CORRELATING RADON ACTIVITY WITH CARBON DIOXIDE CONCENTRATION IN AN IOWA CAVE

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Abstract

Reports of encountering "bad air" have occasionally resulted from humans entering cave passages with high levels of carbon dioxide. Humans breathing high concentrations of carbon dioxide will note obvious physiological impacts, ranging from breathlessness up to death at extreme values. Numerous caves have been shown to feature "bad air" in the form of high levels of radon gas, but instrumentation is required for this to be diagnosed. Although the sources that bring carbon dioxide and radon into the caves are different, it is plausible that poor ventilation of a cave's interior is a contributing factor toward that cave having high levels of either gas. This study was set up to measure the levels of both gases simultaneously in an Iowa cave, and to evaluate how highly correlated these values are to one another.

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Introduction

Stories of caves featuring "bad air" have accumulated over time, most commonly associated with high concentrations of carbon dioxide (CO_2) leading to discomfort and in extreme cases, death (Lewis, 2000). The conventional wisdom in the caving community has been that the higher density of CO_2 compared to air leads to it settling to the floor of the passage via gravitational forces, potentially leading to a "CO_2 sump" or "CO_2 trap", where a dangerously-high level may accumulate. Whereas it is correct that CO_2 is denser than air, calculation of the gravitational enrichment factor illustrates that this type of augmentation is so small that it is unlikely to be detectable (Badino, 2009). The CO_2 concentration in caves is produced by the balance of a handful of sources and sinks; Kowalczk (2010) has produced a mathematical model of the system. James (2010) states that CO_2 enrichment of cave air can be categorized as one of three types. The "Type I" involves injection of CO_2, with subsequent dilution of existing gases. Most caves are formed in carbonate rocks, dissolved by acidic ground water. The water becomes acidic by passing through the soil, which will have a very large partial pressure of CO_2 due to the biomass present. The acidified water can dissolve the carbonate rocks to form the cave, but if this water reaches a subterranean airspace, typically with a much lower CO_2 concentration, the equilibrium will reverse and CO_2 will outgas from the water, creating a major "Type I" CO_2 source (Palmer, 2007). Heavy surface rainfall may lead to an increase in the drip rate into the cave, which will enhance the transport of CO_2 into the cave air (Kowalczk, 2010). Anaerobic respiration via microbes can add CO_2 without consumption of O_2, thus making it another "Type I" source, and some volcanic processes also provide "Type I" sources. The "Type II" CO_2 enrichment involves CO_2 replacement of O_2, which will be caused by aerobic respiration and combustion/decay processes. Organic solids are carried into caves, most typically by water. The solids end up primarily deposited on the floor, where they will undergo oxidative decay and produce CO_2. If the decay takes place during a time and place of very poor ventilation with minimum convection, the CO_2 will accumulate in that region and will lead to elevated concentrations until ventilation improves. The "Type III" CO_2 enrichment only occurs in highly oxygen-deficient caves, and is unlikely to be a significant factor in the caves of northeastern Iowa.

Cave radon has a separate set of sources and sinks compared to CO_2 (Perrier, 2010), but for both of the gases cave ventilation is postulated to be a major factor. Gregorič (2013), working in a Slovenian cave, noted that both radon and CO_2 follow the seasonal pattern of having high levels during the summer, tapering down to much lower levels in the winter season. In the summer, most caves are cooler than the surface conditions, thus containing air of a higher density that doesn't exchange very much with the lower density surface air above it. In the winter, the cave air is less dense than the surface air, which tends to promote better ventilation, lowering both CO_2 and radon in the cave. When the cave entrance is vertical, this latter process has been described as a "cold air avalanche" (Perrier, 2010). Gregorič (2013) noted a very high CO_2-radon correlation in the spring, but less so in the summer. Outside temperature was noted as the biggest influence on radon levels, with atmospheric pressure not a significant factor. Perrier (2010) also observed correlation of these gases in an underground quarry, but felt that pressure was an important factor, producing "barometric pumping" where low pressure pulled greater amounts of the gases from the water and pore space of the rock strata, and high pressure forced the gases into the water and the pore space of the rock strata and prevented the normal diffusional input. Kowalczk (2010) also observed a significant correlation between CO_2 and

radon in a Florida cave, noting the importance of ventilation on both, and further pointing out that the radon sink via radioactive decay was not a major factor as the decay rate was small compared to the rate of air turnover in the study cave.

Kemling Cave lies near Dubuque, IA in limestone of Ordovician Age, and it has 3.51 kilometers of surveyed passages (Klausner, 2019). The cave has a single entrance of cross-section circa 1 m^2 that is gated, but not sealed, to facilitate bat movement in and out of the cave. As such there is open communication with the external atmosphere. Immediately inside the entrance is a 7.19 m vertical pit, followed by the balance of the cave, which can best be described as a horizontal maze. The nature of the entrance would make this cave susceptible to ventilation via cold air avalanches in cold periods, and the presence of significant vegetation in the land above the cave suggests the likelihood of "Type II" production of CO_2 following introduction of organic matter into the cave. Previous studies have shown high and variable radon in Kemling Cave, thus for CO_2 to be correlated with radon in a significant manner, it would also need to be highly variable (Welch, 2015, 2016, 2017, 2018).

Materials and Methods

Integrated average radon activity levels were measured with E-PERM® EIC (Electret Ionization Chamber) sensors, consisting of an electret of either the short-term [ST] or long-term [LT] variety, and a chamber of either the H, S, or L-OO variety, all from Rad Elec Inc. Chamber volumes were 960, 210, and 58 ml respectively, with the E-PERM becoming more sensitive with an increase in chamber volume. Electret voltages were measured with a SPER-1E electret voltage reader (Rad Elec). Calculations were done with WinSper software Version 2.3.21 or with Radon Report Manager software Version 3.8.44 from Rad Elec. Background gamma radiation exposure was evaluated with the Model 2 Gamma Ray Dosimeter manipulated with the Model 909B charger from Arrow-Tech. The EIC units were deployed in Tyvek® envelopes to protect them from mud and water while in the cave.

