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Determinants of personal, indoor and outdoor VOC concentrations: An analysis of the RIOPA data

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Abstract

Community and environmental exposure to volatile organic compounds (VOCs) has been associated with a number of emission sources and activities, e.g., environmental tobacco smoke and pumping gasoline. Such factors have been identified from mostly small studies with relatively limited information regarding influences on VOC levels. This study uses data from the Relationship of Indoor Outdoor and Personal Air (RIOPA) study to investigate environmental, individual and social determinants of VOC concentrations. RIOPA included outdoor, indoor and personal measurements of 18 VOCs from 310 non-smoking households and adults in three cities and two seasons, and collected a wide range of information pertaining to participants, family members, households, and neighborhoods. Exposure determinants were identified using stepwise regressions and linear mixed-effect models. Most VOC exposure (66 to 78% of the total exposure, depending on VOC) occurred indoors, and outdoor VOC sources accounted for 5 (*d*-limonene) to 81% (carbon tetrachloride) of the total exposure. Personal exposure and indoor measurements had similar determinants, which depended on the VOC. Gasoline-related VOCs (e.g., benzene, methyl tertiary butyl ether) were associated with city, residences with attached garages, self-pumping of gas, wind speed, and house air exchange rate (AER). Odorant and cleaning-related VOCs (e.g., 1,4-dichlorobenzene and chloroform) also were associated with city and AER, and with house size and family members showering. Dry-cleaning and industry-related VOCs (e.g., tetrachloroethylene and trichloroethylene) were associated with city, residence water supply type, and dry-cleaner visits. These and other relationships were significant, explained from 10 to 40% of the variation, and are consistent with known emission sources and the literature. Outdoor concentrations had only two common determinants: city and wind speed. Overall, personal exposure was dominated by the home setting, although a large fraction of VOC concentrations were due to outdoor sources. City, personal activities, household characteristics and meteorology were significant determinants.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.envres.2013.08.005.

Keywords

Determinant; Exposure; Indoor air; Time activity; Volatile organic compounds (VOCs)

1. Introduction

Exposure to volatile organic compounds (VOCs) has been associated with a range of adverse health effects, e.g., eye and nose irritation, allergy, liver and kidney dysfunction, neurological impairment, and cancer (Kim and Bernstein, 2009; US EPA, 2012a, 2012b). Emission sources of VOCs are numerous and widespread (Finlayson-Pitts and Pitts, 2000), and include outdoor sources, e.g., vehicles, filling stations, industry and dry cleaners (Ling et al., 2011; MDE, 2010), and indoor sources, e.g., building materials, cleaning products, cigarette smoke, adhesives, paint strippers, air fresheners, moth repellents, cooking, and water chlorination byproducts (ATSDR, 1997a; Brown, 2002; Singer et al., 2006; US EPA, 2012b; Wallace et al., 1987, 1989; Weschler, 2011). Additionally, outdoor sources contribute a portion of indoor concentrations (Gokhale et al., 2008). In the USA and many other countries, indoor concentrations of many VOCs considerably exceed outdoor levels (US EPA, 2012b). Since people spend most of their time indoors (US EPA, 1989), the largest share of VOC exposure often results from the indoor environment. However, exposures are highly variable and affected by many factors.

The phrase “determinants of disease” has been defined as “any factor or variable that can affect the frequency with which a disease occurs in a population” (Putt et al., 1987). Determinants affecting health at individual and community levels can be classified into three groups: social/economic environment, the physical environment, and a person’s individual characteristics and behaviors (WHO, 2012). The present study draws parallels from these definitions by considering determinants of VOC exposures, that is, factors affecting VOC concentrations and exposures. Like health determinants, exposure determinants can be due to socioeconomic factors (e.g., income level and socioeconomic position), factors related to the physical environment (e.g., meteorology and house age), and personal factors (e.g., race/ethnicity, and behavior). While not entirely exclusive, these groupings provide a structure that aids the understanding and analysis of factors affecting exposure.

A review of 12 studies that have examined VOC determinants is summarized in Table 1. (This review emphasized general, i.e., non-occupationally-exposed, populations.) The number of determinants is large and includes many environmental factors. Elevated exposures have been associated with low ventilation rates and closed windows (D’Souza et al., 2009; Riederer et al., 2009; Sexton et al., 2007; Symanski et al., 2009; Wang et al., 2009), house type (Byun et al., 2010; Riederer et al., 2009), years lived in home (D’Souza et al., 2009), and fireplaces in the home (Delgado-Saborit et al., 2009).

A modest number of personal determinants have been identified. VOC exposure has been related to ethnicity (Riederer et al., 2009; Wang et al., 2009). Occupation clearly affects exposure, e.g., benzene, toluene, ethylbenzene, xylene (BTEX) exposure has been linked to service station and vehicle repair jobs (Jo and Song, 2001), and pinene, limonene, toluene, ethylbenzene and styrene have been associated with cleaning jobs (Wolkoff et al., 1998).

Machine-related jobs have been linked to BTEX exposure (D'Souza et al., 2009), and time at work/school has been associated with benzene, ethylbenzene, xylene and tetrachloroethene (PERC) exposure (Wang et al., 2009). However, effects of occupation on VOC exposures for the general public rarely have been observed.

VOC exposures clearly are affected by an individual's activities, as shown by many studies (Table 1). As examples, BTEX and styrene exposure was elevated by smoking and environmental tobacco (D'Souza et al., 2009; Delgado-Saborit et al., 2009; Edwards et al., 2001; Kim et al., 2002; Wallace, 2001; Wallace et al., 1989), and being near vehicles (Delgado-Saborit et al., 2009; Hinwood et al., 2007; Kim et al., 2002; Wallace et al., 1989). Pumping gas, being near gasoline, or living in a home with an attached garage increased gasoline-related VOCs exposures (D'Souza et al., 2009; Delgado-Saborit et al., 2009; Sexton et al., 2007; Symanski et al., 2009; Wang et al., 2009). The use of paint strippers and thinners also has been associated with BTEX exposure (D'Souza et al., 2009; Delgado-Saborit et al., 2009; Symanski et al., 2009). The use of gas heating and gas stoves increased exposure to aromatic VOCs and a gasoline additive, methyl tertiary-butyl ether (MTBE) (Delgado-Saborit et al., 2009; Kim et al., 2002). Participation in arts and crafts hobbies increased exposure to toluene, ethylbenzene and xylene (Hinwood et al., 2007). Deodorizer and mothball use increased exposure of 1, 4-dichlorobenzene (1,4-DCB) (D'Souza et al., 2009; Wallace, 2001; Wallace et al., 1989) and naphthalene (Batterman et al., 2012). Visiting a dry-cleaner or being near dry-cleaned clothes elevated PERC exposure (D'Souza et al., 2009; Wallace, 2001; Wallace et al., 1989). Finally, since chlorine is widely used as a disinfectant to treat public water supplies, contact with chlorinated water through drinking tap water, showering/bathing, swimming, washing dishes/clothes has been shown increase in exposure to chloroform (D'Souza et al., 2009; Sexton et al., 2007; Wallace, 2001; Wallace et al., 1989).

Few socioeconomic determinants have been identified. Education and income has been negatively associated with exposures of benzene, 1,4-DCB, PERC and chloroform (Wang et al., 2009), suggesting that persons of higher socioeconomic position experience fewer high-exposure activities, e.g., house cleaning, reside in cleaner homes and neighborhoods (e.g., distant from traffic), and/or commute and work in cleaner environments. In Korea, children had higher exposure to traffic-related VOCs (e.g., toluene, ethylbenzene, and *m,p*-xylene) in a city with narrow streets and close proximity to vehicle traffic (Byun et al., 2010). In broad terms, many socioeconomic factors are expected to be correlated with environmental factors, which may be considered more direct determinants of concentration or exposure. However, identification of socioeconomic determinants may increase our understanding of the factors affecting exposure.

While many exposure determinants have been identified, the underlying studies have several limitations, the significance and applicability of the determinants are uncertain, and many determinants likely remain undiscovered. First, many of the studies used small samples, e.g., the Birmingham study enrolled only 12 adults (Kim et al., 2002), the New York City study had 46 high school students (Kinney et al., 2002), and the Minneapolis–St. Paul study enrolled 70 adults (Sexton et al., 2007). Observational studies, especially cross-sectional studies, require large sample sizes to disentangle contributions of personal activities and

indoor and outdoor environments. Second, the studies had important data gaps. For example, although the National Health and Nutrition Examination Survey (NHANES) sample was large (646 individuals) and designed to be nationally representative (NCHS, 2012), outdoor and indoor concentrations, time activity, and other information was not collected. However, another large study, the Relationship of Indoor Outdoor and Personal Air (RIOPA) study (Weisel et al., 2005a), collected outdoor, indoor and personal VOC measurements, along with considerable other information, and thus provides a good opportunity to examine potential exposure determinants.

