of Civil and Environmental Engineering, Georgia Institute of Technology, Atlanta, USA

²Department of Environmental Health, Rollins School of Public Health, Emory University, Atlanta, USA

³Department of Public Health, Ben-Gurion University of the Negev, Beer Sheva, Israel

Abstract

Accurately characterizing human exposures to traffic-related air pollutants (TRAPs) is critical to public health protection. However, quantifying exposure to this single source is challenging, given its extremely heterogeneous chemical composition. Efforts using single-species tracers of TRAP are, thus, lacking in their ability to accurately reflect exposures to this complex mixture. There have been recent discussions centered on adopting a multipollutant perspective for sources with many emitted pollutants to maximize the benefits of control expenditures as well as to minimize population and ecosystem exposure. As part of a larger study aimed to assess a complete emissionto-exposure pathway of primary traffic pollution and understand exposure of individuals in the near-road environment, an intensive field campaign measured TRAPs and related data (e.g., meteorology, traffic counts, and regional air pollutant levels) in Atlanta along one of the busiest highway corridors in the US. Given the dynamic nature of the near-road environment, a multipollutant exposure metric, the Integrated Mobile Source Indicator (IMSI), which was generated based on emissions-based ratios, was calculated and compared to traditional singlespecies methods for assessing exposure to mobile source emissions. The current analysis examined how both traditional and non-traditional metrics vary spatially and temporally in the near-road environment, how they compare with each other, and whether they have the potential to offer more accurate means of assigning exposures to primary traffic emissions. The results indicate that compared to the traditional single pollutant specie, the multipollutant IMSI metric provided a

[#] Address correspondence to: Donghai Liang, Ph.D. Department of Environment Health, Rollins School of Public Heath, Emory University, 1518 Clifton Rd NE, Atlanta, GA 30322, USA. Tel: 404-712-9583, donghai.liang@emory.edu. ¶These authors contributed equally to this work.

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Publisher's Disclaimer: This is a PDF file of an unedited manuscript that has been accepted for publication. As a service to our customers we are providing this early version of the manuscript. The manuscript will undergo copyediting, typesetting, and review of the resulting proof before it is published in its final form. Please note that during the production process errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

more spatially stable method for assessing exposure, though variations occurred based on location with varying results among the six sites within a kilometer of the highway.

Keywords

Traffic-related air pollution; Multipollutant exposure metric; Near-road environment; Exposure assessment; Integrated mobile source indicator

INTRODUCTION

On-road vehicles lead to elevated concentrations of traffic-related air pollutants (TRAPs) including nitrogen oxides (NO_x) , volatile organic compounds $(VOCs)$, carbon monoxide (CO), and primary fine particulate matter $(PM_{2.5})$ within the near-road microenvironment. In many studies, single-species TRAPs have been linked to a range of acute and chronic adverse health effects (see (1) and references there in). While co-exposure occurs in ambient conditions, epidemiologic studies commonly utilize measurements of single-species tracers or proxies for understanding the health effects of traffic-related emissions (2–4). Because humans are exposed to a complex mixture of air pollutants simultaneously, multipollutant approaches, including the use of source apportioned measures of primary traffic pollution emissions, have been used to consider health risks from combined exposures to traffic mixtures (6–9).

A multipollutant perspective for sources with many emitted pollutants has also been adopted by recent regulatory intervention to maximize the benefits of control expenditures as well as minimize population and ecosystem exposure (10–12). However, in order to transition to a greater multipollutant air quality regulatory framework, improvements are needed to understand how well multipollutant interventions reduce exposure (13). In addition to setting multipollutant regulations, further development and assessment of statistical methods are needed for understanding the benefits of using multipollutant exposure metrics (14) and assessing health impacts of mixtures. While populations are exposed to multiple pollutants simultaneously, which encourages shifting to a multipollutant approach, several different aspects need to be considered in the assessment of multipollutant metrics (15, 16).

As vehicle emissions decrease, the elevated concentration at the roadway decreases, making it more difficult to distinguish the local emissions source from the background concentration (5). For highly heterogeneous sources like vehicle emissions, a multipollutant framework provides new opportunities to characterize exposure-to-source emissions. While a wide range of multipollutant metrics have been developed, each has benefits and limitations to assessing source impacts and the related health outcomes (17). Single tracer pollutants are easy-to-use and measure, but may share multiple sources and not necessarily indicative of a single source. Emissions-based indicators are simple to calculate using weighted average concentrations based on source contribution, but are limited based on the accuracy of emissions inventories. The Integrated Mobile Source Indicator (IMSI) is an emissions-based, multipollutant metric derived from elemental carbon, carbon monoxide, and nitrogen oxide concentrations, along with the fraction of these species emitted by vehicles, to develop integrated estimates of vehicles impacts (9). Developed using ambient concentrations in

Atlanta, the IMSI has also been applied to two other cities with different emissions profiles to show applicability in a range of city types (8).

Although IMSIs showed promise as a potential multipollutant TRAP indicators in previous work (9), there is growing evidence that the near-road environment is changing rapidly and that traditional source contributions, fate and transport properties, and exposure factors already differ from those reported previously in existing literature (18–22). Substantial gaps now exist in our understanding of how both traditional single-pollutant and non-traditional multipollutant metrics vary spatially and temporally in the near-road environment, how they compare with each other, and whether they offer accurate means of assigning exposures to primary traffic emissions (23, 24).

