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Relationship between vapor intrusion and human exposure to trichloroethylene

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Abstract

Trichloroethylene (TCE) in groundwater has the potential to volatilize through soil into indoor air where it can be inhaled. The purpose of this study was to determine whether individuals living above TCE-contaminated groundwater are exposed to TCE through vapor intrusion. We examined associations between TCE concentrations in various environmental media and TCE concentrations in residents. For this assessment, indoor air, outdoor air, soil gas, and tap water samples were collected in and around 36 randomly selected homes; blood samples were collected from 63 residents of these homes. Additionally, a completed exposure survey was collected from each participant. Environmental and blood samples were analyzed for TCE. Mixed model multiple linear regression analyses were performed to determine associations between TCE in residents' blood and TCE in indoor air, outdoor air, and soil gas. Blood TCE concentrations were above the limit of quantitation (LOQ; ≥0.012 μg/L) in 17.5% of the blood samples. Of the 36 homes, 54.3%, 47.2%, and >84% had detectable concentrations of TCE in indoor air, outdoor air, and soil gas, respectively. Both indoor air and soil gas concentrations were statistically significantly positively associated with participants' blood concentrations (p=0.0002 and p=0.04, respectively). Geometric mean blood concentrations of residents from homes with indoor air concentrations of >1.6 $\mu g/m^3$ were approximately 50 times higher than geometric mean blood TCE concentrations in participants from homes with no detectable TCE in indoor air (p<.0001; 95% CI 10.4 – 236.4).

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This study confirms the occurrence of vapor intrusion and demonstrates the magnitude of exposure from vapor intrusion of TCE in a residential setting.

Keywords

vapor intrusion; trichloroethylene; groundwater plume; indoor air exposure; soil gas exposure; volatile organic compounds

Introduction

The 2013 Priority List of Hazardous Substances ranks trichloroethylene (TCE) 16 out of 275, based on a combination of its frequency, toxicity, and potential for human exposure at National Priority List (NPL) sites. [1] The Environmental Protection Agency (EPA) has characterized TCE as a human carcinogen due to its ability to cause kidney tumors in humans, and there also is evidence that TCE contributes to other cancers, such as non-Hodgkin lymphoma and liver cancer. [2] TCE has been associated with cardiac malformations in animal studies, and findings from several epidemiologic investigations suggest that there might be an association between TCE exposure and congenital cardiac defects. [2–4]

As a volatile organic compound (VOC), TCE has the ability to volatilize from shallow groundwater into soil where it can rise up into homes and other buildings through utility lines, piping, cracks, or other openings in the floor or slab. [5] Transport through such openings can occur by molecular diffusion or convection caused by under-pressurization of a home relative to the soil (e.g., by use of exhaust fans or through temperature differences between indoor air and soil). [6] This process, often referred to as vapor intrusion, can be of concern for buildings located above or near VOC-contaminated groundwater or soil because of its potential ability to impact indoor air quality. [5] Since the early 1970s, it has become increasingly apparent that indoor air quality can negatively impact people's health. [7] In some cases, breathing contaminated air indoors can be a dominant exposure pathway. [7] Between home and work, most people in the U.S. spend 90% or more of their time indoors, and groups potentially most susceptible to the effects of indoor air pollutants, including small children, the elderly, and the chronically ill, often spend even more time indoors. [8, 9] Indoor air pollution can be a serious health risk that may cause both immediate and long-term health effects, ranging from respiratory irritation and neurological symptoms to heart disease and cancer. [9]

Because indoor air pollutants can impact human health, vapor intrusion may be an important but often overlooked exposure pathway. In most instances, contaminant concentrations inside homes due to vapor intrusion may be low. However, efforts to lower energy costs by reducing indoor/outdoor air exchange rates could further exacerbate potential effects from indoor air pollutants by allowing pollutants to accumulate inside the home. [7]

An Agency for Toxic Substances and Disease Registry (ATSDR) review of public health reports at 121 vapor intrusion sites found that TCE was present in the indoor air at more

sites that were deemed to be a public health hazard than any other VOC. [10] Due to the potential for vapor intrusion to result in human exposure, the purpose of this study was to determine whether living above TCE-contaminated groundwater could result in human exposure. We examined associations between TCE concentrations in various environmental media and human exposure as measured by TCE concentrations found in blood.

