

1 **An evaluation of EPA's National-Scale Air Toxics Assessment (NATA): comparison with benzene**
2 **measurements in Detroit, Michigan**

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- 32
- 33 Ambient concentration
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- 35 Personal exposure
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- 37
- 38

39 **Abstract**

40

41 The U.S. EPA periodically evaluates ambient concentrations, human exposures, and health risks for 180

42 hazardous air pollutants plus diesel particulate matter using modeled estimates from the National-Scale

43 Air Toxics Assessment (NATA). NATA publishes estimates at the spatial resolution of U.S. Census

44 tracts, which are subdivisions of a county. These local scale, model-predicted estimates from NATA are

45 used extensively in community-based assessments; however, evaluation of NATA's ambient

46 concentrations and human exposure estimates against measurement data has been limited to date. This

47 paper compares modeled annual average benzene results from the 2002 NATA with measured results

48 from the 2004-2007 Detroit Exposure and Aerosol Research Study (DEARS) as a case study of the quality

49 of NATA results. NATA model estimates support community-scale characterization and assessment.

50 Benzene is particularly important as it was estimated by the 2002 NATA as the largest single air toxic

51 pollutant in terms of cancer risk in the U.S. We found that the average ambient concentrations of benzene

52 predicted by NATA were within 5 percent, on average, of the 24-hour integrated average ambient

53 concentrations measured in DEARS. The NATA human exposure estimates, which include only outdoor

54 sources for benzene, were, on average, approximately half the measured breathing zone concentrations

55 from DEARS. Our analyses support that the factors driving higher DEARS personal benzene

56 concentrations relative to the NATA predicted exposure values are likely due, at least in part, to indoor

57 sources. This work points to further community-scale modeling research to improve characterizations and

58 assessments of human exposures.

59

60 **1. Introduction**

61

62 The National-Scale Air Toxics Assessment (NATA), U.S. EPA's periodic evaluation of air toxics since

63 1996, models ambient concentrations, human inhalation exposures, and health risks for 180 hazardous air

64 pollutants plus diesel particulate matter across the U.S. at a fine geographic scale. It is very popular for
65 community-based environmental assessments by governments, universities, community groups, and the
66 public at large. NATA publishes results at the spatial resolution of U.S. Census tracts, the small and
67 relatively permanent statistical subdivisions of a county that usually contain between 2,500 and 8,000
68 persons (http://www.census.gov/geo/www/cen_tract.html). Given the public interest in NATA, it is
69 important to assess the quality of its modeled estimates of ambient air toxics concentrations and human
70 exposure. Although some work on evaluating the NATA model and estimates has been reported (Lupo
71 and Symanski 2009, Ozkaynak et al. 2008, US EPA 2005, Payne-Sturges et al. 2004), the Detroit
72 Exposure Aerosol Research Study (DEARS) (Williams et al., 2009) provides an extensive data source for
73 a local evaluation; we believe our evaluation of the human exposure estimates is the most intensive such
74 study to date. Local or community-scale characterizations and assessments are of interest for identifying
75 disproportionate impacts and environmental injustices, particularly when the associated health risks are
76 large. In addition to its use as an air toxics tool, NATA is also a template for other research outside of air
77 toxics (Zartarian and Schultz, 2010) in support of community-based and environmental justice
78 assessments.

79
80 NATA model estimates are derived from a variety of information types including meteorological data,
81 emissions inventory data, monitoring data, and modeled point and nonpoint/mobile source data
82 (<http://www.epa.gov/ttn/atw/nata2002/methods.html>). In particular, NATA uses meteorological data from
83 the National Weather Service, monitoring data from, among others, EPA's Air Quality System (AQS),
84 and data from several EPA models/inventories: National Emissions Inventory (NEI;
85 <http://www.epa.gov/ttn/chief/eiinformation.html>), Assessment System for Population Exposure
86 Nationwide (ASPEN; <http://www.epa.gov/ttn/atw/nata/aspem.html>), Human Exposure Model (HEM;
87 http://www.epa.gov/ttn/fera/human_hem.html), and Hazardous Air Pollutant Exposure Model version 5
88 (HAPEM; http://www.epa.gov/ttn/fera/human_hapem.html). NATA's background ambient concentration
89 results for benzene are based on AQS and other monitoring data
90 (http://www.epa.gov/ttn/atw/nata2002/02pdfs/background_conc_county.pdf), with modeled ambient
91 concentration results for area and mobile source emissions from ASPEN and for point source emissions
92 from HEM. NATA's human inhalation exposure results are from HAPEM, which uses ASPEN and HEM
93 dispersion-modeled concentrations as input data. These model results are combined with available unit
94 risk estimates and inhalation reference concentrations to yield NATA estimates for the risk of cancer and
95 other serious health effects from chronic exposure to air toxics from outdoor sources. NATA provides a

96 snapshot of the outdoor air quality and the risks to human health that would result if air toxic emissions
97 levels remained unchanged (<http://www.epa.gov/ttn/atw/nata2002/natafaq.html#A5>).