To address these knowledge gaps, we conducted the current analysis with the primary objective to examine what metrics best capture TRAP exposure for use in short-term studies of exposure in epidemiologic models. Specifically, this analysis aims to characterize the variability in traditional single TRAP indicators and a multipollutant exposure metric that utilized measurements collected along a highly-trafficked highway in Atlanta. The emissions-based metric used national emissions inventory source ratios for primary, easilymeasured mobile emissions to provide accurate estimates of short-term changes in exposures to traffic-related air pollution for use in acute health impact studies without the need for source apportionment (9, 17). We compared spatial and temporal variability of the multipollutant IMSI metric to individual, single-species TRAP measurements at sites with varying distances from a major highway. The dynamics of the metrics were also assessed in relationship to local meteorological and traffic conditions. Understanding how well a multipollutant metric represents exposure to vehicle emissions based on the location of TRAP measurements has important implications for future regulatory and health assessment frameworks and the use of multipollutant indicators.

METHODS

Data for this analysis were collected as part of the Dorm Room Inhalation to Vehicle Emissions (DRIVE) study. The DRIVE study focused on pollutant dynamics and exposures in the near-road environment, with specific attention on an area adjacent to a section of heavily trafficked interstate in urban Atlanta. The study included three extensive monitoring locations on the Georgia Institute of Technology (GIT) campus in Atlanta, Georgia that monitor gaseous and aerosol traffic-related air pollutant concentrations at a distance of 3m, 60m, and 1.4km from the highway. The latter two sites included indoor monitoring to assess infiltration rates and exposures. Measurements of continuous TRAPs included carbon monoxide (CO), nitrogen oxides (NO_x) , and particulate black carbon (BC) and integrated daily $PM_{2.5}$ were collected from September 8, 2014 to January 5, 2015. Additional measurements from a site part of the EPA Near-road Monitoring Network and an urban background site were included in this multipollutant analysis. A detailed description of the DRIVE study location, study design, as well as the sampling methods can be found elsewhere (24–27).

Site description

The DRIVE study domain was centered around a segment of arterial interstate where Interstate 75 and Interstate 85 (I-75/I-85) merge in the center of Atlanta, Georgia. During the study period in 2014, this highway segment supported an annual average daily traffic (AADT) count of approximately 330,000 vehicles of primarily light-duty gasoline passenger cars and trucks. Heavy-duty diesel trucks made up approximately 4% of the total daily vehicles on this portion of the highway.

The DRIVE study sampling locations included a near-road site and two sites located in the main dormitory clusters on the GIT campus (Figure 1). The campus occupies the property expanding 1.5 km west of the highway and contains limited vehicle access roads. The nearroad sampling site (NR DRIVE) was located in a parking lot with less than 85 passenger vehicle spots located about 5m from the west side of the fifteen-lane highway (eight southbound and seven northbound) to the south of 10th Street and to the north of North Avenue. The vertical height from the road to the surrounding land was 0.5m on the west side of the highway. Surface streets on the east side of the highway follow a gridded pattern with an average block length of 140m and an AADT 15 times less than that of the highway. The two sites in student dormitories were located 60m and 1.4km from the highway. The site closer to the highway (Near Dorm or ND) operated out of an occupied administrative office on the ground floor of a five-story building and had an inlet height of 0.5m. The site further from the highway (Far Dorm or FD) operated out of an empty room part of a two bedroomone bathroom suite on the ground floor of a five-story building and had an inlet height of 1.5m. Both dormitory sites included an automated valve on a 15-minute interval to alternate measurement of outdoor (NDO and FDO) and indoor (NDI and FDI) concentrations.

The Near-road Georgia Institute of Technology (NR GIT) site is a part of the EPA Near-road Monitoring Network. The NR GIT site is located on the GIT campus adjacent to I-75/I-85 about 300m north of the NR DRIVE site. Trees were removed from a vegetative barrier along the highway to provide space for the site. Directly west of the site, there is a small limited-access parking lot for about 100 passenger vehicles. Monitoring for CO and NO_x began on July 1, 2014 and for BC on November 3, 2014. The NR GIT site is operated by the Georgia Department of Natural Resources (GA EPD) with in inlet height of 3m and hourly concentration data were downloaded from the GA EPD air quality system (32).

The urban background (UB) site was located 2.3km west of the highway and was part of the Southern Aerosol Research and Characterization (SEARCH) network. Previous studies have assessed this site as representative of Atlanta urban background pollutant concentrations and composition (29–31).

Exposure Assessment

We measured pollutants to provide information related to the particulate and gaseous composition of primary traffic emissions and characterize the regional pollution from September 8, 2014 to January 5, 2015. The pollutants we measured included traditional single-species traffic-related indicators: black carbon (BC), carbon monoxide (CO), nitric oxide (NO), nitrogen dioxide (NO₂), nitrogen oxides (NO_x), and fine particulate matter or

particles with diameters less than 2.5 micrometers $(PM_{2.5})$ (See Appendix, Table S1 and Table S2 for a complete list of measured pollutants and instrumentation). BC, CO, NO, $NO₂$ and NO_x were measured continuously or semi-continuously at each sampling location. Details on the exposure assessment can be found elsewhere (24–27).

All field instrumentation, used to measure continuous pollutant concentrations, including all gas phase instruments, were evaluated, refurbished if needed, and calibrated prior to field sampling. In order to compare concurrent pollutant measurements across the multiple sampling sites and ensure accurate concentrations during the sampling period, instruments measuring the same pollutant parameters were also co-located both before and after the sampling period and consistently calibrated throughout the 13-week intensive field sampling period. Complete information regarding the sampling methods and data quality have been previously published (26).

