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Air Pollutant Mapping with a Mobile Laboratory During the BEE-TEX Field Study

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Supplementary Issue: Ambient Air Quality (B)

ABSTRACT: The Aerodyne Mobile Laboratory was deployed to the Houston Ship Channel and surrounding areas during the Benzene and Other Toxics Exposure field study in February 2015. We evaluated atmospheric concentrations of volatile organic hydrocarbons and other hazardous air pollutants of importance to human health, including benzene, 1,3-butadiene, toluene, xylenes, ethylbenzenes, styrene, and NO_2. Ambient concentration measurements were focused on the neighborhoods of Manchester, Harrisburg, and Galena Park. The most likely measured concentration of 1,3-butadiene in the Manchester neighborhood (0.17 ppb) exceeds the Environmental Protection Agency's E-5 lifetime cancer risk level of 0.14 ppb. In all the three neighborhoods, the measured benzene concentration falls below or within the E-5 lifetime cancer risk levels of 0.4–1.4 ppb for benzene. Pollution maps as a function of wind direction show the impact of nearby sources.

KEYWORDS: 1,3-butadiene, benzene, Houston, cancer risk, air quality

SUPPLEMENT: Ambient Air Quality (B)

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Introduction

The region surrounding the Houston Ship Channel is home to a high density of petrochemical facilities. This unique industrial complex includes crude oil refineries, processing plants, and chemical manufacturers. Together, these facilities enable efficient conversion of raw crude oil and various natural gas liquids into saleable products, including gasoline, diesel, and other distillates. Other specialized facilities produce chemical feedstocks for manufacture of plastic, rubber, and other compounds.

Interspersed within this industrial area are several residential neighborhoods. Industrial plants often purchase land that abuts their facilities in an effort to expand their borders, and thus limit the concentration of pollutants at their inhabited fenceline. Land-use disputes become entangled with social justice issues when the issue of air toxics arises, and the relationship between industry and residents often grows contentious. In this study, we focus on the Manchester neighborhood, bordered by industry on the north, a rail yard on the south, and a highway on the west. Two other neighborhoods, Harrisburg and Galena Park, are sampled to a lesser degree. The geographic bounds of the sampled regions are shown in Figure 1.

The data collected in these neighborhoods are part of the interdisciplinary Benzene and Other Toxics Exposure (BEE-TEX) study directed by the Houston Advanced Research Center. A team of scientists leveraging a variety of measurement and study techniques was deployed to the Houston Ship Channel in February 2015. This study focuses on the measurements made by the Aerodyne Mobile Laboratory[2,3] (AML) of a variety of aromatic toxics such as benzene, toluene, ethylbenzene, and xylene (so-called BTEX compounds); nitrogen dioxide (NO_2); as well as 1,3-butadiene, styrene, and other hazardous air pollutants (HAPs). The concentrations of these toxics in the three neighborhoods along the Houston Ship Channel are also characterized.

NO_2 is a byproduct of combustion and involved in smog chemistry. It is a lung irritant and can lead to an increased incidence of respiratory illness in children and asthmatics.[4,5]

Benzene, the simplest aromatic building block, is used in the synthesis of other chemicals. Benzene, along with toluene and xylene isomers, is also commonly used as a solvent. Ethylbenzene is a feedstock for styrene production, which in turn is used in the manufacture of polystyrene, styrene–butadiene rubbers (SBR), and other polymers. 1,3-Butadiene is also used as a building block in the manufacture of synthetic rubbers including neoprene, nitrile, and SBR, which end up in consumer products such as wet suits, gloves, tubing, and tires.

