Assessment of Mobile Source Air Toxics in an Environmental Justice Denver Community Adjacent to a Freeway

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Abstract:
Air pollutant concentrations are often higher near major roadways than in the surrounding environments owing to emissions from on-road mobile sources. In this study, we quantified the gradient in black carbon (BC) and air toxics concentrations from the I-70 freeway in the Elyria-Swansea environmental justice neighborhood in Denver, CO during three measurement campaigns in 2017-2018. The upwind-downwind gradient of BC concentrations from the roadway was 300-600 ng/m^3, equal to an increment of approximately 20-40% above local background levels within 180 m of the freeway. When integrated over all wind directions, the gradients were smaller, approximately 100-150 ng/m^3 (~10-15%) over the course of nearly four months of measurements. No statistically significant gradient in air toxics (e.g., benzene, formaldehyde, etc.) was found, likely because the uncertainties in the mean concentrations were larger than the magnitude of the gradient (<25%). This finding is in contrast to some earlier studies in which small gradients of benzene and other VOCs were found. We estimate that sample sizes of at least 100 individual measurements would have been required to estimate mean concentrations with sufficient certainty to quantify gradients on the order of ±10% uncertainty. These gradient estimates are smaller than those found in previous studies over the past two decades; more stringent emissions standards and the steady turnover of the vehicle fleet may be reducing the overall impact of roadway emissions on near-road communities.

Keywords: black carbon; air toxics; near-road; environmental justice

1. Introduction

In recent years, near-road air quality has been the focus of many monitoring and modeling studies (Ginzburg et al., 2015; Saha et al., 2018a) [Barzyk, 2015; Parvez 2019; Bates 2018] due to a large portion of the population residing close to major roadways and the associated negative health impacts of air pollution from major roadways (Health Effects Institute, 2010; Health Effects Institute Air Toxics Review Panel, 2007; Brugge et al., 2007; Ghosh et al., 2017; Ghosh et al., 2016; Wilhelm et al., 2011). Several studies have shown that daytime pollutant concentrations can be many times greater than background concentrations within 150 m of a major roadway, while decreasing rapidly with increasing distance from the roadway.[Parvez, 2019; Saha, 2018] (Zhu et al., 2004; Zhu et al., 2002; Baldauf et al., 2008; Hagler et al., 2009; Jeong et al., 2019; Oakes et al., 2016; Riley et al., 2014). Observational studies have shown that there is a significant gradient of black carbon (BC), ultrafine particles, and nitrogen dioxide (NO_2) concentrations next to roadways, and a smaller gradient of fine particulate matter ($PM_{2.5}$) (Karner et al., 2010; Saha et al., 2018b). Furthermore, concentrations of certain pollutants measured in the near-road environment are heavily influenced by the fleet mix of vehicles traveling on the adjacent roadway; for example, ultrafine particles and BC concentrations are positively correlated to diesel-fueled vehicles [Patterson, 2019 (Brown et al., 2014; Dallmann et al., 2012; Dallmann and Harley, 2010; Riddle et al., 2008; Kleeman et al., 2009). Several studies have

also found significant negative health impacts for individuals living within 300 m of major roadways, such as reduced lung function, low birth weights, and increased asthma and risk of heart failure (Ghosh et al., 2016; Urman et al., 2014; Brandt et al., 2014; Health Effects Institute, 2010).

Hazardous air pollutants (HAPs), also called air toxics, are ambient air pollutants that pose a wide range of threats to human health. Section 112 of the Clean Air Act (CAA), as amended in 1990, lists 189 HAPs and mandates regulations to control their emissions. Scientific studies have shown that a number of these air toxics are carcinogenic, and many are associated with a wide variety of negative health impacts, including adverse effects on reproductive, developmental, and neurological health [Clark 2017, Loh 2007, Woodruff 2000](Rosenbaum et al., 1999; Wilhelm et al., 2011; Woodruff et al., 1998; Loh et al., 2007) Mobile source air toxics are air toxics that are emitted through the combustion cycle of motor vehicles. The 2014 National Air Toxics Assessment provides an overview of which HAPs provide the greatest cariogenic exposure risk, and which emissions sources contribute to that risk—showing that mobile source emissions from light duty vehicles, heavy duty vehicles, and diesel vehicles remain a significant contributor to cancer risk (U.S. Environmental Protection Agency, 2015a, 2014b). Benzene is a key hazardous air pollutant and MSAT that is the focus of the measurements and modeling here, as it classified as a known human carcinogen for all routes of exposure under the proposed revised Carcinogen Risk Assessment Guidelines(Fruin et al., 2001; U.S. Environmental Protection Agency, 1996; Skov et al., 2001; Woodruff et al., 1998; Health Effects Institute Air Toxics Review Panel, 2007) (EPA., 2018; Smith, 2010; Halliday, 2016). Other potentially important sources of benzene and other HAPs in the area include emissions from manufacturing, petrochemical and chemical facilities, waste incinerators, non-road mobile sources, and stationary engines.

Mobile source emissions have been a regulatory target of successive rulemaking procedures following the statutory mandate of the 1990 amendments to the CAA (https://www.epa.gov/mobile-source-pollution/regulations-reduce-mobile-source-pollution). Between 2000 and 2014, the U.S. Environmental Protection Agency (EPA) promulgated three major regulations: Tier 2 Motor Vehicle Emissions Standards and Gasoline Sulfur Control Requirements; Control of Hazardous Air Pollution from Mobile Sources; and Tier 3 Motor Vehicle Emission and Fuel standards (U.S. Environmental Protection Agency, 2000, 2014a). Implementation of Tier 3 standards for motor vehicle emissions and fuel composition are ongoing, and tailpipe emissions standards "generally phase in between model years 2017 and 2025." This regulatory framework has led to significantly lower emissions from the traffic sector overall (McDonald et al., 2013; Reid et al., 2016).

A review study by Karner et al. [2010] examined the spatial gradient of pollutant concentrations over a 500-meter distance from the edge of the roadway, relative to the background and relative to the edge of the roadway, synthesizing results from numerous studies prior to 2010 (Karner et al., 2010). They found that some volatile organic compounds (VOCs), including ethane, propane, n-butane and n-hexane, showed a rapid decline in concentration (>50%) starting 150 meters from the roadway. They also present a black carbon gradient of ~50%, 150 meters from the roadway, and show that total $PM_{2.5}$ has a much weaker gradient, 15% over 150 meters. Other studies have focused on the modeled and measured gradient of vehicle emissions versus distance to road, showing gradients of benzene, BC, and ultrafine particles, generally focused on downwind conditions only [Parvez, 2019(Zhu et al., 2004; Zhu et al., 2002; Baldauf et al., 2008; Riley et al., 2014; Saha et al., 2018b).(Chang et al., 2015; Riley et al., 2014).

As the exposure environmental health effects from criteria pollutants in the United States have been substantially reduced over the past four decades, an increased focus has been placed on the equity of exposure and to the environmental and health-related outcomes related to the location of emissions sources as reflected in environmental justice concerns. Nationally, 19% of the population lives within

500 meters of high traffic volume roads, which are defined as having greater than 25,000 annual average daily traffic (AADT) based on 2010 census data (Rowangould, 2013). Studies have found inequities of exposure to air toxics in the near-road population with respect to race, community segregation, and socioeconomic status (Houston et al., 2008)(Morello-Frosch 2006) as well as race and income (Apelberg et al., 2005; Rowangould, 2013)(Miranda et al 2011; Pope et al 2016).

The Elyria-Swansea neighborhood is an environmental justice community located in central Denver near the convergence of Interstates 70 and 25. I-70 runs east-west through the neighborhood and a major rail line runs southwest-northeast through the neighborhood. The community in the vincinity of Swansea Elementary school is the domain of this study and ranks in the 86 perentile for $PM_{2.5}$ exposure, 96 percentile for diesel PM exposure, and in the 97 percentile for traffic proximity and volume, according to EPA's EJScreen EJ Index data (available interactively through https://ejscreen.epa.gov/mapper/). Higher percentiles mean that the census blocks of the Elyria-Swansea neighborhood are among the worst in the U.S. for these pollution categories (U.S. Environmental Protection Agency, 2017). Total AADT on I-70 is 153,000. A major redevelopment project has been proposed to change the roadway configuration of I-70 as it transits this neighborhood (see *https://www.codot.gov/projects/i70east)*. Currently, I-70 is an elevated viaduct approximately 30 feet higher than the surrounding communities. As part of the I-70 redevelopment project beginning in 2019, the I-70 viaduct will be replaced with a 30 foot below grade highway. A 990 foot long stretch adjacent to Swansea Elementary School will be covered by a 4-acre cap and ground-level parkland. One of the major goals of this project is to provide baseline concentration measurements of pollution gradients around the elevated roadway configuration prior to redevelopment. It is anticipated that a second monitoring study will be performed after completion of the I-70 redevelopment project to attempt to quantify the changes in pollution concentrations.

