Evaluation of Radon and Building Pressure Differences as Environmental Indicators for Vapor Intrusion Assessment

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ABSTRACT

Indoor air radon and differential pressure measurements are gaining increasing attention as potential cost-effective indicators for vapor intrusion (VI) assessment and decision-making. To provide better understanding of such environmental indicators, this study evaluates the correlation between indoor air volatile organic compounds (VOCs) sampling results and long-term indoor air radon and out-to-indoor air pressure differential monitoring results from two well-documented study houses. Data from one study house suggest high-level indoor air trichloroethylene (TCE) concentrations are likely to be measured during high-indoor-air-radon concentration periods. The median TCE concentration is 1.02 ug/m^3 for the days during which indoor air radon concentrations were greater than their 95th level, and is about 3x greater than the average TCE level for the whole monitoring period. However, such correlation is not evident in the other building where sewer VI pathway is believed a significant VI contributor. Increase in indoor air VOC concentrations is found in both buildings during periods when outdoor pressures were greater than indoor. Radon and TCE attenuations from subslab soil gas to indoor air are also studied using soil gas sampling data from one study building. The results show generally greater radon attenuations than TCE at same sampling locations over time. About 1/3 radon attenuations are more than 10x greater than TCE attenuations. These findings are expected to provide useful insight for future development of using environment indicators for VI assessment.

INTRODUCTION

Subsurface to indoor air vapor intrusion (VI) risk assessment often follows a multiple lines of evidence (MLE) approach and relies heavily on chemical analysis of point-in-time groundwater, soil gas, indoor air and/or outdoor air samples (ITRC, 2007; USEPA, 2015a; Eklund et al., 2018; Ma et al., 2020). Among these, indoor air sampling results of chemicals of potential concern, such as chlorinated volatile organic compounds (CVOCs), are often the most preferred when it comes to decision making. However, because of the significant range in temporal variability of indoor air CVOC concentrations observed at residential (Folkes et al. 2009; US EPA 2012 and 2015bc; Holton et al. 2013) and commercial and industrial buildings (Hosangadi et al., 2017;

Lutes et al. 2021a), the results of one or a few point-in-time indoor air samples can lead to incorrect decisions (Holton et al. 2013; Weinberg et al. 2014). Increasing indoor air sampling frequency can improve characterization of indoor contaminant exposure (Schuver et al. 2018), yet it quickly becomes unfeasible at sites where multiple buildings need to be evaluated. As such, determining the number of samples required to evaluate CVOC VI (CVI) risks is a common challenge amongst VI practitioners, regulators, and stakeholders. Indoor air radon concentrations, soil gas-to-indoor air radon attenuation factors (AF)s, and cross-building differential pressures have been proposed as potential environmental indicators or tracers for identifying when and/or where CVI exposures occur and for supporting data evaluation and decision making (McHugh et al. 2008; Schuver and Steck, 2015; DoD 2017; Schuver et al., 2018; Lutes et al., 2021b).

Radon intrusion is a common problem in North America and is often compared to CVI due to its similar soil to indoor air migration and exposure pathways (US EPA, 2012). Radon is formed by the decay of radium-226, which is produced from certain rocks with high uranium contents, including granites, volcanic rocks, and dark shales. Real-time, consumer-grade radon monitoring devices capable of hourly sampling frequencies or even higher frequencies are readily available and considerably less expensive than comparable CVOC indoor air monitoring devices. As such, indoor air radon monitoring datasets can be generated prior to CVOC sampling to help guide the location and timing of CVOC samples. Another potential use of radon gas monitoring is to estimate a building-specific attenuation factor for VOCs (McHugh et al., 2008; Schuver and Steck, 2015), when both subslab and indoor air samples are available and certain preconditions are satisfied. However, neither of these approaches have been validated. Positive trends between radon and CVOCs in indoor air have been reported in multiple studies (Mosely et al., 2008; Lutes et al., 2010; USEPA, 2012; Johnson et al., 2016; Lutes et al., 2021), but their positive correlations are not significant. Moreover, the potential limitations of using radon monitoring results are not well understood; spatial variability of subsurface radon and CVOCs, as well as the impact of conduit VI pathways, were not included in previous studies.

Pressure differences across the building envelope is recognized as a driving force for CVOCs entry (Hubbard et al. 1995; Guo et al., 2015; Holton et al., 2015; McHugh et al., 2012; USEPA, 2012, 2015b). Negative indoor pressure conditions can promote CVOC entry from the subsurface, which is why sustained pressure differences are believed to be a potential indicator of active CVI. Long-term building pressure control (BPC) studies concluded that negative building pressurization can create worst-case VI impact any time of the year (Guo et al., 2015; Holton et al., 2015). Yet, evaluation of the long-term correlation between outdoor to indoor air pressure differences and indoor air CVOC concentrations under natural conditions is inadequate. Luo (2009) simulated transient indoor air contaminant concentrations with fluctuating barometric pressure cycles, and suggested contaminant entry rates would increase during periods of negative building pressure. A recent study by Hosangadi et al (Hosangadi et al., 2017) reported field observation of increasing TCE concentration with increasing cross-slab pressure differentials, but it was less than 10 consecutive days of monitoring.

The goal of this paper is to gain a better understanding of the correlations between indoor air radon and CVOC concentrations, outdoor to indoor pressure differences, and indoor air VOCs concentrations, and to determine if these relationships can help evaluate the potential for use these indicators (i.e. IA radon concentrations and cross building envelope pressure differentials) for VI investigation. Therefore, long-term monitoring results of these samples from two well-studied research buildings are evaluated.