98

99 Our retrospective evaluation compares 2002 NATA model-predicted estimates for ambient concentrations
100 and human inhalation exposures to monitor measurements from the 2004-2007 Detroit Exposure Aerosol
101 Research Study (DEARS). This work is a case study for benzene, which was estimated by the 2002
102 NATA as the largest single air toxics pollutant contributor in terms of cancer risk in the U.S.
103 (<http://www.epa.gov/ttn/atw/natamain/>).

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105

106 **2. Methods**

107

108 2.1. Background

109

110 DEARS measurement data comprise monitoring results from the Michigan Department of Natural
111 Resources' Allen Park ambient monitoring site, from outside at participants' residences, from inside
112 participants' residences, and from personal monitors worn by participants. DEARS samples were
113 collected five days a week, always Tuesday through Saturday, for seven weeks each over three summers
114 and three winters in the Detroit area. Measurements were taken for a particular participant during one
115 week in either a summer or winter season, or both for some participants; see Williams et al. 2009 for
116 further details on the study design.

117

118 The spatial resolution of this evaluation is a census block, defined by the U.S. Census as a subdivision of a
119 census tract. A census block is generally small in area, for example, a city block bounded by streets.

120 NATA's census tract-level data are public-use; however, the even finer scale, census-block-level NATA
121 results used in this analysis were developed for this comparison and are not publicly available. The spatial
122 coverage of this evaluation is the 99 census blocks where DEARS measurements were taken. NATA
123 model estimates predict ambient concentrations and human exposures at the geographic center of census
124 blocks. DEARS ambient samples were taken outside participants' residences and at the Allen Park
125 ambient monitoring site. DEARS human exposures were estimated by measuring concentrations from
126 monitors in the personal breathing zone on a vest worn by the participant.

127

128 While the spatial characteristics of the NATA and DEARS data used in this evaluation were similar, the
129 temporal characteristics were not as similar. The temporal scale for both is 24-hour integrated averages;
130 however, the NATA estimates represent an annual average in 2002 while the DEARS results represent
131 daily averages in the 2004-2007 timeframe. Thus, our evaluation is most relevant for describing how well
132 the long-term, annual average may represent sub-annual averages of daily measurements. We interpret our
133 findings with the focus on the overall average comparison.

134 135 2.2. DEARS Sample Collection and Benzene Measurement

136
137 The locations where DEARS participating residences were selected included a background site in
138 Belleville (west of Detroit) and several areas in Detroit, including along the Southfield Freeway and an
139 industrial area near the Ambassador Bridge into Canada. Allen Park was selected as the DEARS central-
140 site monitor location, in part, because it is a permanent, Michigan facility not overly influenced by nearby
141 sources.

142
143 DEARS volatile organic compound (VOC) measurement data, including benzene, were collected by
144 Carbopack-X passive samplers (McClenny et al. 2006, McClenny et al. 2005). This sampling method is
145 reported by McClenny et al. 2005 to potentially have high benzene blank measurements that are
146 attenuated by sufficient preconditioning of the samplers. The samplers used for DEARS were
147 preconditioned, and the benzene data were assessed to assure background levels were appropriate.
148 Approximately 80% of the DEARS benzene measurements ($\mu\text{g}/\text{m}^3$) were above the minimum detection
149 limit (mean 0.5, SD 0.7).