In this work, we present results from measurements of black carbon and air toxics at multiple temporary community sites perpendicular to I-70 in the Elyria-Swansea neighborhood, focused on the area at and around Swansea Elementary School. We use these measurements to determine concentrations of BC and air toxics, and to what extent concentrations are higher with increasing proximity to the freeway. Measured results are compared to modeling of benzene with AERMOD. As part of work to understand the impact of a future expansion of I-70, Denver Department of Public Health and Environment (DDPHE) conducted modeling of $PM_{2.5}$, NO_2 and benzene for the Elyria-Swansea neighborhood. This was presented in The Going One Step Beyond in North Denver study (Thomas et al), which is a combined neighborhood and project scale modeling assessment, to compare a 2011 baseline year with 2035 predicted conditions for the expanded and fully built out I-70 (Central 70) project. Benzene in Denver is mostly emitted from motor vehicle exhaust in addition to other sources such as gasoline services stations, oil and gas activities and refining, and wood burning.

2. Experiments

2.1. Spatial Domain

Air pollution monitoring of air toxics (carbonyls, VOCs, and BC) was conducted over three campaigns on a north-south axis along Elizabeth Street and Thompson Ct. perpendicular to I-70, focused on the area at and around Swansea Elementary School, which has a playground adjacent to I-70. Monitoring stations were sited on or near the Swansea Elementary School campus north of the highway and at sites on rooftops and in parks south of the highway. Figure 1 shows the study domain along with monitoring site locations for Campaigns I, II, and III. Monitoring site locations for Campaigns I and II are shown in blue and for Campign III are shown in red.

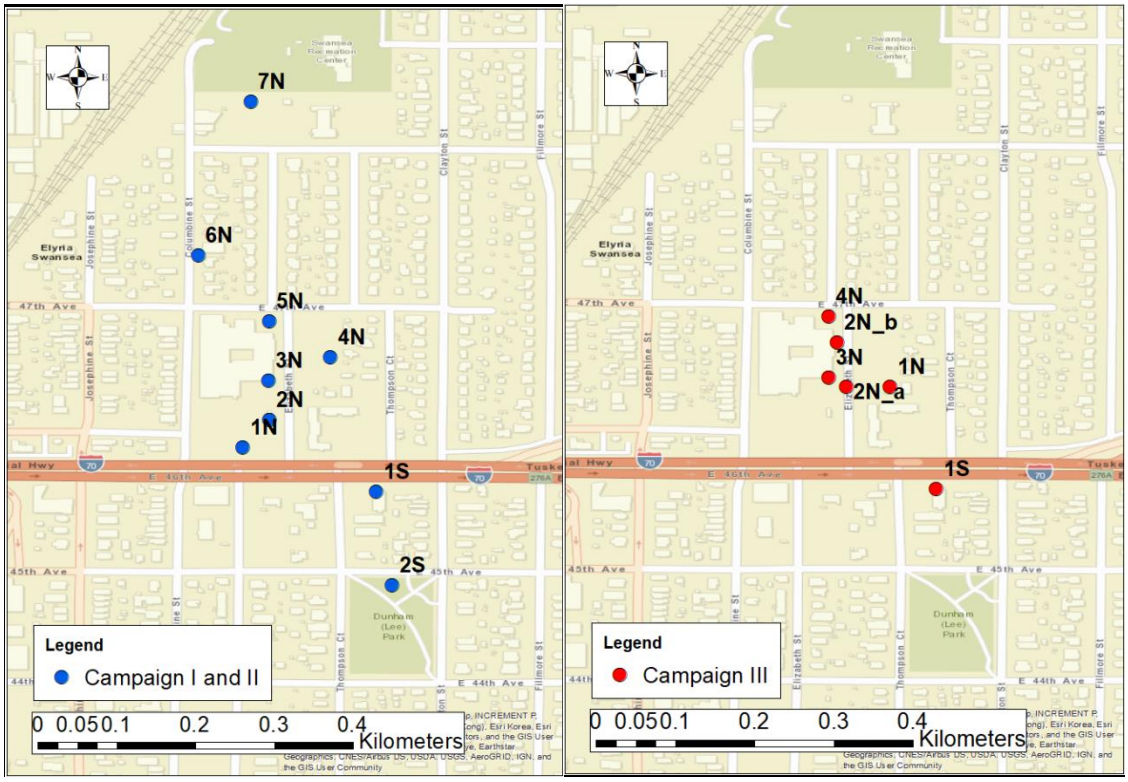


Figure 1. Monitoring site locations for Campaigns I and II (left) and Campaign III (right).

2.2. Monitoring Campaigns

The study involved three monitoring periods. Campaigns I and II had identical monitoring methods and site locations. Campaign III deployed different methods at different site locations.

- Campaign I was run from October 25, 2017, through November 25, 2017.
- Campaign II was run from February 2, 2018, through March 2, 2018.
- Campaign III was run from September 14, 2018, through October 29, 2018.

Table 1 lists the site locations, names, and measurement methods deployed in Campaigns I and II. Table 2 lists the site locations, names, and measurement methods deployed in Campaign III. Note that in Campaign III, site 2N_a only operated from September 14 through 25, 2018. That site was then relocated to the site labeled 2N_b, which operated from September 26 through October 29, 2018. BC was measured by microAeths at five near-roadway sites in Campaigns I and II. Sites 1S and 1N were closest to the roadway (within 20 meters of the nearest lane of traffic on I-70). Sites 2S, 3N, and 5N were all more than 100 meters from the nearest lane on I-70.

Table 1. Site locations and measurements deployed in Campaigns I and II

Site Name	Distance from I-70 (m)	Latitude	Longitude	MicroAeth	Carbonyls	Met
1N	17	39.78034	104.9564	x	x	
1S	16	39.77983	104.95488	x	x	
2N	146	39.78066	104.9561	x	x	
2S	127	39.77877	104.9547	x	x	

3N	102	39.78111	104.95611	x	x	
4N	177	39.78137	104.9554		x	x
5N	177	39.78178	104.9561	x	x	

Table 2. Site locations and measurements deployed in Campaign III

Site Name	Distance from I-70 (m)	Latitude	Longitude	MicroAeth	VOCs	Met
1S	16	39.779834	104.95488	x	x	
1N*	91	39.781	104.9554	x	x	x
2N_a	146	39.781	104.9559	x	x	
3N	102	39.7811	104.9561	x	x	
2N_b	148	39.7815	104.956	x	x	
4N	177	39.7818	104.9561	x	x	

*Site 1N was not at the same location as in Campaigns I and II due to inaccessibility of the location on the school playground during Campaign III.

2.3. Monitoring Methodology

Four unique measurement methods were employed during the campaigns. (1) BC measurements were made in all three campaigns using microAethalometers. (2) Meteorological measurements were made in all three campaigns, most importantly including wind speed and wind direction. (3) Carbonyl cartridge samples were collected in Campaigns I and II using EPA method TO-11. (4) Summa canisters were used to collect VOCs in Campaigns I-III and analyzed using GC-MS EPA method TO-15 (citation). VOC results from Campaign I, identified midway through campaign II (Feb 2018), were suspected to be of questionable accuracy due to a laboratory issue. Three split samples to different laboratories late in campaign II confirmed this. This was a primary driver for conducting Campaign III, since VOC concentrations are important to the surrounding communities.

Black carbon – BC concentrations were determined using the Aethlabs AE51 personal microAeth Aethalometers. The microAeth samplers are fully self-contained instruments with a built-in pump, flow control, and data-storage; see https://aethlabs.com/microaeth/ae51/overview. Data were collected at five-minute intervals in all campaigns but have been aggregated up to hourly means for hours with at least 75% completeness of valid measurements (status indicator = 0, concentration ≥ 0 ng/m^3). Cai et al found high reproducibility among six microAeth units (relative standard deviation of 8%) and good agreement with the rack-mounted Aethalometer AE33 unit (R=0.92, slope=1.01) (Cai 2014). Cheng and Lin found that differences between the AE51 and an AE31 model Aethaometer of up to 14% but overall good agreement (R^2=0.97 without AE31 data treatment) (Cheng 2013). Viana et al found similar results comparing the AE51 to elemental carbon measurements from a Thermo Multi-Angle Absorption Photometer (MAAP), with R^2 values between five AE51s and the MAAP ranging between 0.75 and 0.85 with slopes between 0.75 and 1.15 (Viana et al 2015). The AE51 has been used in multiple studies for determining personal exposure as well as BC concentrations inside residences and in ambient environments (Rivas et al 2015; Lee et al 2017; Lovinsky-Desir et al 2018).

Meteorology – A standard suite of meteorological measurements were collected at one monitoring site during each campaign. Meteorological data collected included resultant wind speed, resultant wind direction, relative humidity, barometric pressure, and temperature. We note that the siting for the meteorological station did not necessarily meet EPA requirements for siting due to a limited fetch with obstructions from nearby trees and buildings. However, these measurements are

representative of the meteorology in the near-road environment for the north side of the study domain during the campaigns.

Carbonyls – Carbonyl compounds were analyzed from samples collected on an every-other-day and weekly schedule using dinitrophenylhydrazine (DNPH)-coated sorbent cartridges following EPA's Method TO-11a (U.S. Environmental Protection Agency, 1999a). These samples were sent to an analytical laboratory for analysis using high-performance liquid chromatography (HPLC).

VOCs – VOCs were collected for 24 hours on an every-other-day schedule using SUMMA stainless steel canisters following EPA method TO-15 (U.S. Environmental Protection Agency, 1999b). Collected samples were sent to a local university laboratory for analysis using gas chromatograph mass spectrometry (GC-MS).

