

1 **Spatial variation in ambient benzene concentrations over a city park**

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4

5 **Abstract**

6 Passive diffusive samplers were used to collect ambient benzene for a one week sampling period
7 (April 27 to May 4, 2011) at eleven locations throughout a city park in the Tampa area.
8 Concentrations were determined through gas chromatography with mass spectrometry. Spatial
9 variability within the park and its contribution to uncertainty in health risk estimates were
10 studied. Measured concentrations ranged from 0.23 – 0.34 $\mu\text{g m}^{-3}$. The relative percent
11 differences for samplers collocated with a regulatory reference monitor and with a duplicate
12 were 3% and 14%, respectively. The spatial variability over the park was small with a
13 coefficient of variation of 11%. The concentration variation due to sampler placement
14 contributes less to uncertainty in health risk estimates than the uncertainty associated with the
15 inhalation unit risk parameter (39% versus 170% relative percent differences over the ranges
16 studied). Result suggest a limit to spatial resolution needed for risk calculations.

17

18 **Keywords:** air pollution, children's exposure, health risk uncertainty, passive sampling

19

20 **Introduction**

21 Benzene is a known human carcinogen (e.g. it is classified as Group 1 by the International
22 Agency for Research on Cancer), and a substantial contributor to estimated health risks of
23 ambient exposures to toxic air pollutants (McCarthy, O'Brien, Charrier, & Hafner, 2009).

1 Current regulatory monitoring networks for hazardous air pollutants, including benzene, use
2 large and expensive systems that require electricity, restricting the location and number of
3 measurement sites. This results in low spatial resolution of concentration data and inadequate
4 characterization of environmental benzene exposures, particularly for children; hence, a high
5 level of uncertainty is present in risk assessments (e.g. Möller, Schuetzle, & Autrup, 1994).

6 Due to continued lung development, higher breathing rates and activity levels, and more
7 time spent outdoors, children are more susceptible to the health effects of air pollution than
8 adults (American Academy of Pediatrics Committee on Environmental Health, 2004). High
9 concentrations of combustion-related pollutants (including benzene) have been associated with
10 higher incidences of acute respiratory infections in children (Romieu, Samet, Smith, & Bruce,
11 2002). Levels of a benzene metabolite in the urine of school children in Bangkok have been
12 found to be comparable to that of adult street vendors who experienced higher ambient
13 concentrations (Ruchirawat, Settachan, Navasumrit, Tuntawiroon, & Autrup, 2007).
14 Toxicological support documents on benzene carcinogenicity for the U.S. Environmental
15 Protection Agency (USEPA, 1998) suggest that both the type of leukemia and susceptibility
16 differ between children and adults; however, not enough data are available to quantify these
17 differences. Higher resolution data would contribute to the understanding of both exposure and
18 health effect differences.

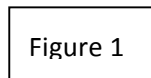
19 To understand children's exposures, concentrations of benzene where children spend
20 time, such as parks and school grounds, are needed. In a review, Mejía et al. (2011) found that
21 most studies on air pollutant exposures at school used data from remote monitoring stations or
22 dispersion modeling. Studies that monitored on school grounds often did not indicate the
23 specific location of measurement. The pollutant focus of previous work has primarily been on

1 NO₂, ozone, SO₂ and particulate matter. Work on spatial variations of benzene in urban areas
2 has investigated impacts of roadway traffic and the relationship between indoor and outdoor
3 concentrations. Thorsson and Eliasson (2006) and Menezes et al. (2009) included measurements
4 of benzene in urban parks, in Sweden and Brazil respectively; they found concentrations in parks
5 to be substantially lower than levels in high traffic areas. Janssen et al. found decreasing outdoor
6 benzene concentrations with distance from the motorway in a study of 24 schools in the
7 Netherlands (Janssen, van Vliet, Aarts, Harssema, & Brunekreef, 2001); indoor concentrations
8 were higher than outdoor concentrations. Godoi et al. (2009) found measured benzene levels to
9 be higher indoors than outdoors in a study of two schools in Brazil. By examining the indoor-
10 outdoor concentration ratio, they concluded that the indoor concentrations were primarily due to
11 outdoor sources. Overall, more measurement data is needed on the spatial variations in benzene
12 concentrations within areas where children may be exposed.

