INFLUENCE OF DECREASING BAROMETRIC PRESSURE FREQUENCIES ON HETEROGENEOUS UNDERGROUND RADON SOURCES

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Abstract

Radon as a fluid is subject to induced flow within a porous medium from vertical pressure gradients. The extent of these gradients is driven by the pneumatic diffusivity of the medium and the perceived frequency or duration of pressure differentials; however, these may be further influenced by variability in local geology. Perceived cycles across different frequencies correspond to induced flow from characteristic depths. Low frequency cycles or events extending days to weeks may promote enhancement of eventual surface emissions from deep underground radon sources if coupled with compatible source configurations; particularly frequencies less than 1E-05 hertz may breach finite low permeability covers and drive predictable intra-seasonal radon cycles that may not be possible for higher frequency signals. Demonstrations of these cycles is explored for consideration in both bounding surface emissions and measuring the resulting air concentrations. Real-world examples and exploratory scenarios modeled in the RnMod3d gas transport code are presented.

Introduction

Radon, specifically radon-222 in this context, is a natural decay product of primordial radium and uranium present in varying degrees across nearly all rock and soil types. In porous media the gas emanates into the pore space and is subject to the transport mechanisms of physical fluids, predominately diffusive and advective processes (Rogers and Nielson, 1991; Nazaroff, 1992). Basic diffusion is broadly considered the driving process for most soil-to-atmosphere gas exchanges (U.S. Nuclear Regulatory Commission [NRC], 1984, 1989); however, advective flows induced from pressure gradients can generate observable enhancements of eventual surface emissions as determined by the geophysical characteristics of the medium (Nilson et al., 1991; Holford et al., 1993). Pressure-driven flow of bulk soil gases in real environments are generally attributable to time-variable barometric pressure fields (Massman, 2006; Perrier and Girault, 2013), though discrete pressure differentials such as those from building structures can also be observed (Turk et al., 1990). The direct influence of barometric pressure on the advection of pore space radon concentrations is often perturbed by time-variable and perhaps cyclical interferences including shrink-swell cycles of the medium (Holtz and Gibbs, 1956), soil temperature (Carslaw and Jaeger, 1959; Kitto, 2005), precipitation and pore space saturation (Nazaroff, 1992; Kitto, 2005), and surficial wind speed (Clarke and Waddington, 1991), among others. Although in ideal cases pore space radon would be expected to vary inversely with barometric pressure (i.e., increase during a decreasing pressure event) (Tsang and Narasimhan, 1992; Holford et al., 1993), this relationship is heavily subject to the compatibility of independent meteorological and geological conditions relative to the radon-generating source configuration.
This work examines the interplay between variable medium gas permeability and barometric pressure cycles across different time scales and the resulting influence on discrete subsurface radon sources at characteristic depths. In particular, the penetration of a pressure field with a given frequency into the pore space influences subsurface gases at characteristic depths determined by the frequency of the signal and the gas permeability of the medium. The relative influence of advective forcing to molecular diffusion varies; however, the enhancement of soil gas emissions may be dramatic for compatible source configurations. As the radon testing paradigm for both surface flux (U.S. Environmental Protection Agency [EPA]) and air concentration (American Association of Radon Scientists and Technologists [AARST], 2019, 2021) continues to utilize short-term measurements as an initial screen or determination, understanding the role of predictable radon cycles extending days to weeks may be valuable to contextualize initial measurements in term of temporal bias.

Pressure-Driven Soil Gas Advection

Modeling the collective behavior and motion of radon soil gas over time within a porous medium (i.e., soil or earthen cover materials) must account for the radiological decay and ingrowth from intrinsic radium, if present, as well as the diffusion and advection properties described by Fick’s Laws and Darcy’s Law, respectively, for physical fluids. Fick’s Laws broadly define the net spatial tendency of individual particles to drift along a concentration gradient from areas of high concentration to a lower concentration. From Fick’s second law in one vertical dimension, \( z \), the medium pore space radon concentration varies over time, \( t \), by:

\[
\frac{\partial C}{\partial t} = D_m \frac{\partial^2 C}{\partial z^2}
\]

where

\( C = \) air-filled pore space radon concentration (Becquerel [Bq] per meter cubed [m³]), and

\( D_m = \) air-filled pore space radon diffusion coefficient (meters squared per second [m²/s]).

Darcy’s Law defines the bulk flow of fluids through a porous medium along a pressure gradient from areas of high pressure to areas of lower pressure. In one vertical dimension, \( z \), the flow is defined as:

\[
q = \frac{-K \partial p}{\nu \partial z}
\]

where

\( q = \) advective flow velocity of air-filled pore space radon (m/s),

\( K = \) bulk medium air permeability (m²),

\( \nu = \) dynamic viscosity of air (pascal [Pa] s), and

\( p = \) pore gas pressure (Pa).