All continuous data were averaged to hourly levels to assess temporal variability differences between pollutants. Traditional single pollutants were measured to generate and compare with multipollutant traffic indicators.

Data Analysis

The Integrated Mobile Source Indicator was originally developed and evaluated using air quality concentration and mobile emissions data from Atlanta, Georgia to construct integrated estimates of vehicle emissions impact (9), with a particular focus on its use for acute health impact analyses. By utilizing multiple single pollutant measurements, the metric provides a more stable value for characterizing exposure from both gasoline and diesel vehicles. The indicator values are derived from elemental carbon (EC), carbon monoxide (CO), and nitrogen oxide (NO_x) concentrations normalized by the standard deviation (σ) of the hourly pollutant concentrations observed during the sampling period in order to combine concentrations of different magnitudes (Eq. 1). The species emissions ratios from vehicles were calculated as the fraction of the specific species emissions from mobile sources to total species emissions estimated from the 4km grid cell that includes all the sampling locations using the Sparse Matrix Operator Kernel Emissions (SMOKE) modelling system (33), which uses the Mobile (MOVES) (34).

$$
IMSI = \frac{\left(\frac{EC_{mob}}{EC_{tot}}\right)_{Emis} \frac{EC}{\sigma_{EC}} + \left(\frac{NOx_{mob}}{NOx_{tot}}\right)_{Emis} \frac{NOx}{\sigma_{NOx}} + \left(\frac{CO_{mob}}{CO_{tot}}\right)_{Emis} \frac{CO}{\sigma_{CO}}}{\left(\frac{EC_{mob}}{EC_{tot}}\right)_{Emis} + \left(\frac{NOx_{mob}}{NOx_{tot}}\right)_{Emis} + \left(\frac{CO_{mob}}{CO_{tot}}\right)_{Emis}}
$$
(Eq. 1)

where the measured concentrations are normalized by the standard deviation (σ) of the pollutant concentration. The emissions estimate ratios are calculated as the fraction of species emissions from mobile sources to the total species emissions. Detailed information on the assumptions, mathematical derivation, and previous use of the metric is published elsewhere (9).

Multivariate regression modeling

We used multivariate linear regression models to compare sources of variability among the metrics and assess the factors that drive near-road exposures and influence the observed strengths of the association. To assess the factors that affected the temporal variability in the concentration of each TRAP as well as the IMSI, this study used a multivariate linear mixed regression model:

$$
P_t = \beta Z_t + \theta_t + \varepsilon_t \tag{Eq. 2}
$$

Where P_t denotes the concentration of BC, CO, NO, NO₂, NO_x, or IMSI measured during hour t and β is the coefficient of interest that describes the influence of factor Z_t on the hourly pollutant level. The factors assessed included time period of the day, temperature, relative humidity, wind speed, wind direction (categorical), weekend (Saturday and Sunday), and hourly traffic counts. The time period of day was divided into five periods: morning rush hour (6 – 9am), mid-day (10am – 3pm), evening rush hour (4 – 8 pm), late evening (9pm – 12am), and early morning $(1 - 5$ am). The wind direction factor was divided into three directions: north $(315 - 45$ degrees), east $(45 - 135$ degrees, which leads to the monitoring sites being downwind of the highway), and south $(135 - 225$ degrees). θ_t represents timespecific random intercepts used to capture potential variations not explained by Z_t and ε_t represents residual random normal error. In interpreting the results from these regression models, a positive coefficient indicates an increase in the pollutant concentration levels as the unit of the corresponding factor increases, while controlling for all other factors included in the models. The regression relationship between pollutant concentrations or the multipollutant metric and driving factors were generated with R (version 3.3.1).

RESULTS

To assess the variability and levels of single pollutant concentrations compared to the multipollutant IMSI traffic exposure indicator, hourly data for CO , NO_x , and BC from the six sampling locations (the near-road (NR DRIVE) site, the Near-Road Network site (NR GIT), the near-highway dormitory outdoor (NDO) and indoor (NDI) sites, the far dormitory outdoor (FDO) and indoor (FDI) sites, and the urban background (UB) site) were compared to the hourly IMSI values at each site. Descriptive statistics and inter-site correlation analyses provided metrics to compare the temporal variability (Table 1 and Figure 2). A complete description of all the pollutants and meteorological parameters measured and analyzed, including personal exposure and biomarkers, can be found elsewhere (26).

Observed air pollutant and multipollutant metric levels

The IMSI metric exhibited similar spatial trends as CO , NO_x , and BC, decreasing in level with increasing distance from the highway. The steepest portion of the gradient occurred within 60m from the highway between the NR DRIVE and NDO sites with a mean difference of 81 ppb (19%) for CO, 11 ppb (22%) for NO_x, and 0.7 ug m⁻³ (41%) for BC. From the NR DRIVE to the FDO site (1.4km), the mean difference was 140 ppb (52%) for CO, 6 ppb (34%) for NO_x, and 0.14 ug m⁻³ (50%) for BC. The IMSI follows a similar trend

with a 14% decrease in value at the NDO site and a 40% decrease at the FDO site relative to the NR DRIVE site (Figure 2).

