

The relationship between local climate and radon concentrations in the Temple of Baal, Jenolan Caves, Australia

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Abstract

Radon measurements were collected over a period of one year in a large chamber known as the Temple of Baal at Jenolan Caves, near Sydney, Australia. Correlation of radon concentrations with rainfall, surface air pressure and temperature confirmed that radon originating from different locations was predominant under different conditions. During periods of low rainfall, radon concentrations varied in strong anti-correlation with the surface air pressure, indicating that most of the radon was coming from remote locations of large pore or void volume in rock of limited permeability. On the other hand, in wet periods the observed radon levels were low and steady, suggesting a local source. In both wet and dry conditions the correlation of radon concentrations with rainfall on a time-scale of a few days was positive, proving that permeability of surface strata affected the ventilation rate in the cave. The study achieved a detailed understanding of radon concentrations in the Temple of Baal, and the main conclusion reached was that the magnitude and variation of radon concentrations in the Temple of Baal were closely related to the degree of water saturation in the local surrounds.

Keywords: karst, radon, cave climate.

Introduction

Climate influences the amount of any gas emanating from the earth, through both air pressure and temperature effects and by determining the movement and quantity of moisture in the sediment and underlying strata. More particularly, water in these strata is believed to be the major factor governing gas movement through the earth (Tanner 1980, Nielson *et al.* 1984, Schery *et al.* 1989). Consequently, if a source of a gas lies within the strata, climate can affect the amount of the gas escaping into the air. Potentially such a gas could be used as a tracer to reveal information about the affect of climate on the degree of water saturation in deep strata. Radon is a suitable gas for such studies.

Radon is an inert radioactive gas emitted by rocks and sediments. It is produced by decay of radium (Figure 1), which is present in trace quantities throughout the Earth's crust. Being a gas, radon can move from its source into the cave¹. Once a given amount of radon is in the cave, its concentration depends on the cave volume and ventilation. An understanding of radon concentrations therefore requires knowledge of the location of the radon sources and the associated transport mechanisms.

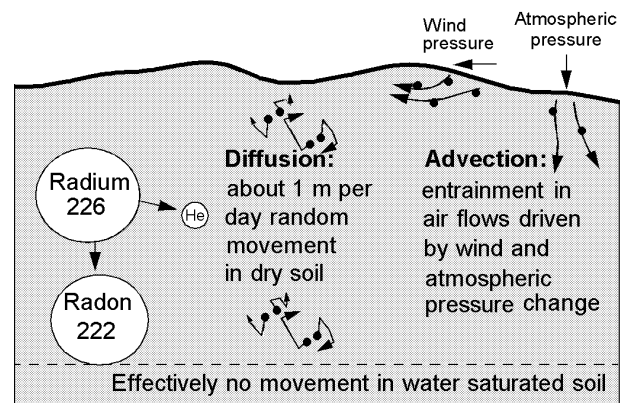


Figure 1. Radon movement in soil

Figure 1 illustrates the main mechanisms by which radon moves through sediment. After radon is produced there are four possibilities:

- It may decay close to where it is produced.
- It may diffuse through air filled pores in the sediment by virtue of the inherent kinetic energy of the radon atoms. Radon can migrate up to a metre a day in dry sediments through such movement, but much less when water fills the pores. Because of its short half-life of 3.8 days, only radon produced within a few metres of the surface of dry sediment can escape in this manner.
- It may be transported by advection, the entrainment of radon in air-flows driven by wind or atmospheric pressure change. Think of this as a form of suction; if

¹ For the purposes of this paper, "cave" includes both underground chambers and passages. The term "cave wall" implies all cave surfaces - floor, ceiling and walls.