Thus, the general material balance transport equation for air-filled pore space radon concentration simplified to one dimension, \( z \), is given (Rogers and Nielson, 1991; Nazaroff, 1992) as:
\[
\frac{\partial C}{\partial t} = D_m \frac{\partial^2 C}{\partial z^2} - \frac{K \partial p \partial C}{\nu \partial z \partial z} - \lambda C + R p \lambda E
\]

where

\( \lambda = \) radon decay constant (s\(^{-1}\)),

\( R = \) bulk medium radium activity concentration (Bq per kilogram [kg]),

\( \rho = \) bulk medium dry density (kg/m\(^3\)), and

\( E = \) radon emanation into the medium pore space (unitless).

Steady state diffusion solutions have been explored (NRC, 1984, 1989); however, the influence of external barometric forcing on subsurface radon gas remains a fairly niche field of study largely due to the site-specific complexity of pore gas pressure gradients and the interplay between the local climate and geology. Independent evaluation of bulk soil gas advection provides a means to explore potential enhancement of surface emissions from defined barometric variability. Restricting the configuration of a vertical column of length, \( L \), to a fixed porosity and isothermal transitions reduces the linear heat flow, and pore space pressure, within the medium (Carslaw and Jaeger, 1959) to:

\[
\frac{\partial p}{\partial t} = D_p \frac{\partial^2 p}{\partial z^2}
\]

where \( D_p \) is pneumatic diffusivity (m\(^2\)/s). Pneumatic diffusivity broadly describes the capacity of an induced pressure wave at the surface to penetrate or “diffuse” into a medium and is predominately dependent on the gas phase permeability of the medium. It is defined (Massman, 2006; Perrier and Girault, 2013) in this context as:

\[
D_p = \frac{K^g P_0}{n^g \nu}
\]

where

\( K^g = \) gas phase permeability within the medium (m\(^2\)),

\( P_0 = \) mean absolute barometric pressure (Pa), and

\( n^g = \) air-filled pore space (i.e., unsaturated) (unitless).

If a vertical column of length, \( L \), is considered with zero initial pressure (at time zero), a fixed pressure at depth, and constant defined periodic barometric pressure variation at the surface, \( p_{atm} \):

\[
p_{atm}(t) = p_1 \sin(\omega t + \phi)
\]

the general solution to (4) with depth, \( z \), over time, \( t \), has been obtained (Carslaw and Jaeger 1959):

\[
p(z, t) = p_1 A \sin(\omega t + \phi + \phi) + \\
2p_1 \pi D_p \sum_{n=1}^{\infty} \frac{n(-1)^n(D_p n^2 \pi^2 \sin \phi - \omega L^2 \cos \phi)}{D_p^2 n^4 \pi^4 + \omega^2 L^4} \sin \left( \frac{n\pi z}{L} \right) \exp \left( -\frac{D_p n^2 \pi^2 t}{L^2} \right)
\]
where

\[ A = \frac{\sinh \theta z (1 + i)}{\sinh \theta L (1 + i)} \]  
and

\[ \varphi = \text{arg} \left( \frac{\sinh \theta z (1 + i)}{\sinh \theta L (1 + i)} \right) \]  

and where

\[ \theta = \left( \frac{\omega}{2D_p} \right)^{\frac{1}{2}} \]  

where

\[ p_1 = \text{amplitude of barometric surface pressure (Pa)}, \]
\[ \omega = \text{angular frequency of the barometric surface pressure (hertz [Hz])}, \]
\[ \phi = \text{phase shift (unitless)}. \]

If the variable barometric pressure at the surface can be expressed as a Fourier series rather than a single periodic component:

\[ p_{atm}(t) = \sum_{i=1}^{\infty} p_i \sin(\omega_i t + \phi_i) \]  

the steady periodic component of (7) can be expressed as:

\[ p(z, t) = \sum_{i=1}^{\infty} p_i A_i \sin(\omega_i t + \phi_i + \phi_i) \]  

Therefore, there is direct relationship between the relative intensity of underlying component barometric frequencies and the induced spatial gradient with depth, \( z \), through the scalar magnitude, \( A \), and the frequency-specific pneumatic attenuation length, \( \theta^{-1} \).

**Identification of Potential Forcing Frequencies**

The U.S. National Oceanic and Atmospheric Administration (NOAA) maintains a central database of local climatological data (LCD) containing hourly, daily, and monthly summaries for approximately 950 Automated Surface Observing System (ASOS) stations throughout the United States. Critical among these data are hourly station barometric pressure measurements spanning decades in many geographic locations. True barometric pressure signals, or time series, consist of a complex superposition of underlying frequencies indicative of competing meteorological processes as well as extensive spectral noise. Figure 1 presents spectral decomposition of hourly pressure measurements at six regionally representative stations from 2011 through 2020 using the Fast Fourier Transform (FFT) algorithm of the Python scientific computing library, NumPy (NumPy Community, 2021), and converted to a period domain in days to illustrate historic tendencies across different geographical and meteorological regions of the United States.
Figure (1): United States regional barometric pressure components in days via Fast Fourier Transform (FFT) from hourly station measurements (inches of mercury) between 2011 and 2020 (NOAA). (A.) Spokane, WA (Pacific Northwest): Identifiable low frequency peak at 13.6 days; (B.) Albuquerque, NM (Central U.S.): Identifiable low frequency peaks at 13.6, 24.6, and 32.6 days; (C.) Chicago, IL (Great Lakes): Identifiable low frequency peak at 33.5 days; (D.) Boston, MA (Northeast): Identifiable low frequency peak at 22.9 days; (E.) Los Angeles, CA (Southwest): No identifiable low frequency peaks, (F.) New Orleans, LA (South/Southeast): No identifiable low frequency peaks
The station data sets within the NOAA LCD database record hourly measurements as well as sporadic additional measurements at irregular frequencies for notable site condition changes (i.e., the onset of a large precipitation event or passing frontal system). Several hourly measurements were also not recorded or recorded as “null” throughout the queried interval. To maintain uniform data spacing for the FFT, extra non-hourly measurements were omitted from the final set and gaps (~1% of the domains considered) were linearly interpolated using the adjacent recorded pressures. If extended gaps were identified spanning more than a few hours which would compromise a reliable interpolation, compatible station data was obtained, when possible, from the corresponding Weather Underground (www.wunderground.com) historical database.