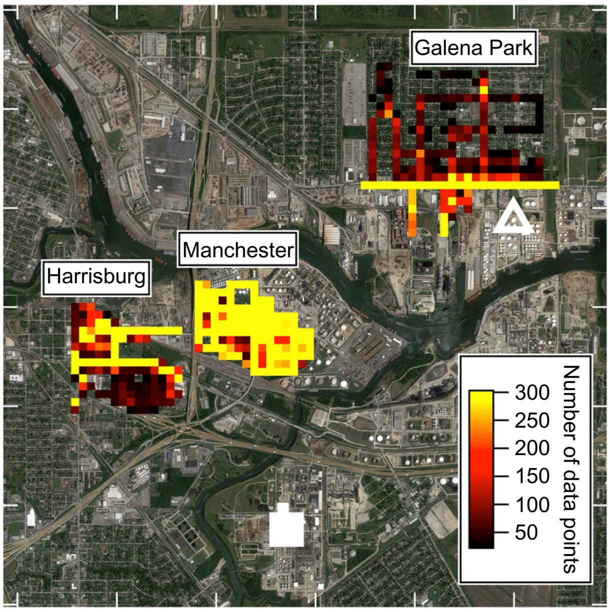

Figure 1. The BEE-TEX study area along the Houston Ship Channel. The neighborhoods of Manchester, Harrisburg, and Galena Park are shown in colors corresponding to the frequency of sampling (see text). The major sources of 1,3-butadiene (squares) and benzene (triangles) are also shown.[1] Map underlay © 2015 Google.

Both benzene and 1,3-butadiene are carcinogenic to humans, causing lymphohematopoietic cancers such as leukemia.[6–9] Other effects include blood disorders such as anemia, and 1,3-butadiene causes ovarian atrophy in mice. These carcinogenicity determinations are based on a combination of animal studies and human exposure studies on factory workers in the shoemaking[10] or rubber-manufacturing[11] industries. More recent studies on ambient levels of HAPs, and specifically on benzene and 1,3-butadiene, have also been performed.[12,13] Whitworth et al used the addresses of childhood leukemia patients along with estimates of ambient concentrations in and around Houston (including the Houston Ship Channel) to deduce an association between childhood leukemia and exposure to 1,3-budatiene and benzene.[14] Some evidence on the carcinogenicity of styrene exists,[15] while epidemiological evidence on other compounds of interest, particularly at low concentrations, is insufficient.[16,17]

Concentration data collected within the neighborhoods of interest are averaged over the BEE-TEX study period and compared to estimated concentrations calculated as part of the United States Environmental Protection Agency's (EPA) National Air Toxics Assessment (NATA). NATA concentrations are based on the best available inventory data. Neighborhood measured concentrations are also compared to exposure and risk limits set by the EPA. The average concentrations measured in this work are smaller than 2005 NATA estimates, but approach the concentrations determined by EPA risk assessments to increase cancer risks over a lifetime. The effect of the dominant wind direction is explored, showing the impact of nearby source regions. Certain sub-neighborhood regions are shown to have higher than average concentrations of pollutants, highlighting the importance of using mobile measurement platforms for monitoring and assessment of HAPs at the neighborhood scale.

Methodology

This paper leverages measurements made by the AML[2,3] equipped with trace gas measurement instrumentation,[18,19] including a Proton Transfer Reaction Mass Spectrometer operated in NO^+ reagent ion mode[20] (NOMS) for the detection of benzene, toluene, ethylbenzene, xylene, 1,3-butadiene, styrene, and other HAPs. The AML continuously samples ambient air at a height of 2.8 m and a rate of 8 standard liters a minute as it travels around the study area. Geographic and meteorological parameters are also measured during the experiment. Position information is obtained from a Hemisphere V100 GPS Compass. A sonic anemometer (Airmar 200 WX) mounted to the AML's mast provides true wind measurement and a redundant GPS measurement. The Airmar 200 WX also measures temperature, and direct water concentration measurements are done with an Aerodyne tunable infrared laser direct absorption spectrometer.[21]

The 2.5-week study period (February 2–18, 2015) was chosen to best allow all interdisciplinary members of the BEE-TEX campaign to participate. Sampling was conducted continuously during the study period, with most data collected during daylight hours (see Supplementary File, for additional discussion of sampling in terms of date, day of week, time of day, and ambient temperature.)

Mobile concentration measurement data for the 2.5-week period were collected, plotted, and averaged geographically. The sampling frequency for some data varied between 1 second and 7 seconds, but all data were interpolated[22] onto a 1-second time base prior to the geographic analysis.

Data points with over 1000 ppb of carbon monoxide were excised from the dataset (2.8% of the data) in order to eliminate data dominated by exhaust plumes from cars, trucks, and other mobile sources. Broader background enhancements due to traffic sources are still represented in the analysis.