MATERIAL AND METHODS

Site description. The data analyzed in this study were collected from two well-studied research buildings: a two-story residential house and an unoccupied, unfurnished duplex. Both are described in detail in previous publications (Holton et al., 2013; Holton et al., 2015; Guo et al., 2015; USEPA, 2012). Important and relevant features are summarized below.

The single residential house, also known as SDM, overlies a groundwater plume with dissolved TCE concentrations ranging from approximately 10-50 μg/L-H_2O. An open-ended land drain lateral pipe connects the subslab area with the neighborhood land drain network present near the street. This important physical feature was discovered and confirmed to be a significant pathway for TCE vapor migration by Holton et al., 2015 and Guo et al., 2015. Elevated subslab TCE soil gas concentrations near the terminus of the land drain were recorded during long-term building negative pressurization testing (Guo et al 2015). This suggested that the alternative VI pathway doesn’t directly connect to building indoor air, but rather contaminant vapors enter the building by migrating through soil and subslab cracks. Figure 1 illustrates the sampling schematic and floor plan of SDM.

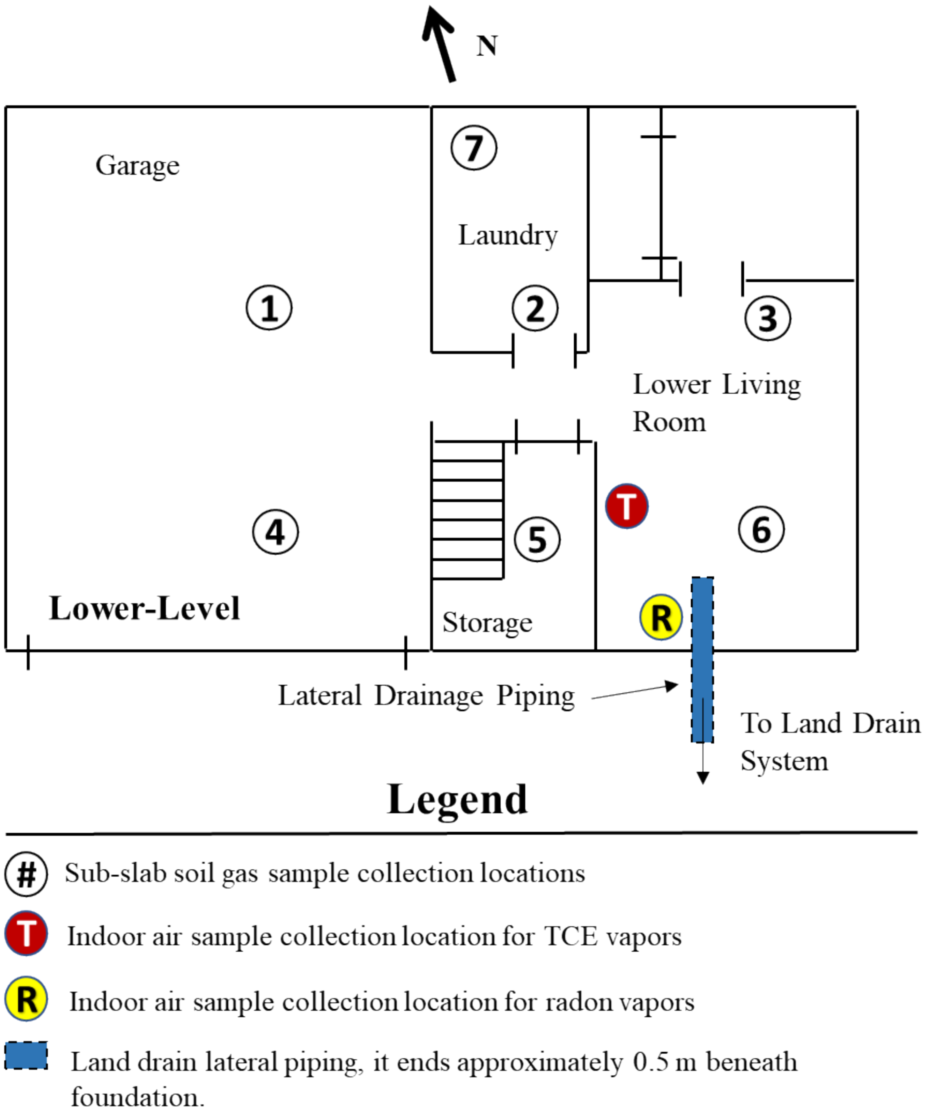

Figure 1. Sampling schematic and floor plan of SDM

The Duplex Research Building (DRB) was located near several potential VOC sources which included historic dry cleaner sites. In addition to the soil vapor intrusion pathway, McHugh et al. (2018) concluded from a tracer study that the sewer lines played an important role in transport of VOCs from subsurface sources into the duplex envelope. A detailed sampling schematic can be found in USEPA, 2015b (Figure 3-14).

Data collection and analytical methods. Both research structures were well-equipped and capable of collecting long-term, high-frequency data. Of interest were outdoor to indoor differential pressures, indoor air VOC concentrations, and indoor air radon concentrations. This study focuses on the data that were collected during periods when the structures were monitored under natural conditions without disturbance of other research activities that could change the natural, near-building and indoor air pressure conditions, such as building pressure manipulations, sub-surface VI mitigation system testing, and alternative pathway manipulations (only in SDM). Table 1 summarizes sampling periods and analytical methods from both sites.

Table 1. Summary of data collection and analytical methods from both study sites.