The IMSI diurnal profile also followed the patterns observed for the other primary pollutants, with a peak in the morning as traffic increases and a decrease mid-day with an increase in the mixing height. The diurnal profile suggests chemical processing and transport influences the levels throughout the day leading to hours from 9am to 12pm with lower metric values near the highway. The mean normalized diurnal profiles for BC, CO , NO_x , as well as the IMSI show the degree of daily variation for each pollutant at the six sites (Figure 3). The normalized CO, NO_x , and BC concentrations have similar diurnal profiles compared to the IMSI with a morning concentration peak at 9am, an evening peak at 9pm, and minimum concentrations observed at 3am and 4pm. With increasing distance from the highway, the diurnal variability of the normalized concentrations of the primary species increased from about 0.8 to 1.5 times the mean NR DRIVE concentration compared to about 0.5 to 2.2 times the mean FDO concentration for all species and the multipollutant metric. With increasing distance from the major source of vehicle emissions, the diurnal variability became more pronounced for CO , NO_x , BC , and the IMSI. This is further reflected in the concentration difference between the diurnal maximum and minimum concentrations. At the NR DRIVE site, the CO, NO_x , BC, and IMSI difference in the diurnal maximum and minimum is 178 ppb, 33 ppb, 0.93 ug m⁻³, and 0.8 compared to the 245 ppb, 44 ppb, 1.62 ug m−3, and 1.6 difference at the FDO site, respectively. While the mixing height varied enough throughout the day to decrease the concentration at the NR site, direct vehicle emissions associated with the consistently high daytime traffic count throughout the day from 7 am to 7 pm maintained a minimum concentration of about 0.8 times the mean concentration at the NR DRIVE site. Chemical processing and transport led to lower normalized minimum concentrations and higher normalized maximum concentrations at the FDO site.

Similar to the outdoor concentrations, the indoor pollutant concentrations peaked during the morning and evening. For BC and CO, the outdoor and indoor peaks occurred at the same time, but for NO_x the morning peak observed an hour lag in the observed maximum indoor morning concentration. Concentrations were also similar suggesting the high infiltration rate for both the gaseous and aerosol species.

Spatial and temporal correlations

Between-site correlations examine how well hourly temporal variability patterns between the six monitoring locations reflected corresponding temporal variability at the other sites with varying distances from each other (Table 1). With increasing distance, the IMSI Spearman's correlation between the NR DRIVE site and the other outdoor sites also decreased. The individual pollutants, however, did not all exhibit this same correlation trend. The BC concentrations at the NR GIT and UB, for example, sites were more strongly correlated with the NR DRIVE site than with the NDO site. The NO_x and CO concentrations at the NDO site, however, were more strongly correlated with the NR DRIVE site than the NR GIT site. Site specific properties likely influenced the concentration spatial gradient by pollutant. $NO₂$ concentrations measured at the NR DRIVE were more temporally correlated

Correlation between the ND and the FD sites were higher for the gaseous NO_x (0.67) and CO (0.70) pollutants than particulate BC (0.46). By using the IMSI as a metric for exposure, the Spearman's correlation (0.72) between the two locations was greater than the correlation for any of the individual species.

In addition to differences in correlations between the dorm buildings, the infiltration rate of the pollutants varied based on species and building. For the ND site, the correlation between the outdoor and indoor measurements were higher for NO_x (0.93) and CO (0.90) compared to BC (0.53). This trend was also observed at the FD site where the correlation was 0.96 for NO_x and 0.97 for CO compared to 0.51 for BC. Building filtration systems are designed to capture particles, even though the gaseous pollutants were captured at a higher efficiency with the newer system installed at the ND building. Compared to the species, the IMSI metric shows a correlation between the ambient and indoor environments of 0.80 at the ND and 0.87 at the FD.

Diurnal site correlations further illustrate how the temporal variability of the vehicle emissions and meteorological conditions affect the concentrations measured at sites with increasing distance from the highway. The diurnal correlation between the measurements at the NR DRIVE site and the other sites showed the correlation was strongest in the morning consistent with the idea that the highway emissions are a major local source impacting concentrations (Figure 4). Further, as traffic increases in the morning, vertical diffusion was limited resulting in higher, more consistent concentration levels across sites.

Decreased correlation was observed throughout the day reflecting both increased photochemical reactions and vertical mixing processes. Decrease in correlation strength between sites for CO occurred earlier in the day than for NO_x . The CO correlations began to decrease at 5am, reached a minimum correlation at 1pm, and then continued to increase again throughout the afternoon. While the CO correlation with the NDO site was between 0.49 and 0.82, the correlation with the FDO site was much wider between 0.12 and 0.74. The NO_x correlations began to decrease later in the day at 10am and remained at a minimum from 2pm to 7pm. The correlations with the NDO site were between 0.74 and 0.91, and the correlations with the FDO site were between 0.23 and 0.81. For CO and NO_x , the correlations between the NR DRIVE and NDO sites were consistently better than the correlations with the FDO site for any given hour, suggesting the correlation decreased with distance. However, the correlations with the NR DRIVE site and the UB site varied such that they were stronger than the NDO or FDO sites at certain hours. While the correlations between the NR DRIVE site and the other sites still decrease throughout the day, correlations involved the IMSI metric is more consistent across the sites. The minimum diurnal IMSI correlation between the NR DRIVE site and the NDO site was 0.50 and for the UB site, it was 0.38.

