

Temporal Variations In Vapor Intrusion-Induced Indoor Air Contaminant Concentrations

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Abstract

Temporal variability in indoor air contaminant concentrations at vapor intrusion (VI) sites has been a concern for some time. We consider the source of the reported variability at VI sites located near Hill Air Force Base (AFB) in Utah, an EPA experimental house in Indiana, and Naval Air Station North Island in California. We focus in particular on how the indoor/outdoor pressure differences and air exchange rates affected indoor air contaminant concentrations at these sites. We investigate how these dynamics differ for a site that is characterized by a preferential pathway (like Hill AFB) and VI sites that are not influenced by such pathways, using three-dimensional fluid dynamics models and statistical analysis of the aforementioned sites. For a preferential pathway to impact a VI site, there must exist a medium allowing effective communication between a contaminant-delivering preferential pathway and the indoor air space, e.g. a permeable subslab space that may be provided by a gravel layer. At sites characterized by significant advective transport from the subslab to the indoor air space, much of the short-term variability in indoor air contaminant concentration can be explained by an impact of fluctuations in indoor/outdoor pressure differences. Meanwhile, air exchange rate variation drives most of the short-term variability at sites characterized by minor variations in advective transport.

Keywords: Vapor intrusion, Preferential pathways, Temporal variability, Finite element modeling, Air exchange rate, Indoor/outdoor pressure difference

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11 1. Introduction

12 Long term vapor intrusion (VI) studies in both residential and larger com-
13 mercial structures have raised concerns regarding significant observed tran-
14 sient behavior in indoor air contaminant concentrations[1, 2, 3, 4, 5, 6, 7].
15 Such variations make it difficult for those charged with protecting human
16 health to formulate a response - should evaluation of the risk of exposure be
17 based upon observed peak concentrations, or long-term averages, or some-
18 thing else? There is even uncertainty within the VI community regarding
19 how to best develop sampling strategies to address this problem[1, 3, 8].
20 What represents a reasonable sampling strategy for a particular site a single
21 8-hour sample? Repeated 8-hour samples? Month-long samples? Continuous
22 monitoring?

23 VI involves the migration of volatilizing contaminants from soil, ground-
24 water or other subsurface sources into overlying structures. The basic nature
25 of VI has been understood for some time and it has been the subject of much
26 study, but some aspects remain poorly understood, such as the causes of the
27 sometimes observed large temporal transients in indoor air concentrations.
28 Results from a house operated by Arizona State University (ASU) near Hill
29 AFB in Utah, an EPA experimental house in Indianapolis, IN and a large
30 warehouse at the Naval Air Station (NAS) North Island, CA have all shown
31 significant transient variations in indoor air contaminant concentrations. All
32 were outfitted with sampling and monitoring equipment that allowed tracking
33 temporal variation in indoor air contaminant concentrations on time scales
34 of hours. All have shown that these concentrations vary significantly with
35 time - orders of magnitude on the timescale of a day or days[9, 10, 5].

36 In one instance the source of the variation was clearly established during
37 the study of the site. At the ASU house a drain pipe (or “land drain”) con-
38 nected to a sewer system was discovered beneath the house. Careful isolation
39 of this source led to a clear conclusion that this “preferential pathway” for
40 contaminant vapor migration significantly contributed to observed indoor air
41 contaminant levels and their fluctuations[10, 11]. While in this case the issue
42 of a contribution from a preferential pathway was clearly resolved, what it
43 left open was a question of whether existence of such a preferential path-
44 way would always be expected to lead to large fluctuations in indoor air
45 contaminant concentrations.

46 Similarly, a sewer pipe has recently been suggested to be a source of the
47 contaminants found in the EPA Indianapolis house. That site was also char-

acterized by large indoor air contaminant concentration fluctuations[12, 7]. Sewer lines have been previously implicated as VI sources at several sites[13, 12, 14, 15]. A Danish study has estimated that roughly 20% of all VI sites in central Denmark involve significant sewer VI pathways[16]. Thus while consideration of sewer or other preferential pathways is now part of normal good practice in VI site investigation[1], it is still not known whether the existence of such pathways automatically means that large temporal fluctuations are necessarily to be expected.

In some of the above cited cases[13, 15], a sewer provided a pathway for direct entry of contaminant into the living space. While potentially important in many instances, this scenario is not further considered here where the focus is on pathways that deliver contaminant via the soil beneath a structure. It is, however, now known that even absent a preferential pathway, there may be significant transient variation in indoor air contaminant concentrations at VI sites[2, 17, 4]. One example is the site at NAS North Island at which no preferential pathways have been identified. Instead, a building at this site is characterized by significant temporal variations in indoor-outdoor pressure differential[5]. It is believed that this is the origin of the observed indoor air contaminant concentration fluctuations at that site.

This paper investigates the sources of the temporal variation in indoor air contaminant concentrations in both the presence and absence of preferential pathways. In this work, the latter scenarios are referred to as “normal” VI scenarios, in which there is typically a groundwater source of the contaminant. Specifically, we pose the question of just how much variation in indoor air contaminant concentration may be expected at such normal VI sites vs. those characterized by preferential pathways within the soil beneath the site. The conditions required for preferential pathways to become significant contributors to temporal variations in indoor air contaminant concentrations are also explored, and the consequences for sampling strategies are discussed.

2. Methods

2.1. Statistical Analysis Of Field Data

To frame the question of just how much variability in indoor air contaminant concentrations is actually observed, field datasets have been analyzed. For this purpose, datasets from the ASU house in Utah, the EPA Indianapolis site and North Island NAS were chosen for analysis. Read-

ers are referred to the original published works for details regarding data acquisition[9, 10, 3, 5, 7].

The ASU house data were obtained over a period of several years. During part of this time, controlled pressure method (CPM) tests were being conducted, in which the house was underpressurized to an extent greater than that characterizing “normal” house operation, thus increasing VI potential[18, 6, 9]. The period of CPM testing is thus excluded from the analysis. Likewise, the existence of a preferential pathway at the ASU house needs to be considered in examining that dataset; during some of the testing at that site, this pathway was cut off, resulting in “normal” VI conditions in which the main source of contaminant was diffusion of contaminant vapor from an underlying groundwater source.

The NAS North Island dataset has not (as far as is known) been influenced by a preferential pathway, but the structure there was subject to “large” internal pressure fluctuations. By “large” is meant still only of order 10-20 Pa, but these were greater than those generally recorded at the ASU house during normal operations. The underlying soil at NAS North Island is sandy[5] and more permeable than that at the ASU site, which will be shown to lead to greater pressure sensitivity in the former case.

The Indianapolis site investigation also spanned a number of years and periodically included the testing of a sub-slab depressurization system (SSD) for VI mitigation. Only the period before the installation of this system was considered in the present analysis. It is likely a sewer line beneath the structure acted as a preferential pathway[12]. Unlike at the ASU house, this preferential pathway was never removed or blocked, making it impossible to isolate the role of the preferential pathway at this site. It is still of interest to consider the data from this site because of the completeness and extensiveness of the data collection. Figure 1 illustrates a typical reported series of indoor air trichloroethylene (TCE) concentration measurements from this site. There is almost a two order of magnitude variation in the concentration data.

