

Indoor and outdoor concentrations of nitrogen dioxide, volatile organic compounds, and polycyclic aromatic hydrocarbons among MICA-Air households in Detroit, Michigan

1 ABSTRACT

2
3 The Mechanistic Indicators of Childhood Asthma (MICA) study in Detroit, Michigan introduced
4 a participant-based approach to reduce the resource burden associated with collection of indoor
5 and outdoor residential air sampling data. A subset of participants designated as MICA-Air
6 conducted indoor and outdoor residential sampling of nitrogen dioxide (NO₂), volatile organic
7 compounds (VOCs), and polycyclic aromatic hydrocarbons (PAHs). This participant-based
8 methodology was subsequently adapted for use in the U.S. National Children's Study. The
9 current paper examines residential indoor and outdoor concentrations of these pollutant species
10 among health study participants in Detroit, Michigan.

11
12 Pollutants measured under MICA-Air agreed well with other studies and continuous monitoring
13 data collected in Detroit. For example, NO₂ and BTEX concentrations reported for other Detroit
14 area monitoring were generally within 10-15% of indoor and outdoor concentrations measured in
15 MICA-Air households. Outdoor NO₂ concentrations were typically higher than indoor NO₂
16 concentration among MICA-Air homes, with a median indoor/outdoor (I/O) ratio of 0.6 in
17 homes that were not impacted by environmental tobacco smoke (ETS) during air sampling.
18 Indoor concentrations generally exceeded outdoor concentrations for VOC and PAH species
19 measured among non-ETS homes in the study. I/O ratios for BTEX species (benzene, toluene,
20 ethylbenzene, and m/p- and o-xylene) ranged from 1.2 for benzene to 3.1 for toluene. Outdoor
21 NO₂ concentrations were approximately 4.5 ppb higher on weekdays versus weekends. As
22 expected, I/O ratios pollutants were generally higher for homes impacted by ETS.

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24 These findings suggest that participant-based air sampling can provide a cost-effective
25 alternative to technician-based approaches for assessing indoor and outdoor residential air
26 pollution in community health studies. We also introduced a technique for estimating daily
27 concentrations at each home by weighting 2- and 7-day integrated concentrations using
28 continuous measurements from regulatory monitoring sites. This approach may be applied to
29 estimate short-term daily or hourly pollutant concentrations in future health studies.

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33 INTRODUCTION

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35 Elevated exposures to air pollutant species commonly found in both indoor and outdoor
36 residential environments have been implicated in a wide spectrum of adverse health outcomes.
37 Volatile organic compounds (VOC) and polycyclic aromatic hydrocarbons (PAH) have been
38 associated with reproductive, developmental, neurological, allergic and respiratory,
39 cardiovascular, and cancer outcomes (ATSDR, 1995 ; ATSDR, 2000; Suh et al., 2000; Miller et
40 al., 2004; ATSDR, 2005; ATSDR, 2007a; ATSDR, 2007b; ATSDR, 2007c; Hertz-Picciotto et
41 al., 2007; Spengler et al., 2007; Bernstein et al., 2008; Hertz-Picciotto et al., 2008). Nitrogen
42 dioxide (NO₂) has been identified as a respiratory irritant responsible for asthma exacerbation
43 (D'Amato et al., 2005; Bernstein et al., 2008).

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45 Concentrations and exposures to these pollutants can be measured by collecting indoor, outdoor
46 and personal measurements, a task typically undertaken by trained technicians (Breysse et al.,
47 2005; Diette et al., 2007; Mukerjee et al., 2009a; Williams et al., 2009). Technician-based air
48 monitoring can be resource intensive and may impose a significant burden on study participants.
49 Estimates of pollution concentrations and personal exposures can also be predicted using
50 empirical statistical models, e.g., land-use regression models (Brauer et al., 2002; Jerrett et al.,
51 2005; Ross et al., 2006; Smith et al., 2006) spatial interpolation techniques, e.g., kriging or
52 splining methods (Jerrett et al., 2005); and physical or mechanistic modeling-based approaches,
53 including atmospheric, indoor / outdoor / personal exposure, and hybrid models (Jerrett et al.,
54 2005; Boothe et al., 2005; Isakov et al., 2006; McConnell et al., 2006; Isakov and Özkaynak
55 2007; Özkaynak et al., 2008). However, modeling studies may require detailed information on
56 emissions, building, and exposure factors, posing technical challenges. In the absence of more
57 comprehensive exposure information, epidemiology studies generally rely on simple surrogates
58 of personal exposures such as central-site monitoring data, proximity to roadways or traffic
59 volume near the home as indicators of exposure (Venn et al., 2001; Janssen et al., 2003; Nicolai
60 et al., 2003; Lewis et al., 2005; Ryan et al., 2005).