2.4. Modeling Methodology

The AERMOD dispersion model was used to estimate benzene concentrations from both polygon (area and nonroad) and link-based (highway) sources. Polygon emissions were calculated by allocating the 2011 NEI county benzene emissions to the polygon transportation analysis zones (TAZ's). The allocation surrogates were obtained from the Denver Regional Council of Governments (DRCOG) data including vehicle miles traveled (VMT), population and/or population density, number of oil and gas wells, and miles of railroad tracks. Most assumptions and model settings were kept the same as the 2016 North Denver study (Thomas 2016). Area source emission rates from polygon sources did not change between 2011 and 2018. The 2004 meteorological data used in this study were collected at the nearby Rocky Mountain Arsenal (surface) and the former Denver Stapleton Airport (upper air) and were again used in this model.

In the 2016 North Denver study, the Motor Vehicle Emission Simulator (MOVES) model was used to estimate county level annual emissions of benzene (as well as PM2.5 and NOx) for 2015, 2020, 2025, 2030, and 2035 (U.S. Environmental Protection Agency, 2015b, 2016). Since the year 2018 was not explicitly modeled, the 2018 county annual emissions of benzene were calculated by interpolation between 2015 and 2020. The MOVES results showed that the 2011 on-road emissions of benzene were reduced by 57% in 2018. This reduction factor was used to estimate the polygon 2018 benzene emissions. Benzene emissions from non-road, railroad and oil and gas sources were assumed to be reduced from 2011 to 2018. Reduction factor based on professional judgment were used; (10%) reduction in non-road, (10%) reduction in railroad emissions, and (25%) reduction in oil and gas emissions. There are relatively few emissions from oil and gas in Denver County as compared to counties to the north (20-90 miles away). The modeled impacts in Denver from oil and gas are negligible when using AERMOD. Given both 2035 and 2018 (interpolated) annual benzene emissions from MOVES, as well as link emissions for 2035, the link emissions for 2018 were estimated using a linear relationship. The study estimated emissions and concentrations of $PM_{2.5}$, NO_2, and benzene for two scenarios: winter (January) and summer (July) concentrations. Benzene results are presented here.

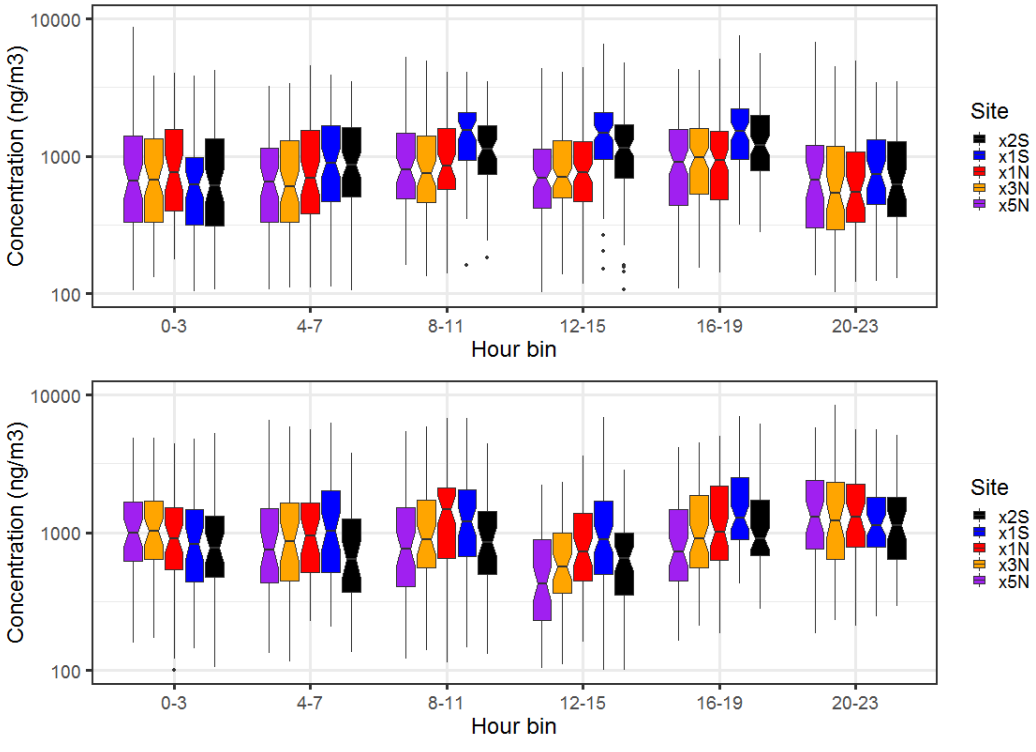
It is important when interpreting the results from this project to understand the local meteorology. The South Platte River is about one mile west of the project boundary shown in Figure 1. Typically, winds are from the south during the overnight and early morning hours, following the river drainage. Therefore locations north of the highway are expected to see higher impacts during those times. As the mountains to the west of Denver absorb sunlight and the temperature rises, a low pressure zone is created (i.e. the Denver Cyclone) and winds start backing from the northeast by early afternoon, especially during the warmer months. On average, it is more likely that a site north is downwind of the highway in the early morning hours and then upwind of the highway in the afternoon and early evening hours. This will mask concentration gradients from the highway unless concentrations are binned by wind direction.

3. Results

3.1. Black Carbon in Campaigns I, II, and III

Near-road gradients can be examined as either acute phenomena that occur within the context of a local wind field, or as a cumulative chronic gradient that integrates over all possible wind directions. Both types of exposures can be important, as people living, working, and going to school next to roadways have different activity patterns that may be affected as a result of their movements. We examined both the chronic integrated exposures and the acute maximum gradients when winds are from the roadway.

Diurnal patterns of BC concentrations segregated into three wind bins are shown in Figure 2. Wind bins are 120 degree arcs—winds from the north, winds in two 60-degree arcs from the east or west parallel to the freeway, and winds from the south. Hours are binned in 4 hour aggregates to increase the sample size, reduce notch sizes and better illustrate central tendencies in patterns. A few patterns are evident. First, BC concentrations show distinct patterns that mirror traffic activity, most importantly showing rush-hour peaks from 0600 to 0800 local time (LT) and from 1600 to 1800 LT. These patterns are more pronounced in Campaign III data, shown in Figure 3, and when winds are either parallel or from the south in both Figures 2 and 3. The concentration gradients were characterized across all hours when segregated by wind direction and are shown in Tables 3 and 4 for Campaigns I and II, and Campaign III, respectively.



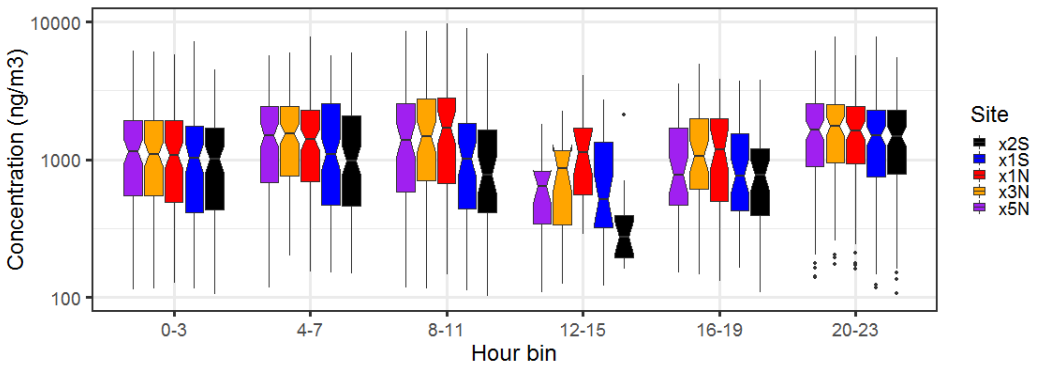


Figure 2. Campaign I and II gradients in hourly average binned BC concentrations (ng/m^3) measured by microAeths when winds are from the north (top), parallel (middle), and south (bottom).