Temporal measurements of radon activity were undertaken using Radon Recon® continuous radon monitors and Recon Download Tool software v0.9.7 (Rad Elec Inc.). Recon measurements were acquired at 10 minute increments rather than the standard 1-hour increment via spreadsheet manipulation of the raw data file. The Recon units were deployed in Tyvek envelopes to protect them from mud and water while in the cave. Once in the Tyvek, the Recon units were packaged in their thermoplastic cases for transport and deployment in the cave. Prior work demonstrated that both the envelopes and the thermoplastic cases were transparent to radon (Stieff, 2012 and Welch, 2015) and did not impact measured levels. The Recon batteries were always recharged immediately prior to deployment, which then permitted up to four full days of data collection in the cave environment.

Carbon dioxide concentration was measured with a CM-0010 sensor using Gaslab 2.1 software from CO2Meter.com. A 60-cm piece of Tygon® tubing was attached to the CM-0010 gas inlet, and a Salter Labs 7000-0-25 water trap and an Alltech 2045 nylon particulate filter with 0.02 μm pores placed in the inlet flow path. The units were transported and deployed in a thermoplastic case repurposed from a Radon Scout Plus (Rad Elec). An external UB645k 6V lead-acid battery from Universal Power Group was used to power the unit for the trials lasting more than a few hours. By shutting off the display and the pump power when in between sample collections, a

CM-0010 with a fully-charged UB645k could run for 48 hours in the cave while collecting samples every 10 minutes. Carbon dioxide from the exhalations of the experimentalists could indeed impact the output of the CM-0010, so the operators moved to a location at least 10 meters away from the unit while it was collecting data. In some of the trials, a portion of the dataset was not used due to the risk of exhalations impacting the measurements.

Method Specifics

Part I, Trial 1

CM-0010: Used a 30-minute collection duration with a 10-minute sampling increment with internal battery power only, then used all the data. E-PERM: Used H chambers and ST electrets with a 30-minute collection duration. The chambers were transported in and out of the cave sealed with a dummy electret while the measuring electrets were kept uninstalled, capped, and in a polyethylene bag until needed. When at the station, the dummy electret was removed and the open chamber was waved around for 60 seconds to ensure it was filled with air specific to the site, at which point the actual electret was installed as quickly as possible, and the trial clock started. The E-PERM was kept in a Tyvek bag during collection. At the end of the collection window, the electret was quickly removed, capped, and bagged, and then it was replaced by the dummy electret for transit out of the cave. Sampling was done in a serial fashion for both sensors, so there was no time overlap whatsoever from station to station. The total collection time span was 315 minutes. A schematic of the experimental timeline is displayed in Figure (1).

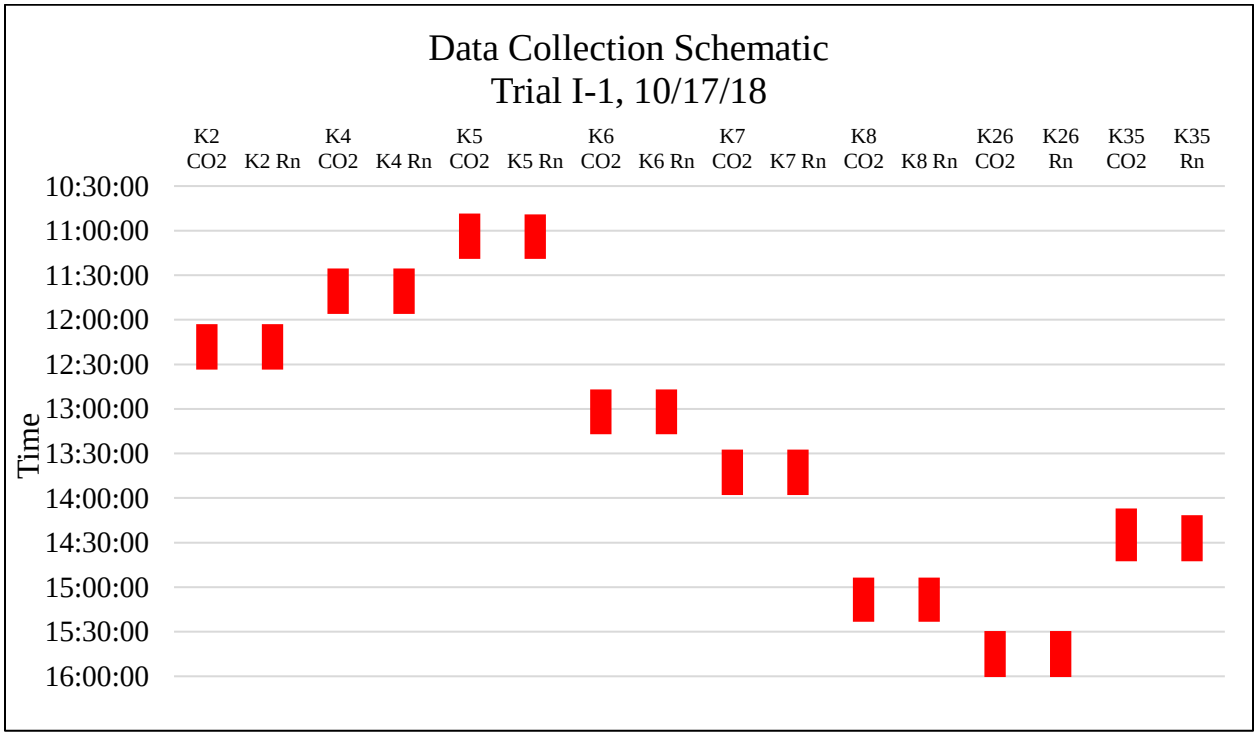

Figure (1): Schematic of data collection as a function of time for CO_2 and radon measurements in Kemling Cave, Trial I-1, 10/17/2018. See Figure (2) for station locations.

