# Climatic and Seasonal Influences on Radon Time Series in an Environment of Low Anthropogenic Activity

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#### Abstract

Between June 2003 and March 2008 (1766 days), radon concentration and ambient temperature were monitored at hourly intervals in an environmentally-stable, rarely-visited partially-subterranean store-room in a public-service building. Sampling periods totalled 1025 days, the longest continuous sampling period and interval between sampling periods being 335 and 271 days respectively.

Mean daily, monthly (calendar) and annual concentrations and temperatures were derived. For each calendar month, averages over corresponding months of at least three separate years were available. Similar means were generated from meteorological data collected 10 km from the measurement site. The mean and standard deviation over the whole radon data-set are 73.0 Bq.m<sup>-3</sup> and 62.7 Bq.m<sup>-3</sup> respectively, corresponding mean and SD internal temperatures being 26.2 C and 1.7 C, respectively.

Radon concentration shows minimal or zero correlation with the three temperature parameters, atmospheric pressure and wind-speed, limited (negative) correlation with rainfall and possible (negative) correlation with relative humidity.

Although statistically-derived Seasonal Correction Factors (SCFs) are conventionally used to convert radon concentration over a sub-year period to a notional annual mean, it is recognised in the literature that this approach is not without its problems, particularly where short-term measurements are concerned. A recent proposal uses external temperature to derive a local SCF, assuming positive temperature-difference between a building interior and its environment. To test this, surrogate SCF sets were derived using monthly mean radon concentration, external temperature and internal-external temperature-difference, and correlated with each other, and with the United Kingdom (UK) 1-month SCF set to quantify goodness-of-fit. Relatively good correlation (|r| = 0.59) exists between radon seasonality and the internal-external temperature difference, but poorer correlation (|r| = 0.18) with absolute external temperature-based formula.

# Key Words

Radon, climate, seasonal variation, time-series

### 1 Introduction

### 1.1 Environmental Radon Gas

Radon (<sup>222</sup>Rn), a naturally occurring  $\alpha$ -emitting noble gas, has variable distribution in the geological environment as a decay product of the uranium occurring in a wide range of rocks and soils and in building materials incorporating or manufactured from these. Radon gas, generated by  $\alpha$ -decay within mineral grains and at their surfaces, reaches air and water-filled interstices by recoil and diffusion, its migration being directed towards lower concentration regions, principally the earth's surface, following the local gradient.

Ionising radiation is well known to have adverse health effects, and inhalation of radon and its radioactive solid progeny <sup>218</sup>Po and <sup>214</sup>Po, both readily adsorbed onto atmospheric particulates, is currently considered [1] to provide the majority of the dose to the respiratory system, leading to chemical and radiological damage to the sensitive inner lining of the lung and increasing the risk of cancer. Total annual mortality in 2006 from this type of cancer in the United Kingdom (UK) was 34150 [2], of which around 1100 (3.3%) are attributed to residential exposure to radon and its progeny [3,4].

### 1.2 Radon Concentration Variability

Domestic concentrations of radon gas, <sup>222</sup>Rn, are subject to numerous influences, being generally higher at night than during the day, and generally higher in winter than in summer. These variations reflect reduced activity and restricted ventilation in the home at night, increased interior/exterior temperature difference at night and during the heating season and, where soil-gas advection provides the predominant radon source, the underlying climatically-modified seasonal variability in soil-gas radon content. Increased

interior/exterior temperature difference results in greater atmospheric pressure differential and enhanced radon ingress *via* the stack effect [5].

In addition to the natural daily cycle, longer periodicities are evident [6], the most significant being the annual seasonal cycle. To compensate for seasonality when assessing radon risk, results from individual short-term measurements are generally converted to equivalent mean annual concentration by application of a Seasonal Correction Factor (SCF). In the UK, and increasingly elsewhere, this is a set of monthly multiplying factors, derived from measurements in a large number of individual homes and reflecting both the length of the measurement and its month of commencement, applied to a radon concentration measurement with duration of one or more months to derive an estimate of annual average radon concentration. The UK SCF set was initially derived from the outcomes of two consecutive 6-month measurements performed in 2300 homes with a geographical distribution reflecting the UK population distribution [7]. The correction process requires knowledge of the start-date and duration of the measurement, and assumes annual, typically sinusoidal, variation [8]. Subsequent refinements, introduced as the database grew, included correction factors for exposure durations ranging from 1 month to 11 months [9].