Radon

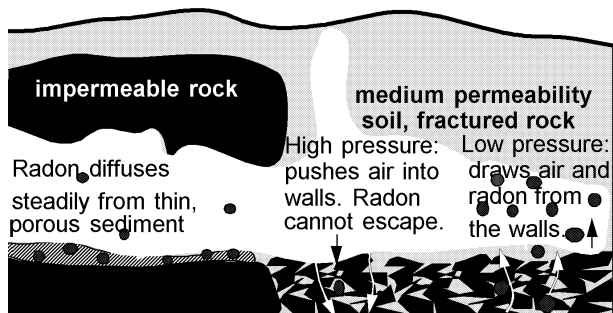


Figure 2. Radon entry into cave air

conditions are such that air is being pulled from the void spaces, radon can be carried along with it.

- If the sediment is water-saturated, there is effectively no radon movement. If there is rapid water flow through the sediments, there is the potential for radon to be transported from its site of generation since radon is nearly as soluble as carbon dioxide. Once removed from the zone of saturation, radon can degas from the water.

The main mechanisms of radon entry into cave air are diffusion (Figure 2, item 1) and advection (Figure 2, items 2 and 3):

1. Radon diffuses into the cave air at a steady rate from thin porous sediment over impermeable rock. In this case, air pressure has virtually no effect on radon concentration.
2. If the cave wall is made of sediment of medium permeability and fractured rock, the process of entry is via advection. In this case, an increase of air pressure can push air into the surfaces of the cave. Even a very small flow of air into the cave surfaces can stop the radon from diffusing out, producing a very low concentration of radon in the cave air.
3. Conversely, a reduced air pressure will pull air from the cave surfaces, carrying radon with it and cause high radon concentrations.

In caves the geologic strata may be complex, but the main effects of pressure can be understood in terms of the simple model shown in Figure 3. The main elements are the cave and the pores and larger voids in the strata containing finely divided rock, such as sediment, which emits radon. If these voids are connected to the cave by thin cracks, it is possible for changes in pressure to affect radon concentrations. Picture these pores and voids as a single large void and the connections as a pipe between the void and the cave.

A change in air pressure forces air through the pipe until the pressure within both the cave and void are again equal. If there is an increase in cave air pressure as the result of an external change, air is pushed through the pipe into the void. While the air pressure in the cave remains higher than that of the void, no air carrying radon from the void can enter the cave. On the other hand, if the cave air pressure is lower than that of the

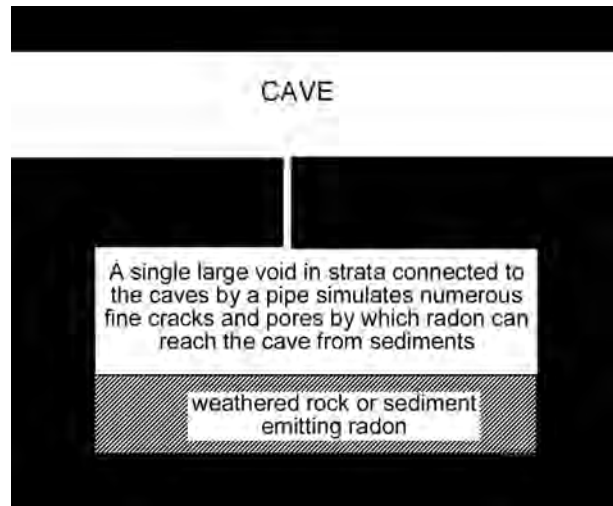


Figure 3. A simple model of radon entry into a cave

void, air laden with radon will be pulled through the pipe into the cave. If the pressures are equal, no advection-driven movement of radon occurs and the only change in radon concentration will be based upon diffusion. The speed at which the pressure differential between the cave and void equilibrates depends upon the size of the pipe connecting the void and cave. If the pipe is narrow, it will take a long time (days or even weeks). Alternatively, if the pipe is large, the pressure quickly equilibrates. In either case, radon can only enter the cave by advection when the pressure is lower than that of the void.