Part I, Trial 2
Same as Trial 1, except a 1-minute sampling increment was used in place of 10 minutes for the CM-0010, and a slightly different set of stations were used for sampling. For CM-0010 data processing, all data points were averaged throughout the 30-minute trial, excluding the first 5 points and the last point of each set, which were not used due to potential contamination from exhaled CO_2.

Part II, Trial 1
CM-0010: Used a 24-hour collection duration with a 10-minute sampling increment and power from an external UB645k 6V battery. The unit was covered with the lid of the thermoplastic case while running, and the inlet hose placed as far away from the Recon unit as possible.
Recon: Same duration and sampling increment as the CM-0010 using internal power. The unit was placed in a Tyvek bag while running.

Part II, Trials 2-6
Same as Trial 1 except that a 48-hour collection duration was used.

Results and Discussion

Part I: Radon vs. CO_2 as a function of location within the cave

Prior studies (Welch 2015, 2016) underlined the fact that radon will vary at different locations throughout Kemling Cave as long as a broad spatial set of sample locations were chosen. Thus, by running a CO_2 sensor side by side with radon sensors spread throughout the cave, a suitable evaluation of their degree of correlation could be observed. Sampling locations for this and all subsequent trials can be seen indexed on a plan view map of the cave in Figure (2). Preliminary trials started off using the small L-OO chambers for ease of transport, but the lower sensitivity of these chambers, even with ST electrets, didn't yield sufficient signal during the one to three hour collection windows utilized. The larger, and more sensitive S chambers, also produced a Δvoltage that was too small, yielding unacceptably large uncertainty values for the radon measurements. Despite these problems, the experiments were promising in that a significant correlation between CO_2 and radon was obvious from the trials.

To improve the experiment, H chambers were adapted for the radon measurements, coupled with ST electrets. These large chambers were normally shunned for in-cave work due to their size making transport difficult, but by designing an experiment whereby the different stations were evaluated sequentially without any time overlap, a single H chamber could be used for all of the stations, making transport less of an issue. This method required installation and removal of the electrets while in the cave, which always proved challenging to avoid getting the sensors muddy. The extreme sensitivity of the E-PERM with ST electret coupled with the H chamber allowed collection over very short temporal spans, which minimized the overall time needed for sequential data collection at multiple stations. Thirty minutes was selected as the sampling time at each station, which was easily enough time for successful CO_2 measurement, but very short

for the E-PERM. Although the match in time windows for the two gases wasn't absolute, it was less than a minute out of the 30-minute trial – as close as could be executed. The sampling

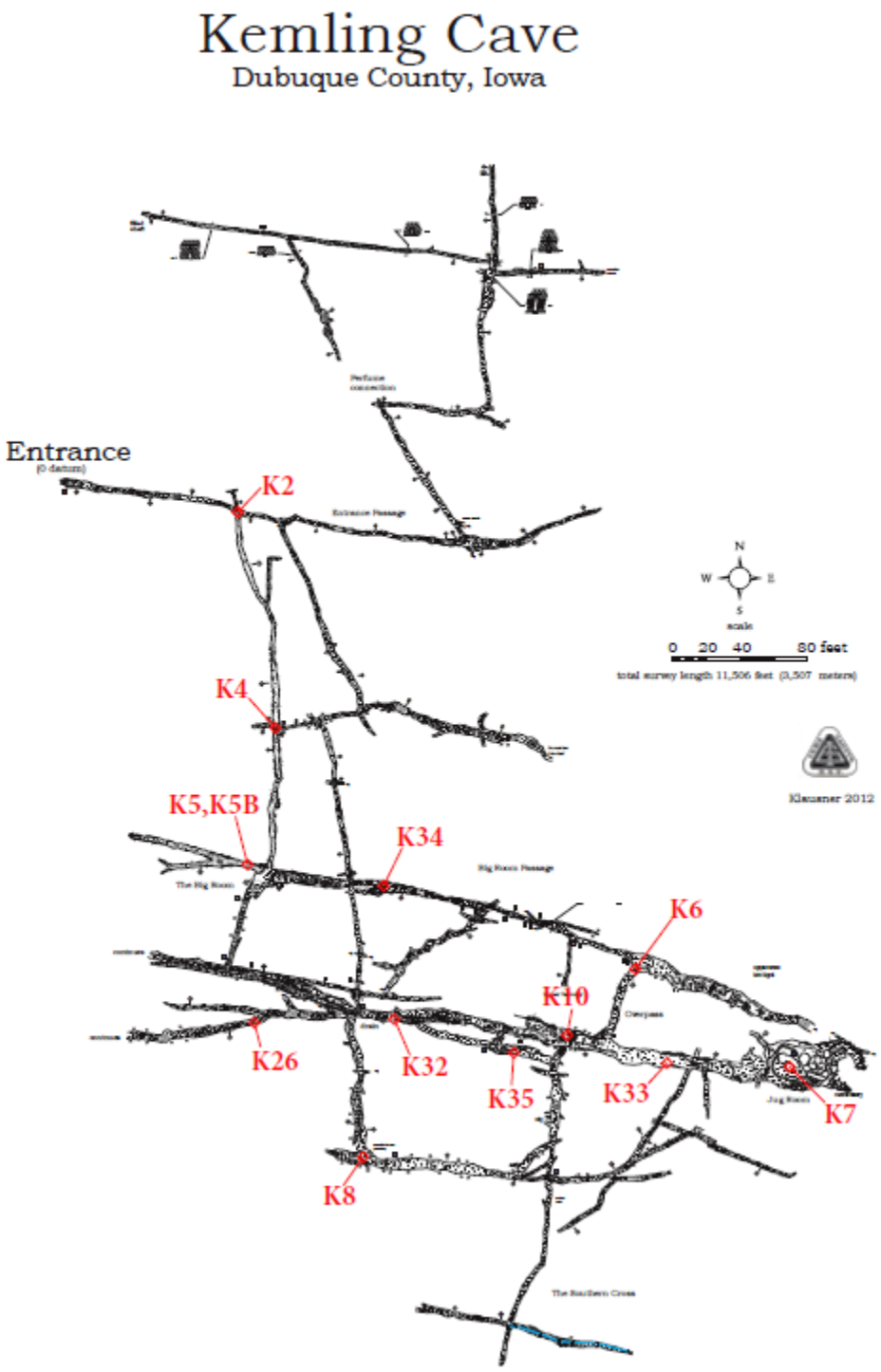

Figure (2): Plan view map of a portion of Kemling Cave showing data collection stations.