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62 The Mechanistic Indicators of Childhood Asthma (MICA) study introduced a participant-based
63 approach to reduce the burden associated with collection of indoor and outdoor residential air
64 monitoring data. Under this approach, a subset of participants designated as MICA-Air collected
65 indoor and outdoor residential air samples. The development and application of participant-
66 based indoor and outdoor air sampling for this study has been described in detail elsewhere
67 (Johnson et al., 2008), and has been adapted for use in the U.S. National Children’s Study. The
68 current report describes indoor and outdoor NO₂, VOCs, and PAHs measured at MICA-Air
69 households and compares air pollution measured under MICA-Air with results from other
70 research and regulatory monitoring in Detroit, Michigan. We also introduce a technique for
71 estimating daily ambient NO₂ concentrations based on 2- and 7-day household measurements
72 coupled with continuous regulatory monitoring data. This approach may be used to estimate
73 short term (daily or hourly) exposure in future health studies.

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METHODS

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MICA-Air Study Design

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78 Gas-phase air sampling was conducted from November 1 – December 29, 2006 in a subset of
79 homes concurrently enrolled in two EPA health studies, MICA and the Detroit Children’s Health
80 Study (Johnson et al., 2008). Passive samplers were shipped to participating households and
81 deployed by the parents of study participants to collect simultaneous indoor and outdoor
82 measurements of NO₂, VOC, and PAH species. Half of the homes deployed VOC and NO₂
83 samplers for a single 7-day sampling event; the other half deployed single event 2-day NO₂
84 samplers as well as 24 and 48 hour PAH samplers. Households were assigned to sampling
85 groups based on several factors—primarily lead time between recruitment and scheduled clinical
86 evaluation for the health studies. Participants received detailed pictorial and written instructions
87 for sampler deployment and retrieval as well as sampling cages in which to set up the indoor and
88 outdoor samplers. Participants were instructed to deploy indoor samplers in the bedroom of the
89 child participating in the health study. Participants recorded start and stop times and dates, as
90 well as indoor temperature based on their indoor thermometer or thermostat, at the beginning and
91 end of the sampling period. Environmental tobacco smoke (ETS) was assessed via

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92 questionnaire. MICA-Air design and protocols have been described in detail elsewhere (Johnson
93 et al., 2008).

Passive Air Sampling

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96 Integrated 2-day and 7 day concentrations of NO₂ were collected using Ogawa passive samplers.
97 Integrated 7 day measures of concentration were collected using Perkin-Elmer tubes packed with
98 Supelco Carbopack B adsorbent for the following VOCs: benzene, ethylbenzene, toluene, m/p-
99 xylene, o-xylene, 2-methylhexane, 2-methylpentane, 2,2,4-trimethylpentane, 2,3-
100 dimethylpentane, 3-methylhexane, methylcyclohexane, 1,1,1-trichloroethane, 1,3-butadiene,
101 1,4-dichlorobenzene, carbon tetrachloride, chloroform, hexane, methylene chloride, methyl t-
102 butyl ether (MTBE), styrene, tetrachloroethene, and trichloroethene. Twenty-four hour
103 concentrations were collected for the following gas-phase PAH species: naphthalene (NAP),
104 acenaphthylene (ACEN), acenaphthene (ACE), anthracene (AN), fluorene (FLN), phenanthrene
105 (PHE), fluoranthene (FL), and pyrene (PY) using Fan-Lioy passive PAH samplers (Fan et al.,
106 2006). Further discussion of passive sampling technology and evaluation is provided in the
107 online supplement.