The objectives of the present study are to characterize determinants of VOC exposures among the RIOPA participants, including the contributions due to indoor and outdoor settings. A wide range of variables are considered as potential determinants, including geographic and meteorological factors, neighborhood and household characteristics, demographics, participant activities, and time activity information. Bivariate and mixed-effect models are used to identify determinants and to construct models that explain the exposure data.

2. Materials and methods

2.1. Data sources

Data from RIOPA included outdoor, indoor and personal measurements of air pollutants in three U.S. cities (Elizabeth, NJ; Houston, TX; Los Angeles, CA), selected on the basis of differences in the emission sources likely to affect pollutant exposure. Homes near outdoor emissions were oversampled in order to estimate outdoor contributions to personal exposures (Weisel et al., 2005b). Due in part to limited resources and conflicts with study goals of evaluating local emission sources, RIOPA used a convenience sample. A total of 310 non-smoking households, 309 adults and 118 children in three cities were recruited and studied from summer 1999 to spring 2001. Exclusion criteria included individuals who were smokers, living in a smoking household, staying at home less than 14-h per day, unable to wear personal samplers, or planning to move within three months. Each household and participant was sampled twice about three months apart. The RIOPA study design, pollutant measurements, and other results are described elsewhere (Turpin et al., 2007; Weisel et al., 2005a. 2005b).

A total of 18 VOCs (benzene, toluene, ethylbenzene, *m*,*p*-xylene, *o*-xylene, MTBE, styrene, 1,4-DCB, methylene chloride (MC), trichloroethylene (TCE), PERC, chloroform, carbon tetrachloride (CTC), *d*-limonene, α-pinene, β-pinene, 1,3-butadiene and chloroprene) were measured using passive samplers (OVM3500, 3M Company, St. Paul, MN, USA), 48-h sampling periods, and gas chromatography-mass spectrometry analysis. Data for 1,3-butadiene and chloroprene were not reported due to low recovery, and MC data were excluded due to inconsistent blank contributions (Weisel et al., 2005b). Method detection limits (MDLs) ranged from 0.21 (α-pinene and PERC) to 7.1 (toluene) μg m^{-3}. Detection frequencies for outdoor measurements ranged from 6 (β-pinene) to 97% (CTC); 26 (TCE) to 96% (CTC) for indoor measurements, and 23 (TCE) to 97% (CTC) for personal measurements (Weisel et al., 2005b). Measurements below the MDLs were replaced with one-half of this value.

RIOPA participants were administered three questionnaires, which included over 500 variables. A baseline questionnaire addressed demographics and lifestyle factors (e.g., ethnicity, employment, opening windows, and use of deodorizer or fresheners); a technician walk-through questionnaire collected neighborhood and household characteristics (e.g., industrial emissions in neighborhood, type of building, and existence of attached garage); and a third questionnaire collected time activity information, e.g., time spent indoors at school/work, pumping gas, bathing or showering, and gardening (Weisel et al., 2005a). Household air exchange rates (AERs), geographic and meteorological information (e.g., city, outdoor temperature, wind speed, and relative humidity) was also obtained.

2.2. Statistical analysis

2.2.1. Data selection and cleanup—The present study uses 544 adult personal measurements (n = 299 and 245 for first and second visits, respectively), 554 indoor measurements (n = 303 and 251), 555 outdoor measurements (n = 302 and 253), and time activity logs (n = 532). Personal measurements from children were excluded to avoid cluster effects in the analysis. (Several households included children; each household included one adult participant.)

2.2.2. Descriptive analyses—Descriptive statistics were calculated for outdoor, indoor and personal VOC concentrations, and for demographic information. Analyses were also stratified by city. Differences among cities were evaluated using one-way ANOVA and chi-square tests for continuous and categorical variables, respectively. The correlation between sample types for each VOC was calculated using Spearman rank correlations.

2.2.3. Time and exposure fractions—The sampling time and time spent in different locations (outdoors in neighborhood, outdoors out of neighborhood, indoors at home, indoors at school/work, other indoors, transportation, and unknown) were calculated for each participant. Participants with missing-time fractions exceeding 0.25 (n = 50), were excluded. The missing time fraction, F_{t_miss}, was calculated as

$$F_{t\_\,\text{miss}}=(T_{\text{total}}-T_{\text{outdoor}}-T_{\text{indoor}}-T_{\text{transit}})/T_{\text{total}} \quad (1)$$

where T_{total} = total time spent (min), T_{outdoor} = time spent outdoors (min), T_{indoor} =time spent indoors (min), and T_{trarisit} = time spent in transit (min).

An individual's total, cumulative or potential exposure is often represented as the sum of the concentration-time product across all compartments or micro-environments in a given time period. We define a compartment-oriented exposure fraction as the fraction of exposure attributable to being outdoors, $F_{\text{outdoor\_}C}$, which was calculated for each participant as

$$F_{\text{outdoor\_}\,C}=(C_{\text{outdoor}}T_{\text{neighborhood}})/(C_{\text{personal}}T_{\text{total}}) \quad (2)$$

where $F_{\text{outdoor\_}C}$ = fraction of a person's exposure due to being outdoors in their neighborhood, C_{outdoor} = residential outdoor VOC concentration (μg m^{-3}), $T_{\text{neighborhood}}$ = time spent outdoors in neighborhood (min), and C_{personal} = personal VOC exposure (μg m^{-3}). Similarly, the indoor exposure fraction is

$$F_{\text{home\_C}} = (C_{\text{home}} T_{\text{home}}) / (C_{\text{personal}} T_{\text{total}}) \quad (3)$$

where $F_{\text{home\_C}}$ = fraction of a person's exposure due to being in their home, C_{home} = indoor VOC concentration (μg m^{-3}) at home, and T_{home} = time (min) spent indoors at home. As discussed later, $F_{\text{outdoor\_C}}$ was generally very small. In contrast, $F_{\text{home\_C}}$ exceeded one for 11 to 20% of the observations (n = 52 to 98, depending on the VOC), 5 to 11% exceeded 1.25 (n = 25 to 53), and 2 to 8% (n = 11 to 39) exceeded 1.5 (Supplemental Fig. S1). Given the importance of the indoor environment for VOC exposures, sampling error might explain a large part of the divergence from the assumptions. Cases where $F_{\text{home\_C}} > 1$ might be excluded, but it seems likely that indoor exposure was important and dominant, and thus appears reasonable to assume that $F_{\text{home\_C}} \approx 1$ and $F_{\text{outdoor\_C}} \approx 0$ in such cases. In the following analysis, we excluded $F_{\text{home\_C}} > 1.25$.

To estimate source (rather than compartment) contributions for VOC exposures, a source-oriented exposure fraction was calculated. This analysis assumed 100% penetration efficiency for outdoor VOCs entering a residence, 0% loss rate (VOC decay), and considered outdoor and home sources. These exposure fractions were calculated as

$$F_{\text{outdoor\_S}} = (C_{\text{outdoor}} (T_{\text{home}} + T_{\text{neighborhood}})) / (C_{\text{personal}} T_{\text{total}}) \quad (4)$$

$$F_{\text{home\_S}} = ((C_{\text{home}} - C_{\text{outdoor}}) (T_{\text{home}})) / (C_{\text{personal}} T_{\text{total}}) \quad (5)$$

where $F_{\text{outdoor\_S}}$ = fraction of a person's exposure attributable to outdoor sources in participant's neighborhood, and $F_{\text{home\_S}}$ = fraction of a person's exposure attributable to indoor sources in the participant's home.

Exposure fractions were stratified by city, and by warm (May to October) and cool (November to April) seasons. Differences were evaluated using Kruskal–Wallis (K–W) tests.

2.2.4. Variable selection and linear mixed-effect models—As an initial step to identify possible exposure determinants, each of the 527 RIOPA variables was used in univariate regression models with outdoor, indoor and personal VOC measurements as dependent variables. These models used six VOCs (benzene, toluene, MTBE, 1,4-DCB, PERC and chloroform), selected to represent a range of VOCs and potential emission sources. Next, variables that attained statistical significance ($p < 0.05$) were used in forward stepwise multivariate regression models with selection based on the Schwarz Bayesian Information Criterion. While reducing the number of variables, the resulting parameter estimates are approximate since these models do not account for correlations due to clustering and nesting, e.g., the two seasonal samples collected for most participants.