Some of the analysis of the above three field data sets relies on a probability density estimation technique called “kernel density estimation” (KDE). KDE is a technique used for estimating the probability distribution of a random variable(s) by using multiple kernels, or weighting functions to characterize the data sets. In this case, Gaussian kernels are used to create the KDEs. This means that it is presumed that the variables of interest (i.e., indoor air contaminant concentrations and indoor-outdoor pressure differ-

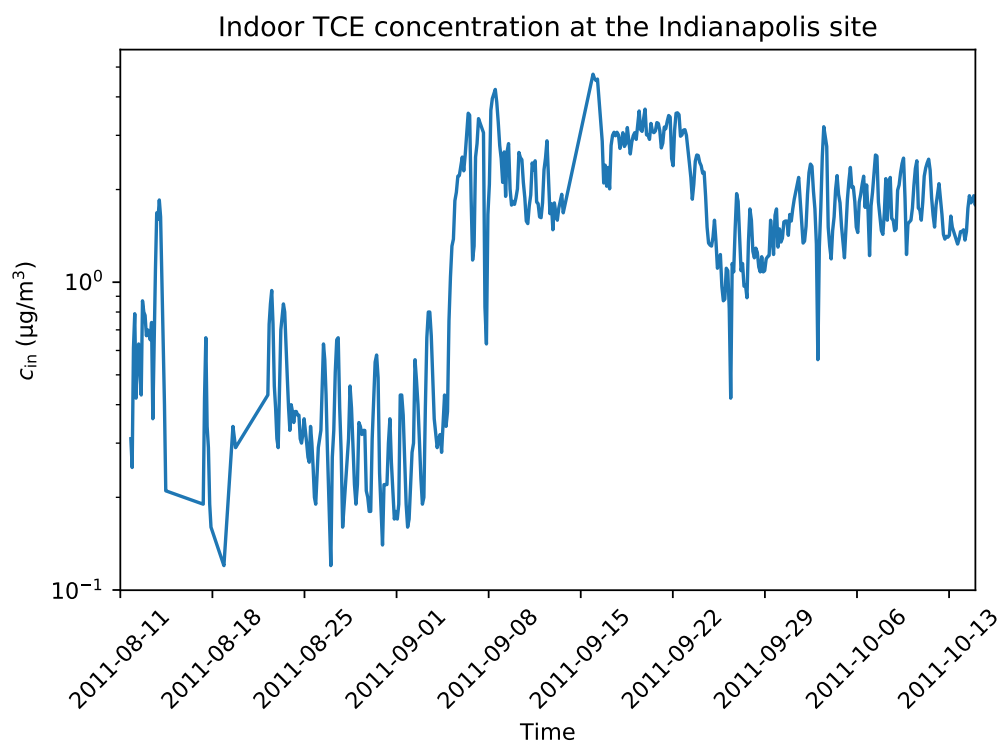


Figure 1: Typical data on indoor air TCE contaminant concentrations at the Indianapolis site[7].

entials, as sampled) are normally distributed around mean values and that there are statistical fluctuations associated with each sampling event. In this instance, the scipy statistical package was used to construct the KDEs, assuming a bandwidth parameter determined by Scott's rule. The SciPy Python library was used to conduct all statistical analysis and data processing[19].

2.2. Modeling Work

A previously described three-dimensional computational fluid dynamics model of a generic VI impacted house has been used to elucidate certain aspects of transient VI processes. In the present work, there has been an addition of a preferential pathway to the "standard" model that has been described before in publications by this group[20, 21, 22]. As in the earlier studies, only the vadose zone soil domain is directly modeled. Figure 2 shows a cutaway view of the relevant modeling domain.

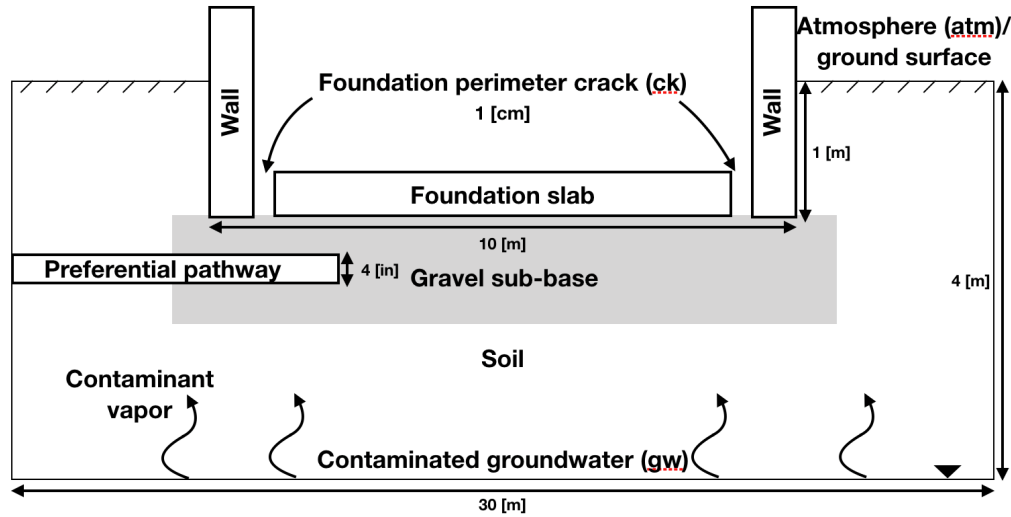


Figure 2: Foundation and vadose zone soil represented in the modeling. Note that here a gravel sub-base material is shown, but in certain simulations, that material is absent and the surrounding soil directly contacts the foundation slab. Different assumptions are made regarding the preferential pathway, here shown as a pipe entering the gravel sub-base. In some cases, the preferential pathway has been "turned off".

The modeled VI impacted structure is assumed to have a 10x10 m foundation footprint, with the bottom of the foundation slab lying 1 m below ground surface (bgs), simulating a house with a basement. The indoor air

space is modeled as a continuously stirred tank (CST)[1] and all of the contaminant entering the house is assumed to enter with soil gas through a 1 cm wide crack located between the foundation walls and the foundation slab around the perimeter of the house. All of the contaminant leaving the indoor air space is assumed to do so via air exchange with the ambient. The indoor control volume is here assumed to consist of only of the basement, having a total volume of 300 m³. Clearly different assumptions could be made regarding the structural features and the size of the crack entry route, but for present purposes, this is unimportant as the intent is only to show for “typical” values what the influence of some critical parameters is.

The modeled surrounding soil domain extends 5 meters from the perimeter of the house and is assumed to consist of sandy loam, except as noted otherwise. Directly beneath the foundation slab, there is assumed to be a 30 cm (one foot) thick gravel layer, except in certain cases here this sub-base material is assumed to be the same as the surrounding soil (termed a “uniform” soil scenario). The groundwater beneath the structure is assumed to be homogeneously contaminated with TCE selected as a prototypical contaminant. The groundwater itself is not modeled, as the bottom of the model domain is defined by the top of the water table. Where relevant, the preferential pathway is modeled as a 10 cm (4”) pipe that opens into the gravel sub-base beneath the structure. The air in the pipe is also assumed to be contaminated with TCE at a vapor concentration equal to the vapor in equilibrium with the groundwater contaminant concentration below the structure, modified by a scaling factor χ (allowing the contaminant concentration in the pipe to be parameterized). This model illustrates the concept of a “preferential pathway”, as the pipe carries contaminant vapor to the immediate vicinity of the foundation, by a path that circumvents the usual soil diffusion pathway.

The ground surface and the pipe are both sources of air to the soil domain. Both are assumed to exist at reference atmospheric pressure. Soil gas transport is governed by Richard’s equation, a modified version of Darcy’s Law, taking the variability of soil moisture in the vadose zone into account[23]. The van Genuchten equations are used to predict the soil moisture content and thus the effective permeability of the soil[24]. The effective diffusivity of contaminant in soil is calculated using the Millington-Quirk model[25]. The transport of contaminant vapor in the soil is assumed to be governed by the advection-diffusion equation, in which either advection or diffusion may dominate depending upon position and particular circumstances. The key

¹⁷⁵ working equations and the boundary conditions are summarized in Table 1f.

176 3. Results & Discussion

177 3.1. Variation In Indoor Air Contaminant Concentration Over Time

178 High frequency measurement of indoor air contaminant concentrations,
179 c_{in} , such as those in Figure 1, took place at both the ASU House and the
180 Indianapolis House over significant periods (Indianapolis: ca 1.7 years, ASU
181 house: ca 3.5 years)[7, 3]. Furthermore, at the Indianapolis site c_{in} for three
182 different contaminants, chloroform, TCE, and tetrachloroethylene (PCE)
183 were all collected, allowing examination of the variability of each VI con-
184 taminant. The NAS North Island NAS dataset was obtained over a much
185 shorter duration (9 days), and is therefore not examined in this portion of
186 the analysis. It should also be noted that the ASU house used 4-hour sorbent
187 tubes, while Indianapolis took instantaneous "grab" samples.