	Single Residential House (SDM)	**Duplex Research Building**
Data collection period(s)	2/1/2011 - 6/8/2012	8/12/2011 - 10/17/2011 12/1/2011 - 12/22/2011 12/29/2011 - 2/7/2012 1/16/2013 - 2/17/2013
Indoor air VOC sampling and analytical method	Indoor air samples were collected every 4 h using sorbent tubes and were analyzed using gas chromatograph (GC) and mass spectroscopy (MS). This method was capable of reporting TCE vapor concentrations as low as 0.034 ug/m^3. Soil gas VOCs concentrations were measured using GC/DELCD. This method was capable of reporting TCE vapor concentrations as low as 0.42	GC with an electron capture detector (ECD) was used to collect indoor air data. EPA (2015). Method was capable of reporting as PCE vapor concentrations as 0.7 ug/m^3..

	ug/m^3.Detailed information was reported by Holton et al.	
Indoor air radon sampling and analytical method	Indoor air radon concentrations were measured every 2 h using a Durridge RAD7 radon detector. Capable of measuring down to 0.5 pCi/L Soil gas radon concentrations were measured using the Durridge RAD 7 in a point-in-time mode	2nd floor indoor air radon concentrations were measured approximately every 10 min using AlphaGUARD radon detector.
Outdoor-to-indoor pressure differential	Differential pressure readings were monitored every 2 min using electronic differential pressure transducers (model P300−0.4″- D, Pace Scientific Inc., Mooresville, NC)	Differential pressure readings were monitored every 15 min by Setra Model 264 low differential pressure transducers.

RESULTS AND DISCUSSION

Long-term indoor air radon and outdoor-to-indoor pressure differences vs indoor air VOCs results. The datasets used in the following analysis are shown in Figures 2 and 3 from the two studies mentioned in previous sections. In the previous publications for these two studies, direct comparisons were made betweenpoint-in-time sampling results for indoor air VOCs concentrations and indoor air radon and outdoor-to-indoor pressure differential results (EPA 2012, SERDP-ER1686). Because air sampling for radon and/or pressure differentials and VOCs were not conducted simultaneously, the point-in-time comparison was done by pairing each VOCs sampling result to the radon and/or pressure monitoring results that were collected within the shortest time difference. This inevitably introduced errors to correlation analysis results. Moreover, indoor air VOC concentrations were not necessarily occurring simultaneously with radon and pressure. For example, Guo et al (2020) reported that during continuous building negative pressurization testing, it took more than nine air exchanges for indoor air TCE concentrations to reach a steady state after pressurization began.. In such instances as this, the 24-h averaged results were used to analyze correlations between radon and pressure differentials vs VOC concentrations. This is also more realistic for practice if using any of these indicators, since the goal is to identify certain days when VI could impose unacceptable risk.

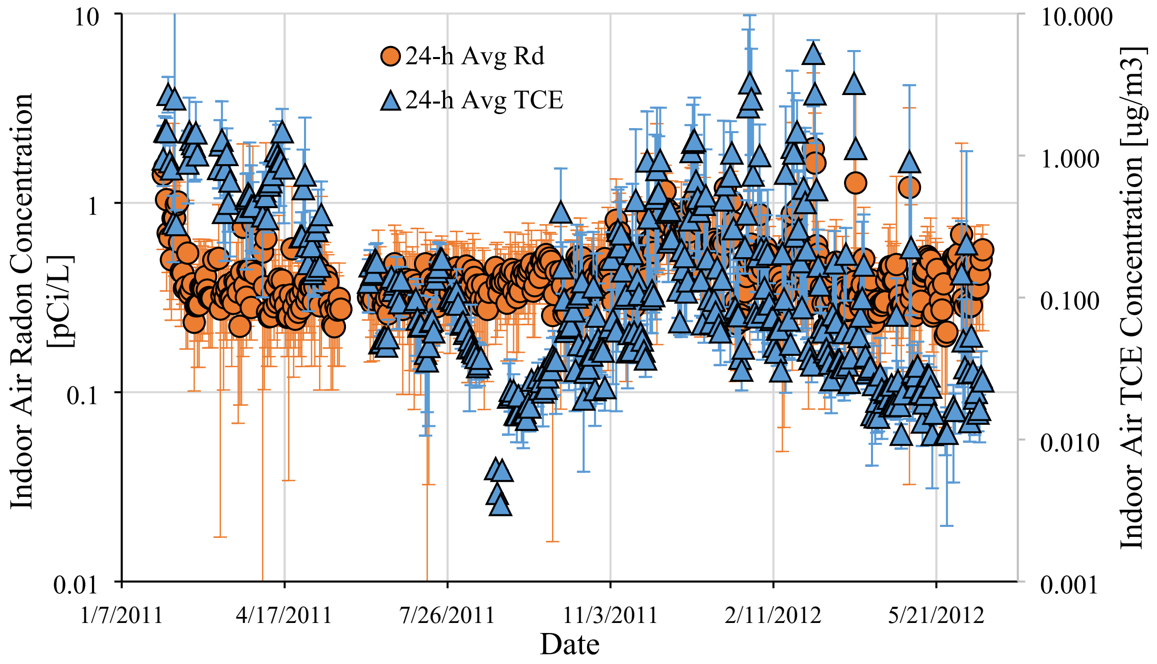

Figure 2. 24-h averaged indoor air radon and TCE concentrations data set from ASU study house.