Materials and methods

Study design and selection criteria

This investigation was an observational community-based multilevel study. Households from four neighborhoods located in Grand Prairie, Texas, were randomly selected for inclusion in the study. Three neighborhoods were located above known TCE groundwater plumes. The fourth neighborhood was not situated above a TCE groundwater plume.

All neighborhoods in the study were fairly similar in terms of housing structure and race/ethnicity, with over 50% of participants living in each of the four neighborhoods being of Hispanic origin. Groundwater in all four neighborhoods was approximately 15 to 25 feet below ground surface, and homes were predominately pier-and-beam construction with a crawlspace under the home. Within each neighborhood, maintenance of ventilation under the homes varied; some homes had crawlspace vents that were well-maintained, others had vents that were blocked or partially blocked.

Stratified random sampling was used to select households in the four neighborhoods to participate in the study. Because many selected households did not reply to the invitation to participate, multiple random samples were generated, for a total of 150 selected households (36 to 40 households from each neighborhood). Letters were mailed to each of the selected households requesting participation in the study. Investigators followed up with non-respondents in person. Households that agreed to participate were asked to provide one or two volunteers (adults or children <6 years of age) to take part in the investigation.

Indoor air, outdoor air, soil gas, and tap water samples were collected from each residence at the same time as blood samples were collected from participants. Participants also completed a brief exposure survey. All biological and environmental samples, as well as the accompanying participant survey, were collected from participants between the dates of April 3 and June 1, 2009.

Informed consent

This study was approved by the Texas Department of State Health Services' (DSHS) Institutional Review Board (IRB#1) to ensure the protection of participants' safety, rights, and welfare. The Center for Disease Control and Prevention's (CDC) role was limited to analysis of coded specimens and was determined to not constitute engagement in human subjects research. Each participant signed a consent form, which included information on the purpose of the investigation, the expected time commitment, the procedures that would be involved, any potential risks or discomforts, and the potential benefits to the participant or the neighborhood. The consent form also stated that any personal identifying information

would be kept confidential. Contact information was provided for participants to contact DSHS investigators with any questions or concerns about the consent form or any of the investigation's procedures.

Sample collection and analysis

A 10-mL venous blood sample was collected from each participant into blood collection tubes that had been extensively cleaned to remove all traces of residual VOCs. [11–12] Samples were stored and shipped cold but not frozen by overnight delivery to the CDC National Center for Environmental Health (NCEH) laboratory in Atlanta, Georgia, for analysis. Samples were analyzed no later than 4 weeks after collection. Blood samples were analyzed for TCE by headspace solid phase microextraction/gas chromatography/mass spectrometry (SPME/GC/MS) using published methods validated to minimize contamination and loss biases. [12–13] This approach, which involves preconcentrating the TCE using headspace SPME, is one of the most sensitive analytical approaches, and the limit of quantitation (LOQ) for TCE achieved with this method was 0.012 μg/L. [13] Raw instrument results were reported down to 0 μg/L. Samples with concentrations below the LOQ were quantified by extrapolating a linear least-squares fit determined using 1/x weighting of the 4 lowest calibrants.

Indoor and outdoor air samples were collected and analyzed using procedures and methods based on US EPA Method TO-17.[14] A pair of sampling systems (a low flow rate sample and a high flow rate sample) was used for both indoor and outdoor air sampling. The two indoor samples and two outdoor samples were collected simultaneously. The air sampling systems consisted of a calibrated air pump, vinyl tubing, a low flow adapter, and a glass sorbent tube filled with a minimum of 0.11 mg of Carbopack™ B. The sample pump for the low flow rate sample system was calibrated for a nominal volumetric flow rate of 12.5 mL/min. The high flow rate sample pump was calibrated for a nominal volumetric flow rate of 50.0 mL/min. The sample time for every air sample was 80 minutes, producing sample volumes of 1.0 and 4.0 liters for each pair of air samples. Air samples were collected indoors, typically on the kitchen table, and outdoors within 15 meters (m) of the house approximately 1 m above the ground. Once sample collection was completed, the flow rate through the sample system was validated using a calibrated volumetric flow meter. Validated sorbent tubes were stored in a protective casing, wrapped in aluminum foil, and stored in an electric cooler to maintain a storage temperature below 4°C. All air samples were analyzed using thermal desorption followed by gas chromatography and mass spectrometry (TD/GC/MS). The GC/MS was operated in selective ion mode (SIMS). By quantifying TCE in samples using this method, detection limits, background noise and spectral interferences were minimized. Both high and low flow rate samples for indoor and outdoor air TCE results were reported as either not detected (below the limit of detection (LOD), <0.12 $\mu g/m^3$), detected but not quantifiable (greater than the LOD but below the limit of quantitation (LOQ), > 0.12 $\mu g/m^3$ but < 0.17 $\mu g/m^3$), or quantifiable (detected above the LOQ, >0.17 $\mu g/m^3$).