In both cases, the actual volume of air moving through the pipe is very small. Because of this, a high or increasing pressure will cut off the source of radon to the cave, but such a small amount of air moves from the cave that all but a few percent of the radon already in the cave stays there. Provided the total void space is not much greater than the volume of the cave, there will not be enough air movement into the void spaces to take up significant amounts of the radon. The radon in the cave either decays *in situ* or is blown out by ventilation processes. If pressure is low, radon starts to enter the cave. Either way, it will take time for the effect of a pressure change to show up in the radon concentration.

In the absence of ventilation, the concentration of radon will reach an equilibrium such that the rate of radon lost by radioactive decay matches the rate at which radon enters from all locations. In a cave with strong ventilation, radon can be diluted with outside air, resulting in a lower concentration. This is an important factor when the air exchange time is less than the radon half-life of 3.8 days.

This paper reports the results of a correlation analysis of radon concentrations with the external climatic variables of surface temperature, air pressure and rainfall, established to explain the behaviour of radon observed in the Temple of Baal, Jenolan Caves.

Site Description

In 1996, a comprehensive air quality monitoring system was established at Jenolan Caves to measure the seasonal variation of a number of parameters including radon gas (Zahorowski *et al.* 1998). The Temple of Baal is a large chamber with active speleothems which lies almost a kilometre away from any natural entrance and was originally selected as a site for radon monitoring because of its low ventilation rate and steady temperature. It appeared to be a relatively simple system, as opposed to another part of Jenolan Caves, Katies Bower, which can experience strongly varying radon concentrations on time scales as short as an hour. The 1996 study showed that, although the Temple of Baal was free from these rapid variations, there were changes on monthly time scales for which there was no immediately obvious explanation.

Instrumentation

The monitoring instrument consisted of a number of sensors interfaced to a computer. For measuring radon levels, a 1 L zinc sulfide coated scintillation cell was employed. The radon detector was calibrated by passing radon from a radon source, model RN1025 (Pylon Electronic Corp, Canada), through the cell. The flow rate for this calibration was determined using a bubble tube system by Gillian Instrument Corporation, USA. The total radon concentration was the sum of that from the source plus ambient radon. The latter was estimated by extrapolating from the time before the radon was injected, using radon progeny measurements to keep track of changes in radon concentration. Rainfall data and surface air pressure and temperature were obtained from Ernst Holland using the Australian Defence Force Academy meteorological station situated in the McKeowns Valley at Jenolan Caves.

Results and Discussion

From previous studies (Barnes *et al.* 2001) of the variation of radon concentrations, at 19 out of 20 sites spread throughout the Jenolan Caves a pattern of high radon levels in summer and low radon levels in winter has been established. In some cases the differential between the two seasons reaches a factor of 20. At all sites, with the exception of the Temple of Baal, the spring and autumn values are intermediate. Moreover, for these 19 sites, the variation in radon levels is largely determined by variations in temperature and air pressure.

Weekly averages of the radon concentrations in the Temple of Baal are shown in Figure 4. In order to show the variability of the weekly average concentrations, the distance between the lines represents the range of the values each week. These data fall naturally into three periods: summer, when radon levels were relatively low

