ORIGINAL RESEARCH

A Passive Sampling Study of Small-Scale Variations in Ambient Acetaldehyde and Formaldehyde Concentrations

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Abstract: Radiello passive diffusive aldehyde samplers were used to measure ambient formaldehyde and acetaldehyde levels, approximately every 0.7 km in a 10 km^2 sampling area in Hillsborough County, Florida from January 21 to 28, 2010. Samples were analyzed for aldehyde-DNPH derivatives via high performance liquid chromatography with ultraviolet detection. Concentrations were compared with values at a regulatory fixed-site monitor. Distribution statistics, concentration ratios, and spatial contours were calculated to investigate spatial variability. Mean aldehyde concentrations were 2.4 and 1.1 µg/m^3 for formaldehyde and acetaldehyde, respectively. Observed spatial concentration patterns were similar for both aldehydes and suggest the influence of nearby roadway emissions. Overall, the spatial variation was small, with coefficients of variation of 13% and 22%, respectively. Results here provide methods and data for understanding exposures to aldehydes at high spatial resolution.

Keywords: passive aldehyde sampling, intra-urban spatial variation, Restek Allure AK column, exposure misclassification

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Introduction

Aldehyde emissions have been noted to increase by as much as two hundred percent with the use of oxygenated fuels.[1,2] However, limited monitoring data exist to evaluate exposures and health impacts associated with such increases. Formaldehyde and acetaldehyde, specifically, are ubiquitous in the environment and have known health effects.[3–6] Formaldehyde has been classified as a human carcinogen associated with increased risk of nasopharyngeal cancers.[7] Acetaldehyde is also suspected to be a human carcinogen.[8,9] In addition, formaldehyde exacerbates asthma and may be associated with incident asthma.[10,11] Both are known irritants (eyes, nose, throat, and skin).[10] Both also contribute to ozone formation and, hence, photochemical smog.[2–4,10,12]

Understanding spatial variations of air toxics, including aldehydes, within urban areas has been identified as a research priority.[13–16] Results from modeling indicate variability of these and other urban air toxics at neighborhood and community scales.[15] However, low-resolution measurement data available from sparsely-located fixed-site regulatory monitors cannot adequately characterize spatial variations over small scales. This has been cited as a significant uncertainty in health effects studies, whose results can be biased due to exposure misclassification.[14,17,18] Furthermore, the magnitude and direction of bias cannot be known without more-resolved data.[17–19]

Models can be informative at small scales, but monitoring data is still necessary to evaluate modeling results.[15,16] Only a few studies have attempted to quantify concentrations of air toxics at high resolution within cities where local sources (e.g., major roadways) can impact personal exposures.[19–21] High-resolution monitoring is often impractical due to the high costs of active sampling instruments. Mobile measurements have been used to gather high-resolution data, but this also requires expensive equipment.[19,22] Spatial characterization is further limited by resulting data that is not coincident in time. Conversely, passive sampling does not require electricity or costly equipment. Therefore, it can be a cost-effective strategy for obtaining high-resolution data and has been successfully used for assessing spatial variations of air toxics within urban areas.[16,21,22]

Here, a pilot passive sampling study was conducted to measure ambient formaldehyde and acetaldehyde in a small sampling area on the approximate scale of a neighborhood. The aims were to assess spatial variations in concentrations at this scale and to evaluate the approach for application in a full campaign.

Methods
Field sampling

This pilot study is part of a larger investigation to understand spatial variations of traffic-related pollutants within Hillsborough County, FL for environmental equity and health effects analyses. The county is located on the west coast of Florida in a growing metropolitan area (Tampa—St. Petersburg-Clearwater Metro Area). It has an estimated 2009 population of over 1 million.[23] A diverse mix of air pollution sources in areas of high population density make this county an interesting case study. Hillsborough county has also been consistently monitoring air toxics for approximately 10 years. Figure 1 shows the location of emission sources and the one active regulatory monitoring site for formaldehyde and acetaldehyde.