13 Studies of environmental equity investigate the distribution of environmental exposures,
14 risks, and effects among subpopulations of different race or socioeconomic status (Brown, 1995).
15 In studies of multiple urban areas, disadvantaged groups have been found to disproportionately
16 live in neighborhoods with high traffic density and poor air quality (Houston, Wu, Ong, &
17 Winer, 2004; Pastor Jr, Sadd, & Morello-Frosch, 2002; Wheeler & Ben-Shlomo, 2005). In the
18 Tampa area, studies have found that census tracts and elementary schools with higher
19 proportions of black, Hispanic and low income groups are located in areas with higher levels of
20 NO₂, air toxics, and traffic counts (Chakraborty, 2009; Stuart, Mudhasakul, & Sriwatanapongse,
21 2009; Stuart & Zeager, 2011). Using neighborhood-scale (block group) census data, Stuart et al.
22 (2009) also found that these same groups disproportionately reside further from regulatory
23 monitors. In a study of southern California, Houston et al. (2004) concluded that the historic

1 urban sprawl growth pattern resulted in minority and poor neighborhoods located in areas with
2 high traffic density and of more affluent neighborhoods located in suburban areas. Hence, it
3 contributed to a disproportionate burden of health effects on poorer populations. Additionally,
4 urban sprawl is considered a less sustainable form of growth than compact, dense cities, due to
5 increased land use and inefficiency of public transportation (Camagni, Gibelli, & Rigamonti,
6 2002). As cities work to manage growth, high resolution monitoring can guide approaches for
7 maximizing exposure equality and urban sustainability.

8 This manuscript presents a pilot study of small-scale variation in benzene concentrations
9 over a city park, and investigates impacts on the uncertainty of cancer risk estimates, particularly
10 for children. We present the methods used for the sampling and analysis of benzene, method
11 evaluation, and data analyses. Results are then discussed on ambient benzene levels, their spatial
12 variation, and the uncertainty in cancer risk estimates introduced by sampler placement.

13  Figure 1

14 **Methods**

15 A city park in Hillsborough County, Florida was chosen as the field site for this pilot study. The
16 park is located adjacent to an elementary school and shares a structured playground with the
17 school. We chose eleven sampling sites with available standing structures for sampler
18 placement (a utility pole or tree), with the goal of spanning the park with approximately
19 equidistant placement. For mapping and analysis, location coordinates were determined using a
20 Garmin GPS device (with accuracy listed as within 10 m). Figure 1a shows the study area and
21 sampling locations. For evaluation of measurement accuracy, we collocated an additional
22 sampler with the only benzene regulatory monitor for the county, operated by the Environmental
23 Protection Commission (EPC) of Hillsborough County; this site is outside the park.

1 To characterize ambient benzene, we exposed passive diffusive samplers [RAD 130,
2 Radiello, Ltd., activated charcoal sorbent] for a one week sampling period (April 27 to May 4,
3 2011) at the sites discussed above. A duplicate sampler was also exposed at site 8 for assessment
4 of measurement precision. Two field blank cartridges accompanied the exposed samplers (one
5 during deployment and the other during retrieval). Each field blank cartridge was uncapped and
6 immediately resealed at one sampling location, as specified by the USEPA (1999b). Upon return
7 to the laboratory, cartridges were stored at 4°C. Each cartridge was submersed for thirty minutes
8 in 2 ml of carbon disulfide [Sigma-Aldrich, 342270-ReagentPlus, low benzene] for extraction.
9 One laboratory blank cartridge was also used for quality control purposes. We quantified
10 benzene via gas chromatography with mass spectrometry (GC/MS) [Varian Saturn 3800-GC,
11 2000-MS] using a polysiloxane capillary column [Varian CP-Sil 8 CB,
12 50 m x 0.25 mm x 0.25 µm]. A 1.0 µl aliquot of the extracted solution was injected on the
13 column at a temperature of 240°C, with helium as the carrier gas at a flow rate of 1.2 ml min⁻¹.
14 The total run time was sixty minutes with the following temperature program: start at 35°C for
15 nine minutes, increase at 5°C min⁻¹ to 60°C, hold at 60°C for 46 minutes. To quantify unknown
16 concentrations, we diluted benzene stock [Fluka, 12540-Analytical Standard] to create five
17 calibration standards ranging from 0.10-1.75 µg ml⁻¹. We used daily calibration curves, with a
18 daily control chart for quality control. All samples and calibration standards were normalized by
19 addition of 2-fluorotoluene [Sigma-Aldrich, F15323] as an internal standard. The analysis
20 protocol was adapted for our specific equipment from the Radiello sampler manual (Fondazione
21 Salvatore Maugeri, 2006), previous studies (Angiuli et al., 2003; Cocheo, Boaretto, & Sacco,
22 1996; Godoi et al., 2009), and established standard methods (USEPA, 1999a; USEPA, 1999b).