Quality Control

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111 To evaluate data quality, the study deployed field duplicates equal to at least 10% of the
112 experimental samplers, and field blanks equal to at least 15% of the experimental samplers
113 deployed in the study. Further details and evaluation of duplicate samplers and blanks is
114 provided in the online supplement. Samples were blank corrected by subtracting the average
115 pollutant concentration measured on field blanks for each chemical species. Pollutant levels
116 reported in this paper represent net concentration. Duration-specific MDL values were
117 calculated for each sample. Calculations for MDL are described in further detail in the online
118 supplement. MDL was used to qualify rather than truncate data; therefore net pollutant
119 concentrations below MDL were not replaced with zero or MDL/(sqrt 2), and values below
120 MDL were included in all analyses reported in this paper unless otherwise noted. However,
121 indoor/outdoor (I/O) ratios were not calculated for households with indoor or outdoor values
122 below zero after blank correction.

Indoor and outdoor concentrations of nitrogen dioxide, volatile organic compounds, and polycyclic aromatic hydrocarbons among MICA-Air households in Detroit, Michigan

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Estimation of Daily NO₂ Concentrations Based on 2- and 7-Day Measurements and Continuous Monitoring Data

We also estimated daily ambient NO₂ at MICA-Air homes by calibrating 2-day or 7-day average NO₂ concentration measured at each home using continuous monitoring data measured at MDEQ sites as follows. The 2-day or 7-day average NO₂ concentration measured at each home was assigned to each day that fell within the sampling period for that home. These daily values were then adjusted for day of the week effect by applying a calibration factor (CF), which was based on daily concentrations at regulatory monitoring sites during the study (Equation 1). Daily estimates for MICA-Air homes were calculated as the product of: daily value, daily calibration factor, and total number of sampling days at the home, divided by daily calibration factors for each of the days on which sampling was conducted at the home (Equation 2).

$$CF_{Sun...Sat} = NO_2_{MDEQ\ Sun...Sat} / NO_2_{MDEQ\ Total} \tag{1}$$

- Where:
- CF_{Sun...Sat} = Daily calibration factors for each day of the week (Sunday...Saturday)
 - NO_{2 MDEQ Sun...Sat} = Average daily NO₂ at MDEQ sites in Detroit for each day of the week (Sunday...Saturday) during MICA-Air study period (Nov 1 - Dec 29, 2006)
 - NO_{2 MDEQ Total} = Average daily NO₂ at MDEQ sites in Detroit for duration of MICA-Air study period (Nov 1 - Dec 29, 2006)

$$NO_2_{Daily} = [NO_2_{MICA-Air} * CF_{Day X} * N] / \sum CF_{1...N} \tag{2}$$

- Where:
- NO_{2 Daily} = Daily NO₂ for Day X based on 2-day or 7-day MICA-Air measurement
 - NO_{2 MICA-Air} = Average NO₂ measured at 2-day or 7-day home
 - CF_{Day X} = Daily calibration factor for date of interest (Sunday...Saturday) from Equation 1
 - CF_{1...N} = Daily calibration factors for each day during which sampling was conducted at the home (Day 1...Day N)
 - N = Number of days in which sampling was conducted at the home

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Regulatory Monitoring Data

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155 Both estimated and unadjusted outdoor NO₂ concentrations for MICA-Air households were
156 compared with continuous monitoring data collected by the Michigan Department of
157 Environmental Quality–Air Quality Division (MDEQ) at MDEQ sites 16 and 19 (Linwood and
158 East 7 Mile) in Detroit, Michigan. Daily concentrations at the two MDEQ sites were similar
159 (mean difference in daily NO₂ = 0.6 ppb; mean standard deviation = 1.2 ppb); therefore mean
160 concentrations at the two sites were used in these comparisons.