methodology ensured that the H chamber was filled with air from the station vicinity at the outset of the trial, so the 30-minute measurement block was sufficient to faithfully respond to radon at each station. However, the short time frame did not allow the level of radon decay products (RDP) present to rise up and reach a steady-state equilibrium value. This would produce calculated radon activities lower than reality, as the software algorithm assumed that the RDP were at a steady-state value. So the radon measurements in Part I should not be considered as highly accurate assessments. A correlation measurement between radon and CO_2 was the primary goal of this study. Given that all the stations had radon measured for the same 30-minute window, and given that this would result in each station having radon underestimated by a constant fraction, the resulting correlation coefficient calculation would find these underestimations all canceling out, and yield a valid R value.

The results from Trial I-1 using the H chambers can be seen in Figure (3), with an experiment schematic given in Figure (1). The error bars represent one standard deviation. A strong correlation (R=0.975) was observed between the two gases. The biggest limitation of

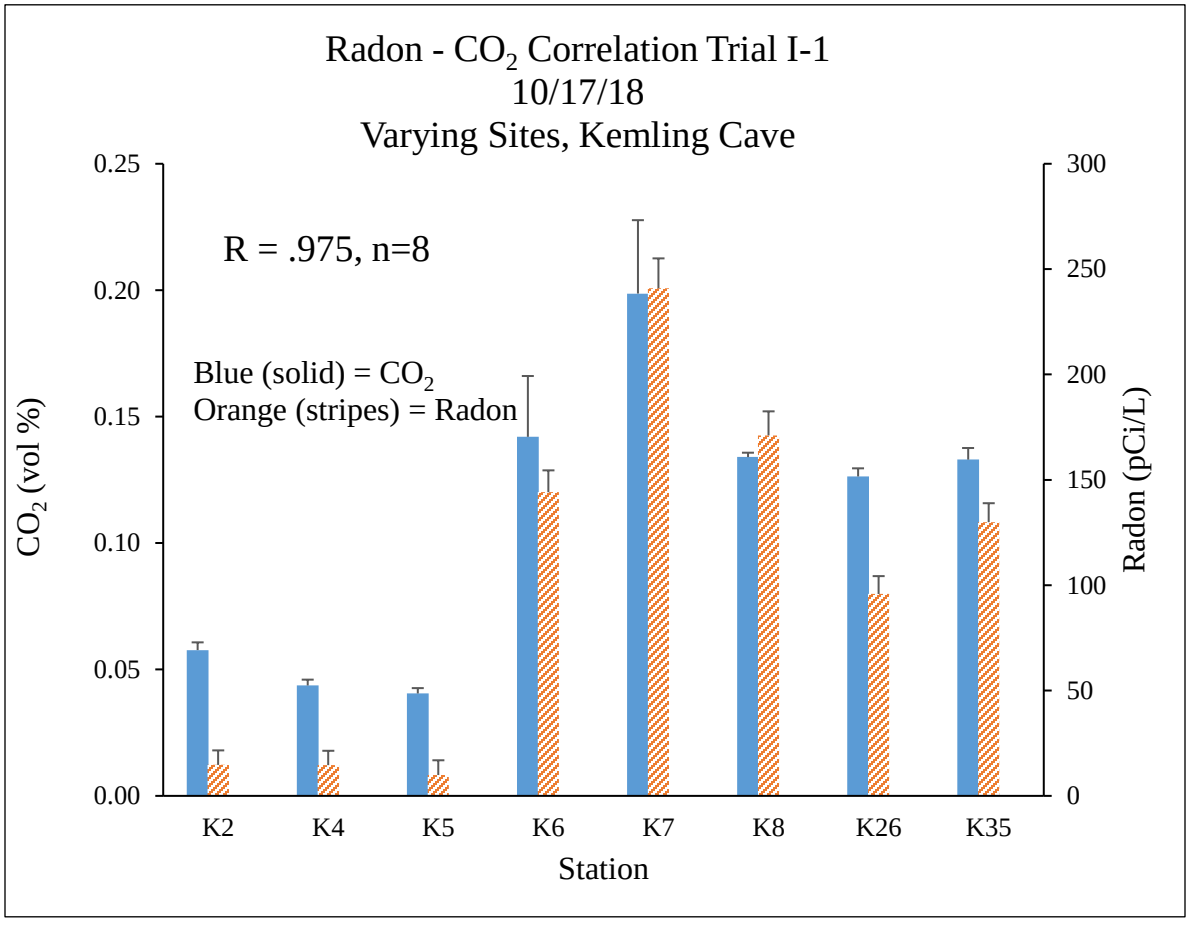

Figure (3): Trial I-1 correlation of radon and CO_2, 10/17/18, Kemling Cave.

Trial I-1 was that the sampling increment for CO_2 was chosen to be 10 minutes, yielding only 3 data points in the 30-minute trial. As such, the uncertainty seen for CO_2 in Figure (3) was much greater than that seen for prior trials. The radon uncertainty was also somewhat larger than seen before, but this was mainly due to having chosen a low-radon day for the trial, as low electret

voltage changes always reduced precision. One more experiment, Trial I-2, was undertaken with the CO_2 sampling interval set from ten minutes to one minute to provide more points and improve the CO_2 precision, and shifting to sampling locations deeper in the cave with presumably higher radon to improve the radon precision, given the 30-minute collection window. The results from Trial I-2 can be seen in Figure (4). The precision of both variables was greater in Trial I-2 compared to Trial I-1. The correlation was slightly weaker at R=0.951 than that measured from Trial I-1, but overall the trials produced pretty consistent results and both feature a strong correlation that would suggest that some of the same factors were impacting the activity level of both of the gases.