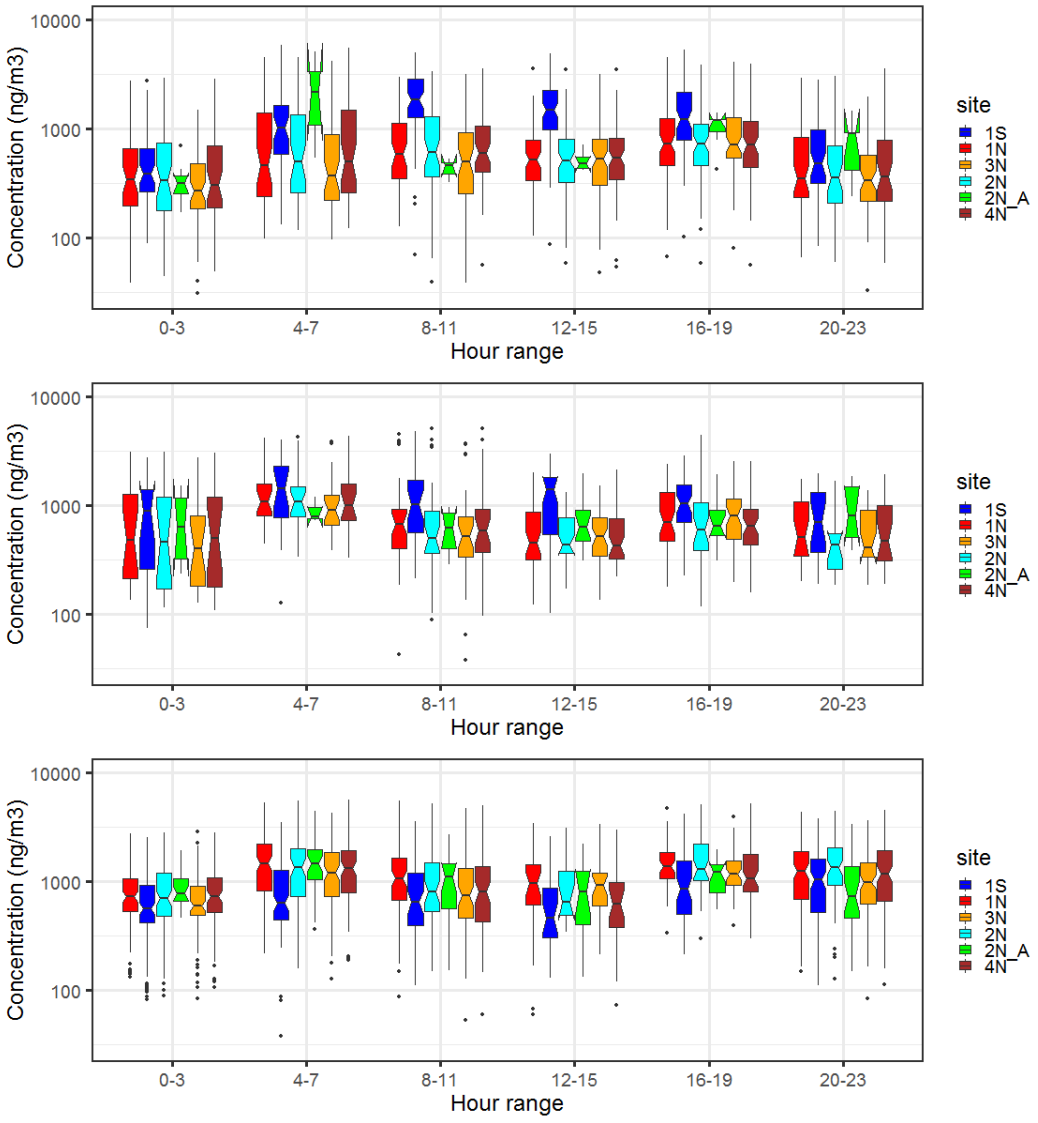


Figure 3. Campaign III gradients in hourly average binned BC concentrations (ng/m^3) measured by microAeths when winds are from the north (top), parallel (middle), and south (bottom).

Table 3. Summary average BC concentrations for Campaigns I and II, segregated by wind direction. Bold values indicate that the distribution of concentrations was statistically significantly higher than those at other sites.

		Average BC Concentration (ng/m^3)			
Site	Distance (m) from I-70	Winds from the South	Parallel Winds	Winds from the North	All Wind Directions
1S	16	1398	**1444**[a]	**1419**	**1418**[a]
2S	127	1308	1103	**1255**	1234
1N*	17	**1692**[c]	1378	1103	1392[b]
3N	102	**1700**[c]	1232	999	1311
5N	177	**1632**[c]	1113	999	1257

[a] The distribution of concentrations for Site 1S was significantly higher at the 95% confidence level than for all other sites except 1N.

[b] The distribution of concentrations for Site 1N was significantly higher at the 95% confidence level than for Site 5N.

[c] The distribution of concentrations for Sites 1N, 3N, and 5N were all higher than for Sites 1S and 2S at the 95% confidence level.

Table 4. Summary average BC concentrations for Campaign III, segregated by wind direction. Bold values indicate that the distribution of concentrations was statistically significantly higher than those at other sites.

		Average BC Concentration (ng/m^3)			
Site	Distance (m) from I-70	Winds from the South	Parallel Winds	Winds from the North	All Wind Directions
1S	16	**970**[a]	**1212**[a]	**1383**[a]	**1169**[a]
1N	91	1335	908	784	1059
2N	146	1363	891	782	1034
3N	102	1114	802	687	903[b]
4N	177	1240	851	796	1007

[a] The distribution of concentrations for Site 1S was significantly higher at the 95% confidence level than for all other sites when winds blew from the north, were parallel, and under All Wind Directions. It was statistically significantly lower when winds blew from the south.

[b] The distribution of concentrations for Site 3N was significantly lower at the 95% confidence level than for all other sites except Site 2N.

Mann-Whitney U-Tests were run on datasets from Campaigns I/II and III by site to determine if the distributions of concentrations were statistically significant at the 95% confidence level. Note that the Mann-Whitney U-Test is non-parametric so does not assume anything about the shape of the concentration distribution. Concentration distributions at Site 1S were significantly higher than those at other sites farther from I-70 in both Campaigns I/II and Campaign III.

Gradients stratified by wind bins in the 120-degree arcs relative to the roadway were statistically significant across many of the comparisons in both the Campaign I/II and Campaign III datasets. When winds were perpendicular to I-70, statistically significant gradients of 300-500 ng/m^3 were found at all downwind sites. The higher downwind concentrations cause higher exposures for people downwind of I-70 during those periods. These gradients were large enough to indicate that

BC was statistically significantly higher than at upwind sites at the downwind sites located more than a hundred meters from I-70.

Thus, we find that proximity to the highway is the most important contributor to exposure over long time periods, but wind direction relative to I-70 is the most important contributor for shorter-term exposure periods. These shorter-term exposure periods can be grouped into activity exposure periods to determine if children attending elementary schools are downwind of I-70 during school hours, for example, and to assess whether their relative exposures will be higher than those of children attending nearby schools located farther from major roadways.

3.2. VOC Sampling in Campaign III

In Campaign III, VOC samples were collected on 22 days. Individual samples from each site were compared to daily mean concentrations to determine the relative increments at each site. The average increments at individual sites were never statistically significantly larger than at other sites, largely as a result of large standard deviations in the population means. Supplemental Table 1 shows the summary statistics for all pollutants collected during Campaign III that had valid above detection limit samples with more than 75% of samples collected. Given that the sample size was only 22 days, the uncertainty in mean concentrations was larger than any concentration differences across sites. For example, the percentages of the standard deviations for most of the hydrocarbons (not the chlorofluorocarbons) were usually larger than 40%, resulting in 95% confidence intervals of around 20% for most site-pollutant means. When compared to 95% confidence intervals, all pollutants' mean increments were statistically significantly indistinguishable from a mean value of 100%; i.e., none of the results are indicative of a population mean that is higher than the overall average with statistical signficance. To detect gradients of less than 15% (i.e., the magnitude calculated for aggregated BC gradients) across sites with the temporal variability in concentrations that were observed day-to-day, at least 80 daily samples at each site would be needed to get confidence intervals down to the 10% level. Note, this is not saying that there is no gradient or that the gradient is ~15% across sites; we are simply stating that a gradient of less than 30% relative magnitude could not be detected with statistical confidence based on the limited number of samples, the uncertainty in the measurement method, and the shifting winds over a 24-hr period of sample collection.

Two independent methods were used to identify potential emissions sources influencing the near-road sites for VOCs. In the first, we used enrichment ratio plots. Enrichment ratio plots normalize concentrations of two pollutants on the x-axis and y-axis by dividing them by the concentration of a third pollutant; this method helps to remove some of the meteorological variability from the standard scatter plot of two pollutants. In the second method, positive matrix factorization (PMF) was run on the combined data from all sites for VOCs to identify "factors" that correspond to emissions sources with covariant pollutant concentrations.

Enrichment ratio plots segregated by wind direction bin provide some evidence that multiple sources are affecting VOC concentrations at the near-road sites. Figure 4 shows the enrichment ratio plots for benzene and n-pentane divided by propane; benzene and 2,2,4-trimethlypentane divided by propane; and benzene and i-butane divided by propane. In the top left panel (benzene, n-pentane), the wind direction bins are clearly separated into a cluster of blue points indicating that higher ratios of benzene/propane occur when the wind blows from the south, whereas lower ratios of benzene/propane occur when the wind blows from the north. In contrast, in the top right panel, the relative ratios remain the same but the wind direction bins are aligned linearly. Finally, the bottom left panel shows benzene/propane and i-butane/propane; similar to n-pentane propane, the samples are clustered by wind direction with clear bifurcation of the patterns between southerly and northerly winds.The contrast between the three sets of figures clearly indicates a second hydrocarbon source in addition to mobile source emissions influencing the near-road sites.