Here, a 10 km^2 area was chosen as the study area to assess small-scale spatial variations of the target aldehydes. Information on variation at this scale is needed to inform sampler placement for the larger study. The area corresponds to three census block groups. The block group is the smallest spatial scale at which detailed demographic information is routinely available and is often used as a proxy for a neighborhood.[24] The specific area here was chosen to allow collocation with the only currently active reference method monitor in the county. To determine spatial variations, a saturation sampling approach, with grid-based sampler placement, was used. This approach has been utilized previously to assess intra-urban spatial variation.[21,25] Here, samplers were placed approximately 0.7 km apart throughout the sampling area, in accessible locations (utility poles) in residential areas. Figure 1 shows the location of the study area and individual sampler placement. Note that two field samplers were collocated.

Ambient aldehydes were collected using Radiello diffusive samplers (Sigma-Aldrich). These samplers are impregnated with 2,4-dinitrophenylhydrazine (DNPH) coated Florisil, to which the aldehydes chemisorb, creating stable aldehyde-DNPH hydrazones. Since short sampling times have limitations for
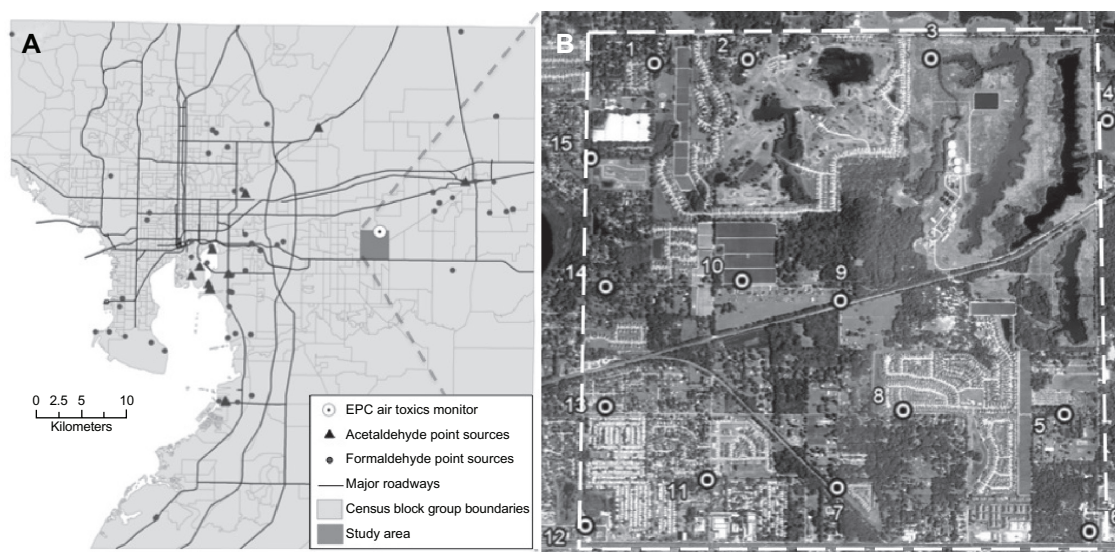
Figure 1. Study area and site locations. (**A**) provides a map of Hillsborough County, Florida, which contains the study area. The reference method air toxics monitor (EPA Air Quality System ID 120573002) is operated by the Hillsborough County Environmental Protection Commission (EPC). **Sources:** Locations of aldehyde point sources are from the US Environmental Protection Agency 2002 National Emissions Inventory. Major highway locations are from the Florida Geographic Data Library, based on 2004 data from the Florida Department of Transportation. Census block group data are from the 2000 US Census Bureau Summary 3 files. The map was created using ArcGIS software by Esri. (**B**) shows the study area (3.2 × 3.2 km), overlaid with the locations (with identifying numbers) of the study samplers. Duplicate passive samplers were collocated at site 2. Site 3 is the location of the reference method fixed-site monitor (and a collocated passive sampler). The sampler at site 6 was found on the ground and excluded from analyses. The white dashed box is the area shown in Figure 3.
Source: Google ©2009. 27° 57′ 11.11″ N, 82° 13′ 32.29″ W, 0 m. Satellite. Imagery Date Apr. 5, 2010.

representing exposures, the maximum recommended sampling interval of seven days was chosen for this pilot study.[26] Fifteen samplers (including one duplicate) were deployed from January 21 to 28, 2010. Samplers were placed at a height of 2.5 m and shelters were used for protection from inclement weather and direct sunlight.[22,26,27] All samplers were successfully deployed and retrieved; one sampler (located at site 6) was found on the ground and was subsequently excluded from analyses.