Initial concern as to the applicability of a single SCF set was raised by Pinel *et al.* [8], their argument being that, due to the significant geological variability across the UK, SCFs derived from earlier nationwide studies [7] might not be appropriate in areas such as south-west England, geologically very different to the remainder of the UK. Studies by the Northamptonshire Radon Research Group (University of Northampton and Northampton General Hospital) of radon concentrations in a set of houses situated on radon-rich Jurassic strata in Northamptonshire, England [10,11], confirmed that the UK SCF set was, indeed, not replicated by the data from the study sample. Other studies have similarly concluded that location-specific SCFs are required for different regions of the UK [12] and Ireland [13]. Moreover, SCFs applicable to the British Isles differ significantly from those derived elsewhere in Europe [14,15,16,17,18,19], Asia [20] and North America [21,22] in both amplitude and phase.

#### **1.3** Radon and External Temperature

Given the twin conclusions that seasonal variability in indoor radon concentration cannot realistically be represented by a single national or international SCF scheme and that different SCFs may be required in localities of differing geological, geographical and climatic characteristics, attention has been directed towards a quantitative understanding of the underlying factors driving soil-gas radon emanation and ingress. Gunby *et al.* [23] demonstrated that many of the physical and constructional characteristics of a house affecting radon concentration could be regarded as surrogates for pressure difference, a proposition extended by Miles [24,25], who argued that the indoor-outdoor pressure difference is itself a function of the indoor-outdoor temperature difference, and that it this temperature difference which is ultimately responsible for the seasonal variation in radon concentration. Based on the initial UK radon survey, Miles [24] showed that the mean monthly domestic radon concentration, *R* (in units of Bq.m<sup>-3</sup>), was a linear function of the mean external temperature, *T* (degrees Celsius), having the form:

$$R = 33 - 1.26T \tag{1}$$

This relationship applies only while average outdoor temperatures are lower than indoors, the necessary condition for the classical driving mechanisms [26,5]. Based on this relationship, Ibrahimi and Miles [27,28] presented a general formula relating mean annual radon concentration, R, in a dwelling to the occupancy-weighted mean radon concentration, C, observed during a 2-week measurement period and the mean external temperature, T, during the same period:

$$R = (C - 4) \times (1/[1.645 - 0.063T]) + 4$$
<sup>(2)</sup>

where units are as previously. Since the mean outdoor radon concentration in the UK of 4 Bq $\cdot$ m<sup>-3</sup> [7] is, to first order, not affected by temperature, this component is subtracted from the measurement prior to applying the correction, and subsequently added back. Use of the relationship presented in Equation (2) is now recommended by HPA in cases where a short, typically 2-week, measurement period is imperative [27].

To investigate temperature-dependent effects further, the present authors commenced an analysis of data on seasonal variability of domestic radon, collected from published and unpublished sources. For consistency,

analysis was limited to situations in which radon concentration or SCF data were available at monthly resolution, although the method is obviously applicable to results presented at quarterly or weekly resolution, with correspondingly lesser, or greater, sensitivity [6,29]. As an extension of this work, attention was turned towards a hitherto unanalysed dataset, gathered during a period extending over several years, although unfortunately not entirely continuous, from an environment in which many anthropomorphic influences commonly affecting radon data collection were, possibly serendipitously, minimised or excluded.

# 2 Method

### 2.1 Radon Concentration Time-Series

During the period 2003 - 2008, a calibrated Durridge<sup>1</sup> RAD-7 radon measuring system was deployed for extended exposure periods in a rarely-visited partially-subterranean store-room in a public institution in the town of Northampton, UK. The room measures approximately 2 metres x 4 metres and is 3 metres high; it is bounded on its two longer sides by similar rooms and forms part of the lowest level of a late 18<sup>th</sup> Century four-storey brick/stone structure built on a sloping site. One of the shorter walls contains a door to the central corridor of the basement, while the other forms the windowless exterior wall of the building, beyond which is a small court-yard, with ground level between and two metres above the floor level in the room. The floor is tile/vinyl on concrete. During the entire investigation period, access to the room was restricted and normally limited to one or two brief visit(s) per week by authorised personnel (totalling no more than 0.5% of the total investigation period), generally at the same time of day on the same weekday.

Measurements of radon concentration, together with ambient temperature and relative humidity of the sampled gas, were made at hourly intervals, during the periods indicated in Table 1. Since the storage capacity of the RAD-7 system is limited to 999 records, data was downloaded at monthly intervals. Gaps in the record correspond to periods where the equipment was deployed elsewhere or absent for calibration, or to electrical power failure.