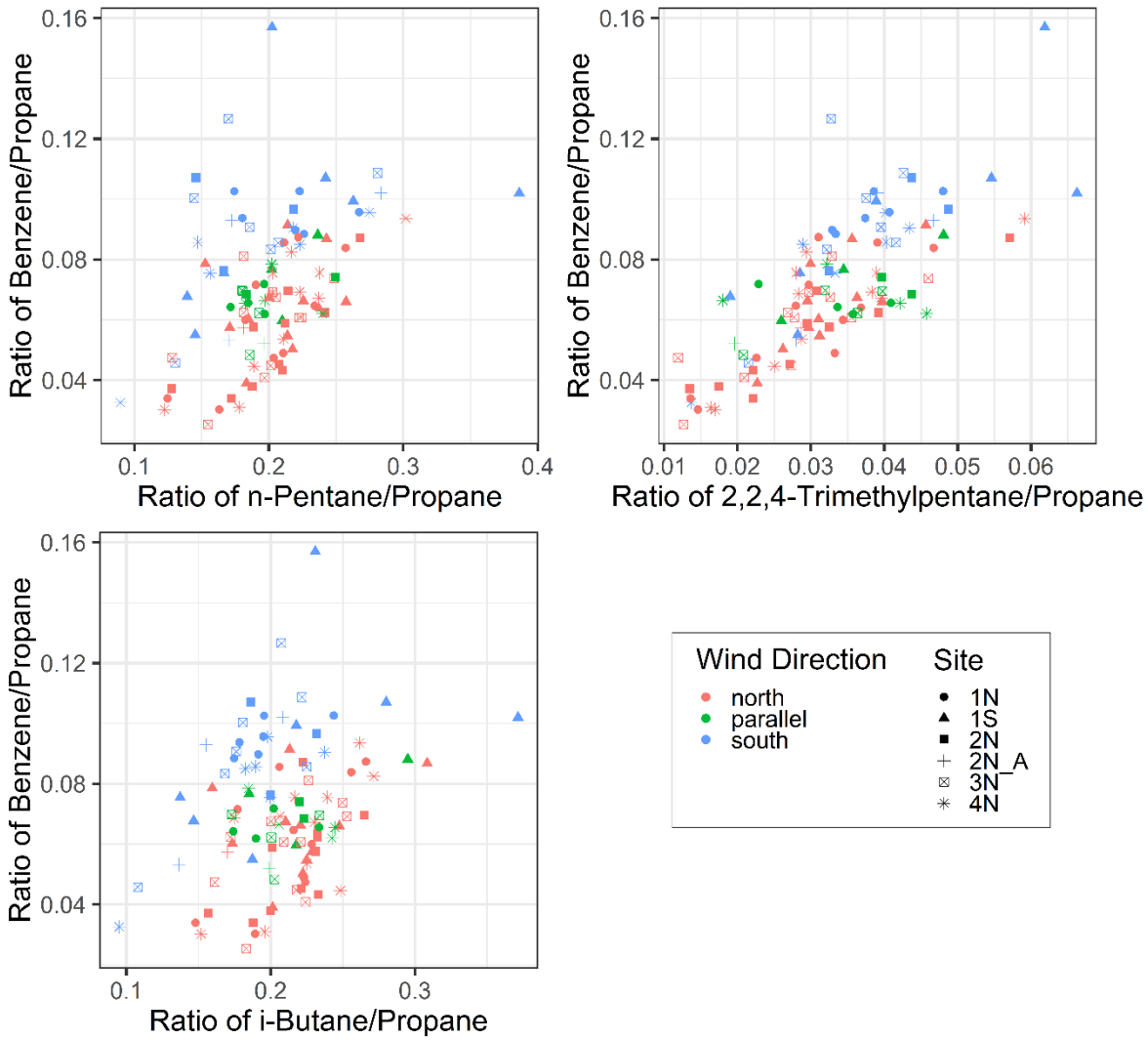


Figure 4. Enrichment ratio scatter plots of (top, left) benzene and n-pentane divided by propane, (top, right) benzene and 2,2,4-trimethylpentane divided by propane, and (bottom, left) benzene and i-butane divided by propane. All figures are colored by wind direction bins, and sites are indicated by different shapes.

Source apportionment using PMF on VOCs has been performed on data collected in areas such as Los Angeles, California; Houston, Texas; and Edmonton, Alberta (Brown et al., 2007; McCarthy et al., 2013; Buzcu and Fraser, 2008; Rappenglück et al., 2013; Miller et al., 2002; Watson et al., 2001; Field et al., 2015). PMF requires only ambient data, and assumptions regarding the numbers or types of sources or specific source profiles are not explicitly needed (Brown et al., 2015; Paatero, 1999). Multiple PMF scenarios were run to examine the potential number of emissions sources that could be identified using PMF. Scenarios were run with 3-, 4-, and 5-factor solutions. Initial runs that included isoprene always included a factor with isoprene as the dominant VOC, representing biogenic emissions. Isoprene was then excluded from the analysis because biogenic emissions sources were not of interest. In subsequent 3- and 4-factor runs, 3-factor solutions were as stable as 4-factor solutions with regard to bootstrapping (88% reproducibility for 3-factor solutions; 86% reproducibility for 4-factor solutions). These bootstrapping results are consistent with a relatively small matrix of total records (~120 samples and 50 species). The 3-factor solution results are classified in Table 5 and shown in Figure 5. Selecting a 4-factor solution split the mobile source factor into two separate categories with a long-chain alkane component with key tracers of n-decane and n-nonane; this may be representative of diesel exhaust emissions.

Table 5. Factors identified and key pollutants used to classify emissions sources in PMF.

Factor Number	Factor Name	Key Pollutants in Factor
Factor 1	Mobile Source	Acetylene, ethene, toluene, 2,2,4-trimethylpentane, xylenes
Factor 2	Background	Carbon tetrachloride, Freons
Factor 3	Short-Chain Alkanes	Ethane, propane, n-butane, i-butane, n-pentane

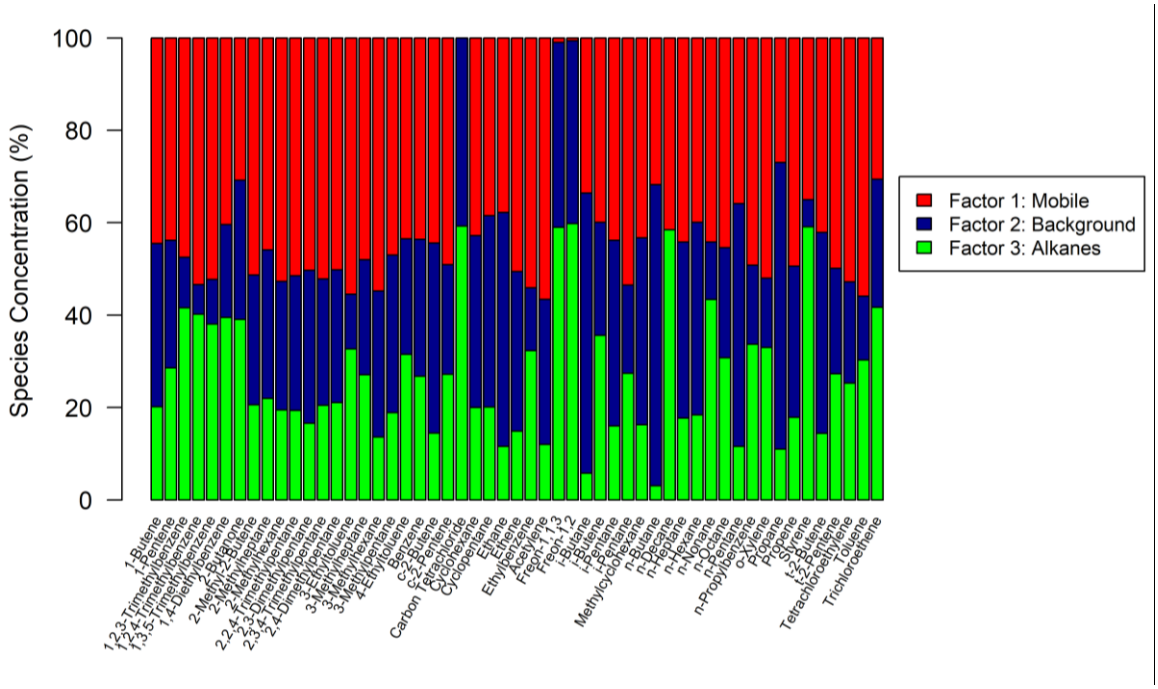


Figure 5. Percent of pollutants identified in each factor with a 3-factor PMF solution excluding isoprene.

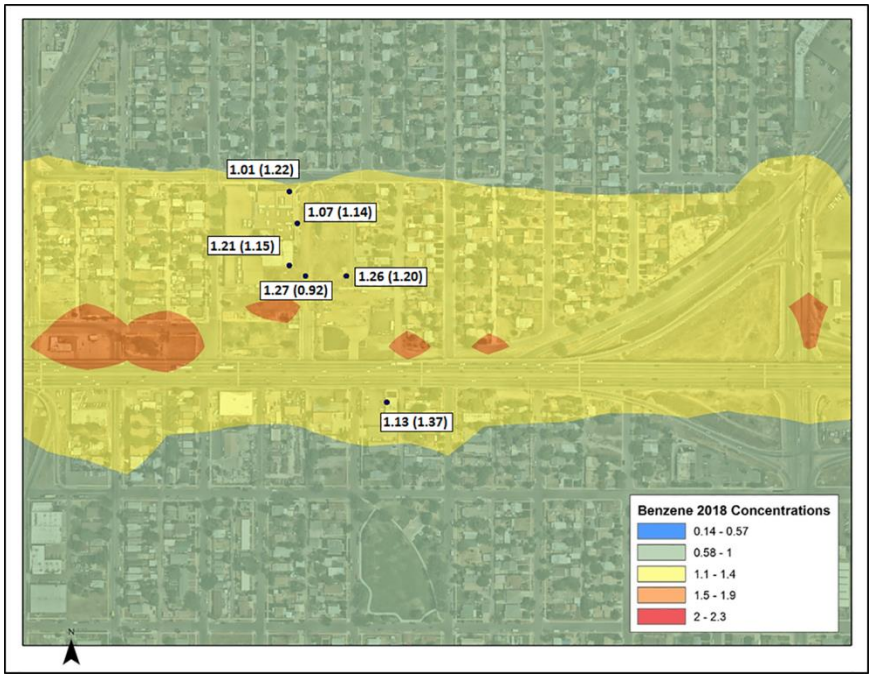
3.3. Aldehyde Sampling in Campaigns I and II

Daily carbonyl samples were collected over 36 days at six sites during Campaigns I and II. Analysis of the aggregate data showed that no gradients were statistically significant, with mean normalized site concentrations of formaldehyde and acetaldehyde varying by less than 15% across sites, significantly lower than the approximately 18-20% 95% confidence interval in the normalized mean. In other words, any gradients would not be statistically distinguishable unless they were much larger in magnitude than the small differences seen. To detect gradients of less than 15% across sites

with the temporal variability in concentrations observed day-to-day, at least 120 daily samples at each site would be needed to get confidence intervals down to the 10% level. Daily concentration statistics for the carbonyls are provided in Supplemental Table 2.