Several notable component frequencies are broadly identifiable across the northern United States. For example, as shown in Figure 1, a discernable peak at 13.6 days is identifiable in the Pacific Northwest (Spokane, Washington), a peak at 33 days is identifiable in the Great Lakes (Chicago, Illinois), and a peak at 23 days is identifiable in the Northeast United States (Boston, MA). Other areas may also exhibit simultaneous influence from neighboring regions such as that seen in the Central United States (Albuquerque, NM). Notable, however, are the lack of low frequency component frequencies (i.e., much less than the diurnal frequency) for the predominately southern United States. Although prominently identifiable and naturally occurring component frequencies may be common, their overall significance to soil gas transport may vary (Harp et al., 2019). If previous historic tendencies may be interpreted as a reasonable expectation of general future trends (i.e., broad meteorological cycles continue locally in the immediate future), then identifying notable component frequencies can assist in predicting pressure-driven soil gas cycles (Harp et al., 2019).

A forcing frequency can be defined in this context as a discrete observable frequency for which non-negligible soil gas transport is induced by barometric forcing, and more specifically as inducing a non-negligible enhancement of surface emissions compared to diffusion alone. This can be interpreted in terms of the Schmidt number (Sc) defined as the ratio of the kinematic viscosity and particle diffusivity of the fluid such that:

$$Sc = \frac{\mu}{\rho_{fluid} D_m}$$  \hspace{1cm} (13)

where $\rho_{fluid}$ is the fluid density (kg/m$^3$). For scenarios with a large Sc (i.e., low molecular diffusivity), soil gas transport by advection may be significant (Webster, 2003; Massman, 2006); however, for low Sc less than unity (i.e., high molecular diffusivity) diffusion tends to dominate over any potentially observable pressure-driven advection (Waddington et al., 1996; Massman, 2006). Lastly, source configuration relative to pressure frequency and the corresponding pneumatic attenuation length may identify advection susceptibility, or most notably, the source configuration relative to the depth of non-negligible pressure gradients. For specific combinations of compatible medium permeability and a legitimate forcing frequency where the overall pneumatic attenuation length breaches into heterogeneous pockets of rich soil gas (i.e., a subsurface source), the possibility of predictable surface emission enhancement may be reasonably anticipated.
Source Configuration and Frequency Sensitivity Cases

The most direct evaluation of a relationship between radon emissions and barometric forcing would be a spectral comparison of the individual time series and Fourier transforms over the same observation interval. While this has been shown in the domain of the diurnal and its harmonics (Porstendörfer et al., 1994; Barbario et al., 2018), adequate spectral resolution in the low frequency domain between the annual and diurnal periods (i.e., roughly between $10^{-8}$ and $10^{-5}$ Hz) requires substantial unbroken data sets. For example, the resolution of an FFT is generally inversely proportional to half of the sample size. Therefore, resolution down to $10^{-8}$ Hz would require at least $2 \times 10^8$ seconds or over 6 years of unbroken hourly radon measurements which is practically and logistically challenging in the age of heating, ventilation, and air conditioning.

RnMod3d is a numerical model of bulk soil-gas and radon transport in porous media developed at Risø National Laboratory (Anderson, 2000) that may be alternatively utilized to simulate total gas transport for a series of sensitivity cases. The code is a finite-volume model capable of running three-dimensional steady-state and transient scenarios considering radioactive decay, anisotropic generation from intrinsic radium, diffusion, and advection through Darcy flow of bulk soil gas. As was assumed in the logic from generic linear heat flow to pressure in (4), RnMod3d also maintains constant porosity and isothermal transitions over time.

Table 1 presents six RnMod3d cases evaluating the sensitivity of surface emission enhancement with respect to source configuration and barometric frequency. Each case assumes a two-layer cover/source configuration with unique diffusivity, permeability, and intrinsic radium content as shown in Figure 2. It should be noted that although the homogenous source cases assume the two-layer configuration, the “cover” and “source” layers are identical. In all cases, both a diurnal and 24-day pressure cycle are run independently for comparison. The mean barometric pressure, $P_0$, and cycle amplitude, $p_1$, were assumed based on NOAA LCD for Niagara Falls, New York between 2011 and 2020.