Start Date	Start Time	Finish Date	Finish Time	Series Count
	16:25	28 Jul 2003	15:08	1296
07 Apr 2004	12:30	18 May 2004	17:17	990
22 May 2004	21:16	28 May 2004	12:14	136
02 Jun 2004	10:11	07 Jun 2004	02:10	113
25 Aug 2004	11:28	27 Aug 2004	10:27	48
20 Sep 2004	14:26	27 Sep 2004	13:26	24
27 Sep 2004	11:34	14 Jun 2005	15:15	6247
21 Jun 2005	10:30	21 May 2006	17:59	8027
23 May 2006	15:12	26 Feb 2007	10:46	6695
04 Apr 2007	14:17	25 May 2007	12:17	218
20 Feb 2008	15:02	03 Mar 2008	04:58	279
18 Mar 2008	18:06	31 Mar 2008	12:02	307
			Total	24594

### Table 1: RAD-7 Data Sampling Periods

Within the overall investigation period of 42284 hours (1761.82 days), radon monitoring equipment was running for 24594 hours (1024.67 days), 58.2% of the total period. Maximum run length was 8026 hours (334.4 days), some 19% of the total period, while maximum gap length was 6504 hours (271.0 days), 15.4% of the total period. These are believed to represent some of the longest continuous UK radon datasets yet collected.

### 2.2 External Climate Time Series

External climatic data, comprising daily Air Temperature, Atmospheric Pressure, Relative Humidity, Rainfall and Wind-speed over the period spanning the radon measurement duration were downloaded from the archive maintained at the Pitsford Hall Weather Station (COL Station No. 91012) and published on the

<sup>&</sup>lt;sup>1</sup> Durridge Company, Inc., 524 Boston Road, Billerica, MA 01821, USA

station web-site [30]. This fully-equipped weather station, operated in conformity with UK Meteorological Office conventions, is maintained by the Department of Geography at Northamptonshire Grammar School, situated in the village of Pitsford, about 10 km north of the study site.

# 3 Results

### 3.1 Radon and Internal Temperature Time Series

Figure 1 shows graphically the full radon concentration time-series for the investigation, showing the measurement periods and the intervening non-measuring intervals.

Raw RAD-7 data was processed to provide mean and standard deviation of radon concentration and ambient temperature on hourly, daily, monthly (calendar) and annual bases. In addition, data for corresponding calendar months throughout the measurement period were aggregated, providing month-by-month statistics, shown in Table 2. Arithmetic mean radon concentration throughout the investigation period was 73.0 Bq.m<sup>-3</sup> (St .Dev. 62.7 Bq.m<sup>-3</sup>), significantly below the UK Workplace Action Level of 400 Bq.m<sup>-3</sup>. Although the data are closely log-normally distributed, as characteristically associated with multiple influencing factors [23], arithmetic mean and standard deviation are reported here for consistency with the other tabulated data. The mean ambient temperature throughout the investigation period was 26.2 C (St .Dev. 1.7 C), confirming the thermal stability of the test environment.

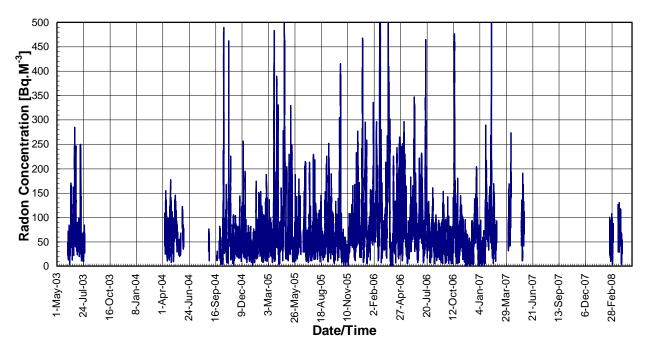


Figure 1: Radon concentration time-series, June 2003 – March 2008.

### 3.2 Climatic Time Series

Figure 2 plots the mean monthly values of the principal climatic parameters (external air temperature, relative humidity, precipitation, atmospheric pressure and wind-speed) over the investigation period, derived from data collected at Pitsford Hall weather station, showing the intrinsic seasonality of these parameters.

Particularly evident are the annual cycles of air temperature (minimum in February, maximum in August), and wind-speed and relative humidity (both approximately out of phase with temperature). Rainfall and atmospheric pressure present less systematic behaviour. Raw data was processed to provide statistical analysis (mean and standard deviation) of climatic parameters. In the absence of hourly data from the weather station, statistical analysis was possible only on daily, monthly (calendar) and annual bases. Again, data for corresponding calendar months throughout the measurement period were aggregated, providing month-by-month statistics, as shown in Table 2.