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Statistical Analyses

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164 Descriptive statistics were generated for indoor and outdoor concentrations of NO₂, VOCs, and
165 PAHs. Percent differences between MICA-Air and regulatory monitoring data were based on
166 unadjusted 2- or 7-day averages from the study homes and MDEQ concentrations averaged over
167 matched time periods. We compared unadjusted NO₂ and BTEX measured under MICA-Air
168 with results from technician-based studies in Detroit. Finally, we performed studentized t-tests
169 to compare weekend versus weekday NO₂ concentrations (for both unadjusted and estimated
170 concentrations) and indoor/outdoor pollutant ratios for ETS versus non-ETS homes. Analyses
171 presented in this paper were limited to households providing complete sampling log data
172 (Johnson et al., 2008). Statistical analyses were performed using SAS 9.1 (SAS Institute, Cary,
173 North Carolina, USA).

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RESULTS

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177 Descriptive statistics for outdoor and indoor NO₂, VOC and PAH concentrations are provided in
178 Tables 1a and 1b, respectively. Mean outdoor NO₂ was approximately 4.0 ppb higher among
179 homes that conducted air sampling for 2 days compared with those that conducted 7-day
180 sampling ($p < 0.05$). There was no observed difference in mean indoor NO₂ concentrations for
181 2-day versus 7-day homes ($p = 0.99$). Mean outdoor concentrations for BTEX species (benzene,
182 toluene, ethylbenzene, and m/p- and o-xylene) ranged from 0.8 $\mu\text{g}/\text{m}^3$ for ethylbenzene to 4.4

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183 $\mu\text{g}/\text{m}^3$ for toluene; 2-methylpentane also contributed a high proportion of the overall pollutant
184 levels measured outside the homes.

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186 For indoor BTEX species, mean concentrations ranged from 2.3 $\mu\text{g}/\text{m}^3$ for ethylbenzene to 18.0
187 $\mu\text{g}/\text{m}^3$ for toluene. Branched alkanes and 1,4-dichlorobenzene were also important contributors
188 to indoor pollution. Standard deviations were generally higher for indoor versus outdoor
189 concentrations for NO_2 and VOC species. NAP was the most predominant of the PAH species
190 for both indoor and outdoor measurements.

Comparison of Unadjusted NO_2 and BTEX Measurements at MICA-Air Homes with Regulatory and Technician-Based Monitoring

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195 Descriptive statistics for NO_2 and BTEX for Detroit area studies including MICA-Air are
196 provided in Table 2. Mean NO_2 measured at continuous MDEQ sites during the same time
197 period as the MICA-Air study (November 1- December 29, 2006) were within 5% of median
198 outdoor concentrations measured under MICA-Air. Mean outdoor NO_2 measurements at
199 DEARS homes in both winter and summer (Williams et al., 2009), and year round regulatory
200 measurements (Rizzo et al., 2002) were also within 10% of outdoor NO_2 concentrations
201 measured at MICA-Air homes.

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203 Mean outdoor BTEX concentrations at MICA-Air homes were similar to outdoor winter
204 measurements at DEARS homes (within 10-15% for benzene, ethylbenzene, and xylenes).
205 Outdoor BTEX concentrations at MICA-Air homes were also consistent with annual average
206 BTEX concentrations (Le et al., 2007) and BTEX measurements collected under Detroit
207 Children's Health Study (DCHS) (Mukerjee et al., 2009b). Mean indoor BTEX concentrations
208 in DEARS homes collected in winter were similar to measurements collected under MICA-Air;
209 ethylbenzene, m/p-xylene and o-xylene concentrations were within 2%, 11% and 13% of MICA-
210 Air measurements, while benzene and toluene concentrations were within 22% and 28%.