Linear mixed-effect models (LMMs) that incorporated fixed and random effects and repeated measures (Krueger and Tian, 2004) were estimated for outdoor, indoor and personal measurements using the variables selected by the stepwise models. These models also incorporated several variables with strong theoretical support or of special interest (e.g.,

city, ethnicity, and presence of an attached garage). Two-way interactions among variables were evaluated. Because few significant interactions between determinants of VOC exposures were found, interaction terms were not retained in the final models. Using log-transformed VOC concentrations, random intercepts, nested effects for city, and interactions, the LMMs are expressed as

$$\log(C_{ti})=(\beta_0+b_{0i})+\beta_1\mathrm{Visit}_t+\beta_2\mathrm{City}+\cdots+\beta_n X_n+\varepsilon_{ti} \quad (6)$$

where C_{ti} = VOC concentration (μg m^{-3}) at time t for individual *i*, β=model coefficients for fixed effects, *b* = random deviation from the overall fixed effects, Visit_t = sample collected at time *t*, *X* = other covariates, and ε_{ti} = random error of the VOC concentrations from the predicted line at time *t* for individual *i*. Since the LMMs used log-transformed VOCs, the effect size for each explanatory variable was calculated as follows,

$$\text{Effect size}=e^{(\beta U)} \quad (7)$$

where *e* = exponential, *U* = 1 for categorical variables, and *U* = interquartile range (IQR) for continuous variables.

Separate LMMs were developed for the 15 VOCs, and grouped into three categories based on common determinants: gasoline-related VOCs (BTEX, MTBE and styrene); odorant and cleaning-related VOCs (1,4-DCB, chloroform, *d*-limonene, α-pinene and β-pinene); and dry-cleaning and industry-related VOCs (TCE, PERC and CTC). To maintain a sufficient sample size, variables with fewer than 400 observations were not included in the final LMMs.

2.2.5. Model assessment—Steps taken to help verily model results included the following: Partial residua] plots were examined to assess linearity and fit of continuous variables, e.g., wind speed and household AERs. Transformations (e.g., log-transformation or reciprocal) were tested for variables showing non-linear relationships. Because the reduction in residual variance (R^2) attributable to fixed effect variables cannot be directly obtained from the SAS procedure, R^2 was estimated as

$$R^2=(\sigma^2_{\mathrm{int}}-\sigma^2_{\mathrm{full}})/\sigma^2_{\mathrm{int}} \quad (8)$$

where $\sigma^2_{\mathrm{int}}=\text{variance of the intercept only model}$, and $\sigma^2_{\mathrm{full}}=\text{variance of full model}$. Here, R^2 indicates the difference in the variance between reduced (i.e., intercept-only) and full (i.e., with predictor variables) models.

2.2.6. Missing data—Candidate variables in the LMMs typically had 50 to 100 missing observations. The effect of missing data was evaluated using multiple imputation (MI), and results were compared to the original dataset (with missing data). Three models for each sample type were selected for this comparison: models with the least missing data (e.g., 3% missing for personal measurements of styrene), models with a modest amount of missing data (e.g., 20% missing for benzene), and models with a high amount of missing data (e.g., 28% missing for *d*-limonene). Differences between the original and MI datasets were computed as the relative change in model estimates of β. This comparison (Supplemental

Tables S1 to S3) demonstrated that while models using imputed data tended to have smaller (more statistically significant) *p*-values, changes were not large. Also, the model parameters themselves did not show obvious biases. Differences tended to increase with the fraction of missing data, although changes remained small. Among the nine models tested, only one (outdoor benzene) had three parameters change by more than 30%. Because missing data did not greatly affect the LMM results, subsequent results do not use MI.

2.2.7. Computation—Most analyses used SAS 9.2 (SAS Institute, Cary, North Carolina, USA). Variable selection used proc glmselect, LMMs used proc mixed, and MI analyses used proc mi and proc mianalyze. Partial residual plots were drawn in R version 2.13.1 (R Development Core Team, Vienna, Austria). Relative changes were calculated using Excel (Microsoft, Redmond, WA).

3. Results and discussion

3.2. Descriptive analyses

3.1.1. VOC measurements—Summary statistics for VOC concentrations show that mean and median concentrations of most compounds were ranked as personal > indoor > outdoor (Table 2). The exceptions, 1,4-DCB and α-pinene, had higher indoor levels due to a number of extreme values (indicated by the 95th percentile concentrations, also shown in the table). These high indoor concentrations suggest that the indoor VOC samplers may have been placed near an emission source, and possibly that indoor concentrations varied in the house. MTBE had the highest mean concentration (8.1 μg m^{-3}) among the VOCs outdoors, and 1,4-DCB had the highest mean among both indoor and personal samples (69.9 and 56.8 μg m^{-3}, respectively).

For most VOCs, the correlation between personal and outdoor, personal and indoor, and indoor and outdoor concentrations was high and significant (Supplemental Table S4). Personal-indoor correlations ranged from $r = 0.63$ (toluene) to $r = 0.88$ (*d*-limo-nene); personal-outdoor and indoor-outdoor correlations were generally lower and spanned a wide range ($0.05 < r < 0.72$). Correlation was negligible between personal and outdoor, and indoor and outdoor samples for chloroform, *d*-limonene and α-pinene, suggesting that outdoor levels of VOCs with strong indoor sources had little effect on personal exposures.

3.1.2. Demographic factors—Many demographic characteristics of the RIOPA participants differed among the three cities (Supplemental Table S5). Most participants were female, but the fraction varied by city (61, 81 and 84% in Los Angeles, Elizabeth and Houston, respectively, $p < 0.001$ for difference). The predominant ethnic group also varied by city (Whites at 55% in Los Angeles; Hispanics at 73% in Elizabeth; Mexicans at 50% in Houston; $p < 0.001$). Participants in Los Angeles had higher educational levels (59% with college or above) and the highest annual household income (20% earned over $75,000). Unemployment rates of participants in Los Angeles and Elizabeth were below 35%, while Houston's rate was 84% in Houston ($p < 0.001$). In contrast to these differences, the average age of RIOPA participants (45 ±17 years) did not vary by city ($p = 0.69$).

3.1.3. Time fractions—Fig. 1 displays the average time fractions spent outdoors, indoors and in transit for the RIOPA participants. Indoor time fractions averaged 89, 92, and 92% in Los Angeles, Elizabeth, and Houston, respectively, $p < 0.001$), and participants in Los Angeles spent the least time at home (71, 80, and 80% for the three cities, $p < 0.001$), likely explained in part by the lower unemployment rate in Los Angeles. Little time was spent outdoors, including time within or out of the neighborhoods (fractions averaged 5.1, 4.5, and 4.3% in Los Angeles, Elizabeth, and Houston, respectively, $p = 0.650$). Similarly, little time was spent in transit (5.5, 3.6 and 3.6% in the three cities, respectively, $p < 0.001$).

Fig. 1 compares the RIOPA time budgets to a nationally representative sample, the National Human Activity Pattern Survey (NHAPS), which is a probability-based telephone interview survey conducted from 1992 to 1994 that collected 24-h time-activity information, demographics, and exposure-related questions from 9196 respondents (Klepeis et al., 2001). NHAPS respondents spent more time outdoors (7.6%) than the RIOPA participants (4.6%), and less time indoors (87%) and at home (69%). RIOPA's eligibility criterion that participants be home at least 10 h per day (Weisel et al., 2005b) may have increased the number of women (75 vs. 54% in NHAPS), age (18% of RIOPA participants over 64 years old vs. 14% in NHAPS), and unemployment rate (53%) of participants in this study. Indeed, the data from NHAPS shows somewhat more time in transit and less time at school/work. Both RIOPA and NHAPS reflect the well know pattern that most individuals spend the overwhelming fraction of time at home.

3.1.4. Compartment and source exposure fractions—Home VOC levels dominated personal exposures, e.g., median F_{home_C} values ranged from 0.66 (MTBE) to 0.78 for α-pinene, and the 95th percentile values approached 1 for all VOCs (Table 3 and Supplemental Fig. S2). The importance of the home concentrations is unsurprising since RIOPA participants spent most (median of 77%) of their time at home, and since indoor concentrations of most VOCs were much higher than outdoors levels.

F_{home_C} differed by season for two VOCs (benzene, MTBE), and by city for most VOCs (except toluene, *o*-xylene, 1,4-DCB, PERC, *d*-limonene and β-pinene) (Supplemental Table S6). The median F_{home_C} was highest in Houston (0.68 to 0.81) for most VOCs (except benzene, styrene, PERC, and *d*-limonene). City effect is likely a result of differences in emission sources, meteorology and household characteristics (e.g., presence of attached garage) among the three cities studied, as discussed later. Seasonal effects on indoor levels of VOCs may be affected by lifestyle factors, e.g., opening windows, and using air conditioners.

Contributions of outdoor VOC levels to personal exposures were very small, e.g., median values of $F_{outdoor\_C}$ less than 0.01 for all VOCs (except CTC) (Table 3 and Supplemental Fig. S2). Even 95th percentile values of $F_{outdoor\_C}$ fell below 0.15. $F_{outdoor\_C}$ was small, a result of both the little time spent outdoors (see previous section) and the low outdoor VOC concentrations. $F_{outdoor\_C}$ differed ($p < 0.05$) by season for all VOCs and by city for over half of the VOCs (benzene, toluene, *m*,*p*-xylene, *o*-xylene, MTBE, TCE, PERC and CTC) (Supplemental Table S6). Because many of VOCs (toluene, styrene, 1,4-DCB, TCE,

chloroform, *d*-limonene, α-pinene, β-pinene) had low detection frequencies (< 60%), the outdoor exposure fractions are approximate.