1 Quality control procedures and metrics were adapted from USEPA Compendium Method TO-15
2 (USEPA, 1999a).

3 We converted to concentrations in air by subtracting the mean blank value and
4 multiplying the blank-adjusted values by a temperature-adjusted sampling rate and the exposure
5 time recorded for each sampler. We used the sampling-period average of hourly temperatures
6 measured at the Tampa International Airport (National Weather Service KTPA station) to adjust
7 the sampling rate (Fondazione Salvatore Maugeri, 2006). The limit of detection was calculated
8 as three times the standard deviation of the blank values, assuming a one week sampling time
9 (10080 minutes).

10 To characterize spatial variations in benzene concentrations over the study area, we
11 mapped the concentration distribution in ArcGIS, followed by spatial interpolation using kriging.
12 Summary statistics, including the coefficient of variation (CV), were used to quantify variation.

13 To investigate the health risks associated with the concentrations measured here, we
14 estimated excess lifetime cancer risk from inhalation exposure by multiplying the exposure
15 concentration (C_E) by the inhalation unit risk (IUR) for benzene. The exposure concentration
16 was calculated as $C_E = C_a t_E f_E d_E / T$ (USEPA, 2009), where C_a is the measured concentration of
17 benzene, t_E is the exposure time (hours day⁻¹), f_E is the exposure frequency (days year⁻¹), d_E is the
18 exposure duration (years), and T is the averaging time (hours). We chose exposure parameters to
19 represent a child who is exposed due to time spent at school, with t_E of 6.5 hours day⁻¹, f_E of 180
20 days year⁻¹, and d_E of 6 years (representing kindergarten through 5th grade) (Hillsborough
21 County Public Schools, 2010; 2011). For T , we applied the value for excess lifetime cancer risk,
22 of 70 years (in units of hours) (USEPA, 2009). The inhalation unit risk represents the upper
23 bound excess cancer risk per unit of exposure concentration determined through review of

1 human and animal studies. USEPA lists a range of 2.2×10^{-6} to 7.8×10^{-6} per $\mu\text{g} / \text{m}^3$ (USEPA,
2 2010), while the California Office of Environmental Health Hazard Assessment lists a value of
3 2.9×10^{-5} per $\mu\text{g} / \text{m}^3$ (OEHHA, 2009). For this work, we estimated risks using the minimum and
4 maximum of these values, i.e. 2.2×10^{-6} and 2.9×10^{-5} . These were combined with the range of
5 measured concentrations to estimate a range of risks.

6 Finally, we investigated the impact of sampler placement (and sampling resolution) on
7 the uncertainty of health risks calculated using the standard methods discussed above. Since
8 actual levels vary over the area represented by a single sample, the choice of sampler location
9 leads to uncertainty in the exposure concentration. When propagated through the risk
10 calculation, proportional uncertainty in the estimated health risk results. Uncertainty in the
11 inhalation unit risk has a similar effect. Hence, we compared the range of measured
12 concentrations (quantified as a relative percent difference) over the study area to that of the
13 inhalation unit risk, in order to assess impacts of sampler placement on health risk assessment.