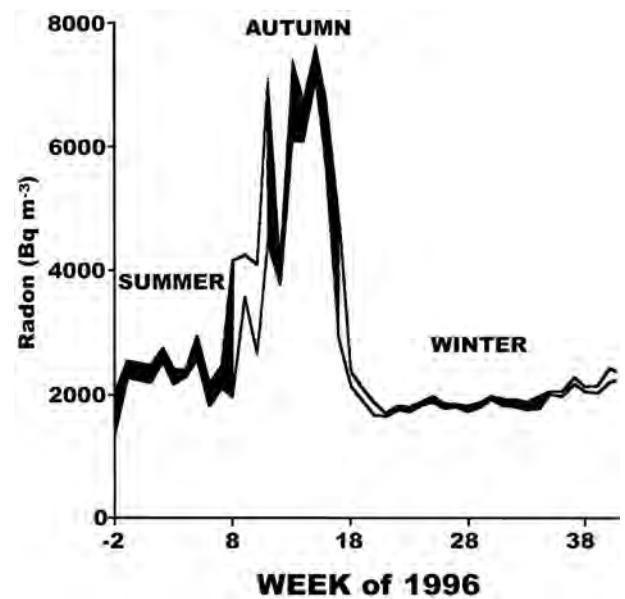


Figure 4. Weekly radon concentrations in the Temple of Baal

and moderately variable; autumn, where the radon levels were high and variable; and winter, when the radon level was lowest and remarkably steady. The shaded zones in Figure 4 are the periods used in this analysis. The data lying between the shaded zones have characteristics intermediate between those of their neighbouring periods and have been excluded. Table 1 summarises the data used in, and the results of, the correlation analyses of radon concentrations and the three major climate variables.

The results in Table 1 for the Temple of Baal show that the summer/winter pattern is only marginally discernible in this chamber (with a differential of just over 1), while there are anomalously high radon levels in autumn. The correlation analyses of radon concentration against climate variables were undertaken in an attempt to obtain an explanation for these results. Furthermore, from the raw data illustrated in Figure 4 and contained in Table 1, the following inferences can be made concerning the ventilation rates and sources of radon in the Temple of Baal during the period of this study.

The steady radon concentration in winter implies a low ventilation rate. In caves with strong ventilation, such as Katies Bower (Zahorowski *et al.* 1998, Barnes *et al.* 2001), the radon concentration can vary by more than an order of magnitude in a day. If there were a complete air exchange within the cave in 3.8 days (the half-life of radon), the loss by dilution would match the loss by radon decay and the radon concentration would drop by a factor of 2. In the Temple of Baal, Table 1 shows that the greatest change in radon concentration over the winter period was 70 Bq m⁻³ from the maximum of 1900 Bq m⁻³, or 1 part in 13. Thus, even if all the variability is attributed to ventilation, there cannot have been more than 1/13th part of the air in the Temple of Baal diluted by surface air. The minimum time for complete

Radon

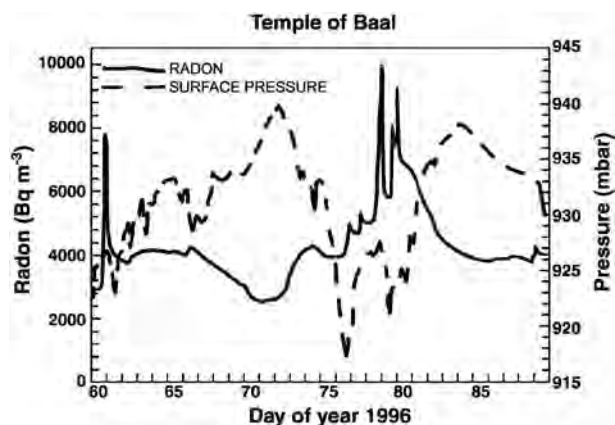


Figure 5. Change in radon levels in the Temple of Baal and in surface air pressure in autumn

air exchange with surface air would be $13 \times 3.8 = 49$ days.

Given the steadiness of the radon concentration within the Temple of Baal during winter, a local source of radon must be postulated. The argument used in the preceding paragraph can be expanded to air exchange with connected caves. However, if radon was being transported from other caves, it would cause a greater variability in the radon concentration in the Temple of Baal than is observed.