188 Figure 1 showed a large degree of temporal variation in one of the com-
189 ponents, and the data for the other components were quite similar. What is
190 apparent upon closer examination of such data is that the actual day-to-day
191 variations are typically not nearly as large as those observed when tracking
192 the data for a longer time. To demonstrate this point, the quotient of the
193 maximum and minimum c_{in} values (denoted as $c_{\text{max}}/c_{\text{min}}$) are shown as a
194 function of time in Figure 3. The values shown in Figure 3 are the means of
195 the quotients calculated for samples separated by the indicated times and the
196 error bars indicate the 95th percentile of all the data points. Hypothetical
197 resampling periods of one, two, three days, and the same number of weeks,
198 and months were chosen.

199 For example, if the data are examined in terms of the mean maximum
200 variation observable over the course of 24 hours (one day) the variation is
201 no greater than about a factor of two for any of the contaminants at the
202 Indianapolis house or for TCE at the ASU house (when the preferential
203 pathway was closed). The mean variability at the latter was only a bit higher
204 (about a factor of 3) when the preferential pathway was open. In other words,
205 a sampling protocol that involves sampling on two consecutive days would
206 typically not uncover the large temporal variations that characterize the site
207 over longer periods of time. As Figure 1 shows, there are certainly isolated
208 days in which a larger daily change was observed, but these were not typical,
209 to the extent that they fall outside of the 95% criteria used in defining the
210 error bars. So while such unusual jumps might be seen (for unknown reasons)
211 in a very small percentage of cases, the expectation is much more represented
212 by what is shown in Figure 3.

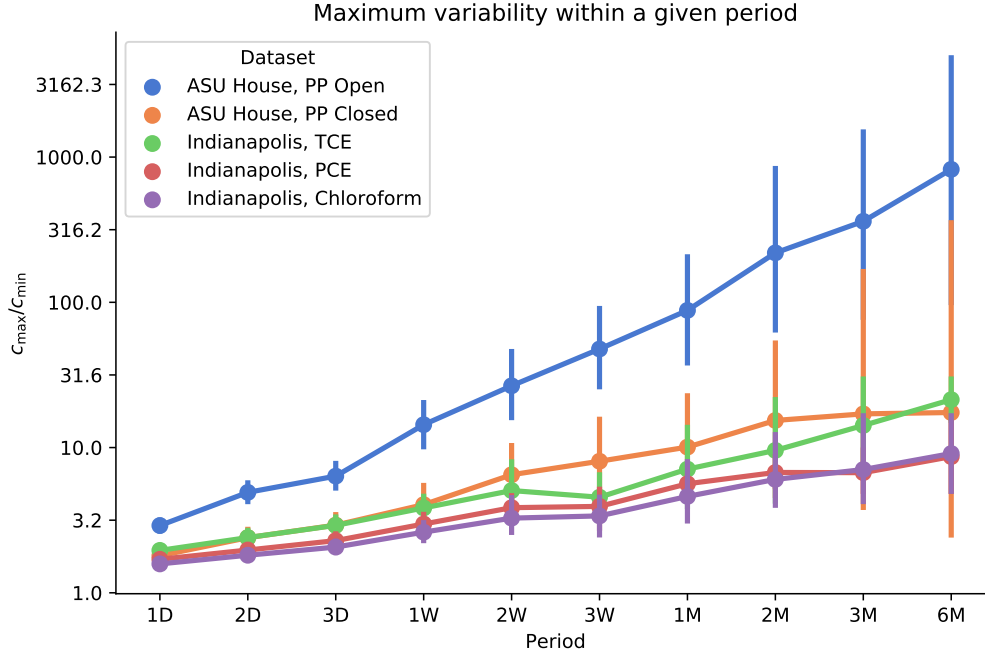


Figure 3: Mean values of the maximum change in indoor air contaminant concentration that may be expected over a given time period. (e.g., 1D is 1 day, 2W is 2 weeks, and 3M is 3 months). The error bars are the 95% confidence intervals.

213 Weeks of temporal separation in sampling events are required to observe
 214 the large variations of concern. Orders of magnitude differences begin to
 215 manifest themselves over the course of months. This is not surprising, since
 216 those who performed the measurements have already reported that there
 217 were seasonal aspects to the values obtained. This would be consistent with
 218 requiring months to see the more significant variations.

219 This analysis also suggests that certain types of preferential pathways
 220 contribute to larger variations on shorter timescales (ASU House). Even
 221 though there was a preferential pathway present at the Indianapolis House,
 222 the transients associated with its presence were of a slower nature and the
 223 behavior was not unlike what was observed at ASU House when the preferential
 224 pathway was closed. This warns that the mere existence of a preferential
 225 pathway is not by itself sufficient to create a situation of large variations over
 226 short sampling times.

227 The longer the resampling period, the larger the maximum variability

228 in observed indoor air contaminant concentrations. In the case of the ASU
 229 House with the preferential pathway open, the variability went from less than
 230 a threefold difference on the timescale of a day, to two to three orders of
 231 magnitude over the course of weeks. Thus there are different timescales that
 232 characterize different extents of variation, again pointing to the existence of
 233 more than a single factor that determines variability.

234 Multiple samples taken over a short time period, e.g. a few days, are
 235 unlikely to uncover significant variation in indoor air contaminant concen-
 236 tration; the larger transient variations typically manifest after longer time
 237 periods.

238 *3.2. Statistical Analysis of Field Data*

239 The data in Figure 1 and Figure 3 raise the question of what then actually
 240 determines the large degree of temporal variation sometimes reported. The
 241 rate of advective entry of soil gas into a structure is frequently cited as playing
 242 an important role in determining entry rate of contaminant. This advective
 243 entry rate is closely linked to the indoor-outdoor pressure difference, as can
 244 be caused by the “stack effect”, for example. Thus we first consider how much
 245 variability there might be in the pressure driving force for advection, and if
 246 this can explain the observed variability in observed indoor air contaminant
 247 concentrations.

248 The pressure difference between the indoor and outdoor/ambient ($p_{\text{in/out}}$)
 249 leads to advection, by which contaminants are drawn into (or prevented from)
 250 entering a structure. Changes in $p_{\text{in/out}}$ can take place quickly, leaving open
 251 the possibility of their impacting VI far more rapidly than can fluctuations in
 252 say groundwater depth or contaminant concentration (these latter processes
 253 take weeks or even months to impact the overlying structure).

254 We examine the relationship between $p_{\text{in/out}}$ and c_{in} by constructing the
 255 two-dimensional kernel density estimation (KDE) plots seen in Figure 4. The
 256 KDE plots allow us to view the measured distributions of $p_{\text{in/out}}$ and c_{in} , and
 257 develop a visual impression of how well these distributions correlate with one
 258 another. For this analysis we considered two VI sites, NAS North Island and
 259 the ASU House. The ASU House dataset was divided into two periods, one
 260 before and the other after the land drain (called the preferential pathway
 261 (PP) from here on) had been closed. By comparing these two periods on a
 262 single plot, the impact of the preferential pathway becomes clearer.