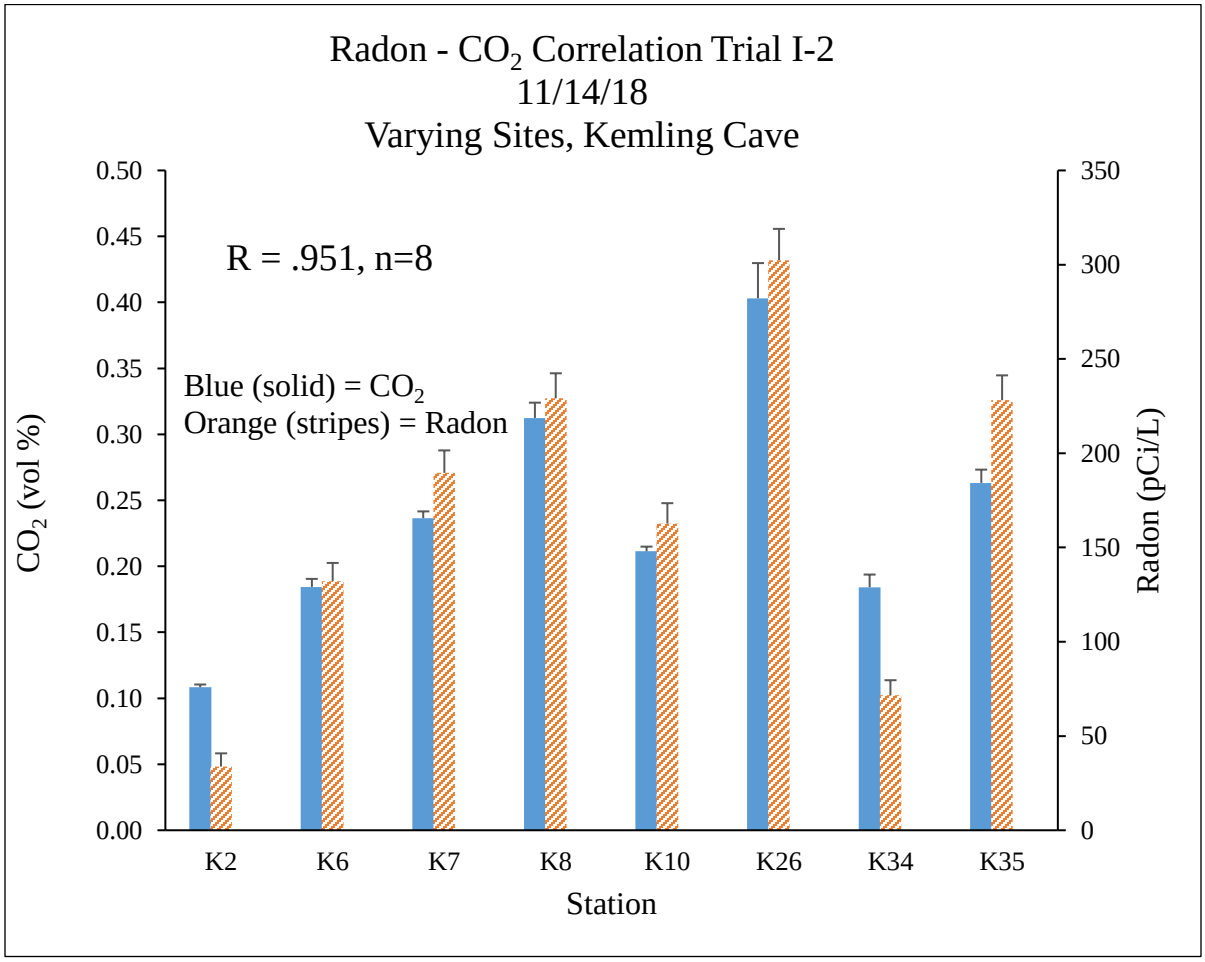

Figure (4): Trial I-2 correlation of radon and CO_2, 11/14/18, Kemling Cave.

Part II: Radon vs. CO_2 as a function of time at a fixed location

Following the significant correlations seen in Part I, temporal comparisons at fixed locations in Kemling Cave were initiated. Prior work (Welch 2018) had shown great variations of radon typically were observed within the time frames of one to seven days, so the same would be expected of CO_2 if it were highly correlated. Trial II-1 was set up at location K5B in Kemling Cave (see Figure (2) for locations, the B suffix denoting that the sensors were elevated off the floor by the height of a 5-gallon bucket, 38 cm), with both gases measured at 10-minute intervals

for a duration of 24 hours. The overlay of the resulting data can be seen in Figure (5), producing an R value of 0.731. Although noteworthy, this correlation was smaller than that observed for