A single soil vapor sample was collected in the yard of each study household. The soil vapor sampling system consisted of a “pushpoint” soil probe, Teflon® tubing, a sorbent tube, and

a calibrated sample pump. Soil vapor samples were collected by hammering an aluminum rod into the ground to a depth of 0.6 m within 15 m of the house. The rod was pulled from the hole leaving a 0.6 m deep sample shaft. The pushpoint sampler was inserted into the test hole and clay was used to seal the top of the sample shaft around the pushpoint probe. For the first nine houses the nominal sample flow rate was 50 mL/min and the sample time was 80 minutes, yielding a total sample volume of 4.0 L. This sampling regimen resulted in a LOD of 0.12 μg/m^3 and an LOQ of 0.17 μg/m^3. Because mass loadings above 2.0 μg were seen in several samples, the method was altered for the remaining 25 houses. The mass loading was reduced by lowering the nominal sampling rate to 15 mL/min and the sample time to 20 minutes. At this reduced flow rate the total sample volume decreased to a nominal value of 300 mL, resulting in an LOD of 1.6 μg/m^3 and an LOQ of 2.3 μg/m^3. Once sample collection was completed, the flow rate through the sample system was validated using a calibrated volumetric flow meter. The sorbent tube was stored and analyzed using the same procedure described above for air samples.

Tap water samples were collected from a non-aerated faucet in each participant's home. Tap water was collected into 20-mL precleaned vials containing 125 μL of a phosphate/thiosulfate solution that quenches residual chlorine and buffers the sample to a pH of 6.5. [15] Before sample collection, the cold water tap was opened fully for approximately 1 minute to flush the supply line. The tap water sample was then collected from that stream with the flow rate reduced. Water vials were filled completely to eliminate any headspace in the sample, in order to prevent volatilization of VOCs. Samples were stored and shipped cold, but not frozen, by overnight delivery to the NCEH laboratory in Atlanta, Georgia, for analysis. Tap water samples were analyzed by SPME/GC/MS for TCE using the same method as the blood samples. [12–13] Samples were analyzed no later than 4 weeks after collection. The LOQ for TCE achieved with this method was 0.012 μg/L.

In addition to collecting environmental and biological samples, participants were asked to complete an exposure survey, which included questions about basic demographic characteristics of each participant, the structure of the house, sources of drinking water, and potential exposures to TCE through hobbies, occupation, or use of chemicals and fuels in the home.

Statistical methods

Descriptive statistics on TCE concentrations from both biological and environmental sample results were performed. For summary statistics, each pair of indoor sample results and outdoor sample results was combined to provide one indoor air value and one outdoor air value per home. For a given air sample type (e.g., indoor air samples), if both low flow rate and high flow rate sample results were below the LOD, the overall result for the home was a “nondetect” (<LOD). Similarly, if TCE was detected (above the LOD) in both samples, but below the LOQ, the overall result was categorized as a “detect” for that home. If the low flow rate and high flow rate samples both yielded quantifiable results (above the LOQ), an average of the two values was taken as the result. However, if the low flow rate and high flow rate results were of different categories (e.g., the low flow rate sample was a nondetect,

while the high flow rate sample was a detect), the high flow rate sample's category/value was taken as the averaged result for the home.

Mixed model multiple linear regression analyses were performed to determine associations between blood TCE concentrations and TCE concentrations in various environmental media (indoor air, outdoor air, soil gas, and tap water), as well as a few other demographic and home-related characteristics obtained from the exposure survey. Because up to two residents from each randomly selected household could participate in the study, mixed models were used to adjust for correlation of the data by household.