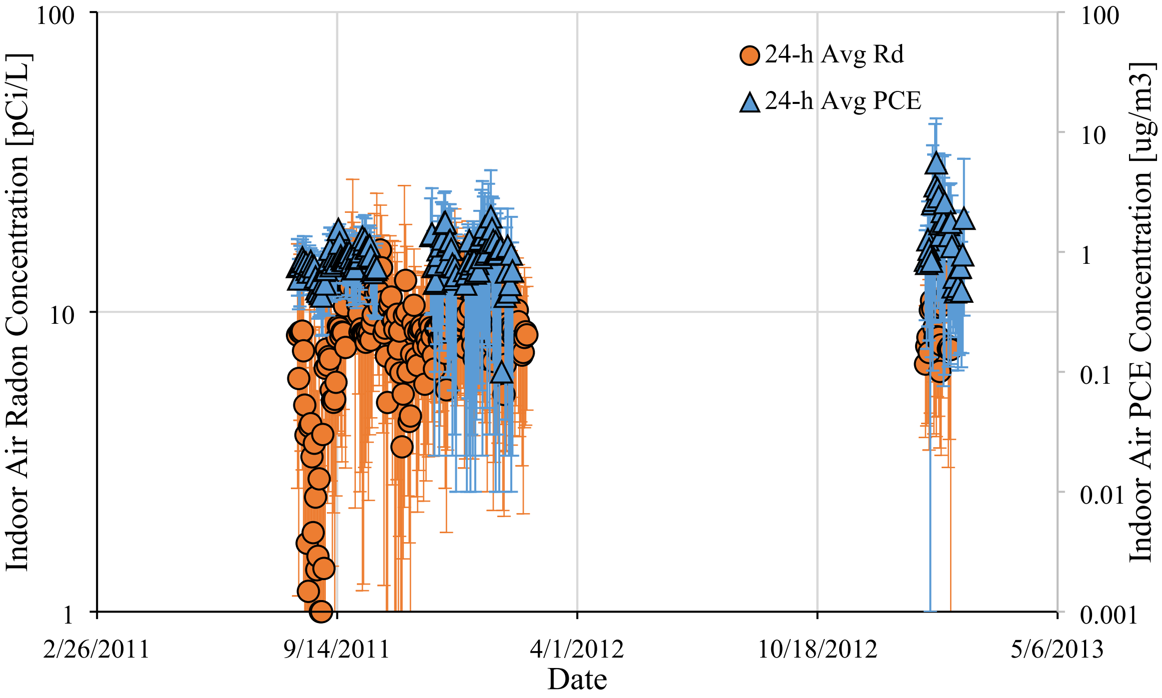

Figure 3. 24-h averaged indoor air radon and PCE concentrations data set from EPA study house.

Statistical analysis of 24-h averaged indoor air TCE concentrations were conducted for the days when the 24-h averaged indoor air radon concentrations exceed the minimum, 50th, 75th, 90th and 95th percentiles of the indoor air radon concentrations for both datasets. Those thresholds were generated based on radon concentration distributions for overall sampling periods in both study buildings. The TCE vs radon and PCE vs radon results are presented in Figure 4 for SDM and DRB, respectively.

The correlation between indoor air TCE and radon concentrations was evident for the SDM data set, as shown in Figure 2. The median TCE concentration increases with the indoor air radon level. The greatest median TCE concentration was 1.02 ug/m^3 for the 22 days during which the 24-h indoor air radon concentrations were all greater than 0.85 pCi/L (95th). In 21 of the 22 days of record, 24-h averaged TCE concentrations were greater than the overall averaged indoor air concentration (0.29 ug/m^3). In contrast, in about 75% of the whole 372 sampled days, 24-h averaged TCE concentrations were less than 0.29 ug/m^3. The results from SDM suggested that greater indoor air TCE concentrations were more likely to be measured when the indoor air samples were collected during high-radon concentration periods.

However, such correlation was not evident in DRB. The median values of indoor PCE concentrations ranged from 0.7 to 1.0 ug/m^3 despite radon concentrations. This was not surprising given the fact that the sewer line, which directly connects subsurface source to building interior, was believed to be a significant pathway for PCE transport (McHugh et al, 2008). Soil gas radon transport, however, did not migrate through the sewer pathway.