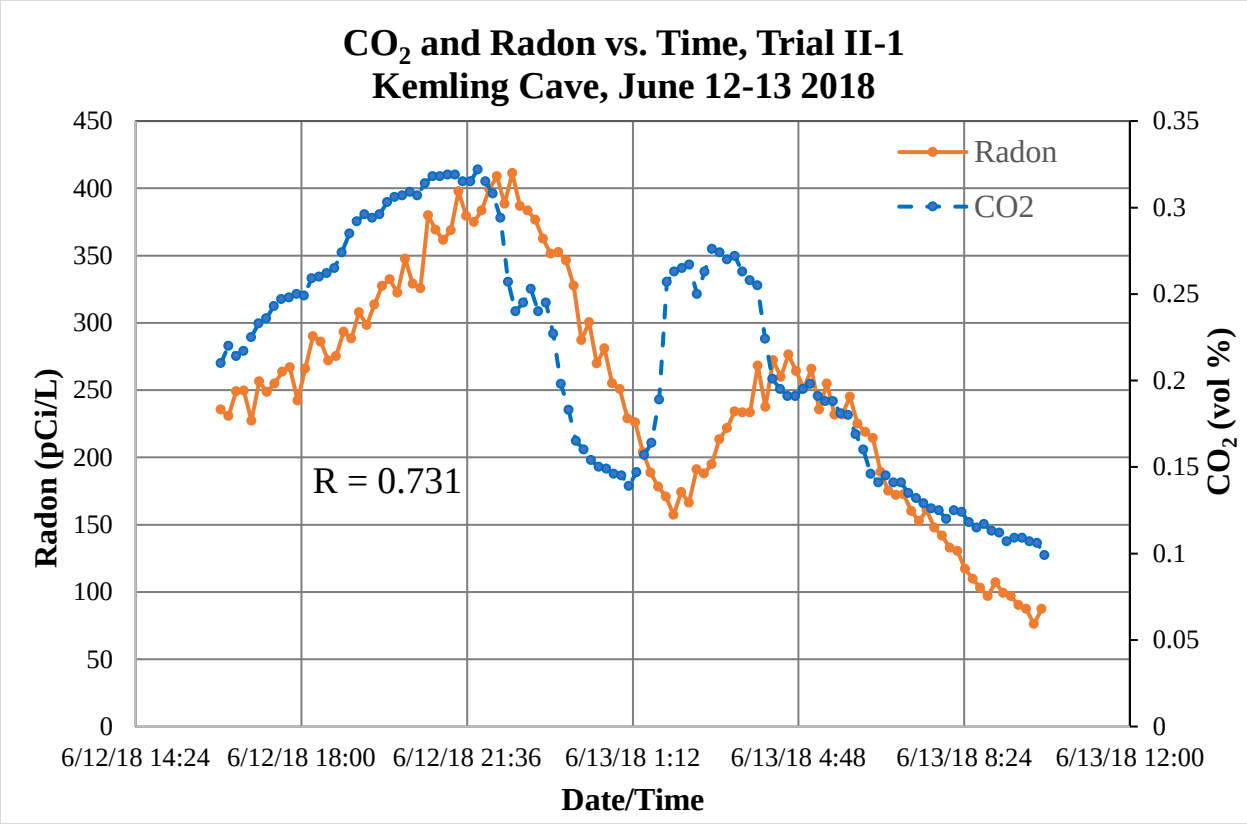

Figure (5): Overlay of radon and CO_2 levels, Trial II-1, June 12-13 2018, station K5B, Kemling Cave.

the Part I trials. However, an examination of Figure (5) suggested that the changes in CO_2 concentration preceded those same changes in radon, and that application of a time offset would likely improve the calculated correlation. The CO_2 measurement was shifted by steps of ten minutes to later times, and then the R value remeasured after each offset. Figure (6) shows the R value function with offset time, which yielded a plot with a clear maximum time. The best correlation of 0.947 was observed when the CO_2 measurement was offset 90 minutes later in time.

The strong correlation was not surprising given the results of Part I, but the offset time was intriguing. The obvious candidate to explain the observed offset was the difference in the response latency of the two sensors. The Radon Recon sensor was passive in design, and required diffusion of the gas samples into the unit. The vendor of the Recon cited an expectation of a 2-hour sensor latency (Stieff, 2018). The CM-0010 had a sampling pump that actively pulled gas samples into the unit, and therefore would be expected to be nimbler in response to gas activity changes than the Recon. Prior CO_2 data collected with the CM-0010 sequentially at different stations in Kemling Cave, with data from one station shown in Figure (7), also suggests a rapid response, but it should be noted that for the Figure (7) trial the CM-0010 pump ran

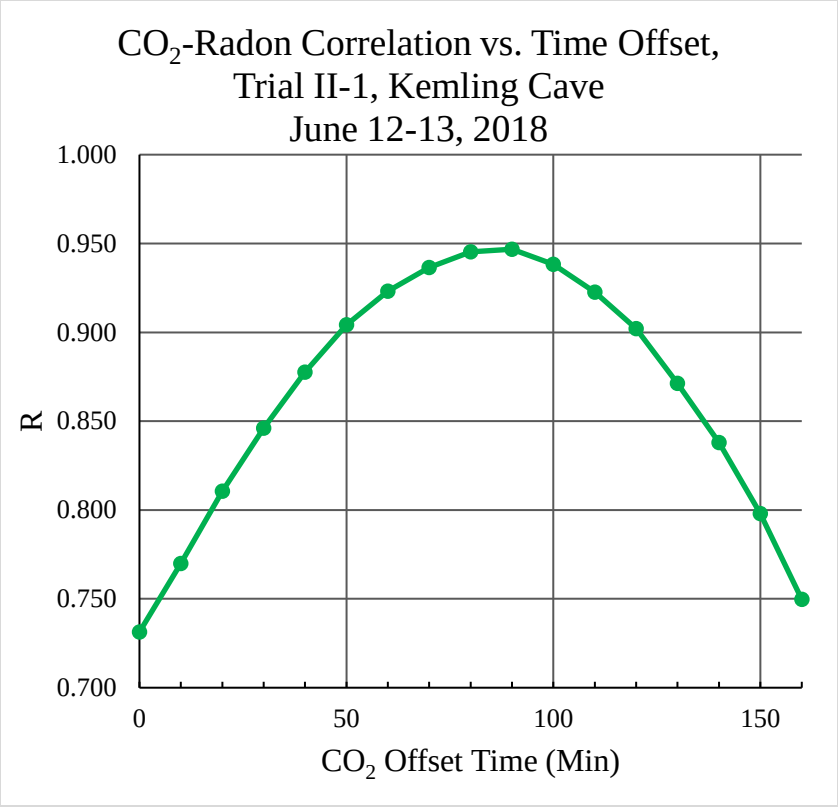

Figure (6): CO_2 – radon R value as a function of time offset (positive offset means CO_2 data offset to later times), Trial II-1, June 12-13 2018, station K5B, Kemling Cave.