The outcome variable in these analyses, blood TCE concentration (in μg/L), was analyzed as a log-transformed continuous variable to meet linear regression assumptions. For this transformation, a value of 0.0001 μg/L was assigned to all 0.000 μg/L blood TCE results, to avoid errors in taking the log of 0. Raw instrument results for blood TCE data were used for all observations, including those values below the LOQ (0.012 μg/L). Although raw instrument values below the LOQ are less precise than values above this limit, they can be more precise than fixed imputed values (e.g., LOD/ √2), which are not representative of the true variation. Potential predictor variables in the multivariable model included soil gas TCE concentration, indoor air TCE concentration, outdoor air TCE concentration, tap water TCE concentration, , as well as variables from the exposure survey (participant age, sex, ethnicity (Hispanic vs. non-Hispanic), home foundation type (cement slab vs. pier-and-beam), and whether the home had been renovated in the past 6 months). The survey variables were included as potential predictor variables to account for basic demographics of the study population as well as relevant home characteristics.

Univariable regression analyses were first conducted using each of these potential predictor variables individually, and variables were only included in the multivariable analysis if their univariable analysis yielded a p-value of 0.25 or less. Soil gas TCE concentration, tap water TCE concentration, and participant age were modeled as continuous variables. Categorical variables were used for both the indoor and outdoor air concentrations rather than continuous variables because a high percentage of indoor air and outdoor air TCE concentration results were below the LOQ (over 50% of results were either nondetects or detects, but had no quantifiable values). The TCE indoor air variable, which included both low and high flow rate sample results, was separated into six categories. The lowest category (reference) contained no detectable results (both low and high flow rate samples had results below the LOD) and the highest category had quantifiable values of ≥1.6 $\mu g/m^3$ for both samples, which is the 95th percentile TCE concentration reported in homes with no known vapor intrusion. [16] The TCE outdoor air variable, which also included both low and high flow rate sample results, was divided into four categories. These categories ranged from no detectable TCE concentrations (< LOD; the reference category) to quantifiable concentrations (≥LOQ; the highest category). The other potential predictor variables were modeled as dichotomous variables. A manual backwards elimination process was performed to arrive at the final model. Predictor variables with a p-value of <0.10 remained in the final model. A linear mixed model likelihood-ratio R^2 value for the final multivariable model was

also obtained, using the `lmmfit' package in R (version 2.14.1). All other analyses were conducted using SAS (version 9.2) and Stata/IC (version 10).

We hypothesized that soil gas, indoor air, and even potentially outdoor air TCE concentrations would be correlated, which would be consistent with the occurrence of vapor intrusion in one or more neighborhoods. Therefore, linear correlations between indoor air, outdoor air, soil gas, and blood TCE concentrations all measured as continuous variables were assessed. Indoor air and outdoor air TCE variables were recoded as continuous measures by imputing values for results that were below LOD or LOQ values. To eliminate duplicate indoor air, outdoor air, and soil gas values in those homes with two participants, a correlation matrix was constructed using the results of only one randomly selected resident per household. Pearson's correlation coefficient values (r) and corresponding p-values were calculated.

Results and discussion

Of the 150 randomly selected households across the four neighborhoods, 36 agreed to participate in the study (24%). A total of 64 individuals took part in the study, which included 57 adults and 7 children. Summary statistics are presented in Table 1.

Blood TCE concentrations

Blood samples were obtained from 63 of the 64 study participants; 1 adult declined to provide blood. Blood TCE concentrations above the LOQ (≥0.012 μg/L) were present in 11 of the 63 blood samples (17.5%). Blood TCE concentrations for participants ranged from below the LOQ (≤0.012 μg/L) to 0.728 μg/L (Table 1).