The outdoor source exposure fractions indicate the significance of outdoor VOC sources. With the exception of VOCs with strong indoor sources (1,4-DCB, chloroform, *d*-limonene and α-pinene), $F_{outdoor\_S}$ exceeded F_{indoor_S}, and $F_{outdoor\_S} > 0.60$ for benzene, MTBE, TCE and CTC (Table 3). Thus, with the exception of VOCs with strong indoor sources, outdoor sources were the major contributor to personal exposures. This conclusion may appear surprising given that many VOC studies have shown elevated indoor/outdoor (*I*/*O*) concentration ratios, thus implicating indoor VOC sources. However, unless *I*/*O* ratios exceed 2, outdoor sources will provide over half of the exposure (the lowest possible estimate based on an individual spending 100% time indoors). For the average RIOPA participant spending 91% of their time indoors, *I*/*O* ratios must exceed 2.1 for indoor sources to dominate exposure. Of the VOCs in RIOPA, median *I*/*O* ratios were 2.6, 4.4, 12.9, and 3.2 for 1,4-DCB, chloroform, *d*-limonene, and α-pinene, respectively. For all other VOCs in RIOPA, outdoor sources contributed most of the exposure. This conclusion parallels prior RIOPA analyses that show indoor and personal $PM_{2.5}$ exposure was mostly due to outdoor sources (Meng et al., 2009, 2007; Polidori et al., 2006).

3.2. Determinants of personal, home, and outdoor VOC concentrations

3.2.1. Gasoline-related VOCs—BTEX, MTBE and styrene, all components of gasoline and vehicle exhaust, shared several exposure determinants (Table 4 and Supplemental Table S7). Increased exposures were associated with living in Houston, homes with attached garages, and self-pumped gas; decreased exposures were associated with higher wind speeds and house AERs. Interestingly, lower exposures of toluene, ethylbenzene and *o*-xylene were found for participants reporting cooking activities during the sampling period. Participants reporting cooking activities spent less time in cars with closed windows (mean time spent=71 min) than those not reporting cooking activities (mean time spent = 88 min, *p*-value of *t* test = 0.04), possibly lowering exposure to traffic-related VOCs. (No differences were seen for time in cars with open windows or for total travel time.)

The literature supports our findings for BTEX, MTBE and styrene (Table 1). In Houston, important VOC sources included petrochemical facilities and vehicles (Weisel et al., 2005b). Attached garages are known sources of gasoline-related aromatics in homes (Batterman et al., 2007; D'Souza et al., 2009; Delgado-Saborit et al., 2009; Sexton et al., 2007; Symanski et al., 2009; Wang et al., 2009). Gasoline pumping has been shown to elevate personal exposures to BTEX in cold weather in Alaska (Backer et al., 1997). The effects of both attached garages and pumping gas on gasoline-related VOCs were also seen in NHANES (Symanski et al., 2009). Concentrations arising from outdoor sources, e.g., vehicle exhaust, are diluted by wind (US EPA, 2010), so higher wind speeds may lower exposures. The AER, which accounts for infiltration and ventilation and which depends on wind speed (US EPA, 2011), influences indoor concentrations and thus personal exposures for those pollutants arising from indoor sources. Cooking-related activities have been shown to increase indoor and personal concentrations of several VOCs, e.g., benzene and toluene (Byun et al., 2010; Clobes et al., 1992). However, RIOPA showed a negative association

between cooking and personal exposures to toluene, ethylbenzene and *o*-xylene. This inconsistency could be due to chance, although the explanation offered above – that participants without cooking activity traveled more to dine out during which time they were exposed to gasoline-related VOCs – appears reasonable. The RIOPA dataset does not allow further analysis, but we speculate that visits to "drive-though" fast-food facilities where vehicles are queued up and idling may be a particularly important source of VOC exposure.

3.2.2. Odorant and cleaning-related VOCs—Four determinants were found for the group of odorant and cleaning-related VOCs (1,4-DCB, chloroform, *d*-limonene, α-pinene and β-pinene) (Table 5 and Supplemental Table S8). Like the gasoline-related VOCs, Houston participants had higher exposures to these VOCs. AERs were negatively associated with VOC exposures, reflecting the dilution of indoor source emissions. Participants in larger houses (more rooms) tended to have lower exposure to 1,4-DCB, chloroform, *d*-limonene and α-pinene. Interestingly, the behaviors of other household members were associated with personal exposure, e.g., non-participants showering during the sampling period was associated with higher exposures of chloroform, *d*-limonene, α-pinene and β-pinene.

The odorant and cleaning-related VOCs are primarily released by indoor sources, e.g., mothballs, air fresheners, cleansers and chlorinated water (ATSDR, 1997a, 2006; Chin et al., 2013; US EPA, 2012a). Thus, use and storage of these products can affect exposure. Also, since these VOCs are due mainly to indoor sources, AER is expected to be a determinant (Mudarri, 2010). The identification of the number of rooms, an indicator of house size, as a determinant may reflect additional mixing in large houses that lowers concentrations compared to levels in smaller houses with comparable product use. In RIOPA, the number of rooms in a household was positively associated with household income ($\beta = 0.79$, p-value < 0.0001), and thus socioeconomic factors may be an indirect or interacting factor associated with high exposures of odorant and cleaning-related VOCs. However, no association with household income and VOC exposures were found. The effect of employment on *d*-limonene exposure might result as unemployed participants spent more time at home (2278 and 2000 min for unemployed and employed participants, respectively; p-value< 0.0001), and possibly engaged in tasks that increased contact with cleaners and odorants.

Chloroform is a water disinfection byproduct of chlorine, thus drinking water, contacting water (e.g., bathing) and inhaling water vapor can increase exposure (ATSDR, 1997a). Elevated chloroform concentrations in a room adjoining a study bathroom during showering have been noted and called "secondary shower exposure" (Gordon et al., 2006). Such secondary exposure is consistent with findings that chloroform exposure in RIOPA increased when other family member showered. However, bathing or showering by the RIOPA participants themselves did not affect exposure. Similar (negative) results with showering were found for the 1999–2000 NHANES dataset, possibly due to a lack of variance in showering-related variables, i.e., most (85%) participants showered during the sampling period (Riederer et al., 2009). The same explanation may apply to the present study (87% of participants showered during the sampling period). Additionally, participants were instructed not to get the samplers wet, and thus they may have removed them outside of the shower or bathroom (Weisel et al., 2005b).

The effect of city can be attributable to several factors, including differences in outdoor emission sources, e.g., industry and traffic (Weisel et al., 2005b), meteorological factors that affect both dispersion and emissions of outdoor pollutants, systematic differences in building AERs, demographic and cultural factors. For example, outdoor temperatures were considerably warmer in Houston during the sampling period, compared to Los Angeles and Elizabeth (respectively averaging 22.3 ± 7.5, 18.6 ± 4.7 and 14.6 ± 8.6 °C, $p <$ value < 0.0001). Higher temperatures increase vapor pressures, permeation rates, and evaporation rates, potentially producing higher concentrations. Since a fraction of odorant and cleaning-related VOCs arise from volatilization and sublimation from indoor sources, indoor temperatures are also important. Indoor temperatures showed less variation and differences were not significant (respectively averaging 23.3 ± 2.6, 23.9 ± 2.6 °C and 24.0 ± 3.4 in Los Angeles, Elizabeth, and Houston, *p*-value = 0.052).

3.2.3. Dry-cleaning and industry-related VOCs—The dry-cleaning and industrial emissions group had three VOCs (TCE, PERC and CTC) affected by city and household water source (Table 6 and Supplemental Table S9). Elizabeth and Los Angeles participants had the highest TCE and PERC exposures, but Houston participants had the highest CTC exposure. Public water supplies were associated with lower TCE exposure, but higher CTC exposure.

As expected, PERC exposures increased by visiting a dry cleaner (Table 6 and Supplemental Table S9). This solvent has been widely used for dry cleaning clothes, and exposures occur when visiting dry cleaning establishments, and storing dry cleaned clothes at home, whether or not clothes are wrapped in plastic (Sherlach et al., 2011), as noted in Table 1. PERC exposures were higher among employed participants. Since PERC has been widely used in industry as a degreaser and also is in products such as adhesives and paint removers (ATSDR, 1997b), exposure among employed participants may be more likely. The city effect may be related to population density: Los Angeles and Elizabeth have higher densities (Weisel et al., 2005b), which may lead to more dry cleaners and elevated ambient concentrations. The outdoor PERC levels were higher in Los Angeles and Elizabeth than in Houston (medians were 1.29, 0.74, and 0.11 μg m^{-3}, respectively, *p*-value < 0.001).