211
212 Monitoring data from two continuous regulatory monitoring sites in Detroit was matched to each
213 MICA-Air home by averaging the daily NO_2 monitoring data for each day during which the

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214 household deployed the passive samplers, and weighting the daily averages by the proportion of
215 sampling time on each day. Unadjusted outdoor NO₂ concentrations at MICA-Air homes were
216 within 15-20% of outdoor NO₂ measured at regulatory monitoring sites (median percent
217 difference: 17%; mean percent difference: 20%). Unadjusted outdoor NO₂ at 7-day homes
218 agreed more closely with concurrent measurements at regulatory monitoring sites, but were more
219 likely to be lower than concentrations at regulatory monitoring sites (mean % difference: 10%;
220 median 15%; range: -81 to 63%), while unadjusted outdoor NO₂ measurements at 2-day homes
221 were generally higher than concurrent measurements at continuous regulatory monitoring sites
222 (mean % difference: 34%; median: 20%; range -35 to 148%).

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Comparison of Estimated Daily NO₂ at MICA-Air Homes with Continuous Regulatory Monitoring

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227 Figure 1 shows daily outdoor NO₂ concentrations during the MICA-Air study period (November
228 1- December 29, 2006) for MICA-Air homes and regulatory monitoring sites in Detroit. MDEQ
229 values reflect the daily averages measured at continuous regulatory monitoring sites in Detroit,
230 while MICA-Air values represent estimated daily concentrations (as described in the methods
231 section). Overall, daily outdoor NO₂ for MICA-Air homes was similar to daily NO₂ at MDEQ
232 sites. The difference between daily NO₂ at MICA-Air and MDEQ monitoring sites was greater
233 during the first and last days of the study period (Nov-1-2, and Dec 27-29), and during the
234 American Thanksgiving holiday weekend (Nov 25-29). Standard error was not reported for
235 these time periods because MICA-Air sampling was conducted at only one household during
236 each of those dates.

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238 Figure 2 shows estimated outdoor NO₂ concentrations at MICA-Air homes and MDEQ sites by
239 day of the week. As with the unadjusted measurements, estimated daily concentrations at 7-day
240 homes were similar to MDEQ sites, while estimated concentrations at 2-day homes were slightly
241 higher. Average outdoor NO₂ was approximately 4.5 ppb higher during weekdays compared
242 with weekends for both MDEQ sites and estimated daily MICA-Air concentrations ($p < 0.05$).
243 Weekend versus weekday comparisons based on unadjusted concentrations for 2-day MICA-Air

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244 homes that conducted sampling on weekends versus weekdays also showed significantly higher
245 concentrations on weekdays versus weekends ($p < 0.05$).

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Indoor/Outdoor Ratios for MICA-Air Homes

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249 Figure 3 depicts I/O ratios for NO₂ and BTEX species. Mean I/O ratios for NO₂ did not vary
250 significantly between ETS and non-ETS homes ($p = 0.79$). Mean I/O ratios for BTEX were
251 greater in ETS homes ($p < 0.05$ for all BTEX species except toluene). Among non-ETS homes,
252 I/O ratios for NO₂ (N=60) ranged from 0.2 to 3.4 with a median of 0.6. Median I/O ratios for
253 BTEX species in non-ETS homes (N=29) were slightly higher, ranging from 1.2 for benzene to
254 3.2 for toluene, while median I/O ratios for ethylbenzene, o-xylene, and m/p-xylene were 1.7,
255 1.7, and 1.6, respectively. I/O ratios for other VOCs and PAHs are provided in the online
256 supplements.

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DISCUSSION

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260 MICA-Air introduced a participant-based approach to exposure characterization in which
261 participants conducted indoor and outdoor air sampling without assistance or oversight from
262 trained technicians. Analyses of participant-based NO₂, VOC, and PAH measurements indicate
263 that concentrations and trends observed in the current study agreed well with concurrent
264 regulatory air monitoring data as well as active and passive monitoring results reported by
265 technician-based studies. These findings suggest that participant-air sampling utilized under
266 MICA-Air was a feasible strategy for measuring indoor and outdoor residential air pollution
267 among health study participants. We also estimated daily ambient concentrations at each home
268 by weighting integrated 2- and 7-day residential measurements with continuous regulatory
269 monitoring data. Trends and associations reported for estimated daily concentrations were
270 consistent with those based on unadjusted measurements, suggesting that this approach may be
271 useful for estimating short-term ambient concentrations in future health studies.