150
151 Fresh passive samplers were deployed daily at Allen Park, outside residences, inside residences, and in the
152 personal breathing zone. The personal breathing zone samplers were clipped on vests which were worn by
153 participants except when they were sleeping, at which time the vests were kept in the sleeping area. The
154 samplers deployed outside, including at Allen Park, or indoors were clipped to sampling stands. All
155 sampling was done by passive collection with the tube's open path end pointed downward. Samplers were
156 analyzed using a multistep automated analytical process. The samplers were initially prepared by purging
157 with dry helium to remove water vapor that could potentially interfere with the analysis. This was
158 followed by thermal desorption of the VOCs from the samplers while using a helium gas stream to
159 transport the desorbed VOCs to a small volume intermediate sorbent trap used to concentrate the

160 compounds and prepare them for introduction to the GC/MS system. Rapid heating of this intermediate
161 trap released the captured VOCs to the GC/MS system for separation, detection, and quantitation.

162

163 2.3. NATA-DEARS Comparison

164

165 Our comparison uses ratios of means for DEARS-measured 24-hour integrated average concentrations to
166 census-block-level NATA-modeled 24-hour integrated average concentrations. The numerator of each
167 ratio is the census-block-level mean concentration for a particular DEARS season, and the denominator is
168 the census-block-level NATA 2002 annual mean concentration. The denominators are constant across the
169 seasons except where the census blocks represented in the DEARS data change as a result of household
170 participation patterns in DEARS. Approximately two-thirds of the DEARS block-level mean
171 concentrations for a particular season represents a single household in a single week, and these means are
172 compared to the corresponding annual mean concentration from NATA.

173

174 Measurement data from samples taken at the DEARS central-site monitor at Allen Park were analyzed
175 with the other outdoor measurements and together compared to NATA-modeled ambient concentrations.
176 DEARS breathing-zone personal monitoring data were compared to NATA-modeled human exposures.
177 Box plots are used to summarize these ratios for each DEARS season. Correlation statistics and regression
178 models are used in additional analyses.

179

180 **3. Results**

181

182 Table 1 presents tract-level DEARS and NATA mean benzene concentrations for all seasons
183 combined. The relatively large number of data values in the DEARS means comes from the day-specific
184 measurements while the NATA means reflect a single model-predicted value per block. This descriptive
185 summary of the data indicates that the mean concentrations for the outdoor measurements are more
186 similar across the two studies than are mean personal concentrations for the human exposure
187 measurements. Each of the DEARS mean concentrations for indoor measurements is lower than the
188 corresponding mean concentrations for personal measurements. The standard errors for the NATA mean
189 concentrations are small (< 0.1), indicating limited ranges for these modeled values within a tract.

190

191 3.1. Outdoor concentration estimates for residences and Allen Park

192

193 The overall mean of the DEARS to NATA ratios for benzene outdoor concentrations is 0.95, and
194 the means of the summer ratios are higher than those for winter (1.7, 1.0, and 1.0 versus 0.8, 0.7, and 0.6,
195 respectively). The ratios range from close to zero to nearly three, although following DEARS Summer
196 2004, the ratios do not exceed two. Figure 1 shows box plots for these ratios (R Development Core Team
197 2007) where the box indicates the interquartile range. There are seasonal differences in the ratios, and
198 most of this variability comes from DEARS measurements since the denominators from NATA
199 repeatedly use the 2002 modeled values (Table 2). The overall mean of the DEARS block-level means
200 was $2.5 \mu\text{g}/\text{m}^3$ with standard error (SE) of 0.1.

201

202 3.2. Human exposure estimates

203

204 The overall mean of the DEARS to NATA ratios for human exposure to benzene is 2.07, and the
205 means of the summer ratios are higher than those for those for winter (3.1, 2.1, and 2.1 versus 1.6, 1.7,
206 and 1.8, respectively). There is more variability in the range of block-season specific ratios for personal
207 measurements than in the outdoor data. The ratios of means from DEARS personal monitoring and NATA
208 personal estimates range from close to zero to more than nine, with greater variability after DEARS
209 Winter 2005 (Figure 2, left panel). As with the outdoor data there are seasonal differences in the personal
210 ratios, and most of this variability again comes from DEARS measurements since the denominators from
211 NATA repeatedly use the 2002 modeled values (Table 3). The overall mean of the DEARS block-level
212 means was $5.5 \mu\text{g}/\text{m}^3$ with SE 0.3.

213 To gain a better understanding of the personal monitor measurements from DEARS, we repeated
214 the analysis replacing DEARS personal data with DEARS indoor data. Figure 2 shows the resulting box
215 plots for these ratios in the right panel. The overall mean of the DEARS to NATA ratios for benzene
216 indoors is 1.64, lower than the 2.07 for personal benzene but still reflecting NATA modeled values being
217 lower than DEARS measured concentrations on average.