Exposures to TCE in this study were greater than what has been observed in the general population; a higher percentage of study participants had blood TCE concentrations above the LOQ (17.5%) than did a sample of US residents who participated in the 2003–2004 National Health and Nutrition Examination Survey (NHANES) data (2.3%). Additionally, the raw median blood TCE level observed among study participants was significantly higher than the raw median concentration observed in the general population (2003–2004 NHANES participants; $p<0.001$). This finding is consistent with study participants' exposure to TCE due to vapor intrusion. However, health effects at the blood TCE levels observed in this investigation are unlikely, because concentrations were all less than the estimated blood TCE biological exposure index (BEI), a concentration below which workers who are repeatedly exposed over a lifetime are not likely to experience any adverse health effects.[17, 18]

Indoor air TCE concentrations

Valid indoor air samples were collected from 35 of the 36 homes; one high flow-rate sample was not collected from a home due to a pump failure. Eleven of these homes (31.4%) had indoor air TCE concentrations that were above the LOQ (0.17 $\mu g/m^3$), ranging from 0.20 to 112 $\mu g/m^3$. Eight homes (22.9%) had concentrations of TCE that were above the LOD (0.12

μg/m^3), but were below the LOQ (0.17 μg/m^3). For the remaining 16 homes (45.7%), indoor air TCE concentrations were below the LOD (0.12 μg/m^3).

While indoor air concentrations of TCE in most of the homes in this study were similar to those reported for homes not impacted by vapor intrusion, [16] some homes had much higher concentrations. In a 2009 evaluation of 18 studies with indoor air data from homes not impacted by vapor intrusion, Dawson and McAlary found the 90th and 95th percentile indoor air TCE concentrations to be 0.9 μg/m^3 and 1.6 μg/m^3, respectively. [16] In our study, the 90th and 95th percentile concentrations were 4.7 μg/m^3 and 22.8 μg/m^3, respectively. In six homes, TCE indoor air levels exceeded the EPA reference concentration (RfC, 2.0 μg/m^3) and ATSDR's minimal risk level (MRL, 2.1 μg/m^3). [2, 3] The EPA offered vapor mitigation systems to potentially affected residents.

Outdoor air TCE concentrations

Outdoor air samples were collected from all 36 homes. Seven of these homes (19.4%) had outdoor air TCE concentrations equal to or greater than the LOQ (0.17 μg/m^3), ranging from 0.17 to 0.61 μg/m^3. One of these homes had both high and low flow rate concentrations above the LOQ; only the high flow rate concentrations were above the LOQ for the remaining six homes. Ten homes (27.8%) had TCE concentrations that were between the LOD (0.12 μg/m^3) and the LOQ (0.17 μg/m^3). The remaining 19 homes (52.8%) had outdoor air TCE concentrations below the LOD (≤ 0.12 μg/m^3).

Although nationwide, statistically based surveys of outdoor air TCE concentrations were not available to provide representative background concentrations, a large amount of data are available on ambient air concentrations of TCE. Based upon data collected in 2006 from 258 monitors in 37 states, ambient air concentrations of TCE range from 0.03–7.73 μg/m^3, with a median of 0.13 μg/m^3. [19] Outdoor air results from our study ranged from below the LOQ (≤ 0.12 μg/m^3) to 0.605 μg/m^3, with a median of ≤ 0.12 μg/m^3, indicating that outdoor TCE air concentrations are comparable to or lower than ambient air levels measured across the United States.

Soil gas TCE concentrations

Valid soil gas samples were obtained for 34 of the 36 homes, with 25 of the homes sampled at a flow rate of 15 mL/min and the other nine sampled at 50 mL/min. Soil gas samples could not be properly collected at two homes because of a significant rainfall event. For the 25 homes sampled at the 15 mL/min flow rate, 18 homes (72%) had soil gas TCE concentrations greater than the LOQ (2.3 μg/m^3), ranging from 2.8 μg/m^3 to 54,300 μg/m^3 with a median concentration of 3.5 μg/m^3. Three homes (12%) had soil gas TCE concentrations between the LOD (1.6 μg/m^3) and the LOQ (2.3 μg/m^3). The remaining four homes had soil gas concentrations of TCE below the LOD. Of the nine homes where soil gas was sampled at a flow rate of 50 mL/min, eight homes (88.9%) had concentrations above the LOQ (≥0.17 μg/m^3), ranging from 0.35 μg/m^3 to 2,090 μg/m^3 with a median concentration of 0.56 μg/m^3. The ninth home had a TCE concentration between the LOD (0.12 μg/m^3) and the LOQ (0.17 μg/m^3).