263 In Figure 4, the indoor air contaminant concentration c_{in} is normalized to
 264 the mean $c_{\text{in,mean}}$ of each dataset, allowing comparison of the impact $p_{\text{in/out}}$

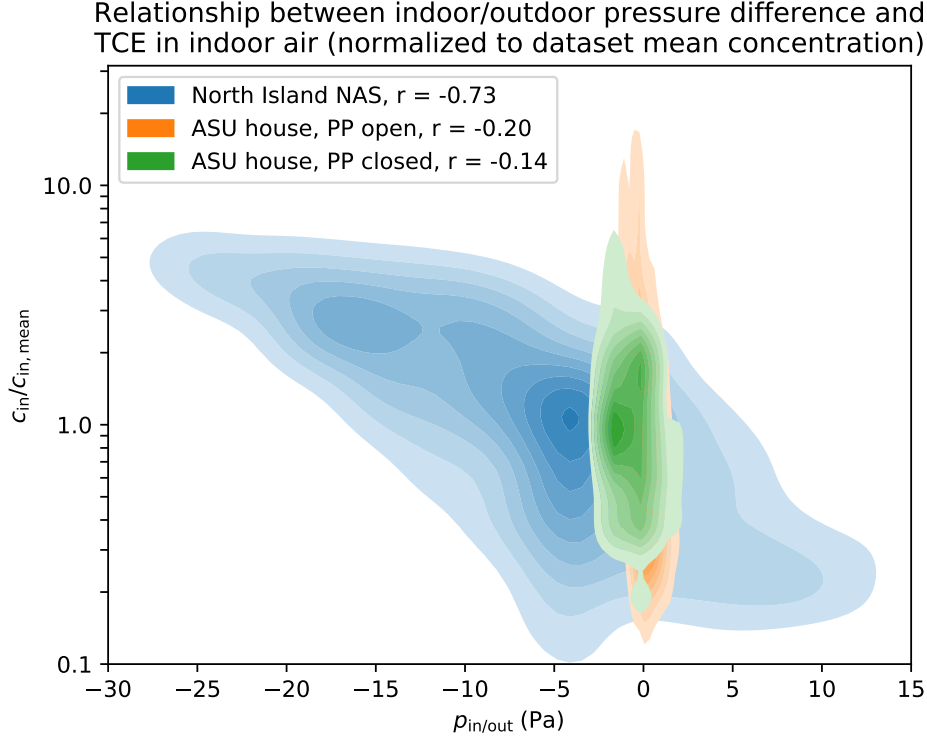


Figure 4: 2D-KDE plot showing the distributions of indoor air contaminant concentration, the indoor/outdoor pressure difference, and how they correlate to each other.

on c_{in} independently from the large differences in absolute values of indoor air concentrations at the different sites. A value of 10 on the y-axis indicates that the corresponding plotted value of c_{in} is 10 times greater than the mean for the dataset, and 0.1 indicate that it is one tenth of the mean.

Inspection of the range of normalized c_{in} values in Figure 4 again shows the two order of magnitude spread in observed values, implying a sampling at one particular time might give a value that is two orders of magnitude different than a result from a different time. Such issues have of course already been pointed out by the investigators who obtained the data.

The power of this KDE representation is that it permits evaluation of the relationship of two independently measured data - the indoor air contaminant concentration and the indoor-outdoor pressure difference. Examining the data in this manner immediately points to an important difference between the data from the ASU House and those from NAS North Island. At NAS

279 North Island site $p_{\text{in/out}}$ varies significantly; the 5th and 95th percentile of
280 $p_{\text{in/out}}$ are -19.9 and 7.4 Pa respectively. This may be contrasted with 5th
281 and 95th percentile $p_{\text{in/out}}$ at the ASU house: -1.4 and 2.1 Pa (with the PP
282 open), and -2.1 and 2.27 Pa (PP closed).

283 The much larger under- and overpressurization of the NAS North Island
284 site compared to the ASU House makes the pressure dependence of indoor
285 air concentration much more visible at the former site. The Pearson's r-value
286 for the correlation between $p_{\text{in/out}}$ and c_{in} for each dataset is shown in the
287 legend, and confirms what is apparent to the eye; the pressure driving force is
288 a determining factor for observed contamination at NAS North Island. But
289 the broadness of the band of the NAS North Island concentration data set
290 suggests that there is still a source of variability in c_{in} that has not been fully
291 captured - this will be addressed below.

292 The ASU house datasets offer a different picture. The variability of c_{in} is
293 just as large, or even larger than at NAS North Island, yet the $p_{\text{in/out}}$ varied
294 far less. The weaker dependence of c_{in} on the pressure difference is confirmed
295 by the much lower r-values for the correlations between the variables. In
296 other words, there is not nearly as strong a correlation between variation in
297 indoor air contaminant concentration and pressure difference for the ASU
298 House as there was for NAS North Island. These results strongly suggest
299 that there are other factors besides indoor pressure determining indoor air
300 contaminant concentrations, and their variations, that may not be accounted
301 for in applying this method.

302 The data for the ASU House also offer an insight into the role of the
303 preferential pathway. At first glance it may seem like the c_{in} values for the
304 periods when the PP is open and closed are relatively comparable. However,
305 the 5th and 95th percentiles values of $c_{\text{in}}/c_{\text{in,mean}}$ differ significantly as may
306 be seen in Table 2. It is clear that existence of the preferential pathway
307 dramatically increases the variability in indoor air contaminant concentra-
308 tion. This again is entirely consistent with what the investigators of that
309 site have already reported[11]. The correlation with indoor-outdoor pressure
310 difference is weak in the ASU house cases, so there are clearly factors other
311 than pressure difference that determine the variability in each. These will be
312 explored with the help of a modeling analysis presented below.

313 3.3. *Variability Of Attenuation to Subslab Concentrations*

314 Observed temporal variations in indoor air contaminant concentrations
315 might be explained by temporal variations in subslab contaminant concen-

trations. To examine how variability in subslab contaminant concentration might contribute to variability in indoor air contaminant concentration, data on the attenuation from subslab ($\alpha_{\text{subslab}} = c_{\text{in}}/c_{\text{subslab}}$) were examined. The dataset utilized for this was that from the ASU House. The c_{subslab} values were taken from a soil gas probe labeled as "6" at the ASU house. This probe was located closest to both the exit of the preferential pathway pipe, and to a reported breach in the foundation that served as a key entry pathway for contaminant getting into the house[11]. The results are shown in Figure 5, which shows the full distributions for both the case in which the preferential pathway was "open" and when it "closed".

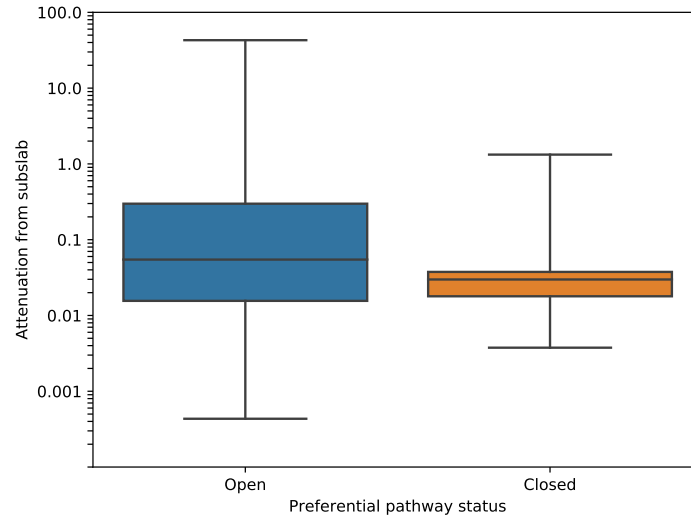


Figure 5: Boxplot of \log_{10} (subslab to indoor air contaminant attenuation) at the ASU house site. The box shows the quartiles of the distribution, the whiskers the extent of the distribution.

It is apparent that during the period when the preferential pathway was closed, α_{subslab} did not vary significantly, and was quite close to the EPA recommended α_{subslab} value of 0.03[1]. Thus during the period when the preferential pathway was closed, large temporal variations in subslab concentrations could not have been driving the variations in indoor air contaminant concentrations.

When the PP was open, there was considerably more variability in the subslab concentration values, and the mean value was higher than in the case where the preferential pathway was closed. It was also not uncommon

335 for the observed α_{subslab} to exceed unity. While large α_{subslab} values may
 336 sometimes indicate indoor sources at a site, there were none at the ASU
 337 house. A more likely explanation is that even though probe "6" was located
 338 in close proximity to the exit of the preferential pathway, there might have
 339 still existed significant spatial variability in c_{subslab} that could not be captured
 340 with a single measurement. This suggests caution is needed in profiling
 341 subslab contaminant concentrations in the presence of preferential pathways
 342 - significant variations are possible.