218 DEARS personal monitors traveled with the wearer, including when the individual traveled
219 beyond the home census block, as they were intended to estimate personal exposure. We repeated the
220 analyses above using DEARS personal and indoor data for the numerators but restricting to cases where
221 the DEARS participant reported staying inside the home all day. Figure 3 gives the resulting box plots,
222 and we note that count of ratios is small, for example, with only 3 ratios in Summer 2004. The patterns are
223 choppy than those in Figure 2, but the story is consistent with that from the unrestricted case. NATA
224 underestimates human exposures, by an average of roughly a third to just under a half, based on DEARS

225 indoor and personal measurements, respectively, and the underestimation is likely due to not adjusting for
226 benzene from indoor sources. By design, NATA incorporates only concentrations from outdoor sources.

227 228 3.3. Additional analyses of indoor benzene exposure estimates

229
230 A potential indoor source of benzene could include gasoline vapors from attached garages
231 although DEARS included few homes with attached garages. Another potential indoor source of benzene
232 in DEARS measurements is environmental tobacco smoke (ETS) even though all participants self-
233 reported to be nonsmokers in nonsmoking households. We examined these and other participant
234 questionnaire responses for possible indoor sources in a descriptive analysis in order to better characterize
235 the large ratios for personal exposure to benzene. Table 4 gives percentages for the top quartile of the
236 ratios versus the percentages for the lower three quartiles of ratios combined, that is, above and below the
237 75th percentile, respectively, for summer and winter separately. The percentages shown are for
238 yes/positive responses to dichotomous questions. For example, 5.9% of those summer participants in the
239 top quartile of ratios reported that they had been around environmental tobacco smoke in their home. This
240 5.9% and each of the other table values reflect multiple responses from each participant (up to five
241 responses per season). Similarly we give results for a composite variable indicating there is a gasoline-
242 powered lawnmower, string trimmer, leaf blower, chain saw, and/or other small engine stored in the
243 garage. Of note, for more households in the top quartile on winter days, respondents reported being close
244 to ETS than did those in the lower quartiles (25.9% versus 10.3, respectively). This pattern reverses what
245 was seen for the summer days where respondents reported being near ETS less of the time for the top
246 quartile (14.7% versus 24.4%). Also, the percentage of residences with small gasoline-powered equipment
247 stored in the garage was higher for those in the top quartile for both summer and winter (24.1% versus
248 14.5% for summer and 20.0% versus 12.3% for winter).

249 For benzene concentrations that were associated with the top quartile of DEARS to NATA ratios,
250 we plotted log-transformed personal measurement concentrations from DEARS against the personal
251 measurement concentrations for toluene and for 1,3-butadiene, usual co-pollutants for benzene; see Figure
252 4. We also plotted log-transformed personal measurement concentrations from DEARS against the
253 personal measurement for an ETS tracer (Williams et al. 2009; Lawless et al., 2004) and against outdoor
254 benzene concentrations. We calculated Spearman correlation coefficients (ρ) for pairings of these
255 variables for the top quartile of DEARS to NATA ratios. The plots and correlation coefficients include
256 multiple days of concentration measurements for DEARS participants corresponding to the DEARS study
257 design. Statistically significant (p -value < 0.05) correlations were seen for log-transformed concentrations

258 for personal benzene with personal toluene ($\rho=0.76$ summer, $\rho=0.73$ winter), personal 1,3-butadiene
259 ($\rho=0.41$ summer, $\rho=0.19$ winter), and outdoor benzene ($\rho=0.33$ summer, $\rho=0.24$ winter).

260 We wanted to assess the correlations taking into account the repeated measures for participants,
261 which were not accounted for in the Spearman correlation coefficients. We performed regression analysis
262 in SAS MIXED to assess the correlations of log-transformed personal benzene concentrations with log-
263 transformed concentrations for personal toluene, personal 1,3-butadiene, personal ETS tracer, and outdoor
264 benzene, taking into account the repeated measures for participants, again for the top quartile of ratios
265 separately for summer and winter seasons in DEARS (SAS 2004). We found that the relationships for
266 personal toluene and outdoor benzene reflect significant correlations with personal benzene in summer
267 with p-values <0.05 , differing from the Spearman correlations where only the ETS tracer was not
268 significant. For winter, we found all except 1,3-butadiene to reflect significant correlations, differing from
269 the Spearman correlations which found the ETS tracer to not be significant.