TCE has been used extensively as a degreaser, paint remover, adhesive, and chemical intermediate (ATSDR, 1997c). Exposure may increase if TCE-containing consumer or home products are present, e.g., vinyl siding, glue and car stain removers (US EPA, 2007). Additionally, TCE is sometimes found in contaminated soils and groundwater, and participants in households near to subsurface or surface contaminated soils may be exposed indoors through soil vapor intrusion and water consumption, if a local well (especially a private well without monitoring or treatment) provides the water source. In the RIOPA dataset, the TCE detection frequency was only 31%, thus, only the higher levels were quantified. In consequence, the results for TCE may not be robust.

Most commercial uses of CTC were phased out by 1986 due to this chemical's toxicity and persistence, and industrial emissions have been limited under the Clean Air Act Amendments of 1990 (ATSDR 2005). (Previously, CTC had been used in medical treatment and as a component in fire extinguishers, fumigants and pesticides.) Currently, CTC use is

permitted only in a few industrial processes for which there are no effective substitutes. CTC is globally distributed at generally low levels with spatial little variation, except near contaminated source areas where levels increase. The variation of CTC exposures among the RIOPA participants was limited, and little was explained by the available variables (see Section 3.4).

In summary, the most common and significant determinants of personal VOC exposures were city (Houston usually had higher levels), wind speed (negative association), AER (negative), number of rooms (negative), presence of an attached garage (positive), self-pumping gas (positive), and employment (varied). Different determinants of personal exposures have been found for other pollutants in RIOPA, e.g., gardening was associated with higher levels of acetaldehyde and acetone, and sweeping with higher $PM_{2.5}$ levels (Liu et al., 2007; Meng et al., 2009).

3.2.4. Determinants of indoor VOC concentrations—An analysis parallel to that performed for personal samples was conducted for the indoor VOC measurements. Given the correlation between indoor and personal exposure measurements, it is not surprising that many of the same factors were identified as determinants (Supplemental Tables S10 to S12). Most of the VOCs were affected by city and several household characteristics. Among household characteristics, AER was negatively associated with indoor levels of toluene, *m*,*p*-xylene, *o*-xylene, styrene, TCE, PERC, chloroform, *d*-limonene, α-pinene and β-pinene. Larger houses (more rooms) were associated with decreased concentrations of benzene, toluene, *m*,*p*-xylene, *o*-xylene, styrene, 1,4-DCB, *d*-limonene and α-pinene. BTEX (except for toluene) and MTBE increased with an attached garage. Again, city effect varied by VOC, although Houston had the highest levels of VOCs except for MTBE, TCE, and PERC. (These were highest in Elizabeth).

Two meteorological factors were negatively associated with indoor VOC levels: ambient relative humidity with toluene, ethyl-benzene, *m*,*p*-xylene, *o*-xylene, styrene, chloroform and β-pinene; and wind speed with ethylbenzene, *m*,*p*-xylene, *o*-xylene, MTBE, styrene and PERC. Wind speed is expected to dilute outdoor concentrations from local sources, and to affect AERs as noted earlier. Outdoor relative humidity may be a surrogate for seasonal effects and weather, e.g., precipitation, possibly representing effect of fronts or low pressure systems with good dispersion or effective cleansing. Another meteorological factor, indoor temperature, showed opposite effects on two indoor VOCs, benzene and chloroform. Higher indoor temperatures were associated with lower benzene, but higher chloroform levels, which may be due increased volatilization (see Section 3.2.2).

3.2.5. Determinants of outdoor VOC concentrations—Outdoor concentrations were affected by city and three meteorological variables (Supplemental Tables S13 to S15). Ambient relative humidity was negatively associated with concentrations of benzene, ethylbenzene, *m*,*p*-xylene, *o*-xylene, MTBE, styrene, and β-pinene levels. Wind speed was negatively associated with concentrations of benzene, toluene, ethylbenzene, *m*,*p*-xylene, *o*-xylene, MTBE, styrene, TCE, PERC, and α-pinene. Effects of city and outdoor temperature depended on the VOC. For example, Houston had the highest concentrations for benzene,

m,p-xylene and β-pinene, which may be due to nearby petrochemical industries (Weisel et al., 2005b).

3.2.6. Common determinants of personal, indoor and outdoor concentrations

—Two factors affected personal, indoor and outdoor levels: city and wind speed. Three factors affected both personal and indoor levels: AER, number of rooms, and attached garage. That five common factors affected concentrations of most personal and indoor VOC measurements suggests the significance of indoor concentrations. However, outdoor sources can be important (Sexton et al., 2007), and the source-oriented exposure fractions showed outdoor sources were responsible for most exposure for all but four VOCs.

3.3. Assumption of linearity

The assumption of linearity for the continuous covariates in the LMMs (wind speed, ambient relative humidity, indoor temperature, AER, and time spent indoors at home) was evaluated using partial residual plots, which account for effects of all other covariates. Plots for wind speed and AER suggested some non-linearities with log-transformed VOC concentrations (Fig. 2 A, C, and E). Several transformations of these variables were attempted, and near-linear relationships were achieved using the reciprocal of wind speed and the logarithm of AER (Fig. 2B, D, and F). Inverse wind speed can be supported based on dilution or mass balance principles (for sources with emission rates that are independent of the wind speed). For buildings with internal emission sources, the AER is proportional to the air flow through the building, so again the reciprocal of the AER is expected to be linearly related to indoor concentrations. However, indoor concentrations are affected by many factors, and AERs are measured with error. The log AER, rather than 1/AER would tend to diminish the effect of both very large and very small AERs, and the fit with this transformation suggests that measured AERs included some outliers. Still, the expected relationship was seen, i.e., indoor concentrations of VOCs with strong indoor sources (e.g., chloroform and *d*-limonene) decreased as AERs increased (Supplemental Table S11).

3.4. Model validation

The estimated fraction of variance (R^2) attributable to fixed-effect variables in the LMMs for each VOC and each sample type (personal, indoor, outdoor) is shown in Supplemental Table S16. For personal exposures, R^2 ranged from 0.003 (CTC) to 0.40 (β-pinene); for indoor measurements, R^2 ranged from 0.09 (toluene) to 0.42 (PERC); and for outdoor concentrations, R^2 values were from 0.17 (1,4-DCB) to 0.65 (PERC). Generally, more variance was explained for the outdoor measurements. VOCs with specific emission sources, e.g., PERC (dry cleaners) and α-pinene (cleaning products and freshener), had the largest R^2 among the 15 VOCs; this applied to all three sample types. In contrast, VOCs used in many commercial products and that were also components of exhaust and other sources, e.g., toluene, had small R^2 across the three sample types. The LMMs explained only a portion of the variance in the dataset. While some of the variance is random and some is due to errors in measurement and model specifications, it is likely that the LMMs are incomplete models in the sense that other (unknown) variables and other (also unknown) interactions among the variables affect exposure. Further, effects of short-term activities,

e.g., cooking, may not be observable with 48-h integrated measurements. However, low R^2 values do not invalidate the identification or significance of the determinants.

3.5. Strengths and limitations

The analysis of the extended and comprehensive RIOPA dataset, which includes outdoor, indoor and personal measurements of 15 VOCs along with over 500 other variables used as candidate factors, advances the understanding of VOC exposure and exposure determinants. Strengths of the analysis included the use of LMMs, the repeated measurements for available participants, and the nested analysis, which allowed estimates of individual differences from average levels for specific variables (Krueger and Tian, 2004; Wu, 1996). The time fractions help to understand the participants' activity pattern, and to estimate the contribution of VOC sources to exposures.

The limitations of the dataset included missing data, which decrease sample size and statistical power. Two methods were used to address these issues. First, variables with sample sizes less than 400 (> 150 missing cases) were excluded from LMMs. Second, the use of multiple imputation was evaluated, and results showed that for the models tested, impacts of missing data would not be substantial. We also note that models for personal exposures explained less variance (lower R^2) than outdoor and indoor models, probably due to the number and complexity of factors (especially behaviors) that affect an individual's exposure. A final limitation of the study is the representativeness of the study sample. RIOPA data were collected in three U.S. cities that have specific emission sources, and homes near outdoor emission sources were over-sampled (Weisel et al., 2005b). Also, RIOPA used a convenience sample, which led to a number of demographic and other differences. However, most findings correspond to those in other studies using regional or national data, and thus most results appear relevant.