343 What the results of Figure 5 do clearly show is that the existence of a
 344 preferential pathway of the kind at ASU House (and idealized in Figure 1)
 345 can influence the temporal variation of subslab concentrations in a much less
 346 predictable way than those observed in "normal" VI scenarios.

347 3.4. Modeling Results

348 3.4.1. Pressure Effects

349 Having established the potential impacts of certain inputs on determining
 350 variability in indoor air contaminant concentrations, the mathematical model
 351 of VI can help further elucidate other key aspects. The results of calculations
 352 on a scenario corresponding to Figure 2 are presented in Figure 6. This
 353 scenario is not intended to exactly represent the situation at ASU House,
 354 but it is similar in the key aspect of having a preferential pathway delivering
 355 contaminant to a gravel sub-base. The full, complex geometry of the ASU
 356 House has not been represented, but the modeled structure is of comparable
 357 size, and will be subject to operational parameters based upon what were
 358 measured at that site. The general modeling conditions are those shown in
 359 Table 1.

360 In the calculation results shown in the top panel of Figure 6, a prefer-
 361 ential pathway is assumed to provide air containing contaminant vapor at
 362 a concentration equivalent to the vapor in equilibrium with the underlying
 363 groundwater source. Here, the indoor air exchange rate A_e was assumed to
 364 be a constant 0.5 per hour, and $p_{\text{in/out}}$ was varied from -5 to 5 Pa. Values
 365 of predicted indoor air contaminant concentrations, c_{in} were obtained from
 366 steady state calculations. The predicted c_{in} values were then normalized by
 367 the assumed vapor concentration in equilibrium with groundwater c_{gw} , giv-
 368 ing the attenuation from groundwater α_{gw} . The predicted values of α_{gw} as a
 369 function of $p_{\text{in/out}}$ are given by the central blue line in the upper panel of Fig-
 370 ure 6. These predicted values are compared to actual measured α_{gw} values

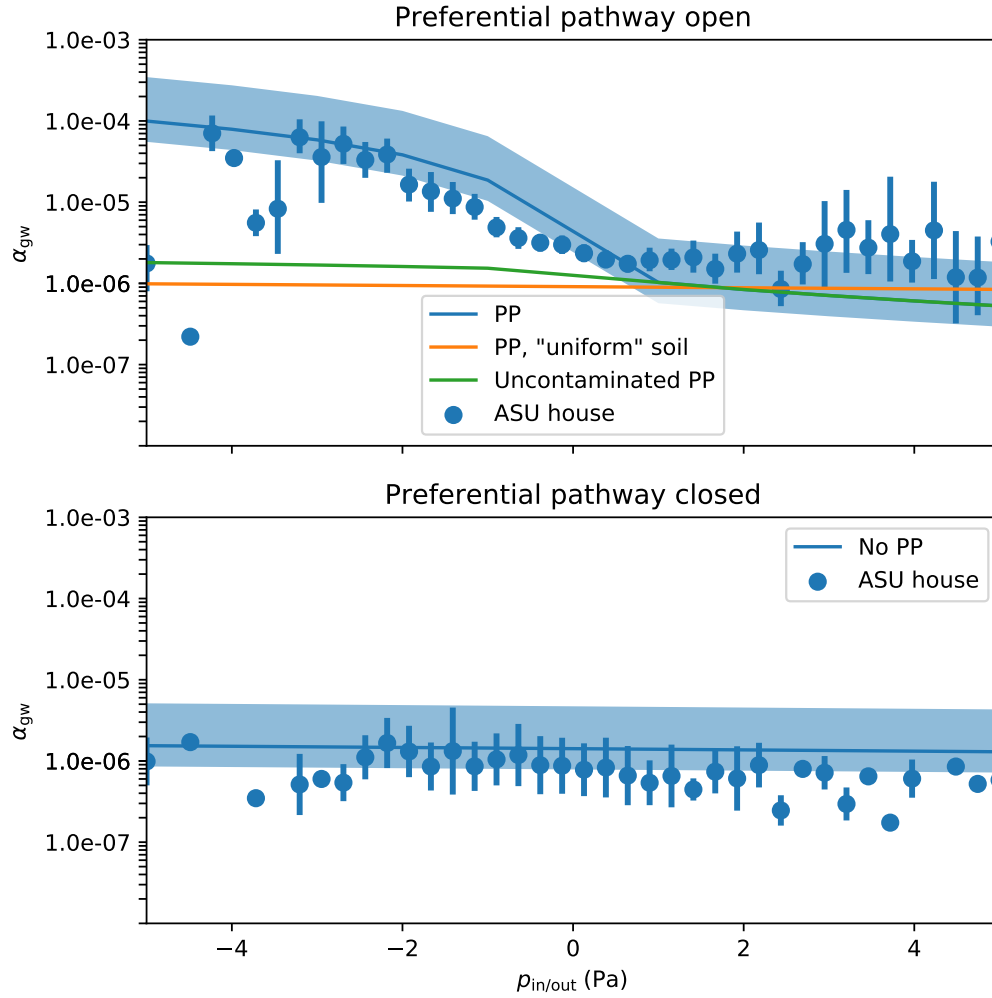


Figure 6: Simulated preferential pathway scenarios compared to actual ASU house field data. Field data are binned in 40 evenly spaced pressure bins, with the dot representing the mean and errors bars the 95% confidence interval of data at a particular pressure range. Shaded blue represent the range of model predictions for the indicated pressure difference, due to air exchange rate variability (using 5th and 95th percentile values of measured exchange rates). Top panel is for various cases representing an "open" preferential pathway, the lower panel with the pathway "closed".

371 from the ASU House for the period during which the preferential pathway
 372 was open (blue points).

373 The model successfully predicts the observed trends in α_{gw} as $p_{in/out}$ de-

374 creases (increased depressurization) but somewhat underpredicts α_{gw} as the
375 house is overpressurized. Most significantly, the model captures that even
376 for a small increase in depressurization (0 to -5 Pa) a very large increase in
377 α_{gw} (two order of magnitude) can occur.

378 The asymmetry relative to the predictions for depressurization and over-
379 pressurization is due to two factors. First, the preferential pathway acts not
380 only as a source of contaminant vapor, but also as a source of air to the
381 subslab. Because of the large resistance to soil gas flow in the surrounding
382 soil, having a local source of air to support the increase of advective flow into
383 the structure from the subslab region makes a large difference.

384 The above was proven by a second simulation, where the model was rerun
385 with the preferential pathway present, but with the permeable (gravel) layer
386 in the subslab removed and replaced by the surrounding soil (sandy loam).
387 This gave a "uniform soil" scenario the results of which are shown as an
388 orange line in the top panel of Figure 6. This simulation demonstrates that
389 without a permeable subslab to effectively allow the "advective potential" to
390 be realized, existence of preferential pathway will actually not impact a VI
391 site very much. In order for a preferential pathway to significantly contribute
392 to VI, this requires a scenario involving good advective communication be-
393 tween it the indoor environment. These requirements were met at the ASU
394 House.

395 A perhaps obvious second requirement is that the preferential pathway
396 must deliver contaminant vapors to be impactful. In another simulation, the
397 permeable (gravel) subslab region was included, but the preferential path-
398 way merely delivered clean air to the subslab. The result of this simulation
399 is shown as the green line in the top panel of Figure 6. This shows that while
400 there was a lightly larger α_{gw} compared to the "uniform soil" scenario, it is
401 nowhere near as significant as when the preferential pathway delivers contam-
402 inant vapors. The contaminated and uncontaminated preferential pathway
403 scenarios (blue and green lines respectively) thus bound the range of α_{gw} that
404 would be observed for a given $p_{\text{in/out}}$ depending on the contaminant vapor
405 concentration in the preferential pathway.