Laboratory analysis

Analytical protocols for analysis with high performance liquid chromatography (HPLC) were adapted from US Environmental Protection Agency (EPA) Method TO-11A and California Air Resources Board's (CARB) standard operating procedure No. 104,[28,29] with modifications for specific equipment and materials. An Allure AK column (200 × 4.6 mm, Restek) was employed to achieve separation of the aldehyde-DNPH derivatives using only two solvents. The mobile phase was a mixture of acetonitrile and water (both HPLC-grade) with gradient elution as follows: 60:40 (acetonitrile: water) for 0–8 min, 70:30 for 8–10 min, and reaching 100% acetonitrile by 10 min. A constant flow rate of 1.0 ml/min was used.

Following Radiello protocols,[26] cartridges were eluted with 2 ml of acetonitrile. 10 μl aliquots of filtered eluate were then analyzed. Absorbance was measured at 360 nm. Calibration was performed over the range of 0.06–6.0 μg/ml using 100 μg/ml aldehyde-DNPH stock (Restek). Calibration curves for each aldehyde had coefficients of determination (R^2) greater than 0.999. After subtraction of the mass measured on a cartridge blank (0.3 μg for formaldehyde and 0.2 μg for acetaldehyde), ambient concentrations were calculated using the recorded sampling time at each sampling site and the sampling rate for each aldehyde. Tabulated default sampling rates were corrected for the local environmental conditions measured at the regulatory reference site,[26] to 101 and 86 ml/min for formaldehyde and acetaldehyde, respectively. Only the local average temperature over the sampling interval (317 K) required a correction, as the measured relative humidity and wind speed were within the range for an invariant sampling rate. Measured ozone levels (22 ppbv on average) were also substantially below values requiring ozonolysis correction.[26]

Data analysis

Descriptive statistics, ratios of acetaldehyde to formaldehyde, and correlations were calculated to summarize

the data and compare results to those from other locations. Concentrations were compared between the duplicate field samples and between the reference method monitor and a collocated passive sampler. To investigate spatial variation in the study area, concentrations were interpolated by kriging to produce contour maps, using ArcGIS (Desktop 9.3, Redlands, CA). Coefficients of variation and concentration differences between sites were also determined.

Results and Discussion
Measured levels of acetaldehyde and formaldehyde in the study area

Table 1 presents the concentrations measured at each sampling site along with descriptive summary

Table 1. Measured concentrations ($\mu g/m^3$) and concentration ratios for each sampling site, with summary statistics.

Site	Formaldehyde[a]	Acetaldehyde[a]	Ratio (A/F)[b]
1	2.4	1.1	0.47
2	2.2	1.1	0.48
2[c]	2.3	1.0	0.44
3[d]	2.2	0.8	0.37
4	2.2	0.9	0.41
5	2.5	1.1	0.43
6[e]	–	–	–
7	1.9	0.8	0.40
8	1.7	0.6	0.33
9	2.7	1.0	0.38
10	2.5	1.2	0.49
11	2.4	1.3	0.53
12	2.4	1.3	0.52
13	2.5	1.4	0.55
14	2.5	1.3	0.52
15	2.8	1.3	0.45
Minimum	1.7	0.6	0.33
Maximum	2.8	1.4	0.55
Mean/median[f]	2.4	1.1	0.45
Standard deviation	0.3	0.2	0.06
Coefficient of variation	13%	22%	14%