14

Table 1

15 **Results and Discussion**

16 Measured benzene concentrations and summary statistics for the study area are shown in Table
17 1. Results are at the low end of the range in weekly outdoor concentrations ($0.3\text{-}5.0 \mu\text{g m}^{-3}$)
18 observed at schools in the Netherlands (Janssen et al., 2001) and weekly levels ($0.3\text{-}1.4 \mu\text{g m}^{-3}$)
19 for an urban park in Gothenburg, Sweden (Thorsson and Eliasson, 2006; Figure 5 therein). This
20 is consistent with previous work showing low concentrations in urban parks as compared with
21 levels in high traffic areas (Upmanis, Eliasson, & Andersson-Skold, 2001). Overall, the values
22 measured here are near the low end of the general range of benzene measured in U.S. studies, of

1 approximately 1-10 $\mu\text{g m}^{-3}$, with peak values up to about 50 $\mu\text{g m}^{-3}$ (Health Effects Institute Air
2 Toxics Review Panel, 2007).

3 The overall range of estimated excess cancer risk for the range of concentrations
4 measured here is from 5.7×10^{-9} to 1.1×10^{-7} . These risks are quite low when compared with other
5 studies (Tam & Neumann, 2004; Payne-Sturges, Burke, Breysse, Diener-West, & Buckley,
6 2004; McCarthy et al., 2009), which reported cancer risk due to benzene of greater than 10^{-6} at
7 almost all sites. Here, we considered only the contribution to the lifetime cancer risk due to
8 elementary school exposure, not an exposure duration of an entire lifetime. Further, we note that
9 the concentrations here are for one week of sampling, and hence, may not be representative of
10 longer-term averages or extremes.

11 Evaluation results indicate a precision from the two collocated duplicate samplers of 14%
12 (as a relative percent difference), which is within the recommended guideline of less than 25%
13 (USEPA, 1999a). A 3% relative percent difference was found between the concentration
14 measured by the reference regulatory monitor and that measured by a collocated passive
15 sampler; this is within the precision of the passive measurement (that ultimately limits the
16 measurable accuracy). We note that the comparison is not direct, since the sampling times are
17 different. The reference monitor takes 24 hour canister samples every six days; the final 13
18 hours of one sampling period and the full second sampling period of the reference monitor
19 overlapped with the sampling period for this study.

20 The spatially-interpolated concentration field is provided in Figure 1b. A gradient can
21 be observed; the highest interpolated concentrations are in the northwest corner of the sampling
22 area, which includes the site near the playground structure (site 2). The highest point
23 measurement value was at site 3. The location of the high area is consistent with emissions

1 expected from the parking lot and vehicular park entrance nearby. Wind data for the Tampa area
2 (at the international airport) do not indicate a dominant wind direction during the study period.
3 However, only 12% of the winds were from the WNW to N quadrant, which may have kept
4 concentrations low overall. It should be noted that, due to the precision of measurement,
5 confidence in real differences is small. Overall, the coefficient of variation for the sampling
6 area was only 11%. Although there is no standard metric for quantifying spatial heterogeneity, a
7 value of 20% has been used to distinguish between a homogeneous and heterogeneous field in
8 previous studies of particulate matter (Blanchard et al., 1999; Wilson, Kingham, Pearce, &
9 Sturman, 2005). The appropriateness of this criteria for a benzene field is unknown, but is
10 explored below.

11 The range of measured concentration values can be used to explore impacts of sampler
12 placement on uncertainties in the estimated health risks of benzene. The relative percent
13 difference between the maximum and minimum measured concentrations was 39%; this results
14 in an equivalent percent difference in estimated excess health risks (if all other parameters are
15 kept constant). As a comparison, a similar value (44%) can be calculated from the range of risks
16 due to toxic air pollutant levels (driven primarily by formaldehyde and benzene) measured at
17 four sites in a study of intra-urban differences in Pittsburgh (Logue, Small, Stern, Maranche, &
18 Robinson, 2010). Conversely, the range of values for inhalation unit risk listed by the USEPA
19 and California OEHHA is much larger, at 170% (as relative percent difference). (Note that this
20 does not represent the full range of risks, just a range of upper bound risk estimates). Hence, the
21 uncertainties associated with sampler placement in this study are small compared with those
22 from the unit risk parameter; the variation in concentration over the sampling area would need to
23 be much larger to have a similar independent effect. This suggests a threshold to the utility of

1 increased spatial resolution of measurement that will depend on the purpose for which the
2 concentration measurements are used. However, we note that the effect on overall risk
3 uncertainty is multiplicative, so even small variations in concentrations could lead to large
4 overall uncertainties in risk. Further, there are uncertainties in other parameters in the
5 calculation that are not analyzed here (e.g. personal activity patterns).