<table>
<thead>
<tr>
<th>Source Radium Concentration</th>
<th>Cover Radium Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>5000 Bq/kg (becquerel per kilogram)</td>
<td>40 Bq/kg (becquerel per kilogram)</td>
</tr>
<tr>
<td>500 Bq/kg (becquerel per kilogram)</td>
<td></td>
</tr>
</tbody>
</table>

Figure (2): Homogenous radium-226 source configuration (left) and heterogeneous radium-226 source configuration (right) for RnMod3D sensitivity cases. Note the assumed geophysical parameters for the cover and source material are presented in Table (1). All model runs assume a two-layer configuration; however, the homogenous configuration cases assume identical cover and source characteristics (i.e., one homogenous source).
<table>
<thead>
<tr>
<th>$^2$RnMod3D \ PARAMETRS</th>
<th>Case 1 Homogenous low-$K$ (F0001)</th>
<th>Case 2 Homogenous high-$K$ (F0002)</th>
<th>Case 3 Heterogeneous 1-meter depth low-$K$ (F0003)</th>
<th>Case 4 Heterogeneous 1-meter depth medium-$K$ (F0004)</th>
<th>Case 5 Heterogeneous 1-meter depth high-$K$ (F0005)</th>
<th>Case 6 Heterogeneous 1-meter depth extreme-$K$ (F0006)</th>
</tr>
</thead>
<tbody>
<tr>
<td>radon half-life, $\lambda$ (s$^{-1}$)</td>
<td>2.10E-06</td>
<td>2.10E-06</td>
<td>2.10E-06</td>
<td>2.10E-06</td>
<td>2.10E-06</td>
<td>2.10E-06</td>
</tr>
<tr>
<td>mean pressure, $P_0$ (Pa)</td>
<td>10,000</td>
<td>10,000</td>
<td>10,000</td>
<td>10,000</td>
<td>10,000</td>
<td>10,000</td>
</tr>
<tr>
<td>cycle amplitude, $p_1$ (Pa)</td>
<td>1,000</td>
<td>1,000</td>
<td>1,000</td>
<td>1,000</td>
<td>1,000</td>
<td>1,000</td>
</tr>
<tr>
<td>default saturation, $S_w$</td>
<td>0.01</td>
<td>0.01</td>
<td>0.01</td>
<td>0.01</td>
<td>0.01</td>
<td>0.01</td>
</tr>
<tr>
<td>Ostwald partition, $L_O$</td>
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<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td>dynamic viscosity, $\nu$ (Pa s)</td>
<td>1.8E-05</td>
<td>1.8E-05</td>
<td>1.8E-05</td>
<td>1.8E-05</td>
<td>1.8E-05</td>
<td>1.8E-05</td>
</tr>
</tbody>
</table>

**Cover Material**

| thickness, $l$ (m) | 1 | 1 | 1 | 1 | 1 | 1 |
| density, $\rho$ (g/cm$^3$) | 1.6 | 1.6 | 1.6 | 1.6 | 1.6 | 1.6 |
| particle diameter, $d_A$ (m) | 1E-06 | 1E-06 | 1E-06 | 1E-06 | 1E-06 | 1E-06 |
| porosity, $n$ | 0.3 | 0.3 | 0.3 | 0.3 | 0.3 | 0.3 |
| saturation, $m$ | 0.7 | 0.7 | 0.7 | 0.7 | 0.7 | 0.7 |
| Ra concentration, $R$ (Bq/kg) | 40 | 40 | 40 | 40 | 40 | 40 |
| diffusivity, $D$ (m$^2$/s) | 1.5E-08 | 3.4E-07 | 3.4E-07 | 3.4E-07 | 3.4E-07 | 3.4E-07 |
| permeability, $K$ (m$^2$) | 1.9E-17 | 5.3E-16 | 4.5E-15 | 4.5E-15 | 1.1E-14 | 1.1E-14 |
| permeability, $K$ effective (m$^2$) | 3.4E-18 | 1.8E-16 | 1.5E-15 | 1.5E-15 | 3.9E-15 | 3.9E-15 |

**Source Material**

| thickness, $l$ (m) | 5 | 5 | 4 | 4 | 4 | 4 |
| density, $\rho$ (g/cm$^3$) | 1.6 | 1.6 | 1.6 | 1.6 | 1.6 | 1.6 |
| particle diameter, $d_A$ (m) | 1E-06 | 5E-06 | 1E-04 | 1E-04 | 1E-04 | 1E-04 |
| porosity, $n$ | 0.3 | 0.33 | 0.3 | 0.3 | 0.3 | 0.3 |
| saturation, $m$ | 0.7 | 0.5 | 0.7 | 0.7 | 0.7 | 0.7 |
| Ra concentration, $R$ (Bq/kg) | 500 | 500 | 5,000 | 5,000 | 5,000 | 5,000 |
| diffusivity, $D$ (m$^2$/s) | 1.5E-08 | 3.4E-07 | 1.5E-08 | 1.5E-08 | 1.5E-08 | 1.5E-08 |
| permeability, $K$ (m$^2$) | 1.9E-17 | 4.5E-15 | 8.9E-15 | 8.9E-15 | 8.9E-15 | 8.9E-15 |
| permeability, $K$ effective (m$^2$) | 3.4E-18 | 1.5E-15 | 1.6E-15 | 1.6E-15 | 1.6E-15 | 1.6E-15 |