Notes: [a]Concentrations are blank corrected. All values exceeded three times the standard deviation of the blanks (0.2 $\mu g/m^3$ for both aldehydes). [b]Ratio of acetaldehyde to formaldehyde concentration. [c]Duplicate collocated sampler. The duplicate precisions (as relative percent differences) were 3.3% and 5.9% for formaldehyde and acetaldehyde, respectively. [d]Reference method comparison site. [e]Data from site 6 were discarded due to possible contamination, as the sampler was found on the ground upon retrieval. [f]The mean and median were equal to the precision shown. Summary statistics and other derived values shown here and in the text were calculated using full numerical precision (in Excel). Hence, the derived values cannot be exactly reproduced using the concentration precision shown here.

statistics. Measured values are similar to the 2009 annual average concentrations from the reference fixed-site monitor for formaldehyde and acetaldehyde, of 2.6 and 1.2 $\mu g/m^3$, respectively.[30] Dasgupta et al. also reported a comparable mean value for formaldehyde, 3.2 $\mu g/m^3$, during May 2002 at a nearby intensive measurement site that was part of the Bay Region Atmospheric Chemistry Experiment.[31] Mean values found at other urban ambient sites in the US range from about 1 to 7 $\mu g/m^3$, with values at rural background sites near the lower end of that range and values at roadside sites near the higher end.[4,10,32–34] Ambient mean levels at sites in some cities outside the US (e.g., Mexico City and Rio de Janeiro), have been found to be about an order of magnitude higher.

In an effort to evaluate the risks associated with chronic exposures to ambient concentrations of pollutants, the US EPA has established comparison concentration levels. Population exposures (including sensitive populations) that occur at or below inhalation reference concentrations (RfC) are not expected to result in adverse non-carcinogenic effects over a lifetime.[6] For carcinogenic effects, measured concentrations can be compared to air concentrations that correspond to a specific risk level for lifetime exposures. For acetaldehyde, the maximum study value (1.4 $\mu g/m^3$) is several times lower than the RfC (9 $\mu g/m^3$),[6] and between the 1 in 1 million and 1 in 100,000 risk level concentrations for carcinogenicity (5×10^{-1} and 5 $\mu g/m^3$, respectively). Hence, health risks associated with lifetime exposure to the levels measured here are expected to be low. For formaldehyde, no RfC has been established,[35] but the established minimum risk level (MRL) for chronic non-carcinogenic inhalation exposures is equivalent to 9.8 $\mu g/m^3$.[5] The maximum study value for formaldehyde (2.8 $\mu g/m^3$) is a few times lower than this value, but is between the current 1 in 100,000 and 1 in 10,000 carcinogenic risk level concentrations (8×10^{-1} and 8 $\mu g/m^3$, respectively). It should be noted that the health risks associated with inhalation exposures to formaldehyde are currently being re-evaluated. Additionally, the levels measured here only represent the time period studied.

Ratios of acetaldehyde to formaldehyde level (by mass) are also presented in Table 1. The acetaldehyde concentration was lower than formaldehyde (by about a factor of 2) at every site in this study, with an

average acetaldehyde to formaldehyde ratio of 0.45 (or 0.31 on a molar basis). Grosjean et al. summarized acetaldehyde to formaldehyde ratio results from multiple studies in urban areas of the US, which ranged from 0.07 to 0.8 (on a molar basis).[12,36] Similar ratios have also been found in more recent work.[33,37–39] Data from Brazil provide an interesting exception. Historically, Brazil has had very high ethanol fuel content, with resulting high acetaldehyde to formaldehyde ratios.[12,40,41] The data here are consistent with urban ratios and ethanol fuel content in the US.