3.4. AERMOD Modeling Results

The AERMOD model was run separately for TAZ and link sources. The benzene concentrations from both models were then summed to calculated the predicted ambient benzene concentrations. Figure 1 displays the spatial pattern of modeled benzene concentrations for the month of January in the study area. The exact model and monitor values are shown on the figure (shown as model(monitor)) and model to monitor ratios range from 0.83 to 1.38. Figure 2 shows the model estimates of average benzene concentrations during the month of July as a representative of summer season. Model-to-monitor ratios range from 0.40 to 0.7 with most ratios less than the factor of 2. It is important to note that even though the benzene measurements were collected during September and October, comparison of these measurements with the model estimates of January and July showed good model performance. When applying what we know about meteorology and climatology by season/month in Denver, the modeled concentration for the Sep-Oct 2018 monitoring study would fall somewhere between January and July, midway would be a reasonable assumption. If that assumption is correct, then model-to-monitor ratios would range from 0.61 to 1.04, which would be considered excellent model performance. Overall, the model gradient (>0.5 ppb) in concentration is much larger than that observed in the measurements.



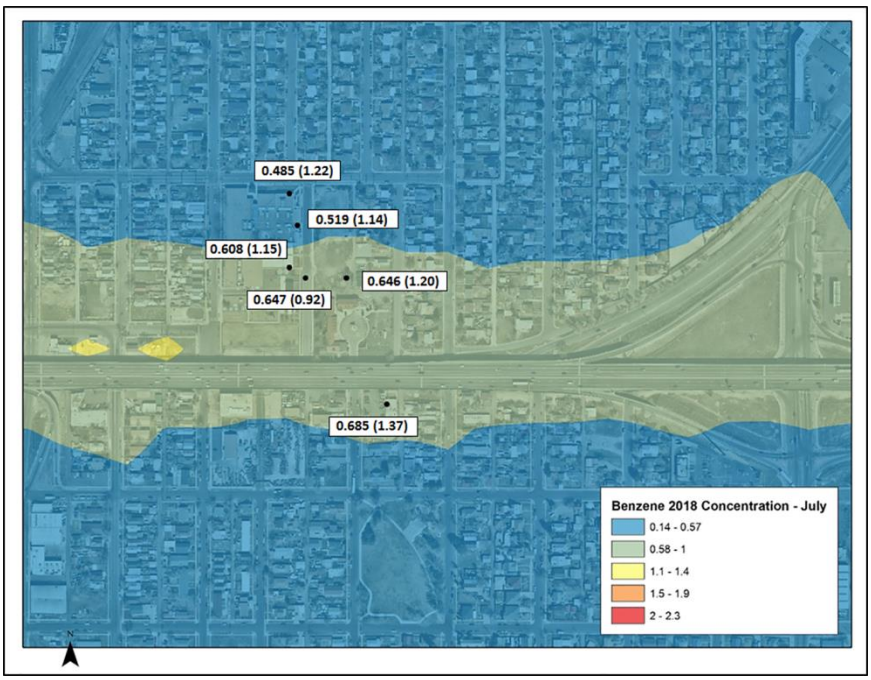


Figure 6. *Predicted 24-hour averages of benzene concentrations during the month of January 2018 (top) or July 2018 (bottom) and the observed benzene measurements (24-hour averages) collected from September 15th through October 28th, 2018. The values on the map are shown as: Model(Monitor).*

4. Discussion

For this study, multiple temporary monitoring stations were deployed in the environmental justice community of Elyria-Swansea in the vicinity of Swansea Elementary School. Monitoring stations were within 200 m of I-70, a large freeway with AADT of 153,000 vehicles.

MicroAethalometer measurements of BC provided statistically significant concentration gradients for the near-road environment in the community. Over the three campaigns, under downwind conditions, hourly average BC concentrations at downwind sites were on the order of 300-600 ng/m^3 higher than those upwind of I-70 during the same hours. This increment was 20-40% higher than average concentrations of BC upwind of I-70. However, aggregation of the concentrations over the entire campaign timeframe with winds alternating between upwind and downwind significantly reduced the total expected exposures of persons living or working next to I-70. Maximum increments observed at sites within 20 m of I-70 were 100-150 ng/m^3 higher than those observed at sites 150 m from I-70 during the same time period. Aggregate increments were reduced relative to the wind-segregated bins due to the shifting winds relative to I-70's east-west orientation. These total increments were only 10-15% higher than concentrations observed 100-150 m from I-70 under the same wind conditions. This apparent reduction may be an important consideration when modeling exposures and expected health impacts for persons living or working in the near-road environment.

In contrast to previous work in this area, we found that integrated gradients in concentrations in the near-road environment were relatively small. The classic work by Zhu et al. (2002) showed an exponential decrease in roadway BC concentrations in Los Angeles, and many follow-up studies in the 2000s showed that BC gradients were large relative to background, i.e., > 80% (Karner et al., 2010). Saha et al. (2018) show a BC gradient around 500-800 ng/m^3 above background (400 m upwind site) in the wintertime, using measurements that are primarily downwind. This study found a comparable BC downwind gradient on the order of 300-600 ng/m^3, relative to the upwind site (16 meters upwind). In contrast to the study by Saha et al. (2018) this study found a small BC gradient of 100-150 ng/m^3, over 150 meters from the roadway. This could be due to differences in dispersion

related to wind speed in Denver, or because the sampling time of the measurements were integrated over longer periods for this study. While total upwind vs. downwind gradients were as large as a 40% increment on the downwind side of the highway, the integrated gradients over all wind directions and samples were small, only 15%. This means that hours when receptors are downwind of a freeway BC concentrations are well above background, and when integrated over multiple months BC concentrations next to the freeway are 15% higher than further away from the freeway. The smaller gradients observed here may be due to cleaner diesel vehicles resulting from tighter emissions standards as the vehicle fleet becomes cleaner over time. This is consistent with an overall observed decreasing trend in black carbon in near-road and urban settings (Rattigan et al., 2013; Dallmann and Harley, 2010; McDonald et al., 2013)(Milando 2016). Alternately, the smaller gradient may just be due to the longer time period and integrated measurement relative to most shorter-term studies.

Following up on the BC comparison, the VOCs and carbonyls provided no quantitative gradients that could be significantly distinguished from the concentrations at other sites. While this may appear to be a null result, we can constrain the possible size of gradient that could be present given our confidence in our ability to measure the concentrations of the VOCs and carbonyls. Our typical 95% confidence intervals for the individual site-parameter combinations were on the order of ±20%. Propagating uncertainty across multiple sites, we can estimate that concentration gradients on the order of 29% would be theoretically detectable with a 95% confidence level. None of the individual sites or parameters displayed a 30% gradient in concentrations, as shown in the supplemental tables. Therefore, we can quantitatively state that if there are gradients in concentrations of these species in the near-road environment, they are smaller than 20% in magnitude, i.e., essentially as small or smaller than the gradients in BC. In addition, we can state that with typical day-to-day variance in meteorology (wind speed and wind direction), we would need at least four times as many samples to theoretically quantify gradients on the order of 10% (similar to the size of the aggregated gradient observed for BC). This may be a useful guideline for future studies considering near-road gradient analyses.

Model performance was excellent in terms of all monitors predicting benzene concentrations at levels within a factor of two of observed measured concentrations. We note that the winds observed at the Rocky Mountain Arsenal site used for the modeling are significantly higher speed than those observed at the Swansea monitoring site and this may bias the model concentration estimates lower than what would be expected with wind speeds more characteristic of the urban area.

5. Conclusions

Air pollutant concentrations are often higher near major roadways than in the surrounding environment owing to the proximity of emissions from on-road mobile sources. In this study, we quantified the gradient in black carbon concentrations in the near-road environment of the Elyria-Swansea environmental justice neighborhood in Denver, Colorado. The gradient in concentrations segregated by wind direction from the roadway was 400-600 ng/m^3, equal to an increment of approximately 30-40% above local background levels. When integrated over all wind directions, the gradients were smaller, approximately 100-150 ng/m^3 (~10-15%) over the course of about four months of measurements. These gradient estimates are smaller than those found in previous studies and may be due to cleaner diesel vehicles due to Tier 2 and 3 emissions standards.