A two-dimensional histogram approach was used to perform a surface-level geographic concentration averaging. Geographic bin sizes measuring 80 m × 80 m (the approximate block size in Manchester) are shown in Figures 2 and 3. Averages are only calculated for geographic bins with a minimum of 25 data points. A color map of the bin counts is included (inset lower right) in order to better represent the variations in sampling frequency across the neighborhoods. Areas of the map shown in yellow have 300 or more sample points.

Two dominant wind directions were present in the Houston Ship Channel during the study period: northerly and southerly. These two wind vectors sample entirely different emission sources, and so the geographic binning was performed after filtering for both north and south winds.

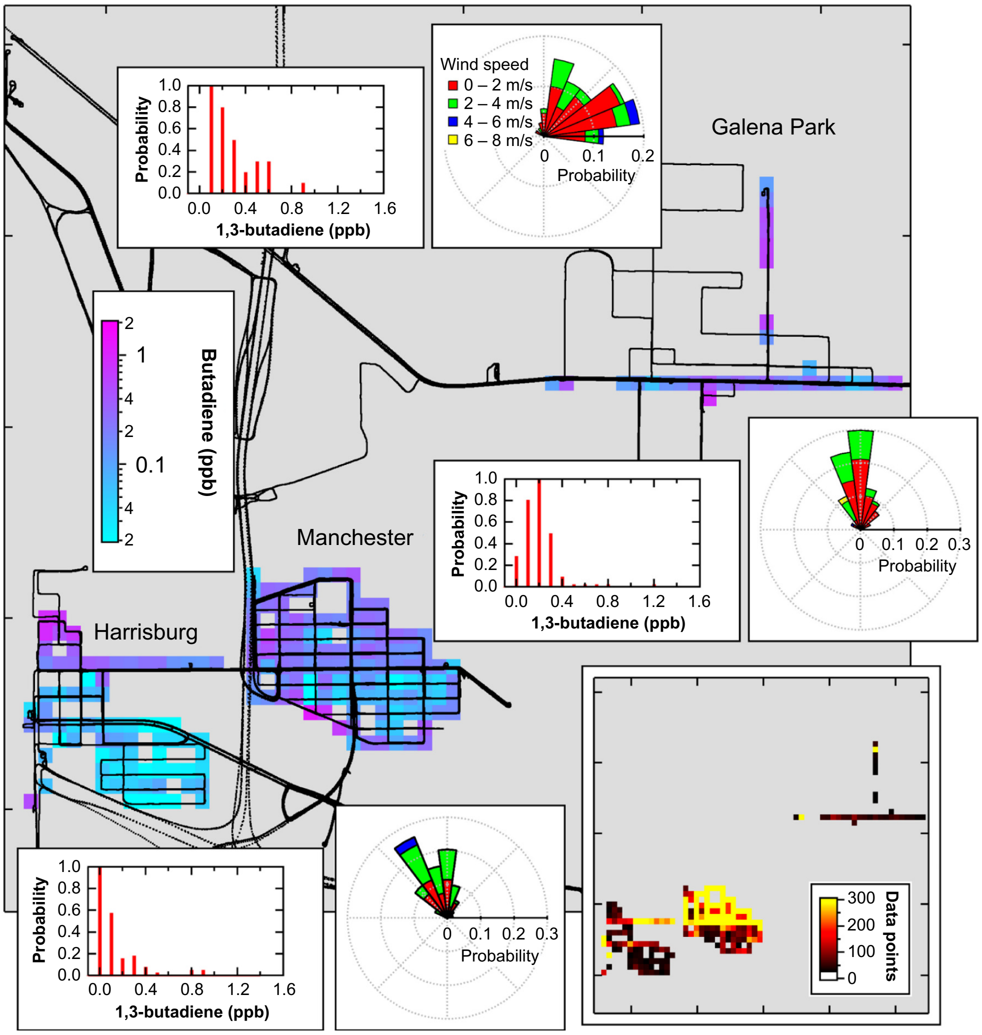

Figure 2. 1,3-Butadiene concentrations with a northerly wind. See text for description of insets.

Wind roses show the dominant wind directions and speeds for the displayed data. The radius of each polar segment indicates a probability, in intervals of 0.1, while the colors indicate wind speed bins: red (0–2 m/second), green (2–4 m/second), blue (4–6 m/second), and yellow (6–8 m/second).