Spatial trends and variability

Spatial trends by site for both aldehydes are shown in Figure 2, with interpolated spatial distributions provided in Figure 3. Similar spatial patterns can be observed for both aldehydes, with a Pearson correlation coefficient of 0.84 between them. Correlations reported elsewhere range from 0.83–0.97.[4,33,37,42] However, little data exist on observed variation of aldehydes at this spatial scale. Based on mobile measurements of formaldehyde for a 16 km^2 area in Wilmington Delaware, Isakov et al. suggested the importance of both local emissions sources and regional photochemical production.[19] High correlations and similar concentration ratios generally suggest similar emissions sources nearby.[33,42,43]

Here, the highest concentrations of both aldehydes were generally found on the more densely populated western side of the sampling area near a busy road on the boundary. For acetaldehyde, the interpolated high area includes an intersection in the southwest corner with high observed traffic volumes relative to the study area. Specific sites with measured levels of both aldehydes in the upper half of each respective distribution are 15, 14, and 13, all of which were located near the roadway (see Fig. 1b). A local high in the interpolated plots is also seen near the southeast corner, adjacent to another relatively high volume roadway intersection. However, similar highs are not seen near all the roadways bounding the study area, including the highest volume roadway to the south (though samplers 7 and 11 were located more than 200 m further from the roadway than the samplers near the western boundary). Local concentration lows were observed in a highly vegetated residential area in the southeast quadrant of the study area in addition to near the reference method monitor to the northeast. However, sites 9, 10 and 5, which were located more centrally in the study area and further from major roadways, also had formaldehyde levels greater than the median formaldehyde level. Conversely, acetaldehyde levels at these sites were
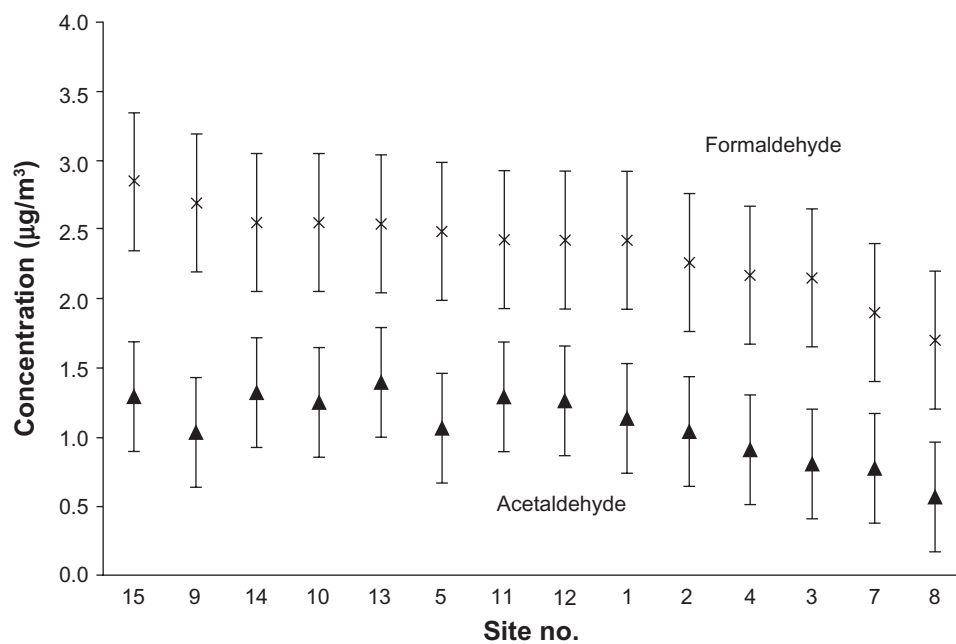