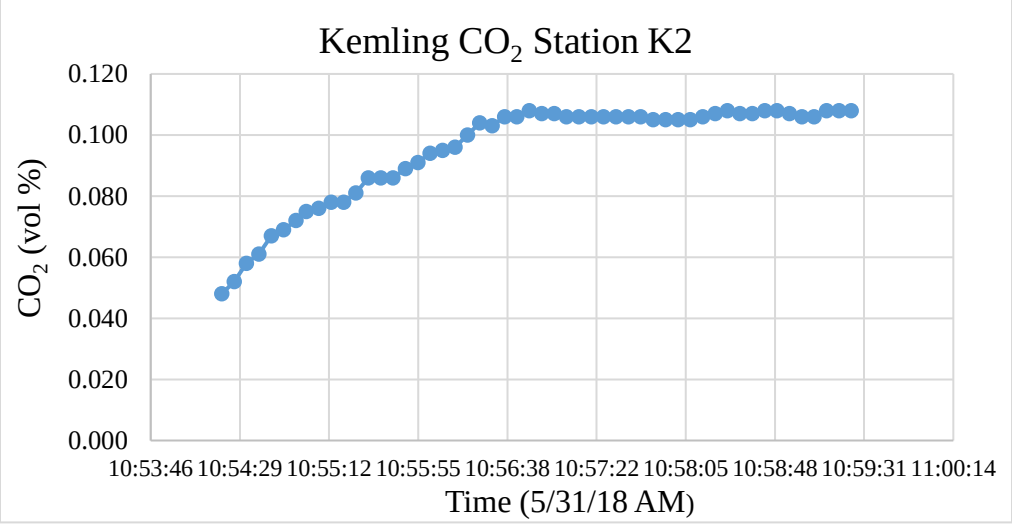

Figure (7): CO_2 level as a function of time at station K2.

constantly, whereas for the Part II trials the sampling was only done once every 10 minutes, and the pump only ran for five seconds per sample and then was deactivated in between to save on

battery power. It should also be noted that the CM-0010 inlet required the gas to move through two filters and a length of Tygon tubing prior to reaching the measuring cell, which would also tend to increase the sensor latency. Given the vicissitudes of instrument setup and deployment, it was felt that the best way to evaluate the sensor latency was to measure it experimentally. Coldwater Cave in Winneshiek County, IA offers a spot just inside the cave entrance that consistently has very high radon and CO_2. Just outside the cave, these gases are at background level. The sensors were started outside the cave and given 4 hours of exposure time to reach equilibrium, then rapidly (circa 1 minute) transitioned into the extremely high radon/CO_2 environment while still collecting, using the 10-minute sampling increment used in the Part II trials. Once the sensors had been given another 4 hours of exposure time inside the cave, the process was reversed. After multiple repeats of this process, the sensor latencies were found to be 40 minutes for the CM-0010, and 120 minutes for the Recon, which was more difficult to gauge given the slower response. So the difference in sensor latency was on the order of 80 minutes, meaning CO_2 response to changing conditions would precede radon by this amount, all other things being equal.

The outcome of Trial II-1, with its 90-minute offset, was similar in magnitude to the measured difference in sensor latency. Were other factors important in the observed offset such as the difference in diffusion rate of the two gases, environmental factors, and passage geometry? A series of follow-up trials were undertaken to gauge the reproducibility of the outcome from Trial II-1 by running duplicate trials at the same location and at other sample sites within Kemling Cave. All of these subsequent trials were designed to run for circa 48 hours, yielding a larger dataset with more features to correlate.

Figure (8) shows the R value plotted vs CO_2 offset time for Trial II-1 and a series of subsequent trials, with tabulated information given in Figure (9). Trial II-2 was collected at the same location as Trial II-1, yielding an optimum R of 0.9926 at a 100-minute offset of CO_2. Given that the sensors were not perfectly synchronized and up to 3-4 minutes of difference could result from this, it was hard to conclude that 100 minutes of offset was significantly different than the maximum of 90 minutes from II-1. The Trial II-2 plot did show a maximum and a very high one at that, but it also had much less curvature and "normal curve" appearance. It should be noted on Figure (9) that Trial II-2 coincided with a very low radon period of time, and subsequent trials illustrated that the outcome of Trial II-2 was typical of low-radon periods: less curvature, very high R values, large relative uncertainties for radon measurements. Trial II-3 was collected deeper in the cave at Station K33, yielding an optimum offset of 70 minutes. Trials II-4 and II-5 were both collected closer to the entrance at Station K2, and both yielded very short offsets of 40 minutes apiece. At this point it was clear that different locations in the cave were leading to different offsets, but whether the offset was constant at a given location was not clear. Trial II-6 was a replicate of II-3 taken at Station K33, and the optimum offset of 90 minutes looked to be different enough from the 70 minute optimum from Trial II-3 to underline the fact that more than just the difference in sensor latency, which would presumably remain constant, was factored into the observed optimum correlation offset. As such, environmental factors look to be impacting these experiments. A further evaluation of the Part II datasets did not reveal a simple explanation for the observed variation of the optimum correlation offset, so an additional series of experiments were planned to probe this in detail, adding further environmental sensors to the package being deployed at each site. The outcome of these experiments and a more definitive

explanation of the variation in optimum correlation offset will be described in a future manuscript.