Measurements of VOCs and carbonyls were unable to statistically quantify gradients because the gradients were too small in magnitude (<30%) compared to the number of samples and uncertainty in measurements. This finding is also in contrast to studies reported in Karner et al 2010, but may be due to significant reductions in emissions over the past ten to twenty years that result in a smaller near-road gradient over an urban background. We estimate that sample sizes of at least 100 individual measurements would have been required to estimate mean concentrations with sufficient

certainty to quantify gradients on the order of 15%. These results suggest that recent decades of improved emissions standards and the steady turnover of the vehicle fleet may be reducing the impact of the roadway environment on near-road communities.

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Supplemental for Assessment of Mobile Source Air Toxics in an Environmental Justice Denver Community Adjacent to a Freeway

Supplemental Table 1. Summary statistics of normalized concentrations of VOCs for Campaign III relative to the daily mean (all statistical values are reported as percentages).

PARAMETER	SITE	count	min	max	mean	median	stdev	CI_95
1,2,4-TRIMETHYLBENZENE	1N	21	23.8	154.6	79.4	78.4	40.4	17.7
1,2,4-TRIMETHYLBENZENE	1S	21	47.1	174.9	103.5	96.6	38.1	16.7
1,2,4-TRIMETHYLBENZENE	3N	21	31.8	186.4	99.6	87.9	45.5	19.9
1,4-DIETHYLBENZENE	1S	22	41.2	288.6	112.0	100.5	57.9	24.8
1,4-DIETHYLBENZENE	3N	21	42.1	159.8	88.0	76.6	34.8	15.3
1,4-DIETHYLBENZENE	4N	21	39.0	448.9	124.7	116.3	87.2	38.2
1-BUTENE	1N	21	47.5	180.6	99.4	93.2	40.5	17.8
1-BUTENE	1S	22	39.4	204.9	115.0	116.9	40.7	17.4
1-BUTENE	3N	22	40.1	168.2	91.4	92.2	36.4	15.6
1-BUTENE	4N	21	30.7	160.4	98.7	105.6	42.3	18.6
1-PENTENE	1S	22	46.5	192.5	105.3	109.5	45.9	19.6
2,2,4-TRIMETHYLPENTANE	1N	21	35.0	178.2	97.2	89.8	45.1	19.7
2,2,4-TRIMETHYLPENTANE	1S	22	38.2	192.1	109.3	109.9	42.6	18.2
2,2,4-TRIMETHYLPENTANE	3N	22	32.5	166.7	92.3	83.4	41.3	17.6
2,2,4-TRIMETHYLPENTANE	4N	21	35.2	191.8	102.5	89.3	49.8	21.8
2,3,4-TRIMETHYLPENTANE	1S	22	24.1	189.3	105.5	108.2	44.4	19.0
2,3-DIMETHYLPENTANE	1N	21	36.7	199.8	97.8	97.8	49.0	21.5
2,3-DIMETHYLPENTANE	1S	22	25.9	197.7	105.2	99.0	43.1	18.4
2,3-DIMETHYLPENTANE	3N	22	36.0	179.6	94.1	94.6	43.0	18.4
2,3-DIMETHYLPENTANE	4N	21	29.4	225.7	102.5	88.5	53.3	23.4
2,4-DIMETHYLPENTANE	1N	21	24.6	193.2	98.2	97.3	45.6	20.0
2,4-DIMETHYLPENTANE	1S	22	40.7	187.5	102.5	104.8	39.2	16.8
2,4-DIMETHYLPENTANE	3N	22	31.2	178.5	98.1	94.7	42.4	18.1
2,4-DIMETHYLPENTANE	4N	21	36.5	186.6	102.1	98.1	47.3	20.7
2-BUTANONE	1N	21	31.2	478.9	120.3	81.6	96.3	42.2
2-BUTANONE	1S	22	41.8	163.1	83.8	71.7	35.1	15.0
2-BUTANONE	3N	22	45.3	217.4	90.0	77.8	44.6	19.1
2-BUTANONE	4N	21	41.1	546.2	111.9	76.7	110.7	48.5
2-METHYL-2-BUTENE	1S	22	36.9	191.3	111.3	109.0	49.6	21.2
2-METHYL-2-BUTENE	3N	22	34.8	172.2	93.4	89.8	43.4	18.6

2-METHYL-2-BUTENE	4N	21	40.9	231.5	101.0	97.8	53.7	23.5
2-METHYLHEPTANE	1S	22	50.1	208.7	102.6	94.1	45.5	19.5
2-METHYLHEPTANE	3N	21	41.4	164.5	93.3	89.7	39.4	17.3
2-METHYLHEXANE	1N	21	35.1	188.7	99.3	95.6	44.1	19.3
2-METHYLHEXANE	1S	22	31.6	184.3	102.7	96.9	38.8	16.6
2-METHYLHEXANE	3N	22	34.0	185.4	97.0	90.5	43.4	18.6
2-METHYLHEXANE	4N	21	28.9	184.3	101.9	93.4	49.2	21.6
3-ETHYLTOLUENE	1N	21	29.4	152.6	88.5	89.0	38.3	16.8
3-ETHYLTOLUENE	1S	22	30.3	178.2	103.2	102.2	42.2	18.1
3-ETHYLTOLUENE	3N	22	35.0	176.4	95.4	91.5	43.0	18.4
3-ETHYLTOLUENE	4N	21	30.1	217.6	107.9	102.7	52.3	22.9
3-METHYLHEPTANE	1N	21	35.6	199.6	87.7	78.6	43.5	19.1
3-METHYLHEPTANE	1S	22	52.1	256.3	112.1	100.8	53.2	22.8
3-METHYLHEPTANE	3N	22	38.0	168.9	95.1	96.1	40.2	17.2
3-METHYLHEPTANE	4N	21	34.6	212.5	109.0	115.8	54.0	23.7
3-METHYLHEXANE	1N	21	35.6	226.5	99.6	88.5	55.9	24.5
3-METHYLHEXANE	1S	22	36.6	213.7	98.7	87.6	48.2	20.6
3-METHYLHEXANE	3N	22	34.2	221.5	97.3	74.3	54.8	23.4
3-METHYLHEXANE	4N	21	30.0	210.0	104.1	103.5	58.9	25.8
3-METHYLPENTANE	1N	21	34.0	225.0	101.2	101.0	48.2	21.1
3-METHYLPENTANE	1S	22	34.1	241.2	104.0	93.3	50.9	21.8
3-METHYLPENTANE	3N	22	41.2	200.2	94.8	83.8	44.5	19.0
3-METHYLPENTANE	4N	21	33.6	219.1	102.7	99.7	54.8	24.0
BENZENE	1N	21	42.1	188.4	99.3	98.1	42.2	18.5
BENZENE	1S	21	58.0	201.4	111.7	108.6	39.6	17.3
BENZENE	3N	22	43.1	176.5	95.6	95.2	35.7	15.3
BENZENE	4N	21	37.1	210.4	101.2	98.8	44.1	19.3
C-2-BUTENE	1S	21	48.7	196.5	109.4	107.7	44.6	19.6
C-2-PENTENE	1S	21	38.9	182.1	106.7	102.3	47.0	20.6
C-2-PENTENE	3N	21	35.9	163.6	89.1	91.7	41.3	18.1
CARBON_TETRACHLORIDE	1N	21	95.6	104.5	100.6	100.4	2.2	1.0
CARBON_TETRACHLORIDE	1S	22	91.4	106.1	99.9	98.9	3.6	1.6
CARBON_TETRACHLORIDE	3N	22	94.0	106.5	100.1	99.7	2.9	1.2
CARBON_TETRACHLORIDE	4N	21	93.7	106.2	99.9	99.2	2.9	1.3
CYCLOHEXANE	1N	21	29.6	335.0	102.8	91.8	68.5	30.0
CYCLOHEXANE	1S	22	45.8	315.1	109.7	96.3	63.2	27.0
CYCLOHEXANE	3N	22	43.1	182.6	90.0	86.5	37.2	15.9
CYCLOHEXANE	4N	21	34.7	277.6	104.5	91.0	58.9	25.8
CYCLOPENTANE	1N	21	45.1	186.0	98.1	93.0	42.3	18.5
CYCLOPENTANE	1S	22	43.6	189.6	101.4	94.7	40.2	17.2
CYCLOPENTANE	3N	22	42.9	182.5	95.6	92.6	42.5	18.2