Figure 2. Measured concentrations of formaldehyde (crosses) and acetaldehyde (triangles) by sampling site. Data are sorted from highest to lowest formaldehyde concentration. Site locations are shown in Figure 1. The value shown for site 2 is the mean of duplicate collocated samplers. Data from site 6 were discarded as discussed in the Table 1 caption. Error bars represent the margin of error for 95% confidence calculated from the duplicate samplers (0.5 and 0.4 µg/m^3, respectively for formaldehyde and acetaldehyde).
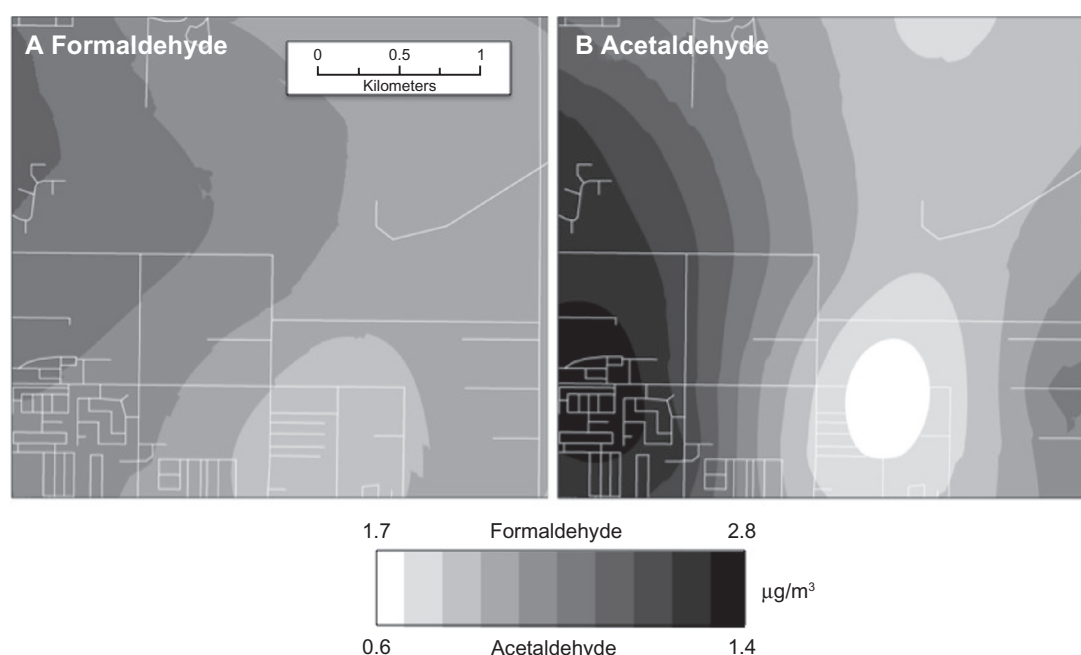
Figure 3. Spatial contours of (**A**) formaldehyde and (**B**) acetaldehyde concentrations in the sampling area. The area shown corresponds to the white dashed box in Figure 1b. Contours are based on kriging interpolation of the measured values at the sites shown in Figure 1b. The white lines provide the basic roadway network.
Sources: The map was created using ArcGIS software by Esri. The roadways locations are from 2000 US Census Bureau data.

near median acetaldehyde levels. Differences in the spatial patterns of formaldehyde and acetaldehyde may indicate differences in source influences.

Although there is no universally accepted method for quantifying spatial variation, the coefficient of variation (CV) has been used.[17,18,20] CVs calculated here indicate that the spatial variation is small in magnitude overall. However, acetaldehyde (CV of 22%) displayed slightly higher variation than formaldehyde (CV of 13%). Isakov et al. reported similar values for formaldehyde (daily morning and afternoon CVs ranged from about 5% to less than 30%) and suggested the importance of photochemical production.[19] Logue et al., in a study in Pittsburg PA, also saw less spatial variation (differences of less than 25%) for formaldehyde and more variation (differences greater than 25%) for acetaldehyde.[20] Higher variations for acetaldehyde versus formaldehyde could indicate larger impacts from local sources on acetaldehyde levels versus impacts of background photochemical production. This is also consistent with the comparative location of the high concentrations for each aldehyde discussed above.

Here, we were particularly interested in assessing overall variability at this small spatial scale, in order to inform a larger scale campaign. A threshold variation

of 20% has been used to indicate a homogenous spatial distribution for particulate matter.[17,25] CVs found here are close to or less than this threshold. This degree of variability suggests that one sampler may be somewhat representative of the study area in a larger-scale campaign. For example, using data from the site at the center of the sampling area (site 9) alone would result in formaldehyde and acetaldehyde concentrations within 15% and 5%, respectively, of the geographic mean. However, the applicability of this threshold to aldehydes requires further study. Additional research is also needed using different sampling areas within the county and multiple times of the year, in order to determine variability for areas with similar spatial scales.

