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Unraveling Radon Transport: Evidence from a Calgary Breathing Well

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Summary

Radon gas inhalation is present at high activities in indoor air, making it the leading cause of lung cancer in people who don't use tobacco. Surprisingly little is known about the specific geogenic sources and subsurface pathways of radon, given its widespread presence and associated risks. The use of ^{210}Pb and ^{226}Ra disequilibrium is an innovative approach, designed to differentiate *in situ* from *ex situ* sources of radon. In this work ^{210}Pb and ^{226}Ra disequilibrium data are used in combination with major and noble gases and their isotopic composition to provide lines of evidence that high activities of radon can be associated with deep (i.e., crustal and mantle) gases, that are potentially transported kilometers in the subsurface before it decays, despite radon's short half-life ($T_{1/2}$ = 3.82 days). If successful, this approach can be used to differentiate different radon sources which could be useful in future radon testing and to further understand radon's migration pathways in the geological subsurface.

Theory / Method / Workflow

^{222}Rn is a gaseous daughter product in the ^{238}U → ^{206}Pb decay chain with a half-life of 3.82 days. Because radon is a gas and has a relatively short half-life, its presence is particularly dangerous. Inhalation of radon atoms increases the likelihood that radon will decay in the lungs and the solid radioactive radon daughter ^{218}Po → ^{210}Po products can become embedded into the lung tissue, further exposing the lungs to radiation for approximately 22 years after inhalation (Pearson et al., 2021). The dangerous nature of radon, as well as its frequent presence in indoor air in elevated activities, explain why radon is the leading cause of lung cancer in nonsmoking individuals and is the seventh leading cause of cancer-related deaths worldwide (Simms et al., 2021).

Despite the clear risks of radon gas in indoor air, its specific geogenic source(s) and subsurface pathways that allow it to reach the shallow subsurface before entering into homes are difficult to determine. Radon is created via the ^{238}U decay chain and is exclusively created by its direct parent isotope ^{226}Ra, which is heterogeneously distributed throughout geological materials (Molla et al., 2021). Efforts to correlate radon in indoor air with surficial and bedrock geology tend to show poor to modest correlations (Rääf et al., 2023). In part this could be because the radon in indoor air measurement is confounded by building architecture (Kahn et al., 2021) but could also be because *ex situ* radon transport is not considered. Also, radon activities observed in soil gas are highly affected by meteorological factors, such as air pressure, wind speed, temperature, precipitation, and humidity (Kashkinbayev et al., 2024). Since radon is a gas, transport in the deeper unsaturated zone is predominantly driven by diffusion. Dissolved radon movement in the saturated zone is primarily driven by groundwater

advection and diffusion which, combined with its 3.8 day half-life effectively limits the transport range <10 meters (Miklyaev et al., 2022). This may have confined the conceptual model for geogenic radon sources *in situ* (i.e., sourced in/near the shallow sediment) as opposed to *ex situ* (i.e., transported from another source) radon transport. Rapid radon transport can occur in the saturated zone under buoyancy if free phase gas, or 'geogas' is present (Mörner and Etiope, 2002). In this case, radon can be transported by a carrier gas (often CO_2) under buoyancy with estimated travel times as great as 7 km/day (Chen et al., 2023). In particular ^{222}Rn-containing geogas can be an *ex situ* source of Rn in the subsurface along its transport pathway.

In an effort to address the complex nature of the sources and pathways of radon, we present a novel approach that combines i) the activities of ^{226}Ra and ^{210}Pb (radon's most immediate daughter product with a relatively long half-life, $T_{1/2}$ = 22.3 years) in sediment samples, and ii) the radon emanation coefficients to estimate the relative *in situ* vs. *ex situ* radon exposure per mass of sample. The ^{226}Ra and ^{210}Pb activity of the sediment samples are measured by gamma spectroscopy (SNOLAB; snolab.ca), and estimated ^{222}Rn activity at secular equilibrium is compared with measured *in situ* radon after a 28 day 'jar test' (Stajic et al., 2023). Since ^{226}Ra has a half-life much longer than that of ^{210}Pb ($T_{1/2}$ = 1602 years), in a closed system their activities would be expected to arrive at secular equilibrium (i.e., the activity of ^{226}Ra ≈ the activity of ^{210}Pb, or $A^{226}Ra \approx A^{210}Pb$) after 150 years.