CYCLOPENTANE	4N	21	43.3	201.0	103.2	100.6	47.6	20.9
ETHANE	1N	21	31.2	164.4	90.5	82.4	41.1	18.0
ETHANE	1S	22	40.6	210.1	112.4	110.4	46.9	20.1
ETHANE	3N	22	32.8	190.0	101.5	92.5	45.8	19.6
ETHANE	4N	21	34.2	173.3	98.4	92.8	42.0	18.4
ETHENE	1N	21	38.7	194.5	95.2	82.0	46.1	20.2
ETHENE	1S	22	54.3	247.7	118.9	110.3	48.3	20.7
ETHENE	3N	22	36.5	188.0	94.1	82.3	43.6	18.7
ETHENE	4N	21	32.4	183.4	98.3	83.2	47.3	20.7
ETHYLBENZENE	1N	21	27.9	168.2	89.6	91.5	39.7	17.4
ETHYLBENZENE	1S	22	39.4	172.9	101.3	102.5	39.4	16.8
ETHYLBENZENE	3N	22	48.3	194.5	114.6	118.4	45.1	19.3
ETHYLBENZENE	4N	21	32.9	205.5	99.9	101.2	50.9	22.3
ETHYNE_(ACETYLENE)	1N	21	28.9	337.9	95.6	83.0	71.1	31.1
ETHYNE_(ACETYLENE)	1S	22	41.9	382.5	111.2	101.1	76.4	32.7
ETHYNE_(ACETYLENE)	3N	22	28.6	327.9	97.8	88.3	67.5	28.9
ETHYNE_(ACETYLENE)	4N	21	26.8	320.8	104.7	89.5	72.1	31.6
FREON-113	1N	21	93.6	107.0	99.7	99.3	3.3	1.5
FREON-113	1S	22	91.9	117.8	100.1	100.0	5.1	2.2
FREON-113	3N	22	95.4	107.1	99.3	98.8	3.0	1.3
FREON-113	4N	21	94.4	109.3	100.7	100.8	3.5	1.5
FREON-12	1N	21	93.7	105.5	99.9	100.3	2.6	1.2
FREON-12	1S	22	91.8	104.5	99.2	99.7	3.2	1.3
FREON-12	3N	22	93.5	106.2	100.2	99.6	3.0	1.3
FREON-12	4N	21	93.9	106.8	100.7	101.0	2.8	1.2
FREON-13	1N	21	90.7	119.7	98.9	96.9	7.0	3.1
FREON-13	1S	22	89.2	116.5	100.1	98.2	6.7	2.9
FREON-13	3N	22	93.5	132.1	100.4	98.1	8.5	3.6
FREON-13	4N	21	91.8	122.4	99.8	97.8	7.2	3.2
I-BUTANE	1N	21	34.4	225.3	97.0	93.6	51.2	22.4
I-BUTANE	1S	22	44.7	216.4	104.7	99.0	47.7	20.4
I-BUTANE	3N	22	33.2	225.0	96.0	88.2	50.0	21.4
I-BUTANE	4N	21	34.1	253.3	103.8	101.8	53.8	23.6
I-BUTENE	1N	21	38.4	165.2	99.8	104.2	39.8	17.4
I-BUTENE	1S	22	64.0	187.0	110.6	104.6	34.0	14.6
I-BUTENE	3N	22	42.1	153.7	94.3	103.0	32.4	13.8
I-BUTENE	4N	21	50.6	161.2	101.5	98.4	37.1	16.3
I-PENTANE	1N	21	39.7	208.0	99.3	92.9	47.1	20.6
I-PENTANE	1S	22	43.0	215.4	107.4	107.2	45.3	19.4
I-PENTANE	3N	22	43.0	196.2	94.8	88.8	44.4	19.0
I-PENTANE	4N	21	39.0	200.1	101.9	96.1	49.8	21.8

M+P-XYLENE	1N	21	32.3	185.6	92.1	94.5	41.4	18.1
M+P-XYLENE	1S	22	42.0	189.7	103.6	97.9	42.7	18.2
M+P-XYLENE	3N	22	42.8	194.2	98.6	98.8	44.1	18.8
M+P-XYLENE	4N	21	27.8	221.9	105.0	105.7	54.4	23.9
METHYLCYCLOHEXANE	1N	21	40.3	198.1	98.5	91.5	47.1	20.7
METHYLCYCLOHEXANE	1S	22	42.3	204.6	105.1	85.3	46.0	19.7
METHYLCYCLOHEXANE	3N	22	39.9	186.6	95.2	89.6	41.2	17.6
METHYLCYCLOHEXANE	4N	21	43.0	219.0	102.9	93.7	50.7	22.2
N-BUTANE	1N	21	36.1	254.3	97.9	85.5	55.6	24.4
N-BUTANE	1S	22	41.7	251.5	103.7	90.1	52.2	22.3
N-BUTANE	3N	22	35.3	252.4	95.9	82.3	54.6	23.3
N-BUTANE	4N	21	35.8	275.8	102.2	90.2	58.9	25.8
N-DECANE	1S	21	20.5	326.1	95.1	76.6	66.0	28.9
N-DECANE	3N	21	35.7	205.0	96.7	81.2	50.5	22.1
N-HEPTANE	1N	21	35.3	190.3	96.5	98.2	45.6	20.0
N-HEPTANE	1S	22	45.1	200.3	104.0	87.8	40.5	17.3
N-HEPTANE	3N	22	39.9	193.5	97.2	84.4	46.1	19.7
N-HEPTANE	4N	21	33.3	181.3	100.1	93.1	46.7	20.4
N-HEXANE	1N	21	45.0	219.8	99.5	86.2	46.4	20.3
N-HEXANE	1S	22	47.0	224.3	104.1	88.5	45.6	19.5
N-HEXANE	3N	22	45.8	204.4	95.3	85.1	44.2	18.9
N-HEXANE	4N	21	41.6	208.8	101.1	92.1	50.6	22.2
N-NONANE	1N	21	32.1	145.8	74.9	70.9	32.2	14.1
N-NONANE	1S	22	19.1	460.1	104.1	90.3	85.4	36.5
N-NONANE	3N	22	20.6	184.8	92.3	82.3	44.1	18.9
N-NONANE	4N	21	31.2	248.1	111.7	96.5	59.6	26.1
N-OCTANE	1N	21	48.4	183.6	90.8	77.3	37.2	16.3
N-OCTANE	1S	22	49.1	240.1	104.3	93.7	53.8	23.0
N-OCTANE	3N	22	41.5	207.4	98.6	90.4	45.8	19.6
N-OCTANE	4N	21	32.3	181.2	99.5	95.5	46.6	20.4
N-PENTANE	1N	21	41.1	221.4	98.3	90.9	51.6	22.6
N-PENTANE	1S	22	44.3	224.4	104.4	97.5	48.7	20.8
N-PENTANE	3N	22	41.4	213.0	94.8	87.7	49.8	21.3
N-PENTANE	4N	21	38.6	216.6	103.1	97.5	51.7	22.7
O-XYLENE	1N	21	30.8	179.7	93.3	93.6	39.3	17.2
O-XYLENE	1S	22	44.0	183.9	102.3	102.1	41.0	17.6
O-XYLENE	3N	22	43.2	179.5	97.3	100.1	42.3	18.1
O-XYLENE	4N	21	32.5	210.4	105.6	101.3	51.7	22.6
PROPANE	1N	21	37.4	209.4	95.7	94.9	45.3	19.9
PROPANE	1S	22	38.0	202.6	99.8	84.4	45.3	19.4
PROPANE	3N	22	41.0	208.9	99.3	93.1	47.1	20.1

PROPANE	4N	21	38.9	248.3	105.4	98.6	56.0	24.5
PROPENE	1N	21	43.5	180.9	96.8	84.1	44.6	19.5
PROPENE	1S	22	44.2	232.7	114.9	99.6	47.4	20.3
PROPENE	3N	22	39.7	167.4	94.1	84.5	40.2	17.2
PROPENE	4N	21	31.8	183.4	99.8	96.9	47.9	21.0
STYRENE	3N	21	59.2	292.8	137.8	135.6	62.3	27.3
T-2-BUTENE	1S	22	52.3	194.9	110.9	110.4	43.6	18.7
T-2-BUTENE	3N	22	37.0	158.9	91.7	84.4	42.4	18.1
T-2-PENTENE	1S	21	49.0	182.8	101.3	98.3	41.7	18.3
TETRACHLOROETHYLENE	1N	21	38.5	234.9	100.2	94.5	52.4	23.0
TETRACHLOROETHYLENE	1S	22	42.1	265.1	99.2	89.4	54.5	23.3
TETRACHLOROETHYLENE	3N	22	37.1	237.3	100.1	87.2	54.4	23.2
TETRACHLOROETHYLENE	4N	21	33.7	244.3	103.1	81.4	57.8	25.3
TOLUENE	1N	21	32.2	187.7	95.8	96.7	45.8	20.1
TOLUENE	1S	22	43.7	203.7	101.3	95.7	45.2	19.3
TOLUENE	3N	22	37.8	191.9	97.2	91.2	46.7	20.0
TOLUENE	4N	21	34.6	188.7	102.4	95.3	54.0	23.7

Supplemental Table 2. Summary statistics of normalized concentrations of carbonyls from Campaigns I and II relative to the daily mean (all statistical values are reported as percentages).

Site	Parameter	count	min	max	mean	median	std dev	range	95th confidence interval
1N	Acetaldehyde	36	26.7	262.7	100.6	88.4	56.2	9.8	18.6
1S	Acetaldehyde	36	4.4	272.5	108.7	93.2	62.9	61.6	20.9
2N	Acetaldehyde	36	4.4	243.8	97.1	77.5	59.4	55.1	19.7
2S	Acetaldehyde	35	31.4	263.4	107.3	86.5	60.3	8.4	20.3
3N	Acetaldehyde	36	28.2	270.2	108.0	87.6	62.0	9.6	20.5
5N	Acetaldehyde	36	23.1	245.4	102.3	89.3	54.9	10.6	18.2
1N	Formaldehyde	36	27.7	232.8	96.9	82.6	53.5	8.4	17.7
1S	Formaldehyde	36	2.1	316.0	110.8	92.1	66.3	151.8	22.0
2N	Formaldehyde	36	2.1	222.0	99.0	89.3	56.3	106.7	18.7
2S	Formaldehyde	35	32.8	270.3	114.3	92.3	63.9	8.2	21.5
3N	Formaldehyde	36	27.7	260.3	112.4	98.2	63.0	9.4	20.9
5N	Formaldehyde	36	28.8	253.0	108.0	94.2	60.6	8.8	20.1