406 The model is also able to capture the weak trend in α_{gw} with $p_{\text{in/out}}$ when
407 a preferential pathway is absent, but when there still exists a permeable
408 subslab region. These results are shown in the bottom panel of Figure 6.
409 These results are again in agreement with what was observed at the ASU
410 House when the preferential pathway was closed, i.e. that there was a much
411 more modest variation in indoor air concentration, irrespective of pressure,

412 when the preferential pathway was cut off.

413 The above simulations capture the trend in α_{gw} with $p_{\text{in/out}}$ but do not yet
414 capture the full variability of the concentration results over the "most prob-
415 able" portion of observed pressure distributions shown in Figure 4 (which
416 tend to be from -2 to +2 Pa). The results of Figure 6 show a spread of
417 almost an order of magnitude over this pressure range for the case of the
418 "open" preferential pathway, and almost no spread at all when the prefer-
419 ential pathway is "closed". Hence the predicted variability is roughly an
420 order of magnitude too low, when considering only the influence of pressure.
421 There is a factor that tends to increase the spread of the data one additional
422 order of magnitude beyond what was predicted by the base calculations of
423 Figure 6. We believe that it is variations in air exchange rate, operating in
424 concert with the natural variations in pressure differential, that explain the
425 remaining variability.

426 3.4.2. Air Exchange Rate Effects

427 Table 3 shows the observed variations in air exchange rates for the ASU
428 House and Indianapolis House, compared with EPA's summary of the dis-
429 tribution of typical residential air exchange rates[29, 30]. Examination of
430 these distributions point in a clear direction for modifying the above model.
431 Instead of using a constant value of air exchange rate, as is customary, its
432 values should be parameterized. A higher air exchange would of course be
433 associated with lower c_{in} and vice versa. Moreover, A_e may sometimes be
434 correlated with $p_{\text{in/out}}$. Determining any general relationship between A_e and
435 $p_{\text{in/out}}$ is difficult: the structure itself and weather phenomena have a signif-
436 icant effect on air exchange. As the data in the supplementary data show
437 (Figure S1), there is no easily discernable correlation between these variables
438 at the ASU site, though there is a hint of slight seasonal dependence. Note:
439 a relationship between A_e and $p_{\text{in/out}}$ may be established for larger $p_{\text{in/out}}$ via
440 the building leakage curves, which are widely used for heating, ventilation
441 and air conditioning systems in construction.

442 To show the influence of possible statistical fluctuations of air exchange
443 rate on the predictions of α_{gw} values, the scenarios of Figure 6 were rerun
444 calculated using the 5th and 95th percentile measured A_e values, 0.17 and
445 0.90 respectively (based upon the actual distributions in Figure S1), provid-
446 ing predicted upper and lower bounds for α_{gw} . These bounds are indicated
447 by the shaded blue regions around the center line calculated for an assumed
448 constant A_e of 0.5 per hour.

449 It is apparent that assuming variability in air exchange rate allows cap-
 450 turing most of the observed variability in α_{gw} . We believe that this explains
 451 the portion of the variation in indoor air contaminant concentration data
 452 that cannot be explained by either existence of preferential pathways or by
 453 the range in indoor depressurization. Thus, we believe that it is the inter-
 454 play of preferential pathway conditions, with indoor pressure variations and
 455 normal air exchange rates that help to explain the observations of significant
 456 variations in reported indoor air contaminant concentrations.

457 3.4.3. Results of Transient Simulations

458 The above analyses have been conducted under simulated steady state
 459 conditions. The conclusions regarding the importance of the different pa-
 460 rameters are now examined in actual transient simulations. The model con-
 461 figuration of Figure 2 is run in 24-hour transient simulations to examine how
 462 c_{in} fluctuates over the course of a "typical" day. The simulations vary $p_{\text{in/out}}$
 463 as one model input, and then assume either a constant or time-varying air
 464 exchange rate, A_e . The ASU House dataset was again the source of the "typ-
 465 ical" $p_{\text{in/out}}$ temporal variation, obtained by examining the median, hourly,
 466 diurnal $p_{\text{in/out}}$ during the non-CPM periods. The statistically "typical" $p_{\text{in/out}}$
 467 cycle may be seen in the upper left panel of Figure 7 (note that values be-
 468 tween the hourly median values are interpolated using cubic splines). The
 469 "typical" air exchange rate is calculated in exactly the same way and is shown
 470 by the blue line in the upper right panel of Figure 7. The orange line is the
 471 air exchange rate value assumed for the calculations at constant air exchange
 472 rate.

473 The result of these simulations are shown in the bottom two panels of
 474 Figure 7, where the left and right panels show the results of open and closed
 475 preferential pathways, respectively. The "max change" value in the legends
 476 is the quotient of the lowest and highest predicted concentrations, i.e. a value
 477 of two indicate that the maximum daily concentration is twice as high as the
 478 lowest. This quantity may be compared with the value that is plotted for
 479 "one day" in Figure 3. When the preferential pathway is open, there is a
 480 maximum daily variation of roughly a factor of 5, irrespective of whether A_e
 481 fluctuates or not, which is somewhat more than the maximum daily varia-
 482 tion shown in Figure 3. The relatively small difference between the variable
 483 and constant A_e cases indicates that most of the variability during a "typi-
 484 cal" day is here attributable to fluctuations in $p_{\text{in/out}}$, i.e. the contaminant
 485 transport into the modeled structure is advection dominated. Even for the

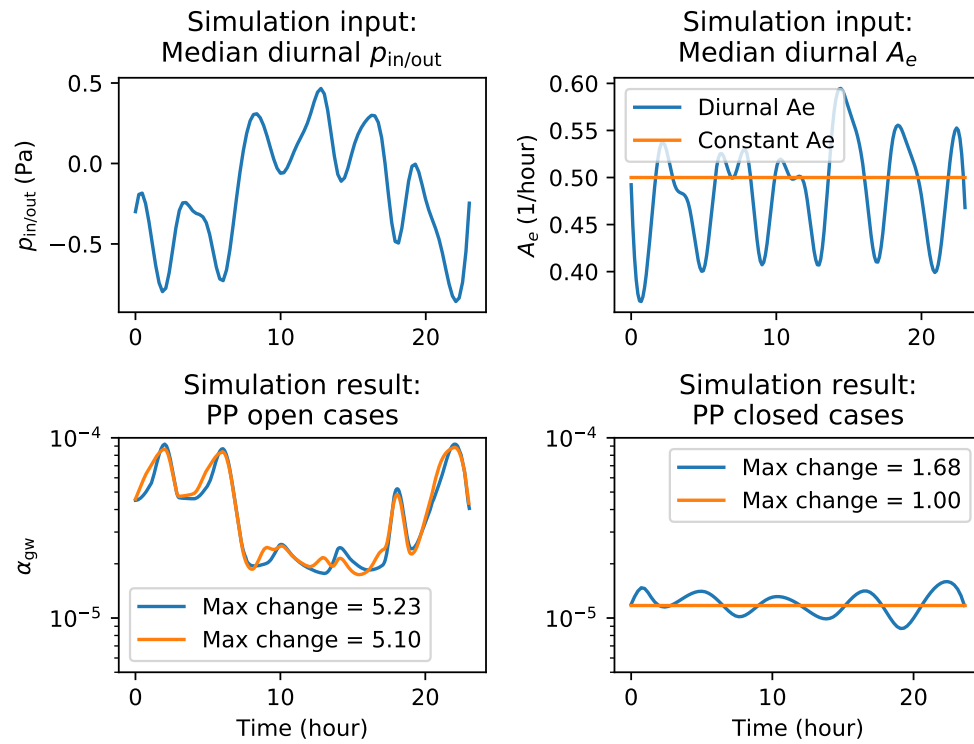


Figure 7: Transient simulation of a "typical" VI day, using diurnal indoor/outdoor pressure difference and air exchange rate as inputs. Effect of preferential pathway considered.

small fluctuations in $p_{\text{in/out}}$ the contaminant entry rate fluctuation drives the observed indoor concentration. When the preferential pathway is closed the story is quite different. When air exchange rate is held constant, there is essentially no variation in c_{in} . This is again not surprising, as Figure 6 demonstrated that when the preferential pathway is closed, the influence of $p_{\text{in/out}}$ on contaminant entry rate (and subsequently c_{in}) is small. Combined with the small $p_{\text{in/out}}$ this indicates that the contaminant transport into the modeled structure in this scenario is dominated by diffusion. When the air exchange rate is allowed to fluctuate, the maximum daily variation in c_{in} is 1.68, which is in line with what is shown in Figure 3. This shows that for a "typical" day, when the preferential pathway is closed off, much of the daily variation in c_{in} is due to daily fluctuations in air exchange rate.