6 Although of limited utility for risk estimation at the very high resolution studied here,
7 monitoring data that characterizes intra-urban variations may be helpful for epidemiological
8 studies, development of better risk assessment values, and city planning. While considering the
9 effects of misclassification of particulate matter exposure on inference from time-series mortality
10 studies, Zeger et al. (2000) found that the largest bias was due to differences between the
11 ambient levels measured by fixed-site monitors and average individual exposures. Spatially-
12 resolved outdoor benzene concentrations may help reduce this bias, particularly given the
13 association between indoor and outdoor concentrations of benzene (Godoi et al., 2009). In
14 combination with personal activity data, higher-resolution concentration data may allow better
15 estimation of individual exposures (Nuckols, Ward, & Jarup, 2004; Whitworth, Symanski, Lai,
16 & Coker, 2011). However, the usefulness of additional increases in spatial resolution may
17 currently be limited both by other uncertainties in risk estimation and the precision of passive
18 measurements. Nonetheless, decreased misclassification of exposure in health effect studies may
19 in turn lower the uncertainty in health risk assessment parameters. This is particularly important
20 for children, as current USEPA risk assessment methods do not differential risk to children for
21 benzene carcinogenicity. Finally, Jensen et al. (2001) suggest that high-resolution concentration
22 data may aid city planners to build sustainability cities and reduce environmental inequity. For

1 these purposes, characterization of concentrations variations over space may be as important than
2 differences in individual activities and exposures.

3

4 **Conclusions**

5 Eleven sampling locations were chosen at a city park to investigate spatial variation in ambient
6 benzene using passive samplers. Precision and accuracy were evaluated through co-location.
7 Excess lifetime cancer risks were estimated, and the magnitude of uncertainty due to sampler
8 placement was compared to the uncertainty due to inhalation unit risk. The precision and
9 accuracy were measured at 14% and 3%, respectively. The spatial variation over the park was
10 found to be low (*CV* of 11%), with an overall range, as a relative percent difference, of 39%.
11 Comparison suggests limits to the use of these methods for very high resolution sampling, but
12 appropriateness for study of larger-scale intra-urban concentration variations of benzene in
13 Hillsborough County.

14 Compared with the range of inhalation unit risk of 170% (as a relative percent
15 difference), these results indicate that one sampler may be sufficient to represent the excess
16 lifetime cancer risk from benzene exposure for the sampling area, using currently recommended
17 methods. Further research is needed to characterize the spatial variation in benzene
18 concentrations and risks over larger intra-urban scales, including neighborhoods. The necessary
19 monitoring resolution will depend on the purpose for which the data will be applied. High-
20 resolution data may lower errors in epidemiological studies resulting from differences between
21 actual and measured concentrations, may improve risk assessment parameters (particularly for
22 less-characterized groups, like children), and may aid city design.

23

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16

17 **Figure Captions**

18 **Figure 1** a) The study area with sampling locations (and corresponding sample IDs) and b) a
19 contour plot of interpolated concentrations ($\mu\text{g}/\text{m}^3$) [generated in ArcGIS]. The white box in a)
20 corresponds to the area plotted in b). Sample ID 12 (not shown) was located at the site of the
21 county reference monitor, outside of the study area. A duplicate sampler was located at site 8.

Tables

Table 1 Benzene concentrations and summary statistics.

Sample ID (or summary statistic)	Concentration ($\mu\text{g m}^{-3}$)
1	0.33
2	0.33
3	0.34
4	0.28
5	0.31
6	0.27
7	0.29
8	0.29
8* (duplicate)	0.34
9	0.29
10	0.23
11	0.31
12	0.26
reference monitor	0.26
mean	0.30
standard deviation	0.03
range	0.23 - 0.34

The limit of detection was $0.18 \mu\text{g}/\text{m}^3$. Sample ID locations are provided in Figure 1. Summary statistics are based on measurements from sites in the study area (1-11), excluding the site 8 duplicate value. The reference monitor value is the average of two 24-hr sample values (from April 27 and May 3) obtained from the Environmental Protection Commission of Hillsborough Co. from every 6th day monitoring during the study period. Reference-method samples are collected in 6 L stainless steel canisters and analyzed by GC/MS, per Compendium Method TO-15 (USEPA, 1999a). All calculations were performed using full precision in Excel; hence summary values cannot necessarily be reproduced from the concentration values reported to two significant digits.