270 We also performed regression analysis in SAS MIXED for an explanatory model for the personal
271 benzene concentrations from DEARS. We modeled log-transformed personal benzene concentrations by
272 regressing it on log-transformed outdoor benzene, the ETS tracer, and the gasoline-powered small engine
273 variable. This model accounted for the repeated measures for participants. We performed the regression
274 analysis on the top quartile of ratios separately for summer and winter seasons of DEARS. The outdoor
275 benzene and small engine variables were statistically significant for the summer model, and outdoor
276 benzene and the ETS tracer were statistically significant for the winter model with p-values <0.05 .
277 DEARS is a relatively small, mostly non-smoking, few-attached-garages study, so there are limited self-
278 response data to substantively parse the various factors which may combine as indoor-sourced benzene.
279 These regression findings, however, strongly suggest that the DEARS personal measurements include
280 benzene from indoor sources.

281

282 3.4. Assessment of the seasonal pattern in DEARS concentrations

283

284 The seasonal pattern seen in the outdoor and personal DEARS to NATA ratios, with summers
285 higher than winters, reflects the DEARS measurements (Tables 2 and 3), not the NATA estimates, which
286 are annual averages. Reports in the literature indicate higher benzene concentrations occur in winter
287 (Stocco et al. 2008, Luecken et al. 2006, Battelle 2003), so it is an unanticipated finding that the season-
288 specific means for DEARS reflect seasonal patterns with summers usually higher than adjacent winters.
289 These seasonal patterns may be, at least in part, a local effect in the DEARS concentrations. We assessed
290 the outdoor pattern by comparing the DEARS means with those reported for other studies and by

291 analyzing benzene data collected by Michigan's Department of Natural Resources at monitors in the
292 Detroit area during the DEARS study period.

293 Luecken et al. (2006) simulated five hazardous air pollutants (HAPs), including benzene, for the
294 continental United States for 2001 using the community multi-scale air quality (CMAQ) modeling system.
295 Their CMAQ model-predicted summer concentrations of benzene were lower than the model-predicted
296 winter concentrations. Luecken et al. attributed this pattern to characteristics of winter: lower benzene
297 decay rates, higher benzene emissions, and lower atmospheric boundary layer heights. They compared
298 their model-predicted concentrations with measured data from the 2001-2002 Air Toxics Pilot Study
299 (Battelle 2003) and noted a relatively large difference in HAP measurements among monitors within a
300 single CMAQ chemical transport model grid in Detroit.

301 The Air Toxics Pilot Study measured benzene and other HAPs at 35 monitors in eight cities across
302 the United States, including Detroit, and focused, in part, on the seasonal effect for benzene in Detroit MI,
303 Providence RI, Seattle WA, and Tampa Bay FL. Battelle (2003) summarized the degree of seasonality
304 across the cities to be related to temperature, sunlight, atmospheric inversion heights, population
305 migration, and gasoline formulation. They reported almost no seasonal effect in Detroit and attributed this
306 lack to the large site-to-site variability in Detroit where, for example, the Yellow Freight monitor had very
307 high levels of benzene and the Houghton Lake monitor had very low levels. At Providence, for example,
308 where there are 5 monitors in a single model grid, they found the concentrations differed by up to a factor
309 of 2 while in Detroit the concentrations differed by up to a factor of 3 in a grid.

310 In a 2005 study in Windsor, Canada, Stocco et al. (2008) did not find a statistically significant
311 difference between the geometric means for winter (0.97) and summer (0.79); however, the mean for
312 winter was higher. Windsor is across the Detroit River from Detroit, and their study was performed during
313 the DEARS study period. The Stocco et al. findings suggest that the Detroit seasonal benzene pattern
314 during 2004-2005 was not widespread.

315 To better characterize the seasonal pattern for benzene in Detroit during 2004-2007, we analyzed
316 additional Detroit-area benzene data provided by the Michigan Department of Natural Resources (DNR).
317 These data were measured during the same time frame as DEARS but were not part of that study. We
318 found a mix of winter-summer patterns when comparing sites and when comparing methods for a
319 particular site in these 2004-2007 benzene concentrations, although the number of samples from a single
320 site in the 3-month seasons of June-August and January-March may be too small for patterns to be
321 reliable. The DNR data were from ambient air monitors at Allen Park, Southwest High School, South
322 Delray, Dearborn, Ypsilanti, and Houghton Lake sites; the sampling used EPA Compendium Methods
323 TO-15 SUMMA canister and TO-14A sub-ambient canister (U.S. EPA 1999a and 1999b, respectively). In