From Table 1, it can be seen that the summer and winter rainfall and pressure ranges are similar. Taking into account the steady local source observed in winter, the total radon concentration in summer must be produced by this steady source plus an additional,

temperature dependent source. That is, the higher temperatures in summer must allow radon from an additional source to reach the Temple of Baal and that this source varies to cause an additional radon concentration between a minimum of 170 Bq m^{-3} ($= 2000 - 1830$) and a maximum of 600 Bq m^{-3} ($= 2500 - 1900$). Such a highly variable source is typical of well-ventilated chambers like Katies Bower. It is therefore reasonable to suggest that there is a small interchange of air between the Temple of Baal and the rest of the Jenolan Southside Caves. Indeed, an intrusion of just 1% of the volume of the Temple of Baal from the adjoining system would result in the observed increase. This cave air exchange could have been present in winter, but because of the low winter radon concentrations in the adjoining cave system (Zahorowski *et al.* 1998) there would not be a significant effect on the radon concentrations in the Temple of Baal.

Table 1 further shows how radon concentration in the Temple of Baal correlates with temperature, pressure and rainfall. The figure in brackets is the time in days it took for the influence of the change to take effect. The period of 6 days for the rainfall period was determined empirically. Shorter periods provided insufficient events to obtain a representative sample for each season, while longer periods resulted in weaker correlations. There are not enough data to fully understand why this is an optimum period, but it is likely that light rain in a single day is only important if there was rain within the few preceding days. On the other hand, if longer periods were taken, the correlation would be lost because the effect, with its lag time of 2.5 days, is short term.

Table 1. Radon concentrations, climate data and associated correlation analyses

	Summer	Autumn	Winter
Radon (Bq m^{-3})	2000 – 2500 Variable	4800 – 7000 Extremely Variable	1830 – 1900 Steady
Range* of surface temperature ($^{\circ}\text{C}$) (average of day and night)	12.5 – 18.5	11.7 – 15.7	6.2 – 8.8
Correlation of radon with surface temperature (lag in days [§] for best correlation)	0.41 (2.3)	< 0.1	0.21 (5)
Range of surface pressure (mbar)	922 – 930	927 – 934	924 – 934
Correlation of radon with surface pressure (lag in days for best correlation)	-0.24 (1.6)	-0.6 (3)	-0.6 (1.5)
Average rainfall (mm d^{-1})	2.8	0.5	2.2
Correlation of radon with average rainfall over a 6 day period [#] prior to radon measurement (lag in days from end of 6 day period for best correlation)	< 0.1	0.5 (2.5)	0.75 (0)

* where ranges are quoted they are for weekly averages

§ time between the end of climate variable measurement and the best correlation with trend in radon level

a 6 day period was chosen to provide enough rainfall data to carry out the correlation analysis

Surface temperature correlates most strongly with radon in summer. In fact, in summer the radon level in the Temple of Baal correlates with the same variables as radon concentrations in chambers like Katies Bower (Zahorowski *et al.* 1998, Barnes *et al.* 2001) where the diurnal radon concentrations are driven by surface temperature. There are two important differences. First, the response time in Katies Bower is about 3 hours, while in the Temple of Baal the response time is over two days. Second, the variation in radon levels in Katies Bower can be a factor of ten in a day but in the Temple of Baal it is only at most 30%. In other words, although there is a temperature effect seen in the Temple of Baal it is not as significant as that seen in other areas like Katies Bower.

The correlations of radon concentration with atmospheric pressure shown in Table 1 are always negative, albeit insignificant in summer. In autumn, the anti-correlation is quite strong and the radon levels indicate a sensitivity to pressure changes. Indeed, Figure 5 shows that during this season the Temple of Baal exhibits the characteristics of a cave connected by moderately permeable strata to a large volume of voids with radon-generating sediment. Although there is also a strong negative correlation of radon concentration with pressure in winter, the changes are relatively small. Evidently, access to the variable source was shut off, allowing a smaller and more steady local source to predominate.