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Indoor and outdoor concentrations of nitrogen dioxide, volatile organic compounds, and polycyclic aromatic hydrocarbons among MICA-Air households in Detroit, Michigan

NO₂, VOC, and PAH Measurements at MICA-Air Homes

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Mean outdoor NO₂ was approximately 4.0 ppb higher among homes that conducted air sampling for 2 days compared with those that conducted 7-day sampling ($p < 0.05$), while indoor NO₂ did not vary between 2-day and 7-day homes. It is unlikely that sampling methodology and analysis could explain the differences between 2 and 7-day homes. Badges were prepared and analyzed using identical procedures, with the exception of sampling duration. NO₂ levels measured in the current study were well below the capacity of the samplers, eliminating the possibility of saturation. Also, Ogawa badges have additional filters and reduced surface area for nitrous acid deposition on tube walls; therefore volatilization, storage loss, and rate of sample accumulation would not be expected to vary with sampling duration as with Palmes tubes.

If air sampling were carried out predominantly during weekends at 2-day homes, higher weekday concentrations could potentially explain the difference in outdoor NO₂ concentrations measured at 2-day versus 7-day homes. However, the number of 2-day households conducting air sampling on weekends versus weekdays was similar, and average outdoor NO₂ measured by 2-day homes that conducted air sampling on weekends was higher than average NO₂ measured by 7-day homes (data not shown). It is also possible that outdoor NO₂ was higher among 2-day homes due to higher levels of ambient pollution near these homes. However, preliminary analysis of spatial land-use variables did not suggest significant differences in source proximity between the two groups (data not shown). Outdoor concentrations are also impacted by seasonality; however it is unlikely that seasonality could explain differences between 2-day and 7-day homes. There was also no evidence to suggest that month of sampler deployment (November versus December) differed between 2-day versus 7-day homes, or that ambient outdoor temperature differed between 2- and 7-day homes ($p=0.88$). Finally, the difference between mean outdoor NO₂ at 2-day versus 7-day homes persisted in sensitivity analyses which assumed constant temperature across households.

Average NO₂ and BTEX concentrations measured under MICA-Air were similar to concentrations measured by continuous regulatory monitoring and technician based studies in Detroit. NO₂ measured at regulatory sites during the same time period as the MICA-Air study

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306 was within 5% of outdoor NO₂ measured under MICA-Air, and ambient NO₂ for most Detroit
307 studies were within 10% of outdoor NO₂ concentrations measured under MICA-Air. Indoor and
308 outdoor BTEX concentrations measured at MICA-Air homes were generally consistent (within
309 15%) with residential DEARS measurements collected during the winter, and lower compared
310 with concentrations reported at DEARS homes during the summer (Williams et al., 2009).
311 Average BTEX concentrations were generally higher in summer (July-August) versus winter
312 (January-March) in other Detroit area studies (Mukerjee et al., 2009b; Williams et al., 2009).
313 MICA-Air conducted air sampling in fall/winter (November-December); therefore mean BTEX
314 concentrations measured under MICA-Air that were similar to, or slightly higher than, winter
315 means in other studies were consistent with the expected influence of seasonality.

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317 To further evaluate the efficacy of participant-based air sampling, unadjusted measurements
318 collected at individual MICA-Air homes were compared with temporally matched (2- and 7-day
319 average) concentrations collected at MDEQ sites in Detroit. The median percent difference
320 between unadjusted outdoor NO₂ concentrations measured at MICA-Air homes and concurrent
321 outdoor NO₂ measured at regulatory monitoring sites was approximately 17%. Percent
322 difference was lower for 7-day homes (13%) compared with 2-day homes (20%).

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Estimated Daily NO₂ at MICA-Air Homes

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326 Estimated daily NO₂ at MICA-Air homes was also compared with daily averages from
327 regulatory monitoring sites. Differences between MICA-Air and MDEQ were greater where
328 daily estimates were based on measurements from only one household. Comparisons between
329 MDEQ monitoring and integrated measurements of NO₂ at MICA-Air homes were consistent
330 with comparisons between MDEQ monitoring and estimated daily NO₂ at the study homes.
331 While some differences between MICA-Air homes and MDEQ sites would be expected due to
332 differences in pollutant concentrations across the urban area, good agreement between
333 continuous monitoring data and MICA-Air (both estimated and unadjusted measurements)
334 suggests that participant based air sampling was reasonable approach for collecting residential
335 monitoring data.