4. Conclusions

Determinants of personal exposures of VOCs in the RIOPA study included city, personal activities (e.g., pumping gas and visiting dry cleaners), household characteristics (e.g., AERs, number of rooms, attached garages), and meteorology (e.g., wind speed). Similar determinants were found for indoor concentrations. Most of these determinants were consistent with previous studies, e.g., BTEX and attached garages, and PERC and visiting dry cleaners. Several new determinants were identified, including city, other family members showering, and residence size. With the exception of four VOCs with strong indoor sources, most exposure resulted from outdoor sources. Further investigation using a more representative population and a wider suite of VOCs would extend and generalize results.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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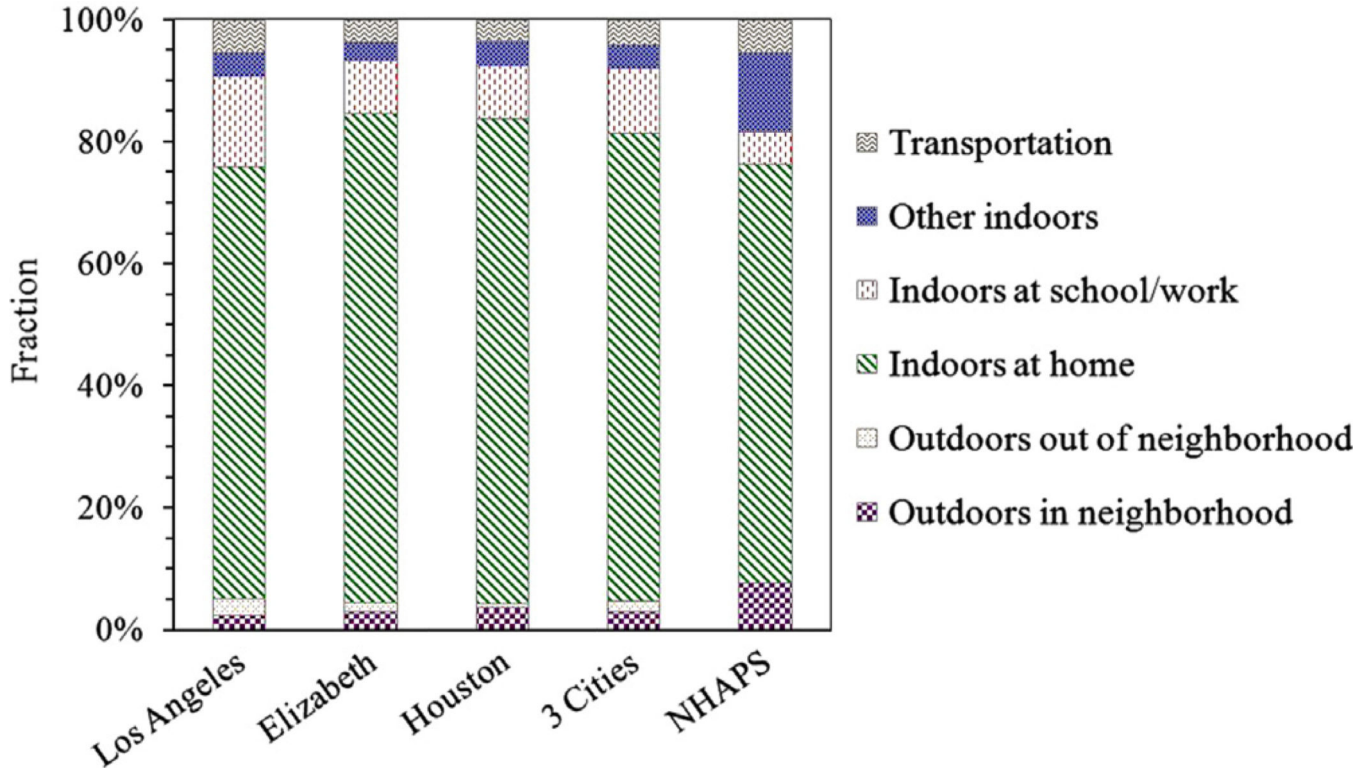

Fig. 1.

Mean time-spent fractions for RIOPA (by city) and National Human Activity Pattern Survey (NHAPS) participants (Klepeis et al., 2001).

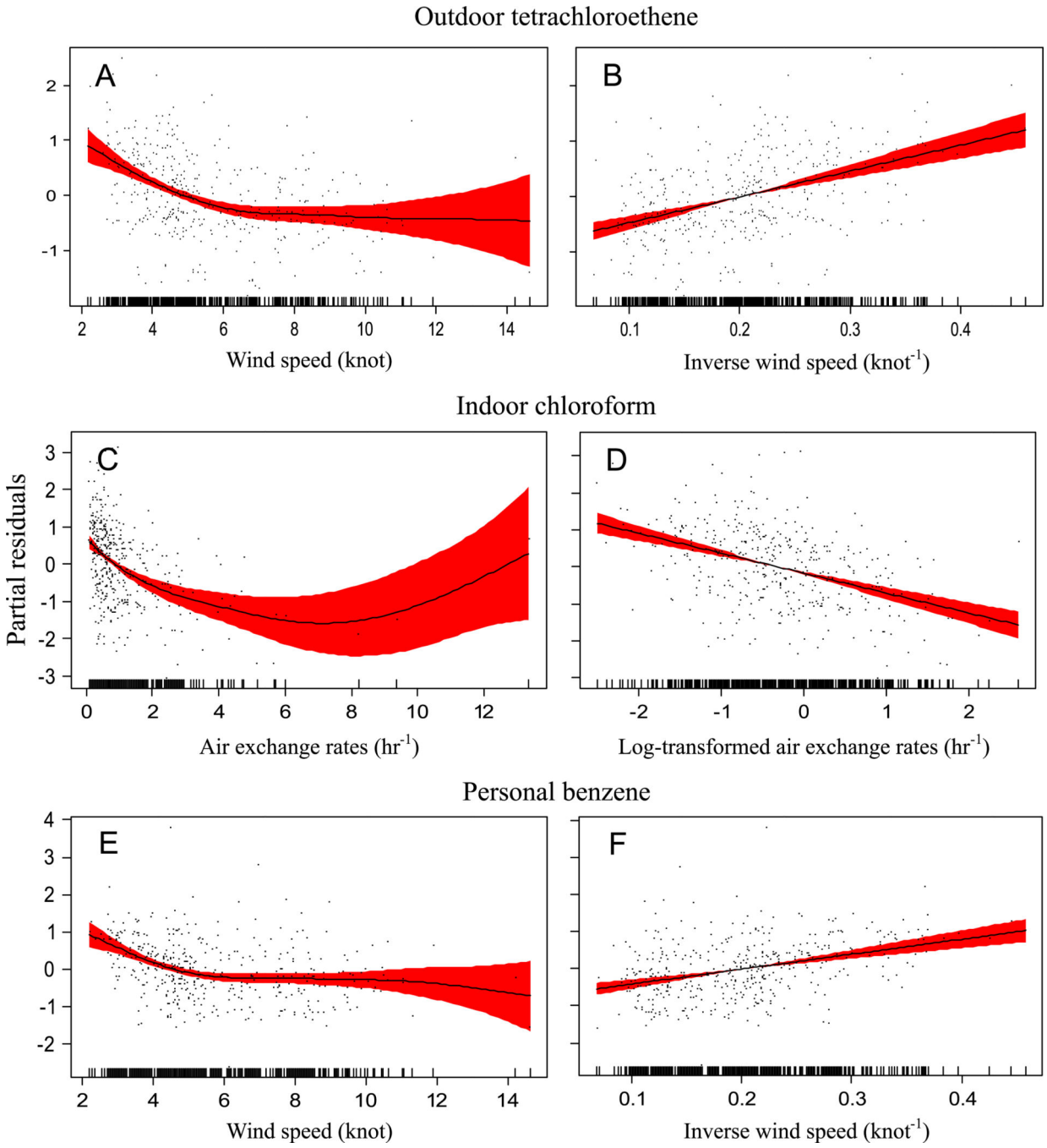

Fig. 2.
Partial residual plots of linear mixed-effect models for selected VOCs.

Table 1

Determinants of VOC exposures in previous and the present studies.