Soil gas TCE concentrations above the LOD were observed in all four study neighborhoods, including the neighborhood not known to be over a TCE groundwater plume. With one exception, however, soil gas concentrations in the neighborhood not above a known TCE plume were low. It is unknown why TCE was found in soil gas in this neighborhood, which was ¼ to 1½ miles from the other neighborhoods in the study.

Tap water TCE concentrations

Tap water samples were collected from all 36 homes. All of the homes were connected to the public water supply system, which was not impacted by the groundwater contamination. Only one tap water sample had a TCE concentration above the LOQ (≥0.012 μg/L); this home had a concentration of 0.04 μg/L, which was slightly above the LOQ value, but well below the EPA's maximum contaminant level (MCL) for TCE in drinking water of 5 μg/L. [20] This tap water sample was taken from the home with the highest indoor air and soil gas TCE concentrations (112.1 $\mu g/m^3$ and 54,300 $\mu g/m^3$, respectively). Based on the relatively high concentrations of TCE in the indoor air, some TCE may have partitioned into tap water at this home. Henry's Law predicts the equilibrium concentration for TCE in the tap water to be 0.35 μg/L. The observed tap water concentration of 0.04 μg/L is well below the equilibrium value and consistent with the rapid transfer of TCE between the air-water interface for water flowing from a kitchen faucet. [21]

Associations with blood TCE concentrations

Mixed model multiple linear regression analyses yielded a final model that included only indoor air and soil gas TCE concentrations as significant predictors of TCE exposure (Table 2). Both indoor air and soil gas TCE concentrations were significantly positively associated with participants' blood TCE concentrations (p=0.0002 and p=0.04, respectively). TCE in tap water was not used as a variable in the analyses because no TCE was found in household water, with the exception of one home where entrainment was suspected. All other potential predictor variables, including exposure survey variables, were removed from the model because they did not contribute to the model at the α=0.1 level (Table 2).

Ratio estimates of geometric mean blood TCE concentrations for each indoor air concentration compared to the reference level (non-detect for both high and low flow samples) are shown in Table 2. After taking correlation of the data by household into account, the geometric mean blood TCE concentration of residents living in homes with indoor air TCE concentrations of >1.6 $\mu g/m^3$ was approximately 50 times higher than the geometric mean blood concentration in participants living in homes with no detectable TCE (concentrations <LOD) in their indoor air (p<.0001; 95% CI 10.4 – 236.4). Residents with indoor air TCE concentrations that were quantifiable (≥0.17 $\mu g/m^3$) but less than 1.6 $\mu g/m^3$ had a geometric mean blood TCE concentration that was approximately 18 times higher than that of participants with no detectable TCE in their indoor air (p=.01; 95% CI 2.1 – 149.3) (Table 2). A test for trend showed that overall, indoor air TCE concentrations were significantly associated with blood TCE concentrations (p=0.0002). Also, after taking correlation of the data by household into account, each 1-$\mu g/m^3$ increase in soil gas TCE

concentration was estimated to result in a 0.006% increase in blood TCE concentration (p=0.04; 95% CI 1.000002 – 1.00012) (Table 2).

When taking correlation by household into account, 56% of the variation in blood TCE concentrations can be explained by indoor air and soil gas TCE concentrations (likelihood-ratio R^2=0.56). Although these results were statistically significant and the magnitude of the association was quite large, confidence intervals around the geometric mean ratio estimates were extremely wide, indicating that the estimates were not very precise (Table 2). These wide confidence intervals likely are due to small sample sizes; only two homes (4 individuals) in the study had indoor air TCE concentrations above the LOQ, but ≤1.6 μg/m^3 for either high or low flow samples, and 6 homes (11 individuals) had indoor air TCE concentrations >1.6 μg/m^3.

Correlation analysis findings

Soil gas and indoor air TCE concentrations were highly correlated when evaluated as continuous variables (r=0.96) (Table 3). In this study, the six highest average indoor air TCE concentrations observed (ranging from 2.5 to 112.0 μg/m^3) all occurred within residences with soil gas TCE concentrations >10 μg/m^3. These results are consistent with the process of vapor intrusion, where TCE vapors in soil can infiltrate homes, thus affecting TCE concentrations in indoor air. [5, 6] Repeated random samples of one resident per household (n=36 residents) indicated that correlation results did not vary much by sample. The strong collinearity observed between soil gas levels and indoor air levels was primarily driven by one household that contained both the highest soil gas and indoor air TCE concentrations. Although soil gas and indoor air TCE concentrations were strongly linearly correlated when analyzed as continuous variables, in our multiple regression analysis, indoor air TCE concentration was measured as a categorical variable (because of the large number of results below the LOQ), which in this instance eliminated any problems with multicollinearity of these two variables in the regression model.