**Steady State**

| Concentration, 50 cm (Bq/m$^3$) | 6.6E+05 | 2.2E+05 | 2.7E+05 | 4.1E+05 | 4.1E+05 | 4.1E+05 |
| Concentration, 150 cm (Bq/m$^3$) | 7.3E+05 | 4.1E+05 | 3.7E+06 | 6.7E+06 | 6.7E+06 | 6.7E+06 |
| Concentration, 350 cm (Bq/m$^3$) | 7.3E+05 | 4.9E+05 | 3.7E+06 | 7.3E+06 | 7.3E+06 | 7.3E+06 |
Table (1): Summary radon flux enhancement from barometric pressure forcing per RnMod3d case scenario

1 All input parameters are realistically assigned per case description unless otherwise defined or calculated
2 Mean barometric pressure ($P_0$) and cycle amplitude ($p_1$) established observationally based on measured station pressure at the Niagara Falls International Airport from 2011 - 2020 per the National Oceanic and Atmospheric Administration (NOAA)
3 As defined by Perrier and Girault (2013) for means of calculating gas-phase permeability (see 8.)
4 The Ostwald water/gas partition coefficient as defined by Anderson (2000)
5 As defined by Anderson (2000) and Perrier and Girault (2013)
6 As calculated empirically per Rogers and Nielson (1991)
7 As calculated empirically per Rogers and Nielson (1991)
8 Effective gas-phase permeability to account for water-filled pore space per Perrier and Girault (2013)
9 As defined by Carslaw and Jaeger (1959) and in this context by Massman (2006) considering gas phase permeability per Perrier and Girault (2013)
10 Flux enhancement defined as the ratio of the maximum flux with advection to the steady state

**NOTE:** In this context permeability descriptions are based on the cover pneumatic attenuation length relative to its thickness for a 24-day periodicity. For example, "low" corresponds to a pneumatic attenuation length less than the cover thickness, "medium" corresponds to a length generally comparable to the cover, "high" corresponds to length greater than the cover, and "extreme" corresponds to a length much greater than the cover or greater than both the cover and source material. Descriptions for the homogenous cases are equivalent to those of the heterogeneous source cases.
Notable conclusions of the sensitivity cases are presented below:

1. *Surface emission (i.e., surface flux) enhancement tends to increase across frequencies as the overall gas permeability increases.* This is direct result of Darcy flow and its relative influence is ultimately limited by decreasing Schmidt numbers; diffusivity tends to increase with permeability until eventually diffusion wholly dominates.

2. *Surface emission (i.e., surface flux) enhancement from homogenous source configurations tends to increase with increasing barometric frequencies.* Case 2 of the homogenous sources demonstrates the influence of the pressure gradient magnitude. Although the pneumatic diffusivity of the 24-day barometric cycle corresponds to a deeper attenuation length and a greater volume of influenced soil gas than the diurnal, the flow is proportional to the magnitude of the pressure gradient from Darcy’s Law. For a constant permeability, the pressure varies more starkly with depth at higher frequencies and thus creates a more intense gradient.

3. *Surface emission (i.e., surface flux) enhancement from discrete or heterogeneous subsurface source configurations tends to increase for barometric frequencies which correspond to pneumatic attenuation lengths generally greater than the depth.* Cases 5 and 6 demonstrate increasing flux enhancement with increasing permeability and decreasing frequency. Collectively these contribute to increasing pneumatic percolation through the cover material into richer subsurface soil gas. As permeability continues to increase, as shown in Case 6, higher frequencies begin to penetrate deeper with more intense pressure gradients than lower frequencies and functions as a limiting factor.

**Discussion and Examples**

The Niagara Falls Storage Site (NFSS) is a federal facility in Niagara County, New York managed and operated by the U.S. Army Corps of Engineers (USACE) under the Formerly Utilized Sites Remedial Action Program. The main feature of the NFSS is a 10-acre earthen Interim Waste Containment Structure containing uranium ore processing residues accumulated by the Manhattan Engineer District and the Atomic Energy Commission pending eventual disposal. These radium-containing residues range from low activity miscellaneous waste to the high activity K-65 residues generated from uranium ore containing 35 to 65 percent U₃O₈. The structure hierarchy shown in Figure 3 is summarized as follows (USACE, 2015):

- 0.15-cm topsoil layer with shallow-rooted turf to minimize erosion and frost damage,
- 0.31-m layer of loosely compacted soil as a protective cover to the underlying clay,
- 0.9-m compacted, low-permeability clay cap designed to a hydraulic conductivity of 2E-8 centimeters per second (or a permeability of approximately 2E-17 m²),
- A large variable layer of miscellaneous wastes (contaminated sands, soils, and debris),
- Ore residues located within legacy building foundations to the south, and low activity residues on the original ground surface to the north.
Figure (3): West to east cross section diagram adapted from Figure 1-7 of the Feasibility Study Report for the Interim Waste Containment Structure (USACE, 2015).