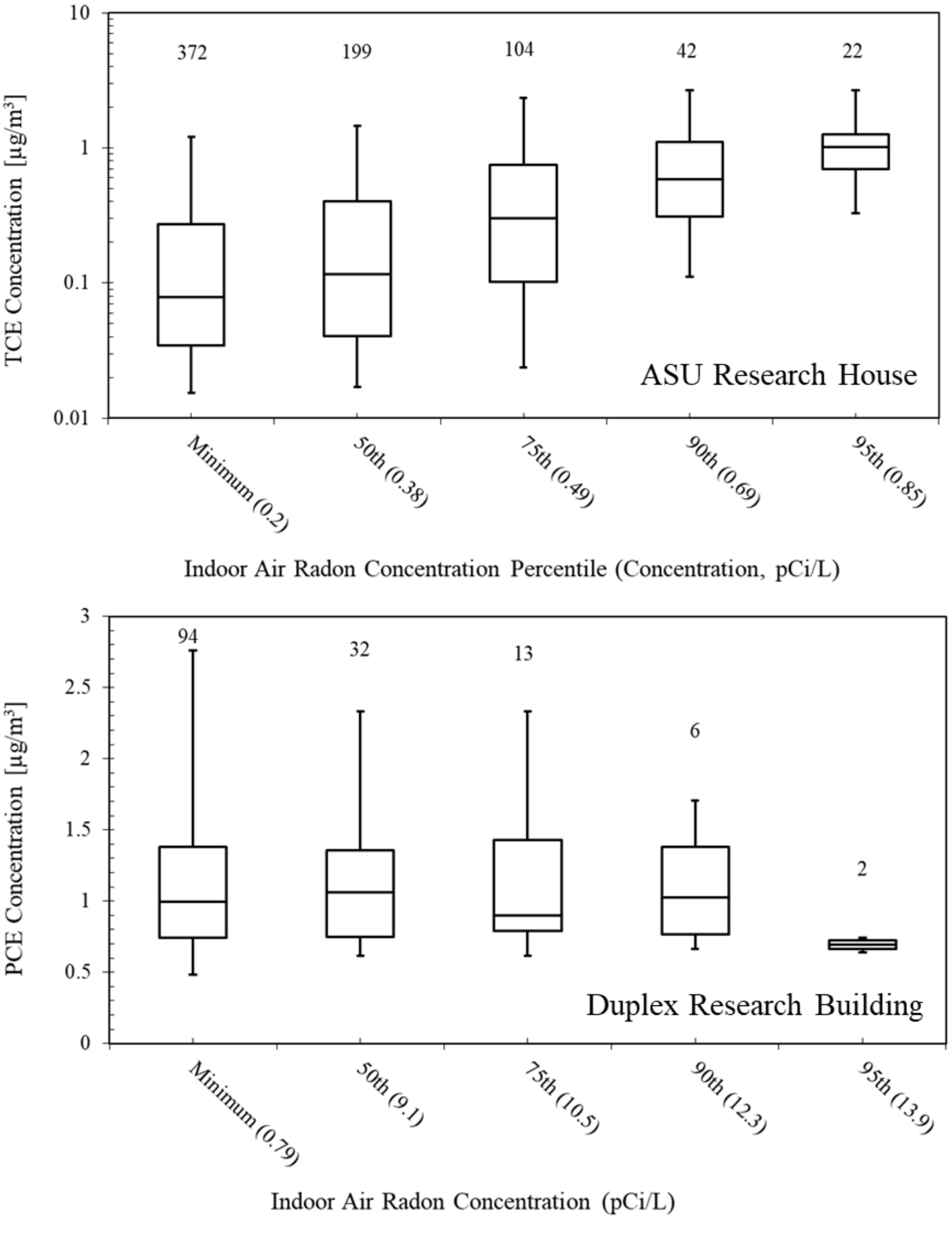

Figure 4. Statistical summary of 24-h average indoor air VOCs sampling results for the ASU research house (upper graph) and Duplex Research Building (lower graph) when 24-h average indoor air radon concentrations were greater than different

percentiles/concentrations. Error bars are 95th, 90th, 75th, 50th, and 5th percentile values for VOCs concentrations. Numbers on top of each box are days that the associated concentrations were observed.

Correlations between indoor air VOCs concentrations and outdoor-to-indoor pressure differentials were evaluated for both study buildings using a similar approach to that discussed above. Figure 5 shows the 24-h average TCE and PCE concentration distributions when 24h average outdoor-to-indoor pressure differentials are < 0 Pa (positive building pressure condition), 0 Pa – 75th percentile, 75th – 90th percentile and > 90th percentile of the whole data collection periods for SDM and DRB, respectively.

SDM data (Figure 5) showed that 24-h average indoor air TCE concentrations increased with building negative pressure differences. Median 24 h TCE indoor air concentration was 1.1 ug/m^3 for the days when 24-h outdoor-to-indoor air pressures differences were at least 1.2 Pa. That was about 20x greater than the median 24-h TCE vapor concentrations of positive building pressure periods under natural condition. This was consistent with the VI conceptual model for SDM which indicated that contaminant vapors were drawn into overlying buildings from the subslab by pressure-driven advective air flows. Increasing indoor air PCE concentrations with increasing outdoor-to-indoor pressure differentials were also found for DRB, as shown in Figure 5. However, median indoor air PCE concentrations under positive building pressure conditions (0.8 ug/m^3) was only about 30% greater than those under negative building pressure conditions. This again suggested the impact of sewer VI pathway in this building, since the migration of PCE vapors from sewer is usually not driven by outdoor-to-indoor pressure differentials, but rather the pressure gradient between building and sewer connection.

In summary, long-term indoor air radon concentration monitoring can be indicative for high-VI-risk periods. This is supported by the SDM data where over 90% of daily averaged TCE concentrations were found to be greater than the true mean when 24-h indoor air radon concentrations were greater than their 95th percentile value. However, this conclusion is only valid when VOCs and radon entry buildings were via a similar route. When preferential pathways like sewer was the primary VI pathway as shown in DRB data, indoor air radon concentration may not be as reliable an indicator for high-VI-risk periods. Similarly, cross-building pressure differences can be another valuable indicator for identifying high-VI-risk periods. Both SDM and DRB monitoring results showed an increase in indoor air VOC concentrations during periods when outdoor pressures were greater than indoor.   However, this correlation was less apparent when the sewer VI pathway existed (DRB data).