Assessment of factors driving metric variability

Since the NAAQS for both $NO₂$ and CO include primary standards based on an hourly concentration, the DRIVE study aimed at understanding exposures to individuals in the nearroad environment with hourly concentrations. The meteorological and traffic conditions were also assessed hourly so that these factors could be linked to the hourly concentrations. Statistical modeling can help identify the significant factors that affect the observations at different monitoring locations. Here, we applied linear mixed modeling to evaluate associations between pollutant concentrations and we used multiple possible contributing predictors to assess factors that drive the temporal variability observed at the different nearroad sites. The regression coefficients for the models developed for the NR DRIVE site and the NR GIT site were compared to assess whether site differences along the same road segment can lead to significant differences in the dominate factors driving primary pollutants or multipollutant metric (Table 2). The regression coefficients for two sites at varying distances from the highway (NR GIT and UB) were also compared to assess how exposure analyses may vary based on proximity to roadways in an urban area (Table 3). Significance of a factor was determined by a p-value less than 0.05.

During the DRIVE study period, the NR DRIVE and NR GIT site regression coefficients for BC, CO, and NO_x concentrations as well as the IMSI metric were negatively associated with wind speed and wind direction from the north, east, and south, indicative of dispersion away from the emissions source (Table 2). Temperature was only associated with a decrease in concentration at the NR GIT site and was associated with an increase in BC and CO concentration at the NR DRIVE site. While mixing height was associated with a significant decrease in BC and NO_x concentration at the NR DRIVE site and in CO and NO_x concentration at the NR GIT site, mixing height did not have a significant coefficient at any site for the IMSI metric. This is because the IMSI is made up of three pollutants, and in each case, mixing height was not strongly correlated with one of the pollutants, leading to a reduced association between the IMSI and that variable (e.g., mixing height). Weekend days showed an association with a significant decreasing concentration for all pollutants (NO, NO2, and BC) except CO at both sites. For the IMSI metric, weekends were significant at the NR GIT site but not at the NR DRIVE site, the latter being driven by the lack of correlation with CO, one of the IMSI components. Traffic count was associated with a significant increase in concentration and multipollutant metric values at both sites, except for CO at the NR GIT site.

Although speculative, we interpret the observed differences in the regression coefficients for the NR DRIVE and NR GIT sites as likely due to the differences in the physical site locations. The NR DRIVE site is located in an open parking lot, while the NR GIT site was located in line with a vegetation barrier along the highway. Although the two sites are within 300m and along the same highway segment, the differences in their regression coefficients has important implications for exposure analyses when relying on a single central monitor.

Discussion

The IMSI values exhibited similar trends compared to ambient CO, BC, and NO_x with an increase in monthly average during the fall DRIVE study. Measurements from the NR GIT

site and the UB site in 2015 show similar trends with the yearly minimum in the summer and increasing concentrations in the fall. At both near-road sites (NR DRIVE and NR GIT), the concentrations measured were low as compared to prior observations, highlighting the impact of emissions reduction policies that have led to a decrease in measured near-road levels. These results were consistent with near-road measurements across the United States in 2015 (38). While vehicle emissions are still a major source for measured concentrations in the near-road environment, regional sources and meteorological conditions are increasingly important for driving concentrations in the near-road environment. Further, steep spatial gradients can lead to significant errors in estimated personal exposure using single pollutant measurements from near-road monitoring sites (24). In addition to the overall spatial variability observed, the concentration gradients as well as the correlations between the sites varied diurnally depending on the single pollutant specie. In particular, correlation for the gaseous NO_x (0.67) and CO (0.70) pollutants between the ND and the FD sites were higher than particulate BC (0.46). These correlation trends are consistent with the spatial gradients observed for the species since gaseous pollutants have more homogenous concentrations and regional sources (35). CO is emitted as a primary pollutant from vehicle and generated as a secondary pollutant from VOC oxidation. Similarly, NO_x is both emitted as a primary pollutant and generated as a secondary pollutant as a result of long-range transport. These additional sources as well as lower deposition rates lead to higher correlations among the sites. BC is a primary pollutant with a lower atmosphere lifetime leading to a greater spatial gradient and a lower correlation between the two dormitory monitoring locations. Due to the complexity of how single pollutants disperse from major highway sources, near-road monitors will lead to exposure measurement errors (as compared to instrument measurement errors) when used to determine exposures for individuals at the far dorm (24). Therefore, there are limitations to using single pollutant measurements in the near-road environment as a proxy for assessing exposure to vehicle emissions. Different exposure assessment strategies should be evaluated for understanding population exposure in the near-road environment.

The IMSI is based on three normalized pollutant concentrations and the mobile source contribution for the CO, NO_x , and BC pollutant concentrations. While the spatial gradient and the diurnal profile for three pollutants were similar, thus leading to an expected multipollutant profile, the multipollutant metric was able to better reflect the spatial gradients of the mobile source impacts by adjusting the normalized species concentrations by the contribution of mobile emissions to total emissions of that species. The IMSI metric also showed greater spatial correlations, i.e., between sites, suggesting it better captures spatial fluctuations in exposure to TRAPs.

Differences between the pollutant concentration levels at the NR DRIVE and the NR GIT sites highlight how site placement can affect measured concentrations. While the two nearroad sites were located along the same highway segment, the NR DRIVE site was located in an open parking lot and the NR GIT site is in line with a vegetative barrier. A vegetative barrier impacts the rate of dispersion, leading to different pollutant dynamics even along the same roadway segment (36, 37). By including three TRAPs in a single multi-pollutant value, the IMSI is not as impacted by differences in site location properties. Therefore, depending on concentrations from a single site could limit the accuracy of using either a single

pollutant proxy or the IMSI metric for exposure of a population living in the near-road environment within a large urban area.