Instead of taking a simple average of all geographic bins, the probability of a given concentration bin is computed (inset histograms, red bars). These one-dimensional histograms then allow for an estimate of the most likely concentration over the neighborhood region. Representative concentrations are determined by taking the average of the most likely concentrations measured under northerly and southerly winds, ie, the average of the peaks of the respective histogram distributions. The most likely concentration x is reported alongside a σ width: x (σ). This width is calculated via error propagation of the Gaussian-fit σ widths from the north and south distributions: $\sigma = \sqrt{\sigma_N^2 + \sigma_S^2}$.

Results and Discussion

Ambient concentration measurements need to be put in context to understand their impacts on human health. Table 1 shows the concentration limits from various sources. EPA values from its Integrated Risk Assessment Software (IRIS) are listed below. These EPA values in μg m^{-3} were converted to ppb using a temperature of 25 °C and a pressure of 760 Torr. The risk assessment values assume a lifetime exposure. The EPA reference concentration (RfC) is a concentration with 1 order of magnitude error bars, below which adverse effects on a population are unlikely. The EPA's lifetime cancer risk number is the lifetime exposure level that leads to an E-5 times increased risk of cancer, or a 1/100,000 increased risk of cancer. Other risk levels (E-6 and E-4) are included in the source EPA material. When "inadequate information" or "no data" is noted, the EPA has deemed studies on humans inconclusive. Finally, the EPA produces census tract level estimates of air toxic concentrations as part of its NATA program.[23,24] The NATA "County-level Pollutants" databases were mined for the three neighborhoods of interest (with specific census tracts noted): Manchester (320300), Harrisburg (311400), and Galena Park (233700).

The most likely ambient concentrations for a number of air toxics for the three neighborhoods of interest are also reported