Since ^{222}Rn is the only intermediate isotope in the U decay chain that can exist in a gaseous state, it will partition into, and be mobile along with, any free phase gas is present. It is thus a good candidate for *ex situ* contributions. This would lead to sediment samples with greater activities of ^{210}Pb than ^{226}Ra compared to those expected at secular equilibrium (i.e., if $A^{210}Pb > A^{226}Ra$). Conversely, sediments where $A^{210}Pb < A^{226}Ra$ could result if any radon that emanated from the sediment is rapidly removed and decays elsewhere. Thus, the $A^{226}Ra:A^{210}Pb$ approach has the potential to differentiate between *in situ* and *ex situ* radon sources. The mapping of ^{226}Ra:^{210}Pb in sediment can provide field evidence if Rn-rich free phase gas transport is occurring, for example around water wells (Khan et al., 2024), fault zones (Etiope and Lombardi, 1995).

This approach was tested at a field site with clear evidence for *ex situ* radon. The site is an anonymous 'breathing well' in the Calgary region that 'exhales' high CO_2 and radon concentrations during periods of low air pressure (Morais et al., 2024). Radon and CO_2 concentrations in the well casing of the breathing well were monitored hourly over a 17-day period (Figure 1), along with air temperature, air pressure, and near-surface soil temperature and moisture. Radon activities were measured Durridge RAD7®, and CO_2 measurements were collected using a Senseair Sunrise® CO_2 detector. Free gas samples were collected from both the background and well bore to use for isotope and combined with major and noble gas analysis, to constrain the sources of gases. Sediment core samples were taken 12 m, 4 m and beside the well bore for $A^{226}Ra:A^{210}Pb$ analysis.

Results, Observations, Conclusions

The breathing action of the well was confirmed by 'whooshing noises' that coincided with air pressure changes. When air pressure rises, atmospheric air is forced into the well; When air pressure falls, gases (mainly CO_2, with high radon activities) rushes out of the well column.