Based on the assessments conducted here comparing sites within 2 km of each other, the IMSI multipollutant metric can provide additional insight for characterizing exposure to primary traffic emissions. However, this evaluation is limited due to the relatively limited study duration. Additional years of similar measurements may be able to provide additional understanding about how the IMSI metric can be used to better quantify exposure. It is also noted, this evaluation is done at one location which will have site specific characteristics. As concentrations for pollutants continue to decrease in the near-road environment, it may be necessary to consider deriving a different equation for an emissions-based metric. When considering exposure to all mobile-source related emissions, future studies should not rely on measuring "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 do include substantial emissions from brake and tire wear as well as resuspended road dust.

Conclusion

Characterizing exposure to primary mobile emissions using near-road monitoring becomes increasingly difficult as vehicle emissions decrease and are no longer the dominant contributing source to measurements. The IMSI is a combination of EC, CO and NO_x observations, so it is not surprising that it was correlated with each of those species individually. The results showed the IMSIs to be less sensitive to elevated levels of any one species due to non-mobile source emissions (e.g., EC/BC or CO due to biomass burning), and is formulated to account for emissions from other sources that contribute to an urban background. The IMSI was more correlated with the IMSIs at other monitoring locations. This, along with prior results showing IMSIs to be slightly more strongly associated with acute cardiovascular outcomes in population time series studies in Atlanta, than individual traffic components (9), and that a prior study found it was more strongly associated between monitoring sites than single pollutants or PMF factors (8), suggest that IMSIs may be useful in future work examining multipollutant mobile source impacts on health. The IMSI will, in particular, take advantage of observations from the recently established USEPA near-road network monitoring. Collectively, we believe our current findings support further analysis to determine spatial and temporal scales appropriate for this approach, or whether a similar or alternative multipollutant indicator, may provide a greater ability to reflect cumulative TRAP exposure within a near-road environment.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgements

Support for this project were provided through a contract with the Health Effects Institute (RFA #4942-RFA13– 1/14–3). The field study conducted as part of this study benefitted greatly from the assistance of many students,

staff, and faculty at both Georgia Tech and Emory. Specific thanks go to C. Cornwell, K. Parada, S. Shim, Dr. K. Johnson and E. Yang for their tremendous help in conducting the field study. We want to thank Dr. R. Weber, Dr. V. Verma, and Ms. D. Gao for their measurements of oxidative potential of ambient fine particles via DTT assay. We are indebted to Dr. J. Schauer (U. Wisconsin) for loaning us several instruments to supplement our sampling network. The Georgia EPD allowed us access to their roadside monitoring site and helped provide data from those monitors, and we particularly thank Ken Buckley for his assistance with this. The study used on the instrumentation assembled for field studies conducted as part of the Southeastern Center for Air Pollution and Epidemiology (SCAPE), which was funded by a US Environmental Protection Agency STAR grant R834799. This publication was also supported by the HERCULES Center P30ES019776 and the Emory Rollins School of Public Health Dean's Pilot and Innovation Grant. The information in this document may not necessarily reflect the views of the Agency and no official endorsement should be inferred. R Golan gratefully acknowledges support by a postdoctoral fellowship from the Environment and Health Fund, Jerusalem, Israel. Dr. J Moutinho acknowledges that this material is based upon work supported by the National Science Foundation Graduate Research Fellowship Program under Grant No. (NSF DGE-1650044). Any opinions, findings, and conclusions or recommendations expressed in this material are those of the author(s) and do not necessarily reflect the views of the National Science Foundation. Dr. A Russell made use of funds provided by a generous gift from Howard T. Tellepson. We owe a debt of gratitude to the numerous administrators at Georgia Tech for allowing us to conduct this study on campus and in their residence hall facilities.

References

- 1. HEI. Traffic-related Air Pollution: A Critical Review of the Literature on Emissions, Exposure, and Health Effects; 2009.
- 2. Hoek G, Brunekreef B, Goldbohm S, Fischer P, van den Brandt PA. Association between mortality and indicators of traffic-related air pollution in the Netherlands: a cohort study. Lancet 2002; 360: 1203–1209. [PubMed: 12401246]
- 3. Levy I, Mihele C, Lu G, Narayan J, Brook JR. Evaluating Multipollutant Exposure and Urban Air Quality: Pollutant Interrelationships, Neighborhood Variability, and Nitrogen Dioxide as a Proxy Pollutant. Environmental Health Perspectives 2014; 122: 65–72. [PubMed: 24225648]
- 4. Brook JR, Burnett RT, Dann TF, Cakmak S, Goldberg MS, Fan XH, Wheeler AJ. Further interpretation of the acute effect of nitrogen dioxide observed in Canadian time-series studies. J Expo Sci Env Epid 2007; 17: S36–S44.
- 5. Ayala A, Brauer M, Mauderly JL, Samet JM. Air pollutants and sources associated with health effects. Air Quality Atmosphere and Health 2012; 5: 151–167.
- 6. Janssen NA, Hoek G, Simic-Lawson M, Fischer P, van Bree L, ten Brink H, Keuken M, Atkinson RW, Anderson HR, Brunekreef B, Cassee FR. Black carbon as an additional indicator of the adverse health effects of airborne particles compared with PM10 and PM2.5. Environ Health Perspect 2011; 119: 1691–1699. [PubMed: 21810552]
- 7. Ostro B, Feng WY, Broadwin R, Green S, Lipsett M. The effects of components of fine particulate air pollution on mortality in California: Results from CALFINE. Environmental Health Perspectives 2007; 115: 13–19. [PubMed: 17366813]
- 8. Oakes MM, Baxter LK, Duvall RM, Madden M, Xie MJ, Hannigan MP, Peel JL, Pachon JE, Balachandran S, Russell A, Long TC. Comparing Multipollutant Emissions-Based Mobile Source Indicators to Other Single Pollutant and Multipollutant Indicators in Different Urban Areas. International Journal of Environmental Research and Public Health 2014; 11: 11727–11752. [PubMed: 25405595]
- 9. Pachon JE, Balachandran S, Hu YT, Mulholland JA, Darrow LA, Sarnat JA, Tolbert PE, Russell AG. Development of outcome-based, multipollutant mobile source indicators. Journal of the Air & Waste Management Association 2012; 62: 431–442. [PubMed: 22616285]
- 10. Dominici F, Peng RD, Barr CD, Bell ML. Protecting Human Health From Air Pollution Shifting From a Single-pollutant to a Multipollutant Approach. Epidemiology 2010; 21: 187–194. [PubMed: 20160561]
- 11. Vedal SJDK. What does Multi-Pollutant Air Pollution Research Mean? AMERICAN JOURNAL OF RESPIRATORY AND CRITICAL CARE MEDICINE 2011; 183: 4–6. [PubMed: 21193783]
- 12. Council NR. Air Quality Management in the United States. Washington, DC; 2004.
- 13. Hidy GM, Pennell WT. Multipollutant Air Quality Management. Journal of the Air & Waste Management Association 2010; 60: 645–674. [PubMed: 20564991]