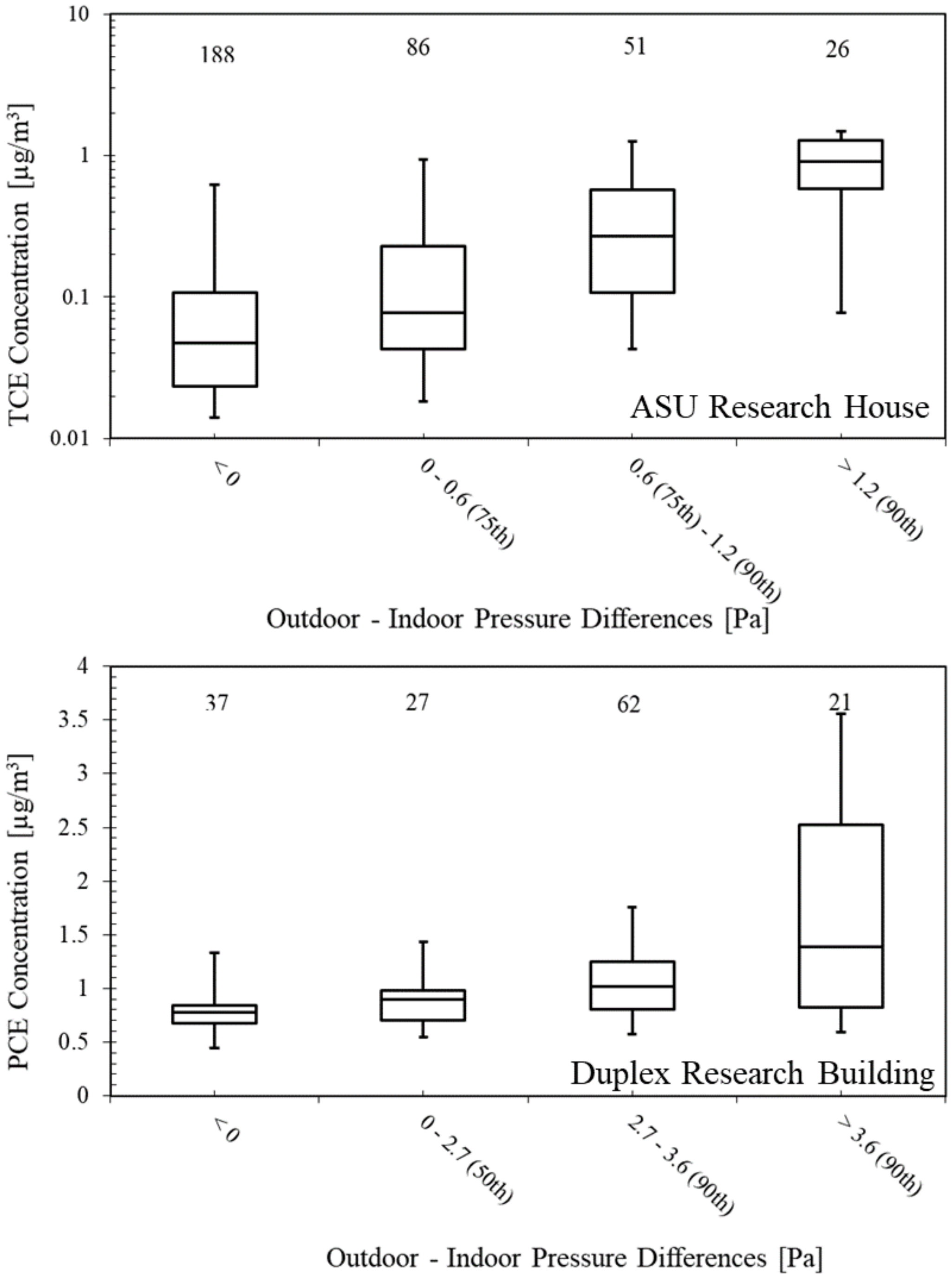

Figure 5. Statistical summary of 24-h average indoor air VOCs concentrations that were collected at the ASU research house (upper graph) and Duplex Research Building (lower

graph) when indoor-to-outdoor pressure differences were within the percentiles/concentrations shown. Error bars are maximum, 75th percentile, median, 25th percentile and minimum values for VOCs concentrations. Numbers on top of each box are days when the associated concentrations were observed.

Radon vs TCE attenuation from subslab to indoor air. The correlation between radon and VOCs attenuation factors from subslab to indoor air is evaluated using soil gas survey results from SDM. Soil gas samples were collected from sub-slab depth during 10 synoptic sampling events from July 2011 to August 2012. In each sampling events, both radon and TCE concentrations were quantified from 7 different locations within the building footprint, a detailed sampling schematic/method and analytical methods for which are reported by Holton et al. (2015). It should be noted that the land drain VI pathway at this site ends in the gravel pack beneath slab of the structure. Thus, TCE vapors from land drain pathway must migrate through the gravel pack and cracks in the slab to enter the building. Soil gas radon must also migrate through the gravel pack and slab cracks. As such, the attenuation factors for both radon and TCE vapors from subslab to indoor air should be comparable, in theory.

The attenuation factors (subslab to indoor air) were calculated by dividing the subslab vapor analytical results from each location by the average 7-day indoor air vapor concentrations during each sampling event. Results are presented in Figure 6.