Strengths and limitations

One of the strengths of this study was the use of stratified random sampling to select participants. Stratified sampling ensured that all four study neighborhoods were represented in fairly equal proportions, and random selection was used as a means to obtain a representative sample from each of the neighborhoods.

Although groundwater data were not collected, blood, indoor air, outdoor air, soil gas, and tap water were examined. By analyzing several elements of the vapor intrusion pathway, this study was able to evaluate the contribution of multiple media on blood TCE concentrations. Vapor intrusion studies that are limited by only looking at soil gas and indoor air concentrations assume exposure concentrations based on a number of participant-dependent variables.

This study is unique in that it incorporated biomonitoring in addition to customary environmental monitoring to quantify TCE concentrations present in people, and provides empirical evidence that vapor intrusion can lead to increased TCE exposure in people living

in affected residences. A further strength of this study was that low detection limits for blood TCE and environmental media concentrations were achieved because of the analytical techniques used. Although 82.5% of study participants did not have blood TCE concentrations above the LOQ (0.012 μg/L), this analysis limit is within a factor of two of the blood TCE level expected in individuals inhaling TCE at the EPA's RfC of 2.0 $\mu g/m^3$; [22] thus, we would expect to see TCE in the blood of people exposed to TCE above the RfC.

One of the limitations of this study was that indoor air concentrations presented are only representative of TCE concentrations during the time the samples were collected. Factors such as changes in weather conditions and subsequent changes in soil gas migration and home ventilation can affect the concentration of TCE in indoor air. Indoor and outdoor air samples for different homes were collected at different times of day and over multiple days that were characterized by different weather conditions.

In addition, guidance documents caution about the contribution of “background sources” of TCE, such as household products and building materials, on indoor air measurements. [23–26] Although participants were asked about potential TCE sources in the exposure survey, these products were not removed from the home in this study. However, the strong correlation between indoor air concentrations and soil gas concentrations suggest that vapor intrusion is a principal source of exposure, at least in several individuals.

Conclusion

Blood, indoor air, and soil gas data collected in this study indicate that vapor intrusion was occurring and that some participants were being exposed to TCE. This study uniquely incorporated biomonitoring in addition to customary environmental monitoring to quantify TCE concentrations in individuals. A strong, clear association was observed between indoor air TCE concentrations and participants' blood TCE concentrations, as well as between soil gas TCE concentrations and blood TCE concentrations. These results are consistent with exposure to TCE resulting from vapor intrusion, and provide novel empirical evidence that vapor intrusion can lead to increased TCE exposure in people living in affected residences.

This study demonstrated that both soil gas and indoor air concentrations were associated with blood TCE concentrations, and also that soil gas concentrations were strongly correlated with indoor air concentrations. Therefore, measuring TCE in soil gas or indoor air could provide valuable information in determining the risk of human exposure to TCE through the vapor intrusion pathway without the need to collect biological specimens.

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Table 1

Summary of TCE concentration results collected from 64 individuals in 36 homes in Grand Prairie, TX.

Sample Matrix (n)	Range	Median	Limit of Detection (LOD)	Number Detected[a] (%)	Limit of Quantitation (LOQ)	Number Quantifiable[b] (%)
Blood (63 individuals)	<LOQ[c]-0.728 μg/L	<LOQ[d]	NA[e]	NA	0.012 μg/L	11 (17.5%)
Indoor Air (35 households)	ND[f]-112.0 μg/m^3	Detect	0.12 μg/m^3	8 (22.9%)	0.17 μg/m^3	11 (31.4%)
Outdoor Air (36 households)	ND-0.605 μg/m^3	ND	0.12 μg/m^3	10 (27.8%)	0.17 μg/m^3	7 (19.4%)
Soil Gas[g]						
9 households	Detect-2,090 μg/m^3	0.56 μg/m^3	0.12 μg/m^3	1 (11.1%)	0.17 μg/m^3	8 (88.9%)
25 households	ND-54,300 μg/m^3	3.5 μg/m^3	1.6 μg/m^3	3 (12%)	2.3 μg/m^3	18 (72%)
Tap Water (36 households)	<LOQ-0.04 μg/L	<LOQ	NA	NA	0.012 μg/L	1 (2.8%)

ND – Not Detected

NA – Not Applicable

For soil gas, the sample method was slightly altered after the first sampling trip, resulting in two different LOD and LOQ values.