It is the effect of rainfall on a seasonal time-scale which is responsible for the generally lower radon concentrations observed in summer and winter as compared to autumn. On a shorter time scale, rainfall in the previous six days had no effect in summer, but in autumn and winter there was a positive correlation with rainfall. In other words, an increase in radon concentration is observed if there is rain in the preceding few days, the opposite of the seasonal trend. The difference in these rainfall effects lies in their differing impacts. Rainfall correlated against radon levels over a season indicates the impact of rainfall on the source of radon. It is the sediment and voids that contain the major radon sources and moisture in the sediment and joints in the epikarst will prevent diffusion of radon into the cave air. On the other hand, sediment on the surface above the cave, while not a major radon source, influences how much air can enter or exit the cave through cracks to the surface. If there is less ventilation, such as when the sediment becomes wet, the radon concentration increases because the radon-laden air becomes trapped in the cave. Soil moisture levels are affected by particular showers of rain only for a few days. Thus, radon

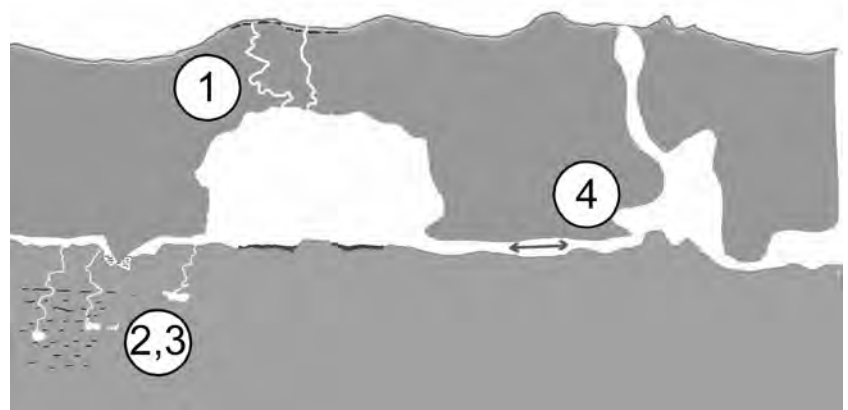


Figure 6. Model of effect of climate variables on radon in the Temple of Baal

1. Rainfall increases radon by reducing ventilation via small cracks to the surface (minor effect)
2. Rainfall reduces radon by blocking radon migration through strata (major effect)
3. Atmospheric pressure affects radon when voids in strata are connected to the cave by fine cracks which restrict air flow (major effect in dry seasons)
4. Temperature causes high, variable radon in chambers near the Temple of Baal in summer. Small air exchanges lead to a small, variable, temperature dependent source in the Temple of Baal

concentrations are positively correlated with recent rain but negatively correlated with rain falling many weeks earlier.

The correlations support the argument, based on the radon variabilities quoted in Table 1, that there is a small interchange of air between the Temple of Baal and the adjoining cave system. In winter the system has a very low radon concentration, so the contribution is negligible and the small effect of pressure and recent rainfall can be revealed by correlation analysis. In summer, the connection to the Jenolan Southside Caves adds a highly variable amount of radon depending on the average ambient temperature. This variable component swamps the small variable component of radon in the Temple of Baal, which itself is pressure and rain dependent. As a result there is no longer a significant correlation with pressure or recent rain.

The autumn pressure range lies within the ranges exhibited in summer and winter so is unlikely to be the cause of the variation in radon levels observed. Average surface temperature is higher in summer and lower in winter, so this also cannot be the cause of the high radon in autumn. However, average rainfall in autumn was one quarter of that for the other periods. Evidently low rainfall on a seasonal time scale allowed the sediment and strata to dry out, opening pores and cracks enough to increase radon movement. Thus, radon was able to reach the Temple of Baal from sources that were closed off in the other periods.

Figure 6 shows a model of the Temple of Baal, which explains the major features of a whole year's observations and illustrates the proposed relationship

Radon

between climate variables and radon concentration. The most important parameter is rainfall. Over periods of weeks to months, rainfall caused substantial decreases in radon concentrations by blocking the migration of radon through strata into the cave. Large passages can be blocked in a sump, while smaller cracks are closed when the water table rises, or when sediment swells as its moisture content increases. Radon concentrations in wet seasons are therefore steady and not variable.