Figure (4): (top) Running hourly barometric pressure from the Niagara Falls International Airport (NOAA) between September 2019 and November 2020 with 10-day running mean (bottom) Niagara Falls, New York barometric pressure components in days via FFT from hourly station measurements (inches of mercury) between 2011 and 2020 (NOAA). Note the peak cluster at 30 and 33 days coinciding with the general periodicity of the running mean.
The southern portion of the Interim Waste Containment Structure is a prime example of a discrete heterogeneous source configuration in the presence of a prominent low frequency barometric pressure component as shown in Figures 3 and 4. Radon emissions are limited under Title 40 of the Code of Federal Regulations (CFR) Part 61, Subpart Z which require annual compliance demonstration through National Emission Standards for Hazardous Air Pollutants (NESHAP) reporting. Historically unique flux observations in 2018 and 2019 spurred a series of investigative measurements as summarized in Figure 5 (USACE, 2019, 2020, 2021). Biased measurements during October 2019 and May 2020 demonstrate not only significant temporal variability beyond that expected from diffusivity fluctuations alone but also significant flux enhancement consistent with low frequency pressure periodicity rather than diurnal or other higher frequency trends. While these higher barometric frequencies may appear significant in terms of direct observation (i.e., weather), they largely function as spectral noise on lower frequency signals for deep subsurface soil gases attributable to the residues.

It should be noted that environmental parameters such as pore space saturation and the resulting diffusivity and permeability may display seasonality. In these cases, seasonal gains in the overall gas permeability may create seasonal susceptibility to non-negligible soil gas advection if the resulting pneumatic diffusivity breaches into the subsurface source (such as sensitivity case transitions from Case 4 to Case 5 to Case 6). Therefore, approximating mean surface emissions or air concentrations from mean annual environmental characteristics may not be wholly representative if the soil gas response behaves more non-linearly according to a threshold rather than a gradient.

Figure (5): (left) Measured surface radon flux (becquerel [Bq] per square meter per second) in context of low frequency barometric pressure cycles (kilopascal [kPa]) highlighted by a 10-day running mean (NOAA). Note the flux increases with decreasing trending pressure (USACE, 2020) and (right) flux decreases with increasing trending pressure (USACE, 2021).
Conclusion

Pressure-driven advection of soil gas from discrete or heterogeneous subsurface sources is sensitive to the specific source configuration and the perceived frequency of barometric fluctuations. As barometric frequency decreases and pneumatic diffusivity increases, susceptible source configurations may experience non-negligible enhancement of surface emissions when compared to molecular diffusion alone. Identifying and understanding local pressure tendencies in the context of pneumatic diffusivity may be valuable for anticipating potential radon cycles per forcing frequency for extended monitoring; however, the conclusions presented for bulk Darcy flow are not specific to radon and may be experienced with other soil gas contaminants with varying degrees of efficiency. The current surface flux and air concentration testing paradigm focus on initial short-term measurements or site screens may introduce predictable bias if the test durations are less than that of a significant radon cycle. Short-term tests strategically performed to coincide with cycle minima or maxima may offer insight into extreme radon flux or air concentrations if desired; however, long-term tests with exposure durations exceeding a reasonably anticipated pressure-driven radon cycle should offer better representation of the mean over time.

References


U.S. Army Corps of Engineers (USACE), 2015. Final (R3) Feasibility Study Report for the Interim Waste Containment Structure at the Niagara Falls Storage Site, Lewiston, NY. Buffalo District, Buffalo.


Attachment: Sensitivity Case RnMod3d Code

program F0001prg; (* Amend program name F000X per sensitivity case, see Table 1 *)

{$MODE Delphi}

(* --------------------- RnMod3d jobfile ---------------------- *)
(* Project: Barometric Flux Enhancement Simulation *)
(* Created: March 13, 2021 *)
(* Revised: March 16, 2021 *)

{$I R3dirs03.pas}
uses R3defi03,R3Main03,R3Writ03;

const
LambdaRn222 = 2.09838e-6; (* 1/s *)
mu = 18.0e-6; (* Pa s *)
rho_g = 1.6e3; (* kg/m^3 *)
L_Ostwald = 0.30; (* water/gas partitioning *)
deltaP = 1000.0; (* Pa *)
Period = 24; (* Days *)
T_per = 24.0*Period*3600; (* Seconds *)
omega = 2*pi/T_per; (* Hertz, variable by case *)
P0 = 10000.0; (* Pa *)
year = 365*24*3600; (* Seconds *)

(* Horizontal (x) dimensions, m *)

(* Vertical (z) dimensions, m *)
L_z_soil = 4.00;
L_z_slab = 1.00;

(* Radium-226 concentration, Bq/kg, variable by case *)
ARa_soil = 500;
ARa_slab = 500;

(* Fraction of emanation, -, variable by case *)
f_soil = 0.2;
f_slab = 0.2;