[a] Number detected indicates the number of samples that had levels above the LOD, but the levels were below the LOQ.

[b] Number quantifiable indicates the number of samples that had levels above the LOQ.

[c] Results below the limit of quantitation (LOQ) are designated as <LOQ.

[d] Although raw instrument values were obtained for all blood TCE samples, the median value was below the LOQ and was thus denoted as <LOQ instead of listing the median raw instrument value.

[e] Only an LOQ was used for blood and tap water results; no LOD threshold was used.

[f] Results below the limit of detection (LOD) are designated as ND.

Table 2

Final model results for mixed model linear regression analysis. Parameter estimates for the two predictor variables left in the model, indoor air and soil gas, are shown.

	Geometric Mean Ratio[a]	95% CI	p
Indoor Air TCE Concentration Category (Low-Flow Sample/High-Flow Sample)			0.0002[b]
Nondetect/Nondetect	1.00 (Referent)	---	---
Nondetect/Detect	0.42	(0.09, 1.91)	0.25
Detect/Detect	0.91	(0.03, 31.6)	0.96
Detect/Quantifiable	0.95	(0.14, 6.30)	0.96
Quantifiable/Quantifiable (at least one value ≤ 1.6 μg/m^3)[c]	17.55	(2.06, 149.30)	0.01
Quantifiable/Quantifiable (both values ≥ 1.6 μg/m^3)	49.55	(10.39, 236.35)	0.00003
Soil Gas TCE Concentration	1.00006	(1.000002, 1.0001)	0.04

Nondetect = indoor air results were below the LOD (≤ 0.12 μg/m^3)

Detect = indoor air results were between the LOD (0.12 μg/m^3) and the LOQ (0.17 μg/m^3)

Quantifiable = indoor air results were above the LOQ (≥ 0.17 μg/m^3)

[a] Correlation within households has been accounted for in all ratio estimates. For indoor air, estimates shown are ratios of geometric mean blood TCE concentrations for each indoor air concentration category, compared to the referent indoor air category (nondetect in both low- and high-flow samples). For soil gas, the ratio increase in geometric mean blood TCE concentration for each 1-μg/m^3 increase in soil gas TCE concentration is shown.

[b] This is the p-value for the indoor air TCE variable as a whole (results from Type 3 analysis, used to test significance of the overall categorical variable).

[c] 1.6 μg/m^3 is the 95th percentile TCE concentration reported in homes with no known vapor intrusion. [16]

Table 3

Correlation matrix between air, soil and blood TCE concentrations. Pearson correlation coefficients and p-values are shown.

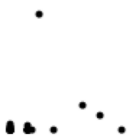
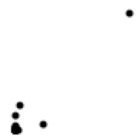
	Indoor Air	Outdoor Air	Soil Gas	Blood
Indoor Air	1.000			
Outdoor Air	0.210 (p=0.227)	1.000		
Soil Gas	0.958 (p<0.0001)	0.074 (p=0.676)	1.000	
Blood	0.999 (p<0.0001)	0.193 (p=0.260)	0.963 (p<0.0001)	1.000

Table 4

Comparison of TCE indoor air concentrations at selected percentiles in this study with percentiles reported by Dawson and McAlary (2009).

Percentiles of sample distribution	Indoor air concentration (μg/m^3)	
	Grand Prairie study	Dawson & McAlary (2009)
25th percentile	< LOD (<0.12)	< LOQ*
50th percentile (median)	< LOQ (<0.17)	0.3
75th percentile	0.2	0.3
90th percentile	4.7	0.9
95th percentile	22.8	1.6
Maximum value	112.0	84.0

*Results from Dawson and McAlary [16] include data from 18 different studies of homes not impacted by vapor intrusion. These studies had various LOQ values ranging from 0.02 to 2.7 μg/m^3