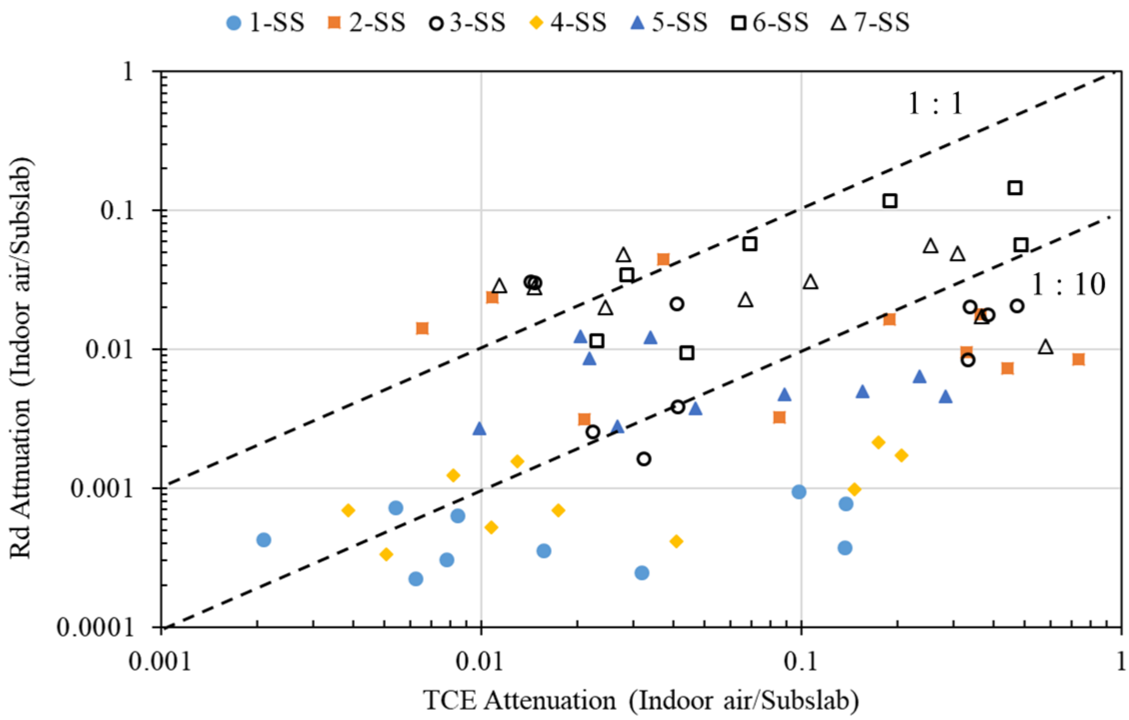

Figure 6. Indoor air to subslab TCE and radon attenuation factors from 7 locations in SDM for during 10 synoptic sampling events.

As shown in Figure 6, subslab to indoor air attenuation for radon is not comparable to TCE at this site. Radon attenuation is generally greater: Attenuation for about 1/3 of the radon numbers are more than 10x greater than those for TCE that were collected during the same sampling event. In about 20% of the paired cases, the difference between radon and TCE attenuation was less than 100% (factor 2x). Moreover, such discrepancies between radon and TCE attenuation varies not only spatially but also temporally. For example, the radon to TCE attenuation ratio ranged from 5 to over 364 at the sampling location 1-SS, whereas, this value was always less than 10 for 6-SS. As such, radon attenuation is not a reliable metric to predict VOCs attenuation from subslab soil gas to indoor air at this site.

The dissimilarity in subslab concentration distributions and the temporal changes of both chemicals can explain such distinct attenuation for both radon and TCE samples. Figures 7 and 8 summarize subslab sampling results for both TCE and radon, respectively. Those figures indicate that subslab TCE soil gas concentrations changed more significantly over time than radon. For example, temporal TCE subslab soil gas concentrations varied more than 85x at Location 2, while it was only 6x for radon. The spatial distributions for both chemicals was also quite different. Median TCE soil gas concentrations varied by less than 10x spatially, however, the spatial variability of median radon soil gas concentrations was greater than 100x. An interesting observation is that radon subslab soil gas concentrations were always much greater at -SS and 4-SS than other locations. This is because subslab soil formation beneath the garage (Figure 1) did not have the gravel pack as did the rest of the sampling locations.

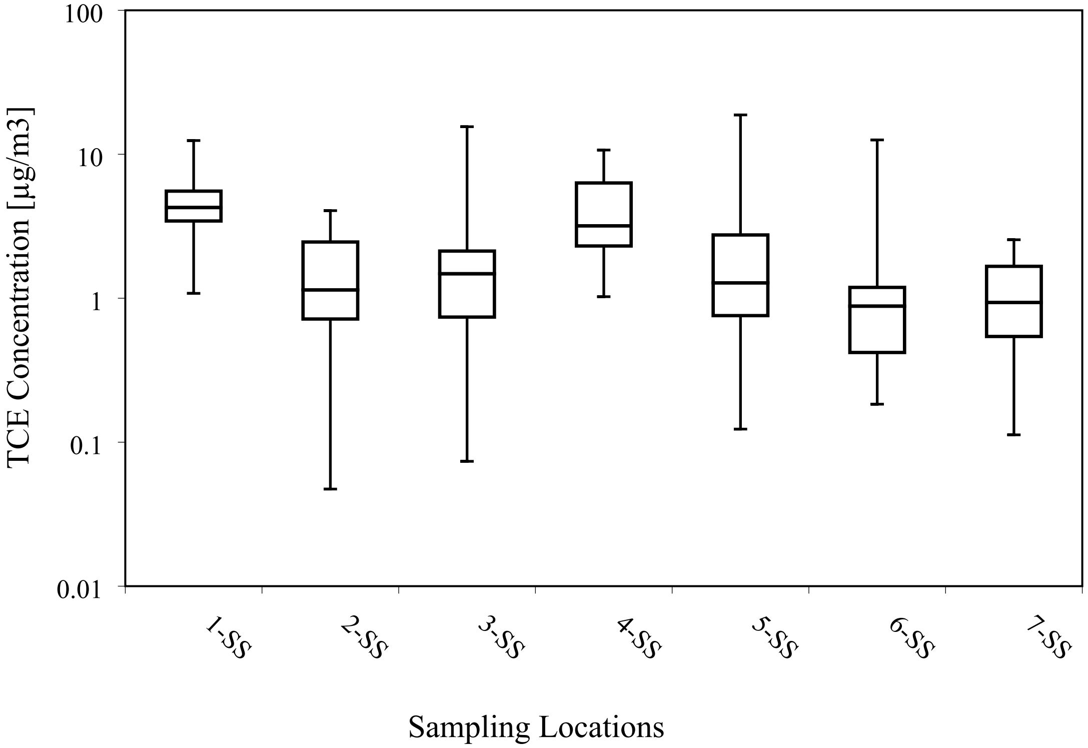

Figure 7. Summary of TCE vapor sampling results from 7 subslab sampling locations. The whisker and box presentation show the maximum, 75th percentile, 50th (median), 25th

percentile, and minimum concentrations, in order from top to bottom.