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Indoor and outdoor concentrations of nitrogen dioxide, volatile organic compounds, and polycyclic aromatic hydrocarbons among MICA-Air households in Detroit, Michigan

337 Outdoor NO₂ concentrations at study homes were higher on weekdays compared with weekends
338 for both daily estimated concentrations at all MICA-Air homes and for unadjusted measurements
339 at 2-day homes that conducted sampling on weekdays versus weekends. These findings are
340 consistent with patterns observed in MDEQ data for Detroit, and in previous studies in the U.S.
341 (Marr and Harley, 2002; Thoma et al., 2008) and abroad (Karar et al., 2005; Tsai et al., 2007;
342 Khoder, 2008) which reported higher levels of NO₂ in urban areas during weekdays versus
343 weekends due to rush hour and commercial truck traffic. In addition, the agreement between
344 weekend versus weekday trends in estimated and unadjusted values suggests that the approach
345 used in this paper to estimate daily concentrations by weighting integrated measurements with
346 continuous monitoring data could be used to estimate short-term air pollution levels in future
347 health studies. In this paper we demonstrate the use of this approach to estimate daily
348 concentrations. However, the technique could potentially be used to estimate hourly pollutant
349 concentrations based on 1-day measurements.

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Indoor/Outdoor Ratios at MICA-Air Homes

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353 I/O ratios showed greater concentrations of outdoor versus indoor NO₂ for most MICA-Air
354 households. In contrast, indoor BTEX concentrations were typically greater than outdoor
355 concentrations. Relationships between indoor and outdoor NO₂ reported in previous studies
356 varied considerably; studies in southern California and Boston have reported I/O ratios between
357 1 and 2 for NO₂ (Lee et al., 1998; Baxter et al., 2007). I/O ratios for BTEX among non-ETS
358 homes in MICA-Air were comparable to I/O ratios reported by the DEARS study in Detroit. For
359 example, median I/O ratios for benzene, ethylbenzene, m/p- and o-xylene among non-ETS
360 MICA-Air homes fell within 10% of median I/O ratios for non-ETS homes in DEARS (Williams
361 et al., 2009). I/O ratios for other VOC species and PAHs are discussed in greater detail in the
362 online supplement.

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364 I/O ratios for BTEX species among non-ETS homes in MICA-Air were slightly higher than I/O
365 ratios reported in other geographic areas. Relationships of Indoor, Outdoor, and Personal Air
366 (RIOPA) reported median I/O ratios for BTEX species ranging from 1.12 benzene to 1.54 for
367 toluene for multi-season air sampling in Los Angeles, CA; Houston, TX; and Elizabeth, NJ

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368 (Weisel et al., 2005); while the Toxic Exposure Assessment Columbia/Harvard (TEACH)
369 reported median winter I/O ratios between 1 and 2 for most BTEX species in New York, NY,
370 with a median I/O ratio of approximately 2.5 for toluene (Kinney et al., 2002). Median I/O ratios
371 were even lower for TEACH homes in Los Angeles (Sax et al., 2004). Although average BTEX
372 concentrations varied between the RIOPA cities, median outdoor concentrations at RIOPA
373 homes were higher compared with MICA-Air homes while indoor concentrations in RIOPA
374 were lower than indoor concentrations reported by non-ETS homes in MICA-Air (Weisel et al.,
375 2005). These results suggest that indoor sources had a greater impact on indoor concentration
376 among MICA-Air homes compared with households in previous studies. I/O ratios may vary
377 between cities due to differences in indoor sources, housing stock and factors that influence
378 penetration of outdoor pollutants. Differences between urban sources and spatial distribution of
379 study homes in relation to those pollutant sources can also contribute to inter-city differences in
380 I/O ratios. However, because ETS was assessed using questionnaire versus analytical methods
381 in this study, it is also possible that higher I/O ratios in MICA-Air were due to misclassification
382 of some ETS homes.