These results demonstrate the complicated nature of temporal variability in c_{in} . It is important to recall that only the effects of indoor/outdoor pressure difference and air exchange rate have been considered here, but slower processes, e.g. changes in groundwater contaminant concentration or various seasonal effects can also have a significant impact on VI over time. For the shorter time periods of concern in recent studies of temporal variability in indoor contaminant concentrations we believe that these are dominated by combinations of indoor/outdoor pressure differentials and air exchange rate. For a site where advective communication between the subsurface and the indoor is good, $p_{\text{in/out}}$ is likely a significant determinant of c_{in} and its temporal variability. We have shown that that such a scenario may arise due to a preferential pathway entering a permeable sub-base, but may also exist even in the absence of a preferential pathway just as the results from NAS North Island demonstrate. At sites where advective transport into the structure is limited, much of the temporal variability in c_{in} may be attributed to natural fluctuations in air exchange rate.

4. Conclusions

Based on the statistical analysis of the field data presented, as well as the modeling efforts the following conclusions may be drawn.

- Indoor air contaminant concentrations are unlikely to vary by more than a factor of three over a week-long period if the site is not characterized by a preferential pathway, such as the one found at the ASU house site; else more than an order magnitude variability within the same period may be expected.

- 522 • Preferential pathways can cause significant spatial variability in the
523 subslab. Subsequently, subslab vapor samples may reveal that the at-
524 tenuation from subslab may significantly exceed the EPA recommended
525 value of 0.03 and even exceed unity - potentially leading to the erro-
526 neous conclusion that an indoor source is present.
- 527 • Preferential pathways such as the one found at the ASU house signif-
528 icantly increase the advective transport potential through the founda-
529 tion breaches at a site. This relies on effective communication between
530 the indoor air space and the preferential pathway, e.g. a permeable
531 subslab region (gravel layer) must exist.
- 532 • Sites characterized by significant advective potential, such as North
533 Island NAS or the ASU house with the preferential pathway open,
534 most of the short-term indoor air contaminant concentration variability
535 may be attributed to fluctuations in indoor/outdoor pressure difference;
536 at sites where the advective potential is low, e.g. ASU house with
537 the preferential pathway closed, short-term variability is dominated by
538 fluctuations in air exchange rate.

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(a) Governing equations

Unsteady-CST	$V \frac{dc_{in}}{dt} = \int_{A_{ck}} j_{ck} dA - c_{in} A_e V_{slab}$
Richard's equation	$\nabla \cdot \rho \left(-\frac{\kappa_s}{\mu} k_r \nabla p \right) = 0$
Millington-Quirk	$D_{eff} = D_{air} \frac{\theta_g^{10/3}}{\theta_t^2} + \frac{D_{water}}{K_H} \frac{\theta_w^{10/3}}{\theta_t^2}$
Advection-diffusion equation	$\frac{\partial}{\partial t} (\theta_w c_w + \theta_g c) = \nabla (D_{eff} \cdot \nabla c) - \vec{u} \cdot \nabla c$
van Genuchten equations	$Se = \frac{\theta_w - \theta_r}{\theta_t - \theta_r} = [1 + \alpha z ^n]^{-m}$
	$k_r = (1 - Se)^l [1 - (Se^{-m})^m]^2$
	$m = 1 - 1/n$

(b) Boundary conditions

Boundary	Richard's equation	Transport equation
Foundation crack	$p = p_{in/out} \text{ (Pa)}$	$j_{ck} = \frac{uc}{1 - \exp(uL_{slab}/D_{air})}$
Groundwater source	<i>No flow</i>	$c = c_{gw} K_H \text{ (}\mu\text{g/m}^3\text{)}$
Groundsurface	$p = 0 \text{ (Pa)}$	$c = 0 \text{ (}\mu\text{g/m}^3\text{)}$
Preferential pathway	$p = 0 \text{ (Pa)}$	$c = c_{gw} K_H \chi \text{ (}\mu\text{g/m}^3\text{)}$

(c) Soil & gravel properties[26, 27, 28]

Soil	$\kappa_s \text{ (m}^2\text{)}$	$\rho \text{ (kg/m}^3\text{)}$	θ_s	θ_r	$\alpha \text{ (1/m)}$	n
Gravel	$1.3 \cdot 10^{-9}$	1680	0.42	0.005	100	3.1
Sandy Loam	$5.9 \cdot 10^{-13}$	1460	0.39	0.039	2.7	1.4

(d) Trichloroethylene (diluted in air) properties[27, 28]

$D_{air} \text{ (m}^2\text{/h)}$	$D_{water} \text{ (m}^2\text{/h)}$	$\rho \text{ (kg/m}^3\text{)}$	$\mu \text{ (Pa} \cdot \text{s)}$	K_H	$M \text{ (g/mol)}$
$2.47 \cdot 10^{-2}$	$3.67 \cdot 10^{-6}$	1.614	$1.86 \cdot 10^{-5}$	0.403	131.39

(e) Building properties

$V_{base} \text{ (m}^3\text{)}$	$L_{slab} \text{ (cm)}$	$A_e \text{ (1/hr)}$
300	15	0.5

(f) Governing equations, boundary conditions & model input parameters. (See below for table of nomenclature).

	North Island		ASU, PP Open		ASU, PP Closed	
Percentile	5th	95th	5th	95th	5th	95th
$p_{\text{in/out}}$ (Pa)	-19.9	7.4	-1.4	2.1	-2.1	2.27
$c_{\text{in}}/c_{\text{in,mean}}$	4.1	0.2	13.5	0.2	3.3	0.4

Table 2: 5th and 95th percentile values of $p_{\text{in/out}}$ and $c_{\text{in}}/c_{\text{in,mean}}$ in Figure 4.

Percentile	10th	50th	90th
EPA[29, 30]	0.16-0.2	0.35-0.49	1.21-1.49
ASU house[3, 11]	0.21	0.43	0.78
Indianapolis[7]	0.34	0.74	1.27

Table 3: Air exchange rate values (1/hr)

Table .4: List of abbreviations

A_{ck}	Crack area
A_e	Air exchange rate
α, n, m, l	van Genuchten parameters
α_{gw}	Attenuation from groundwater contaminant vapor source
c_{in}	Indoor air contaminant concentration
c	Soil-gas contaminant concentration
c_w	Soil-water contaminant concentration
c_{gw}	Contaminant groundwater concentration
χ	PP contaminant concentration scaling parameter
D_{eff}	Effective diffusion coefficient
D_{air}	Diffusion coefficient in air
D_{water}	Diffusion coefficient in water
j_{ck}	Contaminant molar flux through the foundation crack
κ_s	Saturated soil permeability
K_H	Dimensionless Henry's law constant
k_r	Relative permeability
L_{slab}	Thickness of the foundation slab
M	Molar mass
μ	Contaminant vapor viscosity
NAS	Naval Air Stations
p	Pressure in soil
$p_{\text{in/out}}$	Indoor/outdoor pressure difference
PP	Preferential pathway
ρ	Density
Se	Soil water saturation
t	time
θ_g	Vapor/gas filled porosity
θ_w	Water filled porosity
θ_r	Residual water filled porosity
θ_t	Total porosity
\vec{u}	Soil-gas velocity (vector quantity)
VI	Vapor intrusion
V_{base}	Basement volume
z	Elevation above groundwater