(* Porosity, -, variable by case *)
etot_soil = 0.30;
etot_slab = 0.30;
(* Volumetric water content,  \( - \), variable by case *)
msat_soil  \( = 0.70; \)
msat_slab  \( = 0.70; \)

(* Bulk diffusivity, m\(^2\)/s, variable by case *)
D_soil  \( = 1.5 \times 10^{-8}; \)
D_slab  \( = 1.5 \times 10^{-8}; \)

(* Gas permeability, m\(^2\), variable by case *)
k_soil  \( = 3.4 \times 10^{-18}; \)
k_slab  \( = 3.4 \times 10^{-18}; \)

(* Probe Depths *)
pr1  \( = -0.5; \)
pr2  \( = -1.5; \)
pr3  \( = -3.5; \)
pr4  \( = -4.0; \)
pr5  \( = -4.5; \)

procedure grid;
begin
(* x-axis *)
set_FixVal(xFix1,0.000);
set_FixVal(xFix2,1.000);

set_axis_single(xFix1,xFix2,5,FocusA,1.0);

(* y-axis *)
set_FixVal(yFix1,0.000);
set_FixVal(yFix2,1.000);

set_axis_single(yFix1,yFix2,5,FocusA,1.0);

(* z-axis *)
set_FixVal(zFix1,-Lz_slab-Lz_soil);
set_FixVal(zFix2,-Lz_slab);
set_FixVal(zFix3, 0.00);
set_axis_double(zFix1,zFix2,14,14,FocusA,FocusB,2.0,2.0,0.5);
set_axis_double(zFix2,zFix3,6,8,FocusA,FocusB,1.8,1.8,0.5);
end;
procedure boundary_conditions_soilgas(i:itype;j:jtype;k:ktype);
begin
  cBC[fixed1]:=deltaP*cos(omega*tim); (* sinusoidal pressure variation with frequency, omega *)
  cBC[fixed2]:=0;
  if in_plane([inside,eqAB], (* Observe full slab *))
    i,xFix1,xFix2,
    j,yFix1,yFix2,
    k,zFix3,zFix3) then
    change_node(i,j,k,fixed1,ConX,ConX,ConX,ConX,ConX,ConX);
  if in_plane([inside,eqAB], (* Bottom *))
    i,xFix1,xFix2,
    j,yFix1,yFix2,
    k,zFix1,zFix1) then
    change_node(i,j,k,fixed2,ConX,ConX,ConX,ConX,ConX,ConX);
end;

procedure boundary_conditions_radon(i:itype;j:jtype;k:ktype);
begin
  (* cBC[fixed1]:=0; *)
  cBC[fixed2]:=0;
  if in_plane([inside,eqAB], (* Observe full slab *))
    i,xFix1,xFix2,
    j,yFix1,yFix2,
    k,zFix3,zFix3) then
    change_node(i,j,k,fixed2,ConX,ConX,ConX,ConX,ConX,ConX);
end;

procedure fluxes(i:itype;j:jtype;k:ktype);
begin
  if in_plane([inside,eqAB],
    i,xFix1,xFix2,
    j,yFix1,yFix2,
    k,zFix3,zFix3) then
    begin
      update_flxval(Flx1,bottom,i,j,k,plus); (* atmosphere *)
    end;
  if in_plane([inside,eqAB],
    i,xFix1,xFix2,
    j,yFix1,yFix2,
    k,zFix2,zFix2) then
    begin
      update_flxval(Flx2,bottom,i,j,k,plus); (* Leaving source *)
    end;
end;
if in_plane([inside,eqAB],
   i,xFix1,xFix2,
   j,yFix1,yFix2,
   k,zFix1,zFix2) then
   update_flxval(Flx3,top,i,j,k,plus); (* bottom *)
end; (* fluxes *)

procedure probes;
  var valid:boolean;
  begin
    obsval[obs1]:=fieldvalue(0.5,0.5,pr1,valid);
    obsval[obs2]:=fieldvalue(0.5,0.5,pr2,valid);
    obsval[obs3]:=fieldvalue(0.5,0.5,pr3,valid);
    obsval[obs4]:=fieldvalue(0.5,0.5,pr4,valid);
    obsval[obs5]:=fieldvalue(0.5,0.5,pr5,valid);
  end; (* probes *)

function materials(i:itype;j:jtype;k:ktype):mattype;
  var mat:mattype;
  begin
    mat:=mat1; (* soil *)
    if in_region(i,xFix1,xFix2,[inside,eqab],
       j,yFix1,yFix2,[inside,eqab],
       k,zFix2,zFix3,[inside,eqab]) then mat:=mat2; (* slab *)
    materials:=mat;
  end; (* materials *)

function m(i:itype;j:jtype;k:ktype):datatype;
  var mm:datatype;
  begin
    mm:=0;
    case materials(i,j,k) of
      mat1: mm:=msat_soil;
      mat2: mm:=msat_slab;
    else
      error_std('m','Unknown material');
    end;
    m:=mm;
  end;