324 addition to examining patterns across the sites, we compared DEARS means with means from DNR
325 monitors located at the same site or in the immediate neighborhood. With the exception of DEARS
326 measurements at Allen Park, there is spatial uncertainty in this comparison resulting from the DEARS
327 design where residential monitor locations mostly differ from one week to another and from one season to
328 the next. There is also temporal uncertainty since the 24-hour sampling periods differed with DNR
329 sampling starting at midnight and DEARS sampling starting at approximately 9:00 am. Our analysis of
330 the DNR data included 283 samples, but only 13 of these were from Allen Park. We focused our
331 comparison on Allen Park and the neighborhood of Southwestern High School because the DEARS and
332 DNR monitor sites were in closer proximity than were other sites. We found patterns in the DNR means
333 that were consistent with the patterns in means from DEARS (Table 5). We note that the DNR data for
334 Allen Park were only collected in summer and winter 2005. We also note that consistency of patterns is
335 seen in both comparisons despite differences in sampling methods and in sampling periods, in addition to
336 other uncertainties. All DEARS benzene data were collected by the Carbopack-X passive sampler. The
337 DNR benzene data were collected by TO-15 SUMMA canister at Southwestern High School and by sub-
338 ambient canister at Allen Park. Further analysis of seasonal patterns in the Detroit area could consider
339 variability seen for benzene sampling methods and for local effects.

340

341 **4. Discussion**

342

343 It is impractical to perform expensive measurements in every part of the country for every
344 pollutant and on a regular basis, so the availability of modeled values from NATA is inherently important.
345 The census block-level NATA results used in this paper were prepared specifically for our analysis based
346 on the most current NATA modeling available at the time. In order to strengthen the comparison of the
347 2002 NATA to the 2005 DEARS data, we examined the local emissions and meteorological parameters
348 used in the 2002 NATA relative to those used in the now-available 2005 NATA. The emissions estimated
349 by NATA for the Wayne County, MI area show that the overall 2005 benzene emissions are about 20%
350 lower than those included in the 2002 NATA analysis. The meteorological patterns for these two years
351 had almost identical mean temperature and wind. The 2005 NATA results also reflect several
352 improvements to the ambient modeling approach relative to that of the 2002 NATA. These changes and
353 updates in the 2005 NATA yield an approximate 30% decrease from the 2002 NATA predicted ambient
354 levels of benzene at most locations across Wayne County, MI. The human exposure predictions for
355 NATA are a direct function of the ambient levels, so we expect similar reductions in the human exposure

356 levels from NATA as well. We did not attempt to apply these updates to our analysis since the overall
357 conclusions would not be greatly affected.

358 NATA's model estimates are used extensively for informing monitoring programs, prioritizing
359 pollutants and emission sources, and providing input for finer-scale assessments, for example. NATA
360 model estimates reflect chronic exposures resulting from inhalation of ambient air toxics from outdoor
361 sources—they do not consider indoor benzene sources or contributions to total inhalation exposure from
362 indoor sources (<http://www.epa.gov/nata/>). The results of this study clearly confirm that indoor sources of
363 benzene are quite important to human exposure and may well be at least as important as outdoor sources.
364 For an initial screening of hundreds of chemicals, a factor of two difference may not be particularly
365 important as the uncertainties in exposure and risk estimates are large, and the difference in risk between
366 chemicals may be orders of magnitude. At the point a community attempts to improve community health
367 and reduce risks, however, the results of this analysis clearly point to the importance of considering indoor
368 sources when reducing total benzene exposure.