Determinants	Benzene	Toluene	Ethylbenzene	*m,p*-Xylene	*o*-Xylene	MTBE	Styrene	1,4-DCB	TCE	PERC	Chloroform	CTC	*d*-Limonene	α-Pinene	β-Pinene
Personal activities															
Contact with chlorinated water								m			A, C,M		M	M	M
Cooking	L	L, m	m		m						L				
Cycling/walking		E	E	E	E										
Keep pets											m				m
Near vehicle or engines	D, E, G	D	A, D, E	A, D, E	A, D, E		D			A					
Polish/wax furniture				j	j			M							
Pump gas/near gasoline	E, K, M	J.K	E, J, K, M	E, J, K, M	E, J, K, M	M									
Renovate house		M											M		
Smoke or near ETS	A, B, C, D, G, H, k	B, D, e, H	B, D, H	B, D, H	B, D, H		A, B, D								
Start in/presence of attached garages	F, G, H, J, K, M	F, H, J, M	F, G, H, J, K, M	F, H, J, K, M	F, H, J, K, M	H, M				H					
Time spent at home		m					m								
Time spent in closed cars									M						
Undertake arts and crafts		E	E	E	E										
Use air cleaning devices				M	M							M			
Use deodorizers and mothballs								A, C, H				m			
Use gas heating/gas stove	D, G, M	D.j	D	D	D	D	D						M		
Use paint and other solvents	H	H, K	G, H, K, M	H, J, K, M	H,J, K						K				
Use perfume						m									
Visit dry-cleaner or near dry-cleaned clothes										A, C, H, K, M					
Socioeconomic factors															
Age											i, k				
City/region*	1, m	1	1, m	1, m		m	m	m	m	m	m	m	m	m	m
Education/parental education	k				1			k							
Non-Hispanic white	h, k	h	h	h	h	h		h, k			h, i, k				

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Determinants	Benzene	Toluene	Ethylbenzene	*m,p*-Xylene	*o*-Xylene	MTBE	Styrene	1,4-DCB	TCE	PERC	Chloroform	CTC	*d*-Limonene	α-Pinene	β-Pinene
Male	K		K	K	K						k				
Machine-related jobs/work in a factory	H	H	G, H	H	H										
Ownership of the house											m				
Unemployed										m			M		
Environmental factors															
AER		m	m	m	m					m	m		m	m	m
Ambient RH										m	m				m
Furniture refinisher in neighborhood								M							
Existence of a fireplace							G					M			
Existence of a swim pool											H, 1			M	
Existence of a well/use well water									M		h	m			
Indoor temperature	m								m						
Live in an apartment/mobile home	L										1				
Near commercial street/highway						H		H		H					
Number of floors	m					m									
Number of rooms	m						m	m			m		m	m	
Open windows/doors	f, h, j, k	f, h, j, k	f, h	f, h	f, h, m	f	f, m	f, m		f, h	f, h, i, k		f	f	f
Restaurants or bakery in neighborhood								M	m						
Vinyl, asbestos or other siding									M						
Wind speed	m		m	m	m	m				m					
Years lived in home	h	h	h	h	h										

A, Wallace et al. (1989); b, Edwards et al. (2001); c, Wallace (2001); d, Kim et al. (2002); e, Hinwood et al. (2007); f, Sexton et al. (2007); g, Delgado-Saborit et al. (2009); h, D'Souza et al. (2009); i, Riederer et al. (2009); j, Symanski et al. (2009); k, Wang et al. (2009); 1, Byun et al. (2010); m, the present study.

Capital letters indicate increased exposure, and lower case indicates decreased exposure; *, no increasing or decreasing trends.

MTBE, methyl tert-butyl ether; 1,4-DCB, 1,4-dichlorobenzene; TCE, trichloroethylene; PERC, tetrachloroethene; CTC, carbon tetrachloride.

Table 2

Outdoor, indoor and personal VOC concentrations in RIOPA (μg m^{-3}).

voc	Outdoor (*n* = 540)				Indoor (*n* = 539)				Personal (*n* = 544)			
	Mean	SD	50th	95th	Mean	SD	50th	95th	Mean	SD	50th	95th
Benzene	2.13	2.02	1.68	5.13	3.54	5.22	2.19	10.20	3.64	5.31	2.39	10.73
Toluene	6.78	6.48	3.56	18.93	15.26	24.74	10.39	39.55	19.12	37.31	12.42	50.10
Ethylbenzene	1.22	1.15	0.91	2.99	2.55	4.80	1.47	7.77	2.78	5.13	1.68	7.45
m,*p*-Xylene	3.47	3.67	2.42	9.98	7.39	16.07	4.05	22.49	8.07	15.49	4.42	22.60
o-Xylene	1.40	3.63	0.96	3.22	2.49	4.84	1.47	7.44	2.87	5.59	1.72	7.92
MTBE	8.13	10.08	5.32	22.32	11.96	27.63	6.02	36.14	14.77	42.67	7.14	42.05
Styrene	0.49	0.60	0.42	1.19	1.48	4.29	0.42	5.12	1.55	4.31	0.42	5.32
1,4-DCB	2.09	17.25	0.46	3.59	69.88	307.48	1.40	334.28	56.83	229.37	1.88	304.56
TCE	0.28	0.29	0.22	0.80	0.99	7.29	0.22	1.74	1.44	10.74	0.22	2.37
PERC	0.95	1.32	0.61	3.11	1.85	4.53	0.82	6.03	7.17	112.35	0.89	6.82
Chloroform	0.32	0.99	0.21	0.75	1.86	2.97	0.94	5.97	4.25	52.49	1.04	6.36
CTC	0.66	0.21	0.64	0.99	0.71	0.99	0.62	1.11	0.80	2.44	0.62	1.06
d-Limonene	1.90	6.22	0.64	5.77	31.59	108.47	9.95	103.05	41.14	238.90	11.77	112.15
α-Pinene	1.24	3.82	1.02	1.80	7.14	14.76	2.67	26.83	6.85	16.25	2.88	22.48
β-Pinene	0.86	0.96	0.51	1.10	4.95	11.08	1.27	20.77	5.53	13.07	1.52	21.46

SD, standard deviation; 50th, the 50th percentile; 95th, the 95th percentile; MTBE, methyl tert-butyl ether; 1,4-DCB, 1,4-dichlorobenzene; TCE, trichloroethylene; PERC, tetrachloroethene; CTC, carbon tetrachloride.

Table 3

Fractions of personal VOC exposures in RIOPA *(n—484)*.

voc	Median F_{home_C}				Median $F_{outdoot\_C}$				Median F_{home_S}	Median $F_{outdoor\_S}$
	All	CA	NJ	TX	All	CA	NJ	TX	All	All
Benzene	0.72	0.64	0.76	0.73	<0.01	<0.01	<0.01	<0.01	0.05	0.61
Toluene	0.66	0.63	0.67	0.68	<0.01	<0.01	<0.01	<0.01	0.18	0.38
Ethylbenzene	0.69	0.64	0.68	0.73	<0.01	<0.01	<0.01	<0.01	0.15	0.48
m,*p*-Xylene	0.68	0.64	0.67	0.75	<0.01	<0.01	<0.01	<0.01	0.13	0.48
o-Xylene	0.69	0.65	0.67	0.71	<0.01	<0.01	<0.01	<0.01	0.1	0.5
MTBE	0.66	0.63	0.58	0.72	<0.01	<0.01	<0.01	<0.01	<0.01	0.67
Styrene	0.74	0.72	0.79	0.72	<0.01	<0.01	<0.01	<0.01	<0.01	0.58
1,4-DCB	0.72	0.67	0.73	0.76	<0.01	<0.01	<0.01	<0.01	0.29	0.23
TCE	0.74	0.66	0.74	0.8	<0.01	<0.01	<0.01	0.01	<0.01	0.76
PERC	0.71	0.69	0.75	0.71	<0.01	<0.01	<0.01	<0.01	0.03	0.56
Chloroform	0.74	0.74	0.7	0.81	<0.01	<0.01	<0.01	<0.01	0.57	0.16
CTC	0.75	0.72	0.74	0.79	0.01	<0.01	0.01	0.02	<0.01	0.81
d-Limonene	0.71	0.72	0.67	0.71	<0.01	<0.01	<0.01	<0.01	0.6	0.05
α-Pinene	0.78	0.79	0.74	0.81	<0.01	<0.01	<0.01	<0.01	0.45	0.22
β-Pinene	0.76	0.76	.0.73	0.78	<0.01	<0.01	<0.01	<0.01	0.19	0.34

$F_{home\_C} = (C_{home} \times T_{home})/(C_{personal} \times T_{total})$; $F_{outdoor\_C} = (C_{outdoor} \times T_{neighborhood})/(C_{personal} \times T_{total})$; $F_{home\_S} = ((C_{home}–C_{outdoor}) \times (T_{home}))/(C\text{-personal} \times T_{total})$; $F_{outdoor\_S} = (C_{outdoor} \times (T_{home}+T_{neighborhood}))/(C_{personal} \times T_{total})$.

CA, Los Angeles in California; NJ, Elizabeth in New Jersey; TX, Houston in Texas; MTBE, methyl tert-butyl ether; 1,4-DCB, 1,4-dichlorobenzene; TCE, trichloroethylene; PERC, tetrachloroethene; CTC, carbon tetrachloride.

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Table 4

Results of linear mixed-effect models for personal exposure to gasoline-related VOCs.