Conclusions

Concentrations of radon in the Temple of Baal are correlated with the climate variables, rainfall, surface air pressure and surface temperature, on time scales from hours to months. In the short term, a couple of days, rain causes a slight increase in radon concentrations because it reduces the permeability of the surface sediment and slightly reduces the cave ventilation rate (Figure 6.1). This effect was apparent in both dry and wet seasons. In the longer (seasonal) term, high rainfall saturates the epikarst preventing radon emission into the cave atmosphere (Figure 6.2).

Atmospheric pressure can have a large effect on radon concentrations, but only when the strata are dry to the point that radon can diffuse large distances (Figure 6.3). Some of the voids in the strata are then connected to the cave by cracks so fine that it takes days for the void pressure to equilibrate with the pressure at the surface. A high radon concentration and strong negative correlation with pressure is proof of relatively dry strata surrounding the cave.

The least important factor influencing radon concentrations in the Temple of Baal is temperature. Although neighbouring chambers are strongly affected by temperature, there are only small air exchanges with the Temple of Baal (Figure 6.4). This results in a small temperature dependent variation of radon in the Temple of Baal in summer.

Acknowledgments

We are grateful to the Jenolan Caves Reserve Trust for permission to carry out the work, and to the many guides who assisted with the project. Particular thanks are due to Ernst Holland and Karen Jones of the Trust for their weekly checks of the equipment and provision of the surface temperatures and rainfall data. The instrument recording the local meteorological data was supplied by Professor David Gillieson of the Department of Geography, James Cook University, Queensland. Wlodek Zahorowski, Bryan Stenhouse and Michael Hyde of ANSTO contributed to the construction and calibration of the instruments and at times provided a partial assessment of the data obtained.

References.

- Barnes, C.M., James, J.M. and Whittlestone, S., 2001: Radon Studies in Jenolan Tourist Caves, NSW, Australia. *Proceedings of the 13th International Union of Speleology Congress, Brazil*. Sociedade Brasileira de Espeleologica, Campiras, SP Brazil, Vol 1, pp.51-56.
- Nazaroff, W.W., 1992: Radon transport from soil to air, *Reviews of Geophysics*, **30**, 137-160.
- Nielson, K.K., Rogers, V.C., and Gee, G.W., 1984: Diffusion of radon through soils: a pore distribution model, *Soil Soc. Am. J.*, **48**, 482-487.
- Schery, S.D., Whittlestone, S., Hart, K.P. and Hill, S.E., 1989: The flux of radon and thoron from Australian soils, *J. Geophys. Res.* **94**, no. D6, June 20, 1989.
- Tanner, A.B., 1980: Radon migration in the ground: A supplementary review, *The Natural Radiation Environment*, vol. 3, pp. 5-56. National Technical Information Service, Springfield, Va.
- Zahorowski, W., Whittlestone, S., and James, J.M., 1998: Continuous measurements of radon and radon progeny as a basis for management of radon as a hazard in a tourist cave, *J. Radioanalytical and Nuclear Chemistry*, **236(1-2)**, 219-225.

Appendix: Statistical methods.

The statistical methods used in this discussion involve deductions from simple correlations, lagged correlations and the evaluation of concentration variabilities resulting from factors which are known to affect radon concentrations in other situations.

Correlation analysis is used to establish that two parameters tend to increase and decrease at the same time. This does not establish cause and effect, but is a powerful constraint on hypotheses.

Lagged correlations are used to test whether a change in one parameter tends to follow a change in another. This is valuable in ruling out certain cause-effect relationships since a cause cannot follow an effect.

Variability of data over a time period is gauged from the range of values, maximum and minimum. This is important in assessing the consistency of hypotheses. When different factors affect radon concentration, there can be times when one source is dominant and produces so much variability that a second factor is not apparent. In this case a correlation analysis will not show the second factor, even though it is still operating.