369

370 4.1. Conclusions

371

372 We found that the 2002 NATA predictions compared well with outdoor measurements and
373 outdoor sources of benzene as collected in the 2004-2007 DEARS study in Detroit, even though these
374 NATA predictions predate the DEARS measurements by several years. Our findings, however, indicate
375 that the two studies are not as comparable for human exposures. Personal concentration measurements
376 from DEARS were, on average, twice the predicted human exposure estimates from NATA. We conclude
377 that this finding is not an artifact of the differences in years covered by NATA and DEARS given that the
378 2005 NATA predictions are not substantially different from their 2002 NATA counterparts. Our analyses
379 considered personal monitoring data, indoor monitoring data, and known co-pollutants for benzene, and
380 the findings from these analyses are consistent. All these analyses support that the factors driving higher
381 DEARS personal benzene concentrations relative to the NATA predicted exposure values are likely due,
382 at least in part, to indoor sources containing toluene and 1,3-butadiene. The box plots, descriptive
383 analysis, and results from our explanatory regression model also all point to a seasonal effect in the
384 DEARS data with concentrations higher in summer than winter, on average, and this seasonal pattern,
385 while unanticipated, may well reflect a local effect. Given that DEARS had few participants with
386 residences that had attached garages and no participants from self-reported smoking households, the
387 potential for gasoline vapors from attached garages or ETS contributing to personal benzene is lower than
388 that for many households in Detroit and elsewhere. The model findings that the variables for small engine

389 stored in a garage in the summer model and the ETS tracer in the winter model were statistically
390 significant, and these may be viewed as strong findings for seasonality in light of the participants' housing
391 and smoking characteristics. Given the increasing focus on fine geographical scale estimates for
392 community assessments, such as direct use of NATA estimates or through use of the Community-Focused
393 Exposure and Risk Screening Tool (Zartarian and Schultz 2009), this paper also presents an important
394 evaluation of human exposure estimates at this fine scale. These results point to further modeling research
395 for improved characterizations of total human exposures.

396

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406

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411

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478 Table 1. Tract-level mean benzene concentrations for 2004-2007 DEARS measured data and 2002 NATA
 479 modeled estimates ($\mu\text{g}/\text{m}^3$).
 480

Tract	DEARS						NATA								
	Outdoor			Indoor			Personal			Outdoor			Personal		
	n	Mean	SE	n	Mean	SE	n	Mean	SE	n	Mean	SE	n	Mean	SE
5003	223	2.4	0.1	190	4.7	0.3	174	5.7	0.3	23	2.0	<0.1	23	1.9	<0.1
5209	89	1.3	0.1	0	-	-	0	-	-	2	3.0	<0.1	2	2.7	<0.1
5211	151	1.9	0.1	73	3.6	0.3	70	7.6	1.5	6	3.8	<0.1	6	3.8	<0.1
5234	62	3.2	0.2	61	5.2	0.5	57	6.0	0.5	7	4.0	<0.1	7	4.1	<0.1
5238	208	2.8	0.1	192	4.3	0.2	167	5.4	0.3	17	3.1	<0.1	17	3.3	<0.1
5241	142	2.3	0.1	136	4.8	0.4	120	5.1	0.4	15	2.5	<0.1	15	2.7	<0.1
5401	49	1.8	0.1	44	3.1	0.3	38	3.3	0.3	4	2.7	<0.1	4	2.9	<0.1
5402	5	3.5	0.5	5	4.0	0.5	5	4.6	0.4	1	2.2	-	1	2.3	-
5403	30	2.3	0.2	25	6.0	1.2	24	7.7	2.3	2	2.2	<0.1	2	2.3	<0.1
5404	10	1.9	0.5	2	2.3	0.2	2	2.7	<0.1	1	2.2	-	1	2.3	-
5421	63	2.1	0.2	62	3.6	0.4	54	3.8	0.3	5	2.2	<0.1	5	2.5	<0.1
5422	24	2.7	0.3	25	3.6	0.4	24	4.8	0.7	2	2.2	<0.1	2	2.3	<0.1
5426	66	4.1	0.3	63	5.2	0.3	68	6.9	0.6	6	2.6	<0.1	6	2.6	<0.1
5766	181	1.7	0.1	0	-	-	0	-	-	1	2.8	-	1	2.8	-
5870	180	1.3	0.1	56	4.6	0.6	55	5.4	0.6	7	2.5	<0.1	7	2.5	<0.1

481

482

483 Table 2. Descriptive statistics for the numerators from DEARS outdoor concentrations ($\mu\text{g}/\text{m}^3$) and for the
 484 denominators from NATA modeled ambient estimates ($\mu\text{g}/\text{m}^3$) reflected in Figure 1; n is the number of
 485 census blocks.