Variable	Group/unit	Benzene		Toluene		Ethylbenzene		*m,p*-Xylene		*o*-Xylene		MTBE		Styrene	
		β	SE	β	SE	β	SE	β	SE	β	SE	β	SE	β	SE
Intercept		**2.21**	**0.41**	**3.74**	**0.37**	**1.41**	**0.42**	**2.23**	**0.37**	**0.78**	**0.29**	**1.82**	**0.32**	**1.09**	**0.33**
Visit	1	−0.03	0.07	0.12	0.09	−0.14	0.08	−0.08	0.08	−0.07	0.07	0.06	0.10	0.07	0.08
	2	Reference		Reference		Reference		Reference		Reference		Reference		Reference	
City	Los Angeles	**0.83**	**0.12**	0.08	0.11	**0.37**	**0.14**	**0.29**	**0.14**	−0.06	0.13	**0.35**	**0.16**	**0.23**	**0.11**
	Elizabeth	**0.37**	**0.14**	0.06	0.13	−0.16	0.18	−0.25	0.19	−0.17	0.17	0.07	0.20	−0.11	0.10
	Houston	Reference		Reference		Reference		Reference		Reference		Reference		Reference	
Attached garage	No	**0.19**	**0.09**	**0.72**	**0.25**	**0.36**	**0.12**	**0.36**	**0.12**	**0.35**	**0.11**	**0.36**	**0.12**	−0.42	0.25
Cooking	No			**0.22**	**0.09**	**0.17**	**0.08**	0.15	0.09	**0.20**	**0.08**				
Education	Less than HS	0.15	0.12												
	High school	−0.08	0.10												
	> College	Reference													
Ethnicity	White					−0.13	0.15	−0.23	0.16	−0.21	0.14				
	Mexican					0.19	0.19	0.07	0.19	0.12	0.17				
	Hispanic					0.30	0.19	0.27	0.20	**0.35**	**0.18**				
	Other					Reference		Reference		Reference					
Heating fuel	Electricity	0.20	0.18												
	Gas	**0.42**	**0.16**												
	Oil and wood	Reference													
Indoor temperature	°C	**0.04**	**0.01**												
Inverse wind speed	$knot^{-1}$	**4.20**	**0.53**			**3.16**	**0.69**	**2.84**	**0.71**	**2.54**	**0.62**	**5.86**	**0.84**		
Log-transformed AER	h^{-1}			**0.30**	**0.05**	**0.17**	**0.06**	**0.21**	**0.06**	**0.14**	**0.05**	−0.09	0.07		
Number of floors		**0.15**	**0.04**									**0.20**	**0.06**		
Number of rooms		**0.10**	**0.03**											**0.09**	**0.02**
Open doors or windows	No									**0.22**	**0.10**			**0.20**	**0.09**
Pumping gas	No	**0.16**	**0.08**			**0.24**	**0.11**	**0.22**	**0.11**	**0.28**	**0.10**	**0.34**	**0.13**		
Renovation in the past year	No			**0.30**	**0.10**										
Time spent in home	min			**0.0002**	**0.0001**	−0.0002	0.0001							**0.0003**	**0.0001**

Variable	Group/unit	Benzene		Toluene		Ethylbenzene		*m,p*-Xylene		*o*-Xylene		MTBE		Styrene	
		β	SE	β	SE	β	SE	β	SE	β	SE	β	SE	β	SE
Unemployed	No											0.23	0.12		
Using air cleaning devices	No					−0.27	0.18	**0.42**	**0.18**	**0.38**	**0.16**	−0.35	0.20		
Using nail polish remover	No			−0.29	0.17	**0.39**	**0.16**	**0.33**	**0.17**						
Wore powder, spray or perfume	No											**0.41**	**0.12**		

n = 400 to 530 depending on models.

AER, air exchange rate; HS, high school; MTBE, methyl tert-butyl ether.

For dichotomous variables, the reference group is “Yes”.

p-Value < 0.05 shown in bold type.

Table 5

Results of linear mixed-effect models for personal exposure to odorant-related VOCs.

Variable	Group/unit	1,4-DCB		Chloroform		*d*-Limonene		α-Pinene		β-Pinene	
		β	SE	β	SE	β	SE	β	SE	β	SE
Intercept		**3.50**	**0.78**	**1.34**	**0.47**	**3.62**	**0.39**	**2.42**	**0.25**	**1.57**	**0.44**
Visit	1	**0.33**	**0.14**	0.15	0.09	0.10	0.15	**0.18**	**0.07**	0.08	0.10
	2	Reference		Reference		Reference		Reference		Reference	
City	Los Angeles	**1.10**	**0.30**	**0.45**	**0.16**	**0.82**	**0.19**	**0.71**	**0.13**	**1.16**	**0.15**
	Elizabeth	**0.81**	**0.31**	−0.06	0.17	**1.12**	**0.22**	**0.59**	**0.14**	**1.06**	**0.17**
	Houston	Reference		Reference		Reference		Reference		Reference	
Air conditioning	No	**0.54**	**0.23**					**0.51**	**0.10**	−0.20	0.13
Ambient relative humidity	%			**0.010**	**0.005**					**0.011**	**0.005**
Furniture refinisher in neighborhood	No	**1.30**	**0.50**								
Waxing or polishing furniture	No	**0.81**	**033**								
Keeping dogs or cats	No							0.15	0.10	**0.29**	**0.11**
Log-transformed AER	h^{-1}			**0.41**	**0.06**	**0.33**	**0.08**	**0.40**	**0.05**	**0.31**	**0.07**
Not using fresheners or candles	No									0.32	0.18
Number of rooms		**0.14**	**0.07**	**0.12**	**0.04**	**0.13**	**0.04**	**0.10**	**0.03**		
Open doors or windows	No	**0.42**	**0.20**							0.22	0.12
Other family members took showers	No			**0.39**	**0.15**	**0.80**	**0.18**	**0.41**	**0.12**	**0.35**	**0.14**
Outdoor swimming pool or hot tub	No							**0.31**	**0.13**		
Using heating at	<64°F	**0.76**	**0.26**								
	64 to 70 °F	−0.03	0.24								
	>70°F	Reference									
Ownership of the house	No			**0.30**	**0.14**						
Pets indoors	No			**0.32**	**0.12**						
Renovation in the past year	No					**0.45**	**0.15**				
Restaurants or bakery in neighborhood	No	**0.63**	**0.27**								
Unemployed	No					**0.35**	**0.16**				
Using a clothes washer	No	**0.53**	**0.19**								
Using dishwashers	No			**0.25**	**0.13**						

Variable	Group/unit	1,4-DCB		Chloroform		*d*-Limonene		α-Pinene		β-Pinene	
		β	SE	β	SE	β	SE	β	SE	β	SE
Using other heaters (non-CHS)	No					**0.55**	**0.27**				

n = 393 to 433 depending on models.

AER, air exchange rate; CHS, central heating system; 1,4-DCB, 1,4-dichlorobenzene.

For dichotomous variables, the reference group is “Yes”.

p-Value < 0.05 shown in bold type.

Table 6

Results of linear mixed-effect models for personal exposure to dry-cleaning and Industrial-related VOCs.

Variable	Group/unit	TCE		PERC		CTC	
		β	SE	β	SE	β	SE
Intercept		**0.79**	**0.42**	−0.48	0.49	**0.64**	**0.23**
Visit	1	**0.18**	**0.07**	**0.19**	**0.10**	−0.01	0.03
	2	Reference		Reference		Reference	
City	Los Angeles	**0.66**	**0.14**	**0.58**	**0.18**	**0.17**	**0.07**
	Elizabeth	**1.23**	**0.14**	**0.54**	**0.24**	−0.11	0.07
	Houston	Reference		Reference		Reference	
Ambient relative humidity	%			**0.01**	**0.01**		
Ethnicity	White			−0.12	0.19		
	Mexican			**0.48**	**0.23**		
	Hispanic			0.06	0.24		
	Other			Reference			
Having a fireplace	No					**0.13**	**0.07**
Indoor temperature	°C	**0.03**	**0.01**			0.01	0.01
Inverse wind speed	$knot^{-1}$			**4.87**	**0.83**		
Log-transformed AER	h^{-1}			**0.20**	**0.07**		
Not using fresheners or candles	No					**0.20**	**0.08**
Restaurants or bakery in neighborhood	No	**0.26**	**0.13**				
Source of household water	Public	**0.58**	**0.27**			**0.50**	**0.14**
Sweeping indoors	No			0.19	0.12		
Time spent at closed cars	min	**0.0018**	**0.0005**				
Unemployed	No			**0.42**	**0.13**		
Using air cleaning devices	No					**0.19**	**0.08**
Vinyl, asbestos or other siding	No	**0.25**	**0.13**				
Visited dry cleaners during past week	No			**0.63**	**0.15**		

n = 400 to 446 depending on models.

AER, air exchange rate; TCE, trichloroethylene; PERC, tetrachloroethene; CTC, carbon tetrachloride.

For dichotomous variables, the reference group is “Yes”.

p-Value < 0.05 shown in bold type.