function beta_soilgas(i:itype;j:jtype;k:ktype):datatype;
  var ea:datatype;
  begin
    ea:=0;
case materials(i,j,k) of
   mat1: ea:= etot_soil;
   mat2: ea:= etot_slab;
else
   error_std('e','Unknown material');
end;
beta_soilgas:=ea/P0;
end;

function e_soilgas(i:itype;j:jtype;k:ktype):datatype;
begin
  e_soilgas:=0;
end;

function G_soilgas(i:itype;j:jtype;k:ktype):datatype;
begin
  G_soilgas:=0;
end;

function Lambda_soilgas(i:itype;j:jtype;k:ktype):datatype;
begin
  Lambda_soilgas:=0;
end;

function D_soilgas(dir:dirtype;i:itype;j:jtype;k:ktype):datatype;
var kk:datatype;
begin
  kk:=0;
case materials(i,j,k) of
    mat1: kk:=k_soil;
    mat2: kk:=k_slab;
else
    error_std('D_soilgas','Unknown material');
end;
D_soilgas:=kk/\mu
end;

function e_radon(i:itype;j:jtype;k:ktype):datatype;
var ee:datatype;
begin
  ee:=0;
case materials(i,j,k) of
    mat1: ee:=etot_soil;
    mat2: ee:=etot_slab;
else
  error_std('e','Unknown material');
end;
e_radon:=ee;
end;

function beta_radon(i:itype;j:jtype;k:ktype):datatype;
var ea,ew:datatype;
begin
  ew:=m(i,j,k)*e_radon(i,j,k);
ea:=e_radon(i,j,k)-ew;
beta_radon:=ea+LOstwald*ew;
end;

function G_radon(i:itype;j:jtype;k:ktype):datatype;
var GG,ee,lam:datatype;
begin
  GG:=0;
ee:=e_radon(i,j,k);
lam:=lambdaRn222;
case materials(i,j,k) of
    mat1: GG:=rho_g*(1-ee)/ee*lam*f_soil * ARa_soil;
    mat2: GG:=rho_g*(1-ee)/ee*lam*f_slab * ARa_slab;
    else
      error_std('G','Unknown material');
    end;
G_radon:=GG;
end;

function Lambda_radon(i:itype;j:jtype;k:ktype):datatype;
begin
  Lambda_radon:=LambdaRn222;
end;

function D_radon(dir:dirtype;i:itype;j:jtype;k:ktype):datatype;
var DD:datatype;
begin
  DD:=0;
case materials(i,j,k) of
    mat1: DD:=D_soil;
    mat2: DD:=D_slab;
    else
      error_std('D_radon','Unknown material');
    end;
D_radon:=DD;
end;

(* soil-gas simulation *)
procedure Soil_gas_run;
begin
boundary_conditions_def := boundary_conditions_soilgas;
e_def := e_soilgas;
beta_def := beta_soilgas;
G_def := G_soilgas;
lambda_def := lambda_soilgas;
D_def := D_soilgas;
import_finalfield_guess := true;
flowfield := export;
use_fieldbuffer := cBUF1;
(*
export_field := true;
import_field_name := 'PRES00.dat';
export_field_name := import_field_name;
*)
relax_factor := 1.98;
flux_convset := [flx1..flx3];
probe_convset := [obs1..obs5];
conv_evaluation_period := 300;
min_iterations := 150;
max_iterations := 20000;
max_time := 60*60;
max_change := 1e-10;
max_residual_sum := 1e-8;
end;

(* radon simulation *)
procedure Radon_run;
begin
boundary_conditions_def := boundary_conditions_radon;
e_def := e_radon;
beta_def := beta_radon;
G_def := G_radon;
lambda_def := lambda_radon;
D_def := D_radon;
import_finalfield_guess := true;
flowfield := import;
use_fieldbuffer := cBUF2;
\texttt{(*
export\_field := \texttt{true};
import\_field\_name := \texttt{'Rn0000.dat'};
export\_field\_name := import\_field\_name;
*)
\texttt{relax\_factor := 1.0;
flux\_convset := \{flx1..flx3\};
probe\_convset := \{obs1..obs5\};
conv\_evaluation\_period := 300;
min\_iterations := 150;
max\_iterations := 20000;
max\_time := 60*60;
max\_change := 1e-10;
max\_residual\_sum := 1e-8;
end;

begin (* main *)
runid := \texttt{'0001'; (* Change runid per sensitivity case *)
solution := steady;
geometry := cartesian3d;
grid\_def := grid;
flux\_def := fluxes;
probe\_def := probes;
materials\_def := materials;
wr\_iteration\_line\_screen := false;
wr\_flux\_during\_calc\_screen := false;
wr\_axes := false;
tim :=0;
dtim :=3600; (* Hour increments *)

Soil\_gas\_run;
run\_model;

Radon\_run;
run\_model;

solution := unsteady;

repeat
tim:=tim+dtim;

Soil\_gas\_run;
run\_model;\texttt{)}
Radon_run;
run_model;

writeln(RES,tim/3600:16,',',
cBC[Fixed1]:16,',',
FlxVal[Flx1].j:16,',',
obsval[obs1]:16,',',
obsval[obs2]:16,',',
obsval[obs3]:16);

until tim>Tper;
close_model;
end.