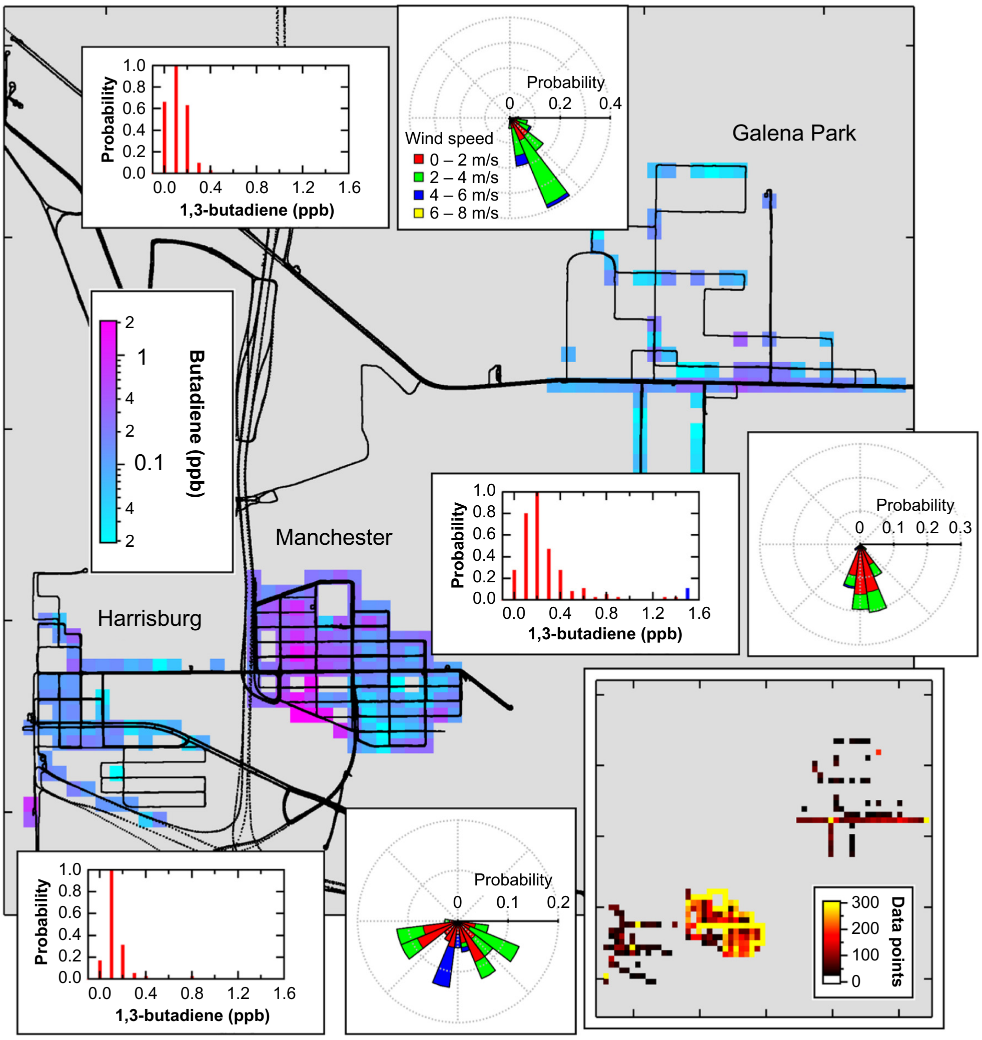

Figure 3. 1,3-Butadiene concentrations with a southerly wind. See text for description of insets.

in Table 1. Of the three neighborhoods, the Manchester neighborhood has the best data coverage over space (Fig. 1) and time (see Supplementary File). Data in the Harrisburg and Galena Park neighborhoods are significantly sparser, and the tabulated concentrations thus less representative.

Benzene concentrations are highest in the Galena Park neighborhood at 0.78 (0.78) ppb. The benzene distributions are significantly narrower and peak at lower concentrations for the Manchester and Harrisburg neighborhoods, 0.37 (0.25) ppb and 0.33 (0.15) ppb, respectively.

1,3-Butadiene concentrations are highest in the Manchester neighborhood at 0.17 (0.15) ppb and are consistent across both north and south winds. The Galena Park and Harrisburg neighborhoods have concentrations of 0.13 (0.14) ppb and 0.07 (0.08) ppb, respectively, but both show a difference of >0.06 ppb between the northerly and southerly wind concentrations.

Other chemical compounds of interest are shown alongside various regulatory and risk assessment concentration limits. These concentrations are in the same ballpark as the Texas Commission on Environmental Quality hourly automated gas chromatographic sampling sites[26] (see Supplementary File). In these comparisons, the Galena Park dataset has sparser geographic coverage than others, with many measurements occurring on a major road. Concentrations from this dataset may thus be more heavily influenced by traffic than those measurements taken on the low-traffic residential streets of Manchester and Harrisburg.

Comparison of the measured concentrations with the risk level concentrations, shown in Table 1, indicates that risk levels for lifetime exposure are met or exceeded only for certain air toxics. Of the compounds investigated here, 1,3-butadiene is the species that poses the highest risk, and as such has the lowest E-5 lifetime cancer risk value at 0.14 ppb. The measured concentration of 0.17 ppb in the Manchester neighborhood exceeds this limit, and the Galena Park neighborhood comes close at 0.13 ppb. Benzene has an E-5 lifetime cancer risk level of 0.4–1.4 ppb, making it the second highest cancer risk species among those under investigation. This 0.4–1.4 ppb range accounts for the fact that there is a wealth of benzene data available that allows for better bounds on this measure of risk. In all cases, the measured benzene concentration in

Table 1. Exposure limits and measured concentrations, in ppb, for selected hazardous air pollutants.

SOURCE	DESCRIPTION OF CONCENTRATION [PPB]	BENZENE	TOLUENE	XYLENES	ETHYL-BENZENE	XYLENES + ETHYL-BENZENE	STYRENE	1,3-BUTADIENE	NITROGEN DIOXIDE (NO_2)
EPA IRIS (Refs[8,9,16,17,25])	RfC	9	1,300	9,000			230	0.9	
	Lifetime cancer risk E-5	0.4–1.4	Inadequate information	Inadequate information			No data	0.14	
EPA NATA (Ref[23])	Estimated, Manchester (320300)	4.64	5.28	3.23	0.71	3.94	1.14	0.58	
	Estimated, Harrisburg (311400)	2.24	2.94	1.74	0.39	2.13	0.86	0.44	
	Estimated, Galena Park (233700)	5.39	5.34	1.96	0.43	2.39	0.26	0.76	
This paper[a]	Measured, Manchester	0.37 (0.25)	0.54 (0.39)			0.52 (0.37)	0.21 (0.12)	0.17 (0.15)	25 (9)
	Measured, Harrisburg	0.33 (0.15)	0.77 (0.69)			0.89 (1.01)	0.16 (0.14)	0.07 (0.08)	22 (3)
	Measured, Galena Park	0.78 (0.78)	0.66 (0.52)			0.53 (0.47)	0.12 (0.11)	0.13 (0.14)	24 (9)