- 14. HEI. Methods to Investigate the Effects of Multiple Air Pollutation Constituents: Health Effects Institute; 2009.
- 15. Greenbaum D, Shaikh R. First Steps Toward Multipollutant Science for Air Quality Decisions. Epidemiology 2010; 21: 195–197. [PubMed: 20160562]
- 16. Mauderly JL, Burnett RT, Castillejos M, Ozkaynak H, Samet JM, Stieb DM, Vedal S, Wyzga RE. Is the air pollution health research community prepared to support a multipollutant air quality management framework? Inhalation Toxicology 2010; 22: 1–19.
- 17. Oakes M, Baxter L, Long TC. Evaluating the application of multipollutant exposure metrics in air pollution health studies. Environment International 2014; 69: 90–99. [PubMed: 24815342]
- 18. Blanchard CL, Hidy GM, Tanenbaum S, Edgerton ES, Hartsell BE. The Southeastern Aerosol Research and Characterization (SEARCH) study: Temporal trends in gas and PM concentrations and composition, 1999–2010. Journal of the Air & Waste Management Association 2013; 63: 247–259. [PubMed: 23556235]
- 19. Blanchard CL, Hidy GM, Tanenbaum S, Edgerton ES, Hartsell BE. The Southeastern Aerosol Research and Characterization (SEARCH) study: Spatial variations and chemical climatology, 1999–2010. Journal of the Air & Waste Management Association 2013; 63: 260–275. [PubMed: 23556236]
- 20. Blanchard CL, Tanenbaum S, Hidy GM. Source Attribution of Air Pollutant Concentrations and Trends in the Southeastern Aerosol Research and Characterization (SEARCH) Network. Environmental Science & Technology 2013; 47: 13536–13545. [PubMed: 24180677]
- 21. Henneman LRF, Holmes HA, Mulholland JA, Russell AG. Meteorological detrending of primary and secondary pollutant concentrations: Method application and evaluation using long-term (2000–2012) data in Atlanta. Atmospheric Environment 2015; 119: 201–210.
- 22. Vijayaraghavan K, DenBleyker A, Ma L, Lindhjem C, Yarwood G. Trends in on-road vehicle emissions and ambient air quality in Atlanta, Georgia, USA, from the late 1990s through 2009. Journal of the Air & Waste Management Association 2014; 64: 808–816. [PubMed: 25122954]
- 23. Baldauf R, Watkins N, Heist D, Bailey C, Rowley P, Shores R. Near-road air quality monitoring: Factors affecting network design and interpretation of data. Air Quality Atmosphere and Health 2009; 2: 1–9.
- 24. Liang D, Golan R, Moutinho JL, Chang HH, Greenwald R, Sarnat SE, Russell AG, Sarnat JA. Errors associated with the use of roadside monitoring in the estimation of acute traffic pollutantrelated health effects. Environmental research 2018; 165: 210–219. [PubMed: 29727821]
- 25. Liang D, Moutinho JL, Golan R, Yu T, Ladva CN, Niedzwiecki M, Walker DI, Sarnat SE, Chang HH, Greenwald R, Jones DP, Russell AG, Sarnat JA. Use of high-resolution metabolomics for the identification of metabolic signals associated with traffic-related air pollution. Environment International 2018; 120: 145–154. [PubMed: 30092452]
- 26. Sarnat JA, Russell AG, Liang D, Moutinho JL, Golan R, Weber R, Gao D, Sarnat SE, Chang HH, Greenwald R, Yu T. Developing Multipollutant Exposure Indicators of Traffic Pollution: The Dorm Room Inhalation to Vehicle Emissions (DRIVE) Study. Health Effects Institute; 2018.
- 27. Golan R, Ladva C, Greenwald R, Krall JR, Raysoni AU, Kewada P, Winquist A, Flanders WD, Liang D, Sarnat JA. Acute pulmonary and inflammatory response in young adults following a scripted car commute. Air Quality, Atmosphere & Health 2018; 11: 123–136.
- 28. Parrish DD, Holloway JS, Fehsenfeld FC. Routine, continuous measurement of carbon monoxide with parts per billion precision. Environmental science & technology 1994; 28: 1615–1618. [PubMed: 22176363]
- 29. Edgerton ES, Hartsell BE, Saylor RD, Jansen JJ, Hansen DA, Hidy GM. The southeastern aerosol research and characterization study: Part II. Filter-based measurements of fine and coarse particulate matter mass and composition. Journal of the Air & Waste Management Association 2005; 55: 1527–1542. [PubMed: 16295278]
- 30. Liu W, Wang YH, Russell A, Edgerton ES. Atmospheric aerosol over two urban-rural pairs in the southeastern United States: Chemical composition and possible sources. Atmospheric Environment 2005; 39: 4453–4470.
- 31. Solomon PA, Chameides W, Weber R, Middlebrook A, Kiang CS, Russell AG, Butler A, Turpin B, Mikel D, Scheffe R, Cowling E, Edgerton E, John JS, Jansen J, McMurry P, Hering S, Bahadori T.