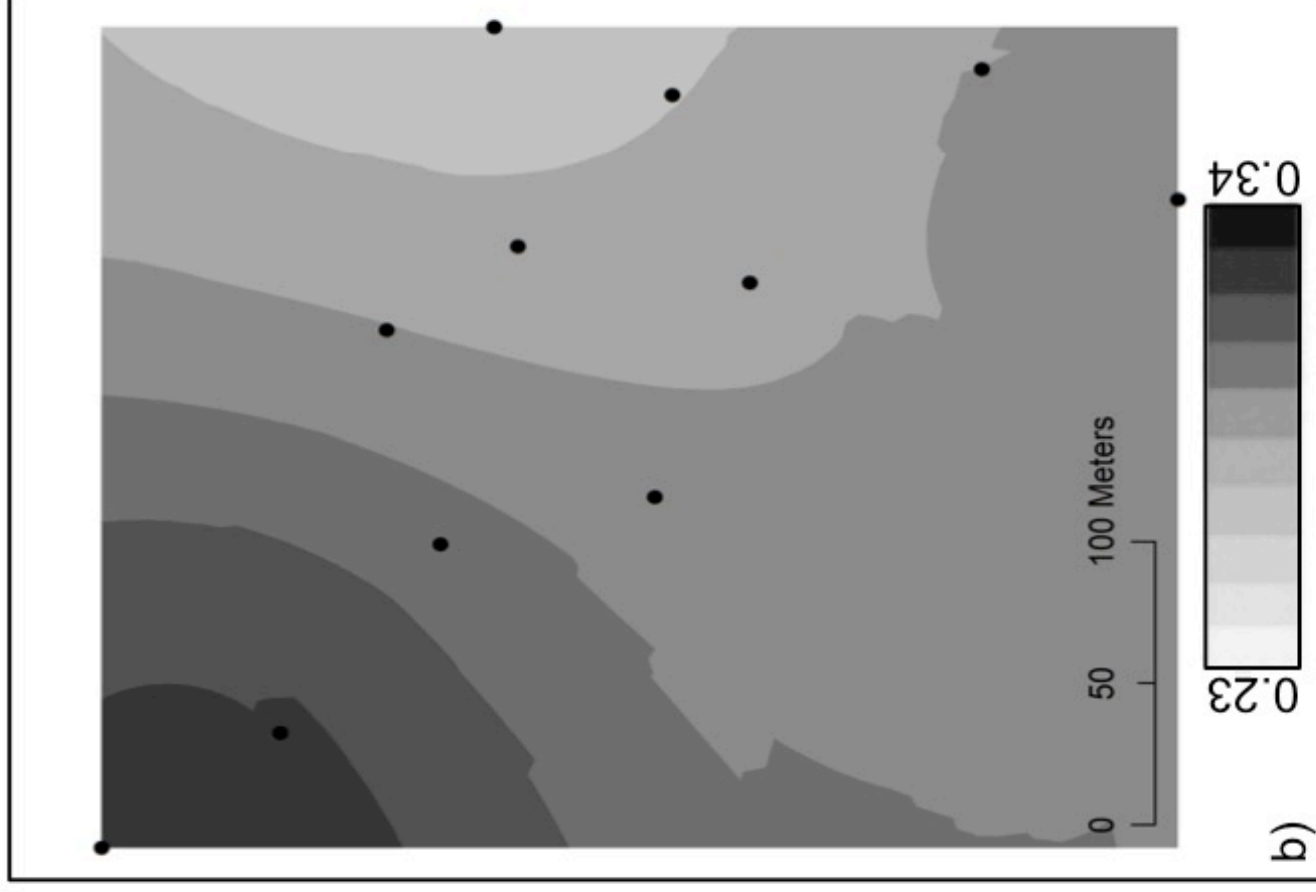


Figure 1. (Caption text provided with the manuscript text)