- 543 [1] U.S. Environmental Protection Agency, OSWER Technical Guide for
544 Assessing and Mitigating the Vapor Intrusion Pathway From Subsurface
545 Vapor Sources To Indoor Air, 2015.
- 546 [2] D. Folkes, W. Wertz, J. Kurtz, T. Kuehster, Observed Spatial and
547 Temporal Distributions of CVOCs at Colorado and New York Vapor
548 Intrusion Sites, *Ground Water Monitoring & Remediation* 29 (2009)
549 70–80.
- 550 [3] C. Holton, H. Luo, P. Dahlen, K. Gorder, E. Dettenmaier, P. C. Johnson,
551 Temporal Variability of Indoor Air Concentrations under Natural Con-
552 ditions in a House Overlying a Dilute Chlorinated Solvent Groundwater
553 Plume, *Environmental Science & Technology* 47 (2013) 13347–13354.
- 554 [4] J. E. Johnston, J. M. Gibson, Spatiotemporal variability of tetra-
555 chloroethylene in residential indoor air due to vapor intrusion: A lon-
556 gitudinal, community-based study, *Journal of Exposure Science and*
557 *Environmental Epidemiology* 24 (2014) 564.
- 558 [5] V. Hosangadi, B. Shaver, B. Hartman, M. Pound, M. L. Kram, C. Fres-
559 cura, High-Frequency Continuous Monitoring to Track Vapor Intrusion
560 Resulting From Naturally Occurring Pressure Dynamics, *Remediation*
561 *Journal* 27 (2017) 9–25.
- 562 [6] T. McHugh, P. Loll, B. Eklund, Recent advances in vapor intrusion
563 site investigations, *Journal of Environmental Management* 204 (2017)
564 783–792.
- 565 [7] U.S. Environmental Protection Agency, Assessment of Mitigation Sys-
566 tems on Vapor Intrusion: Temporal Trends, Attenuation Factors, and
567 Contaminant Migration Routes under Mitigated And Non-mitigated
568 Conditions, 2015.
- 569 [8] P. C. Johnson, C. W. Holton, Y. Guo, P. Dahlen, E. H. Luo, K. Gorder,
570 E. Dettenmaier, R. E. Hinchee, Integrated Field-Scale, Lab-Scale, and
571 Modeling Studies for Improving Our Ability to Assess the Groundwater
572 to Indoor Air Pathway at Chlorinated Solvent-Impacted Groundwater
573 Sites, 2016.

- 574 [9] C. W. Holton, Evaluation of Vapor Intrusion Pathway Assessment
575 Through Long-Term Monitoring Studies, PhD Thesis, Arizona State
576 University, 2015.
- 577 [10] Y. Guo, Vapor Intrusion at a Site with an Alternative Pathway and a
578 Fluctuating Groundwater Table, PhD Thesis, Arizona State University,
579 2015.
- 580 [11] Y. Guo, C. Holton, H. Luo, P. Dahlen, K. Gorder, E. Dettenmaier,
581 P. C. Johnson, Identification of Alternative Vapor Intrusion Pathways
582 Using Controlled Pressure Testing, Soil Gas Monitoring, and Screening
583 Model Calculations, *Environmental Science & Technology* 49 (2015)
584 13472–13482.
- 585 [12] T. McHugh, L. Beckley, T. Sullivan, C. Lutes, R. Truesdale, R. Uppen-
586 camp, B. Cosky, J. Zimmerman, B. Schumacher, Evidence of a sewer
587 vapor transport pathway at the USEPA vapor intrusion research duplex,
588 *Science of The Total Environment* 598 (2017) 772–779.
- 589 [13] K. G. Pennell, M. K. Scammell, M. D. McClean, J. Ames, B. Weldon,
590 L. Friguglietti, E. M. Suuberg, R. Shen, P. A. Indeglia, W. J. Heiger-
591 Bernays, Sewer Gas: An Indoor Air Source of PCE to Consider During
592 Vapor Intrusion Investigations, *Groundwater Monitoring & Remedia-*
593 *tion* 33 (2013) 119–126.
- 594 [14] M. Roghani, O. P. Jacobs, A. Miller, E. J. Willett, J. A. Jacobs, C. R.
595 Viteri, E. Shirazi, K. G. Pennell, Occurrence of chlorinated volatile
596 organic compounds (VOCs) in a sanitary sewer system: Implications
597 for assessing vapor intrusion alternative pathways, *Science of The Total*
598 *Environment* 616-617 (2018) 1149–1162.
- 599 [15] C. E. Riis, A. G. Christensen, M. H. Hansen, H. Husum, M. Terkelsen,
600 Vapor intrusion through sewer systems: Migration pathways of chlori-
601 nated solvents from groundwater to indoor air, 2010.
- 602 [16] K. B. Nielsen, B. Hvidberg, Remediation techniques for mitigating vapor
603 intrusion from sewer systems to indoor air, *Remediation Journal* 27
604 (2017) 67–73.

- 605 [17] D. Brenner, Results of a Long-Term Study of Vapor Intrusion at Four
606 Large Buildings at the NASA Ames Research Center, *Journal of the*
607 *Air & Waste Management Association* 60 (2010) 747–758.
- 608 [18] T. E. McHugh, L. Beckley, D. Bailey, K. Gorder, E. Dettenmaier,
609 I. Rivera-Duarte, S. Brock, I. C. MacGregor, Evaluation of Vapor In-
610 trusion Using Controlled Building Pressure, *Environmental Science &*
611 *Technology* 46 (2012) 4792–4799.
- 612 [19] E. Jones, T. Oliphant, Pearu Peterson, *SciPy: Open source scientific*
613 *tools for Python*, 2011.
- 614 [20] R. Shen, K. G. Pennell, E. M. Suuberg, Influence of Soil Moisture
615 on Soil Gas Vapor Concentration for Vapor Intrusion, *Environmental*
616 *Engineering Science* 30 (2013) 628–637.
- 617 [21] Y. Yao, Y. Wang, Z. Zhong, M. Tang, E. M. Suuberg, Investigating
618 the Role of Soil Texture in Vapor Intrusion from Groundwater Sources,
619 *Journal of Environmental Quality* 46 (2017) 776–784.
- 620 [22] Y. Yao, F. Mao, S. Ma, Y. Yao, E. M. Suuberg, X. Tang, Three-
621 Dimensional Simulation of Land Drains as a Preferential Pathway for
622 Vapor Intrusion into Buildings, *Journal of Environmental Quality* 46
623 (2017) 1424–1433.
- 624 [23] L. A. Richards, Capillary conduction of liquids through porous mediums,
625 *Physics* 1 (1931) 318–333.
- 626 [24] M. T. van Genuchten, A Closed-form Equation for Predicting the Hy-
627 draulic Conductivity of Unsaturated Soils, *Soil Science Society of Amer-*
628 *ican* 44 (1980) 892–898.
- 629 [25] R. J. Millington, J. P. Quirk, Permeability of porous solids, *Transactions*
630 *of the Faraday Society* 57 (1961) 1200.
- 631 [26] H.-C. Dan, P. Xin, L. Li, L. Li, D. Lockington, Capillary effect on flow
632 in the drainage layer of highway pavement, *Canadian Journal of Civil*
633 *Engineering* 39 (2012) 654–666.
- 634 [27] L. D. V. Abreu, H. Schuver, *Conceptual Model Scenarios for the Vapor*
635 *Intrusion Pathway*, 2012.

- 636 [28] U.S. Environmental Protection Agency, Users's Guide For Evaluating
637 Subsurface Vapor Intrusion Into Buildings, 2004.
- 638 [29] U.S. EPA, Exposure Factors Handbook 2011 Edition, Technical Report,
639 U.S. Environmental Protection Agency, 2011.
- 640 [30] M. D. Koontz, H. E. Rector, Estimation of Distributions for Residential
641 Air Exchange Rates, Technical Report, U.S. Environmental Protection
642 Agency, 1995.