Notes: [a]The most likely concentration is taken from concentration histograms of the measured data. The width (σ) of a Gaussian fit of this histogram is shown in parentheses. See text and Supplementary File for further details.

these neighborhood analyses falls below this upper bound of 1.4 ppb, with the Galena Park neighborhood falling within these bounds at 0.78 ppb. The calculations of these E-5 risk levels have inherent levels of uncertainty.[27] However, these results suggest that additional emission reduction strategies, particularly for the highly toxic 1,3-butadiene, are required.

The 2005 NATA data shown in Table 1 highlight the fact that these real-world measurements can be used to test inventory-based estimates. NATA concentrations of hazardous species include contributions from individual facilities, vehicular emissions, and background levels. The concentrations estimated by NATA are greater than the measured values in all cases. These discrepancies may be partly due to the 10-year delay between the inventory date and the measurement campaign. Ozone exceedances have declined in the Houston area in the last decade, which might partly be attributed to reductions in the emissions of industrial volatile organic hydrocarbons.[28,29] Updated NATA data from 2011 are in preparation and can be used to test this hypothesis.

In order to better understand the potential causes and emission sources contributing to these neighborhood-scale average concentrations, we look in more detail at the maps of data as a function of wind direction, comparing northerly and southerly winds. In this analysis, we focus on 1,3-butadiene, the species of greatest relevance to human health. Maps for other species of interest are shown in the Supplementary File.

Averaged binned 1,3-butadiene concentrations in the neighborhoods of Harrisburg, Manchester, and Galena Park are shown in Figures 2 and 3 (blue-pink color scale). The concentrations are colored on a log scale. Histograms showing the occurrence probability of any given concentration are also shown (red bars) with the final bin including all higher concentration data (blue bar). Additional descriptions of these plots are detailed in the methods section. Histograms of the time coverage of these maps are presented in the Supplementary File.

The most striking difference in these plots of 1,3-butadiene occurs in the Manchester neighborhood. With wind from the south (Fig. 3), a hotspot with average concentrations up to 1.9 ppb is observed in the western part of the residential neighborhood, a value that is 10 times higher than the EPA E-5 risk level. Interestingly, the most commonly observed concentration does not change much with wind direction (0.17 ppb with a north wind vs. 0.18 ppb with a south wind, see Supplementary Table 1), but rather the southerly wind results in a distribution with more of a tail at higher concentrations; in fact, the highest concentration 80 m × 80 m bin is at 1.98 ppb.

The 2005 NATA data include a map of facility point sources of HAPs[24] and lists the identities of chemicals comprising 90% of emissions (toxicity weighted). In this region, the TX Petrochemicals LP, Goodyear Houston Chemical Plant, and the PL Propylene LLC facilities account for emissions of 82, 1.3, and 8.5 tons per year of 1,3-butadiene emissions, respectively. These 1,3-butadiene sources, shown in Figure 1, are from the same cluster of facilities investigated by Knighton et al.[30] Other sources in the area are listed at less than 0.002 tons/year. The main cluster of emitters is located 2 km SSE of the region of enhanced 1,3-butadiene, and in the same direction as the dominant southerly wind. Polar concentration plots for 1,3-butadiene in each neighborhood provide another way of identifying the direction of major sources, as shown in Figure 4. Indeed, a distinct ray of 1,3-butadiene appears at 165 degrees compass in the Manchester neighborhood (Fig. 4, left most plot), which points at this cluster of 1,3-butadiene emitters.