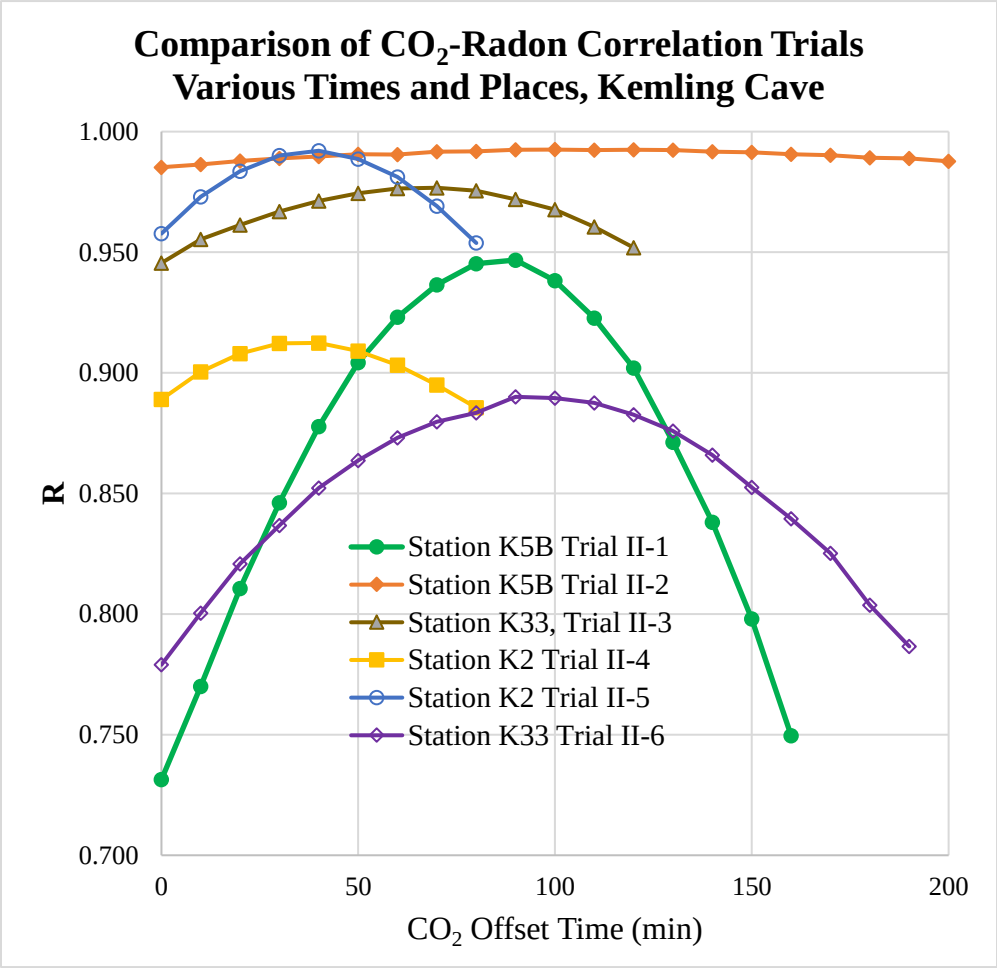

Figure (8): CO_2 – radon R values as a function of time offset (positive offset means CO_2 data offset to later times), Trials II-1 through II-6, Kemling Cave.

Conclusions

The concentrations of radon and CO_2 in the cave air of Kemling Cave are correlated to a high degree, both when tested at various sites in the cave during the same experiment, and when tested as a function of time at a single site. The time correlation can be optimized by inserting an offset time to one of the sensors. The difference in sensor latency has to factor into this offset time, but since the optimum offset time varies greatly from station to station in the cave and to at least some degree trial to trial, other environmental factors were also playing a role here. The strong correlation underlines the importance of cave ventilation on the respective concentrations of these gases. If the cave is poorly ventilated, both gases will accumulate in the cave air. Higher rates of ventilation created by either temperature differentials with the surface or via

barometric pumping will act on both gases as well, reducing their levels. The levels of both gases tend to mirror one another in terms of increasing or decreasing during the trials, but due to the differing sources for each gas, the degree of these changes were not identical.

Compendium						
Radon vs CO_2	Exp 100	Exp 102	Exp 103	Exp 104	Exp 106	Exp 107
Correlation R	Jun 12-13	Aug 21-23	Aug 26-28	Sept 7-9	Sep 14-16	Sep 16-18
Time	Sta K5B	Sta K5B	Sta K33	Sta K2	Sta K2	Sta K33
	Trial II-1	Trial II-2	Trial II-3	Trial II-4	Trial II-5	Trial II-6
real time	0.7314	0.9852	0.9455	0.8929	0.9572	0.7790
CO2 add 10 min	0.7700	0.9863	0.9553	0.9045	0.9728	0.8004
CO2 add 20 min	0.8107	0.9879	0.9613	0.9124	0.9835	0.8208
CO2 add 30 min	0.8461	0.9889	0.9669	0.9166	0.9900	0.8367
CO2 add 40 min	0.8777	0.9897	0.9712	**0.9169**	**0.9919**	0.8523
CO2 add 50 min	0.9042	0.9906	0.9744	0.9137	0.9884	0.8637
CO2 add 60 min	0.9231	0.9905	0.9765	0.9080	0.9809	0.8731
CO2 add 70 min	0.9366	0.9918	**0.9767**	0.9005	0.9686	0.8797
CO2 add 80 min	0.9453	0.9918	0.9756	0.8915	0.9530	0.8835
CO2 add 90 min	**0.9468**	0.9925	0.9719			**0.8901**
CO2 add 100 min	0.9383	**0.9926**	0.9677			0.8896
CO2 add 110 min	0.9227	0.9924	0.9605			0.8876
CO2 add 120 min	0.9020	0.9925	0.9519			0.8827
CO2 add 130 min	0.8713	0.9923				0.8759
CO2 add 140 min	0.8380	0.9916				0.8659
CO2 add 150 min	0.7980	0.9915				0.8525
CO2 add 160 min	0.7496	0.9906				0.8395
CO2 add 170 min		0.9902				0.8252
CO2 add 180 min		0.9892				0.8038
CO2 add 190 min		0.9889				0.7866
CO2 add 200 min		0.9877				
Avg Radon pCi/L	249.68	40.19	456.49	411.70	145.82	625.07
SD Radon pCi/L	88.56	41.11	129.79	151.32	101.77	89.02

Figure (9): CO_2 – radon R values as a function of time offset (positive offset means CO_2 data offset to later times), Trials II-1 through II-6, Kemling Cave. R value maxima in bold.

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