Sampler evaluation

Table 2 provides data comparing concentrations from the reference method monitor with that from a collocated passive sampler (site 3). Note that the averaging times are different (the reference monitor takes 24 hour samples every 6 days, while concentrations from this study are 7 day samples), so the comparison is not direct. Nonetheless, it does provide some confidence in passively measured levels for use in exposure estimation over the longer averaging time of interest here. The measured values from both

Table 2. Comparison of passively sampled concentrations (μg/m^3) to values from the reference method fixed-site monitor.

	Passive sampler	Reference method monitor[a]			
	January 21–28	Jan 20	Jan 26[b]	Jan 26[b]	Weekly average[c]
Formaldehyde	2.2	2.4	1.7	1.9	2.1
Acetaldehyde	0.8	1.9	1.4	1.5	1.7

Notes: [a]Method TO-11A. Data provided by the Hillsborough County Environmental Protection Commission (EPC). [b]Duplicate measurements for quality control purposes, as per regulatory requirements. [c]Average of values from January 20 and January 26 (where that on the 26 is the duplicate mean).

methods were similar, though the passively-sampled acetaldehyde value is somewhat low compared with the reference monitor. Percent differences (referenced to the fixed-site monitor weekly average value) were 3% for formaldehyde and −52% for acetaldehyde. Similar comparative values were observed in a study by Mason.[44] Additionally, low passive acetaldehyde values are consistent with results by Herrington et al.,[45] who found low acetaldehyde collection efficiencies on DNPH-coated solid sorbents for sampling intervals of 24 hr.

Duplicate passive samplers at site 2 had good precision, with relative percent differences of 3% and 6% for formaldehyde and acetaldehyde, respectively. These precision values are similar to those from the January 26 regulatory fixed-site duplicate samples, shown in Table 2. They are also similar to precisions reported in other passive sampling field studies.[43,46]

Summary and Implications

A one-week pilot study using passive samplers was conducted to evaluate the measurement approach and to investigate spatial variations of acetaldehyde and formaldehyde on the neighborhood scale. Measured concentrations of both aldehydes in the study area were found to be in the range of values found in other urban areas in the US. Additionally, values collocated with a reference monitor were similar, though the collection efficiency by the samplers for acetaldehyde for this sampling period (one week) may be low. This work provides an ambient field application of the use of Radiello aldehyde passive samplers for high spatial resolution measurement. Use of these samplers for ambient studies has been limited in the US. Additionally, the method here demonstrates the use of the Allure AK HPLC column, which simplifies the laboratory analysis. Results suggest that the passive sampling and analysis approach used here can effectively characterize concentrations at high spatial resolution, particularly for formaldehyde.

Spatial variations and concentration ratios found here suggest the potential influence of nearby mobile sources for both aldehydes. However, some differences in spatial patterns are seen between the two pollutants. Variations in concentration over the sampling area were small overall, potentially indicating relative homogeneity at this spatial scale (and temporal sampling interval). As few data are available on spatial variation of aldehydes at this scale, results here provide a case study. In addition, the data provide near baseline values regarding fuel ethanol content, as its use is increasing in the area.

However, in order for these results to be generalizable, further work is needed in different areas at similar scales. A sampling area close to the city center is suggested, to assess whether increased local mobile source emissions result in higher spatial variations. Sampling all census block groups within a tract could also lead to better understanding of the spatial scale needed for future sampling (block group versus tract) and lead to better placement of future regulatory fixed monitoring sites. High-resolution data on air toxics concentrations, such as that produced in this study, can help improve exposure assessment, inform city planners and policymakers, and ensure public health.

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Disclosures

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and ethical obligations in respect to declaration of conflicts of interest, funding, authorship and contributorship, and compliance with ethical requirements in respect to treatment of human and animal test subjects. If this article contains identifiable human subject(s) author(s) were required to supply signed patient consent prior to publication. Author(s) have confirmed that the published article is unique and not under consideration nor published by any other publication and that they have consent to reproduce any copyrighted material. The peer reviewers declared no conflicts of interest.

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