Conclusions

This study demonstrates the utility of using a mobile laboratory for neighborhood-scale pollutant mapping. The variations

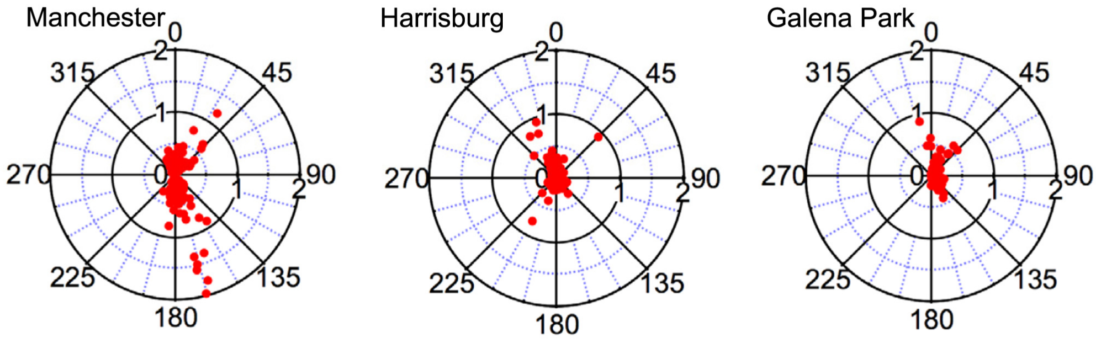

Figure 4. 1,3-Butadiene polar plots for the three neighborhoods. Geographically binned concentrations are plotted radially (in ppb) against the associated average wind direction.

and hot spots of pollution within a neighborhood may not be well represented by a monitoring station style of sampling, which must depend only on wind to sample different source emitters.

Results of ambient concentration measurements of air toxics including benzene and 1,3-butadiene in three neighborhoods near the Houston Ship Channel are presented. Measured concentrations are lower than concentration estimates from NATA 2005, but approach and, in some cases, exceed lifetime cancer risk levels, particularly for 1,3-butadiene. Investigation of concentration maps as a function of wind direction shows the impact of large point sources of 1,3-butadiene on sub-neighborhood scale concentration measurements. These results suggest that further emission mitigation work is needed to bring ambient concentrations of the HAP 1,3-butadiene below the EPA's calculated E-5 lifetime cancer risk level.

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Author Contributions

Collected the data: CF, JRR, SCH, WBK. Analyzed the data: TIY, SCH, JRR, WBK. Wrote the first draft of the manuscript: TIY. Contributed to the writing of the manuscript: TIY, SCH, CEK. Agree with manuscript results and conclusions: All authors. Jointly developed the structure and arguments for the paper: TIY, SCH, JRR. Made critical revisions and approved final version: All authors. All authors reviewed and approved of the final manuscript.

Supplementary File

Supplementary File for this manuscript is available online and includes an expanded table of most likely concentrations, a discussion of sampling representativeness, concentration maps, and histograms for additional HAP species, and further discussions of the 1,3-butadiene polar concentration plot. The tables and figures contained in the Supplementary File are described below.

Supplementary Table 1. Most likely concentrations of various hazardous air pollutants by neighborhood and wind direction.

Supplementary Figure 1. Map of the study area.

Supplementary Figure 2. Histogram plots of the number of data points used in the neighborhood most likely concentrations as a function of date and time of day.

Supplementary Figure 3. Histogram plots of ambient temperature (left) as a function of wind direction.

Supplementary Figure 4. Benzene concentration map (North winds).

Supplementary Figure 5. Benzene concentration map (South winds).

Supplementary Figure 6. 1,3-Butadiene concentration map (North winds).

Supplementary Figure 7. 1,3-Butadiene map (South winds).

Supplementary Figure 8. NO_2 map (North winds).

Supplementary Figure 9. NO_2 map (South winds).

Supplementary Figure 10. Toluene map (North winds).

Supplementary Figure 11. Toluene map (South winds).

Supplementary Figure 12. Styrene map (North winds).

Supplementary Figure 13. Styrene map (South winds).

Supplementary Figure 14. C2-Benzene map (North winds).

Supplementary Figure 15. C2-Benzene map (South winds).

Supplementary Figure 16. 1,3-Butadiene polar plots for the Manchester neighborhood.

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