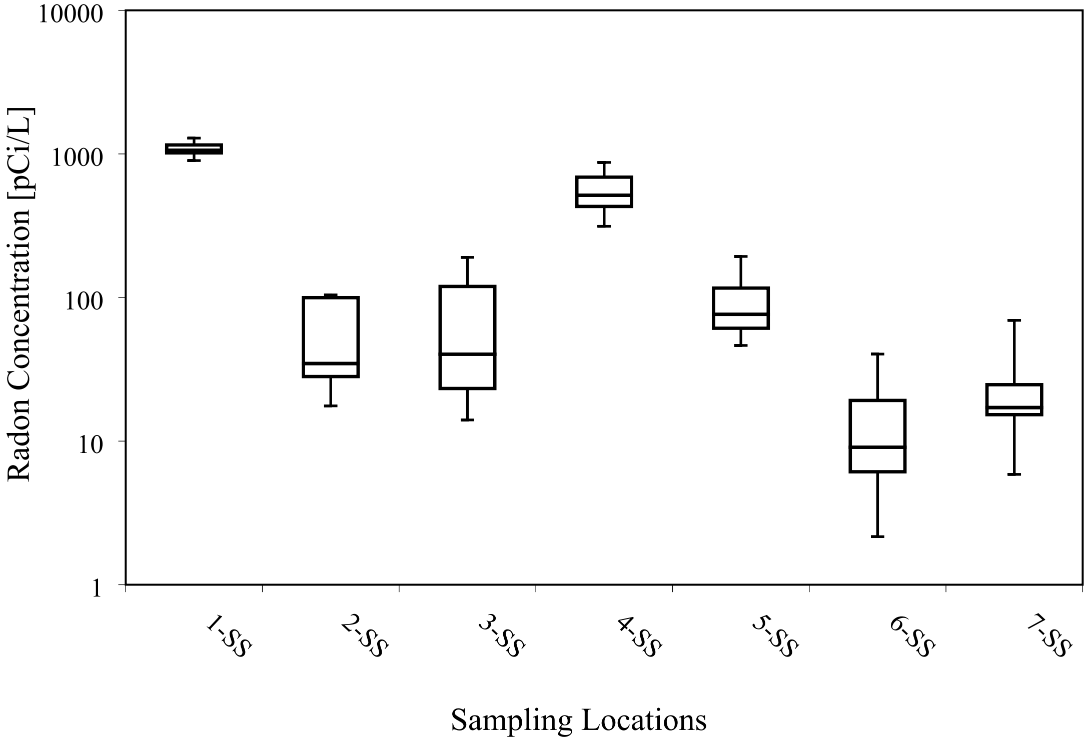

Figure 8. Summary of radon vapor sampling results from 7 sub-slab sampling locations. The whisker and box presentation show the maximum, 75th percentile, 50th (median), 25th percentile, and minimum concentrations, in order from top to bottom.

In contradiction to the hypothesis that indoor air to subslab radon attenuation can be used as an indicator for building-specific VOCs attenuation, the results from SDM show distinct differences between those two metrics. Although SDM is not a site dominated by the soil VI pathway, lessons learned from that site raise concerns when using subslab to indoor air radon attenuation as a proxy for VOC cross-slab AF. Radon and VOCs sources are conceptually different. Radon is generated in the soil/geology formation and/or some building materials. As such, radon source is often adjacent to the building foundation and its' spatial distribution reflects the geologic formation where it is generated. In contrast, the VOC source-to-building separation is often larger than radon, as it originates either from groundwater or contaminated soil. Together, these can create significant enough differences in radon and VOCs vapor concentrations in the subslab environment and could affect the reliability of radon attenuation to predict VOC attenuations. Making things additionally problematic are other factors, such as the presence of indoor air VOC sources and alternative/preferential VI pathways. As such, we recommend the comparison between indoor air and subslab radon concentrations as an indicator of soil gas movement from subslab areas into the structure., rather than a quantitative metric for VI risk evaluation.

Implications and future research

Thanks to the development of improved and cost-effective radon and differential pressure monitoring equipment, radon and pressure differences can be effectively monitored over long periods. The use of indoor air radon concentrations and outdoor to indoor pressure differences as indicators for VI investigation shows promise. Based on the analysis of long-term, high-frequency sampling results of indoor air and soil vapor VOC concentrations, indoor air and soil vapor radon concentrations, and outdoor to indoor pressure differentials from two well-instrumented study buildings, this paper suggests that indoor air radon concentrations and indoor-to-outdoor pressure differences should be used as qualitative indicators for VOC migrations. In another word, they could indicate if soil VOC VI pathway exist and when such pathways may result in high-level exposures, but they are not reliable for quantitative risk assessment.

Although indoor air radon concentrations and outdoor-to-indoor pressure differences show great potential as cost-effective environmental indicators that could help guide VOCs VI investigations, further researches are still needed. One challenge is the occurrence of alternative/preferential pathways. Alternative/preferential VI pathways are subsurface conduits (e.g. sewer and land drain) that only allow VOC vapors transporting through but not radon vapors. In such cases, the use of indoor air radon concentrations and outdoor-to-indoor pressure differences as environmental indicators is questionable.

Another barrier for using indoor air radon concentrations and outdoor-to-indoor pressure differences to improve VI assessment is lacking practical guidance. This study indicated high-levels indoor air VOCs concentrations would likely occur when indoor air radon concentrations and outdoor-to-indoor pressure differences exceed certain thresholds (e.g. 90th of indoor air radon concentrations). However, the values of these thresholds were building specific, and were obtained by analyzing years of long-term monitoring results from two well-instrumented research buildings. As such, future studies are necessary to validate the conclusions from this work to vet recommendations that include valid durations for radon and pressure monitoring, how to identify high risk periods using short-term or even real-time radon or pressure monitoring results, and whether a plausible approach for setting building-specific thresholds for indoor air radon and pressure differences is possible.

Acknowledgments

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