Overview of the 1999 Atlanta Supersites Project. J Geophys Res 2003; 108: 10.1029/2001JD001458.

- 32. EPD G. Ambient Air Monitoring Program. Available from: [https://airgeorgia.org/.](https://airgeorgia.org/)
- 33. CMAS. SMOKE (Sparse Matrix Operator Kerner Emissions) Modeling System. Available from: [https://www.cmascenter.org/smoke/.](https://www.cmascenter.org/smoke/)
- 34. EPA U. Motor Vehicle Emission Simulator (MOVES), Office of Transportation and Air Quality, US Environmental Protection Agency; 2010.
- 35. HEI. Traffic-related air pollution: a critical review of the literature on emissions, exposure, and health effects. Boston, MA: Health Effect Institute; 2010.
- 36. Janhall S Review on urban vegetation and particle air pollution Deposition and dispersion. Atmospheric Environment 2015; 105: 130–137.
- 37. Baldauf R Roadside vegetation design characteristics that can improve local, near-road air quality. Transportation Research Part D-Transport and Environment 2017; 52: 354–361.
- 38. DeWinter JL, Brown SG, Seagram AF, Landsberg K, Eisinger DS. A national-scale review of air pollutant concentrations measured in the US near-road monitoring network during 2014 and 2015. Atmospheric Environment 2018; 183: 94–105.
- **•** Decreasing impact from highway vehicle emissions on surrounding pollutant levels
- **•** Integrated Mobile Source Indicator (IMSI) more spatially stable compared to single pollutant indicator
- **•** IMSIs may be useful in future work examining multipollutant mobile source impacts on health

Moutinho et al. Page 16

Figure 1. Sampling Map.

NR GIT: Near-road Monitoring Network monitor on the GIT campus; NR-DRIVE: Nearroad DRIVE site; ND: Near highway dorm outdoor and indoor sampling; FD: Far dorm outdoor and indoor sampling; UB: Urban background Jefferson St SEARCH site.

Moutinho et al. Page 17

Figure 2. Normalized boxplot presenting the distribution of hourly BC, CO, NOx, and IMSI from Sept 8, 2014 to Jan 5, 2015, ordered in increasing distance from the highway source. NR – DRIVE Near-road (3m), NDO - Near Dorm outside (60 m), NDI - Near Dorm inside (60 m), FDO - Far Dorm outside (1.4 km), FDI - Far Dorm inside (1.4 km), UB - Urban background (2.3 km)

Moutinho et al. Page 18

Figure 3. Normalized concentration from Sept 8, 2014 to Jan 5, 2015, ordered in increasing distance from the highway source.

RD – NR DRIVE (3m), NDO - Near Dorm outside (60 m), NDI - Near Dorm inside (60 m), FDO - Far Dorm outside (1.4 km), FDI - Far Dorm inside (1.4 km), UB - Urban background (2.3 km)

Moutinho et al. Page 19

Figure 4. Diurnal profile of Spearman's correlation between the NR DRIVE site and the other sites from September 8, 2014 to January 5, 2015.

NR GIT – EPA Near-road Monitoring (3m), NDO - Near Dorm outside (60 m), NDI - Near Dorm inside (60 m), FDO - Far Dorm outside (1.4 km), FDI - Far Dorm inside (1.4 km), UB - Urban background (2.3 km)

Author Manuscript

Author Manuscript

Table 1

Table 2
Regression coefficients from multivariate models examining associations between multiple factors and hourly pollutant concentrations from **Regression coefficients from multivariate models examining associations between multiple factors and hourly pollutant concentrations from** September 8, 2014 to January 5, 2015. **September 8, 2014 to January 5, 2015.**

All covariates were included simultaneously in the model for each pollutant of interest All covariates were included simultaneously in the model for each pollutant of interest

Author Manuscript

Author Manuscript

Author Manuscript

Author Manuscript

Author Manuscript

Author Manuscript

Author Manuscript

Author Manuscript

Author Manuscript

Author Manuscript

Regression coefficients from multivariate models examining associations between multiple factors and hourly pollutant concentrations from Table 3
Regression coefficients from multivariate models examining associations between multiple factors and hourly pollutant concentrations from Jan 1, 2015 to Dec 31, 2015. **Jan 1, 2015 to Dec 31, 2015.**

All covariates were included simultaneously in the model for each pollutant of interest *All covariates were included simultaneously in the model for each pollutant of interest

ï

 \overline{a}

* p-Value<0.05