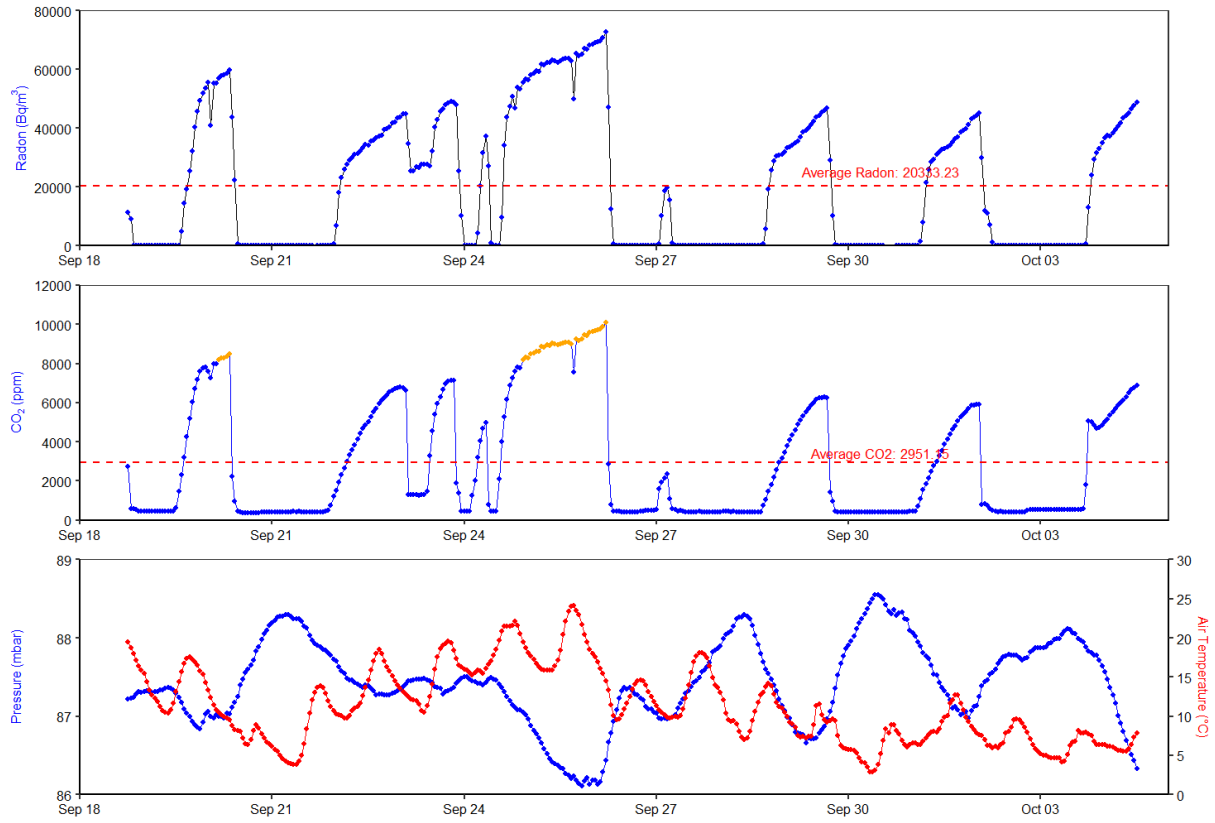

Figure 1: Time series of radon (Bq/m^3) and CO_2 in gas sampled hourly from the inside the well casing at a depth of 5.8 m, air pressure and temperature outside the well. CO_2 concentrations above the detector limit of 8000 ppm were extrapolated (and thus estimated) from the correlation between CO_2 and radon.

The radon and measured CO_2 data from this site follow very similar trends, with matching peaks and troughs, suggesting a common source. Their levels are visually related to air pressure, strongly increasing rates of air pressure changes occur when atmospheric concentrations of CO2 and Rn are observed in the well, while strongly decreasing rates of air pressure are correlated with much higher values. The breathing well data is substantially higher than the background location, with breathing well radon and CO_2 maximums of 72,753 Bq/m^3 and > 8000 ppm of CO_2 (10,074 ppm extrapolated), respectively (compared to background maxima of 2225 Bq/m^3 and 1229 ppm of CO_2). This difference in the activities and concentrations between the well bore values and background location suggests the radon and CO_2 in the well are of a different source than the background, and the extremely high radon suggests a source with a high concentration of radon's parent ^{226}Ra.

Additional data from gas grab samples, including composition, ^{13}C-CO_2 isotopes, and noble gas analysis, help suggest the sources of the radon and CO_2 through its associated gases

(N_2, O_2, Ne_{21}/Ne_{22}. All radon gas present was produced via decay of ^{226}Ra, making it impossible to determine its source directly. Due to this, we rely on the associated gases to infer the source and assume they originate from the same geogenic source, travelling together through the permeable subsurface. A summary of gases observed in free-gas samples is shown in Table 1, which, like Figure 1, shows a correlation between CO_2 and radon concentration and activity.

	Rn	O_2	N_2	CO_2
Rn	-	-	-	-
O^2	-0.26	-	-	-
N_2	0.61	-0.44	-	-
CO_2	0.94	-0.21	0.60	-

Table 1: Pearson correlation coefficients between gas species (n = 16). N_2 and O_2 concentrations are from the grab samples, Rn is measured with the Durridge RAD 7®, and CO_2 with the Senseair Sunrise® CO_2 sensor. Grey font indicates coefficients with P > 0.05 on the Pearson correlation significance test.

The gas data so far has not been able to identify a conclusive source but does show a statistically significant correlations between radon, CO_2 and N_2, all of which are commonly present in geogas. Conversely, inverse correlations between radon and oxygen suggest elevated radon activities are not associated with atmospheric gases. Noble gas analysis may provide further insight into the source of the observed gas in the well bore and will be presented from a related site if laboratory analysis are not available.

In conclusion, the sediment ^{210}Pb and ^{226}Ra disequilibrium, radon emanation tests, and gas composition data will be used in combination to differentiate *in situ* and *ex situ* radon sources in breathing wells.

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