Season	DEARS					NATA				
	n	Mean	SE	Min	Max	n	Mean	SE	Min	Max
Summer 04	30	4.4	0.2	1.5	6.6	30	2.8	0.1	2.0	4.0
Winter 05	31	2.4	0.1	0.8	3.9	31	2.9	0.1	2.0	4.0
Summer 05	39	2.4	0.1	0.9	3.6	39	2.6	0.1	2.0	4.0
Winter 06	38	1.7	0.2	0.4	5.7	38	2.7	0.1	2.0	4.0
Summer 06	37	2.6	0.2	1.0	5.5	37	2.6	0.1	2.0	3.8
Winter 07	30	1.5	0.1	0.9	2.2	30	2.6	0.1	2.0	3.8

486

487 Table 3. Descriptive statistics for the numerators from DEARS personal concentrations ($\mu\text{g}/\text{m}^3$) and for
 488 the denominators from NATA modeled human exposure estimates ($\mu\text{g}/\text{m}^3$) reflected in Figure 2; n is the
 489 number of census blocks.

Season	DEARS					NATA				
	n	Mean	SE	Min	Max	n	Mean	SE	Min	Max
Summer 04	29	8.4	0.6	3.8	14.0	29	2.9	0.1	1.9	4.1
Winter 05	29	4.5	0.4	2.1	11.6	29	3.1	0.1	1.9	4.1
Summer 05	36	5.8	1.0	1.9	35.5	36	2.8	0.1	1.9	4.1
Winter 06	34	4.5	0.5	1.7	14.0	34	2.8	0.1	1.9	4.1
Summer 06	31	5.2	0.5	2.3	12.4	31	2.7	0.1	1.9	3.4
Winter 07	16	4.2	0.8	1.7	13.3	16	2.6	0.1	1.9	3.4

490

Table 4. Participant reports of possible exposures and their housing characteristics

	Summer				Winter			
	n	missing	Below 75th Percentile	Above 75th Percentile	n	missing	Below 75th Percentile	Above 75th Percentile
Day-specific Reports								
Someone smoked inside residence	537	17	2.2	5.9	402	51	3.4	8.0
Around ETS at some location	537	17	24.4	14.7	402	51	10.3	25.9
Around cooking	537	17	83.3	86.0	402	51	87.6	87.5
Housecleaning performed	537	17	59.6	46.3	402	51	50.7	44.6
Petroleum-based solvents used	537	17	4.5	6.6	402	51	2.4	1.8
Traveled in motor vehicle	537	17	66.6	70.6	402	51	63.1	78.6
Refueled motor vehicle	537	17	15.5	19.9	402	51	10.7	17.0
Housing Characteristics								
Residence has attached garage	111	2	2.4	6.9	89	2	7.8	4.0
Gasoline engine equipment in garage	112	1	14.5	24.1	90	1	12.3	20.0

Table 5. Site-specific outdoor mean concentrations ($\mu\text{g}/\text{m}^3$) and number, n, of samples from DEARS and from Michigan Department of Natural Resources (DNR)

Season	n	Mean	n	Mean
	DEARS EMA 1		DNR Southwestern High School	
Summer 04	26	5.3	8	2.7
Winter 05	29	2.6	8	1.6
Summer 05	33	2.4	5	1.5
Winter 06	34	2.4	4	2.5
Summer 06	44	3.3	7	1.7
Winter 07	40	1.6	7	1.2
	DEARS Allen Park		DNR Allen Park	
Summer 04	28	3.3		
Winter 05	30	2.1	6	1.7
Summer 05	31	1.7	7	2.4
Winter 06	32	0.8		
Summer 06	32	1.5		
Winter 07	25	1.2		

¹ Carbopack-X method

² TO-15 SUMMA canister method

³ Sub-ambient canister method

Figure 1. Ratios of means for DEARS outdoor concentrations to 2002 NATA modeled ambient estimates for the six 2004-2007 seasons from DEARS. S is summer, W is winter, and 04-07 is 2004-2007.

Figure 2. Ratios of means for DEARS concentrations to 2002 NATA modeled exposure estimates for the six 2004-2007 seasons from DEARS. DEARS means are for (a) personal and (b) indoors concentrations. S is summer, W is winter, and 04-07 is 2004-2007.

Figure 3. Ratios of means for DEARS concentrations to 2002 NATA modeled exposure estimates for the six 2004-2007 seasons from DEARS. DEARS means are where the participant reported staying home all day for (a) personal and (b) indoor concentrations. S is summer, W is winter, and 04-07 is 2004-2007.

Figure 4. Scatter plots of log-transformed DEARS concentration data ($\mu\text{g}/\text{m}^3$) reflected in the top quartile of DEARS to NATA ratios for benzene for (a) combined summer seasons and (b) combined winter seasons.