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384 Seasonality can have a major influence on the contribution of outdoor pollution to indoor
385 concentration. Outdoor concentrations may exert a greater impact on indoor concentrations
386 during the summer due to increased air exchange, while indoor contributions may be lower due
387 to decreased use of indoor sources such as gas appliances and portable heaters. Higher I/O ratios
388 are expected in winter versus summer due to reduced clearance of pollutants generated inside the
389 home (Kinney et al., 2002). For example, Zhu et al. (2005) reported much higher I/O ratios for
390 BTEX species (ranging from 7.7 for benzene to 16 for m/p-xylene) based on air sampling
391 conducted in Ottawa, Canada during fall and winter seasons (November-March); this study also
392 included homes impacted by ETS. MICA-Air measurements were collected during the winter,
393 while the DEARS, RIOPA and TEACH measurements were collected during multiple seasons.
394 Although seasonal variation may have contributed to differences in average I/O ratios between
395 the studies, I/O ratios in MICA-Air were elevated compared to winter I/O ratios for TEACH.
396 Finally, I/O ratios in RIOPA may have been lower than MICA-Air because the RIOPA study
397 over-sampled homes that were heavily impacted by ambient air pollution sources, while MICA-
398 Air sampled homes of participants in a health study (Weisel et al., 2005).

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Limitations

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The analyses in this paper were limited by several factors. MICA-Air participants conducted air sampling without oversight from trained technicians, and preliminary analyses suggest that participants were able to conduct air sampling according to study protocol and provide useful data (Johnson et al., 2008); however, there may be greater uncertainty associated with these measurements compared with data collected by trained technicians. Other design factors such as small sample size, particularly for PAH measurements, and non-synchronization of the sampling periods may also have impacted the analyses. Also, ETS was assessed through questionnaire rather than air sampling which may have led to misclassification of smoking households.

Comparison of MICA-Air results with other Detroit area monitoring data was limited by disparate sampling technology (e.g., active versus passive), integration periods, sampler analysis and sampling seasons. Furthermore, co-location of samplers by technicians was not possible in MICA-Air because technicians did not visit the homes. Thus, while the current results are promising, further evaluation is needed to elucidate the strengths and limitations participant-based air sampling.

Conclusions

MICA-Air collected indoor and outdoor air sampling data among participants of a health study conducted in Detroit, Michigan using a participant-based approach that has been adapted for use in the U.S. National Children's Study. The current paper characterizes indoor and outdoor concentrations of NO₂, VOC and PAH species in MICA-Air homes. Indoor concentrations generally exceeded outdoor concentrations for most VOC and PAH species measured in the study, and outdoor NO₂ concentrations were higher among homes that conducted air sampling on weekdays compared with weekends. Participant-based NO₂, VOC, and PAH measurements agreed well with previous studies and continuous monitoring data collected in Dearborn and Detroit. For example, average NO₂ and BTEX concentrations reported for other Detroit area monitoring generally fell within 10-15% of average indoor and outdoor concentrations measured at MICA-Air households. These findings suggest that participant-based air sampling might

Indoor and outdoor concentrations of nitrogen dioxide, volatile organic compounds, and polycyclic aromatic hydrocarbons among MICA-Air households in Detroit, Michigan

430 provide a cost-effective alternative to technician-based approaches for assessing indoor and
431 outdoor residential air pollution in health studies among diverse populations.

432
433 We also introduced an approach for estimating short term outdoor pollutant concentrations by
434 weighting residential measurements using continuous regulatory monitoring data. Trends
435 observed in estimated NO₂ concentrations were similar to trends based on unadjusted residential
436 concentrations at MICA-Air homes (e.g., comparisons between weekend and weekday
437 concentrations). Further research is needed to fully evaluate this approach, but preliminary
438 findings suggest that this technique may be useful for estimating short term (e.g., daily or hourly)
439 ambient concentrations in future health studies.

440

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449

DISCLAIMER

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