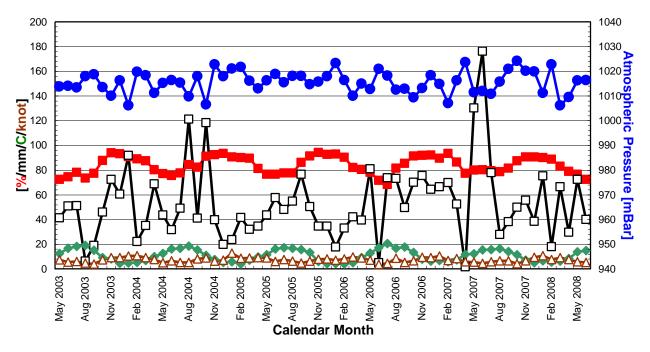


 Figure 2: Climatic parameter time-series, June 2003 – March 2008: Pitsford Hall Weather Station [30]

 Closed square – Mean Relative Humidity
 Open Square – Total Rainfall

 Closed diamond – Air Temperature
 Open triangle – Wind Speed

 Closed circle – Atmospheric Pressure (Right-hand axis)

#### 3.3 Statistical Analysis

Table 2 summarises the aggregated calendar month mean radon concentrations and climatic parameters (Air Temperature, Atmospheric Pressure, Rainfall, Relative Humidity and Wind Speed), with their principal statistical features over the measurement period.

Month	Radon	Internal	Ambient Air	r Temp.	Atmos.	Wind-speed	Rainfall	R.H.
		Temp.	Temp.	Difference	Pressure			
	[Bq.m <sup>-3</sup> ]	[C]	[C]	[C]	[mBar]	[knot]	[mm]	[%]
Jan	66.21	25.32	5.3	19.86	1015.07	10.43	57.45	90.97
Feb	85.32	26.51	4.7	21.76	1017.75	8.07	33.62	90.40
Mar	89.51	26.07	6.5	20.11	1013.65	8.95	42.80	86.32
Apr	84.41	26.82	9.7	16.77	1014.73	8.12	32.03	79.43
May	74.80	27.57	12.5	15.35	1014.28	6.58	68.65	76.75
Jun	81.30	27.20	16.0	10.75	1016.43	5.22	60.15	75.08
Jul	73.46	27.44	17.5	8.84	1014.40	5.75	60.33	76.38
Aug	51.37	27.00	17.2	9.83	1013.97	6.10	61.52	80.18
Sep	54.34	26.22	15.2	10.12	1017.68	5.43	53.13	83.40
Oct	68.84	25.48	11.3	13.04	1013.83	6.73	64.02	89.20
Nov	49.05	25.52	7.2	18.38	1015.87	7.73	58.37	92.20
Dec	68.24	25.23	4.8	20.03	1018.17	8.22	41.55	92.52
Mean	73.04	26.23	10.57	8.60	1014.69	6.74	55.61	85.27
St. Dev.	62.68	1.66	4.81	8.50	4.93	1.99	30.04	6.55

Table 2: Monthly mean radon concentrations and climatic parameters, and statistical analysis

#### 3.4 Correlation Analysis

Calendar month mean radon concentrations for the investigation period were cross-correlated against the corresponding sets of climatic parameters, together with the UK 1-month SCF set, via the Pearson Correlation coefficient, r, the results being summarised in Table 3.

	Radon	Temp. Internal	Temp. Difference	Temp. External	Atmos. Pressure	Rainfall	Wind- speed	Relative Humidity	1-month SCF
Radon	1.00	-0.16	0.23	-0.24	0.07	-0.36	-0.15	-0.19	-0.17
Temp. Internal	-0.16	1.00	-0.43	0.61	-0.37	0.65	-0.48	-0.41	0.61
Temp. Diff.	0.23	-0.43	1.00	-0.98	0.08	-0.32	0.66	0.71	-0.88
Temp. External	-0.24	0.61	-0.98	1.00	-0.16	0.43	-0.69	-0.72	0.92
Atmos. Pressure	0.07	-0.37	0.08	-0.16	1.00	-0.46	-0.10	0.02	-0.09
Rainfall	-0.36	0.65	-0.32	0.43	-0.46	1.00	-0.18	-0.13	0.39
Wind-speed	-0.15	-0.48	0.66	-0.69	-0.10	-0.18	1.00	0.42	-0.58
Rel. Humidity	-0.19	-0.41	0.71	-0.72	0.02	-0.13	0.42	1.00	-0.78
1-month SCF	-0.17	0.61	-0.88	0.92	-0.09	0.39	-0.58	-0.78	1.00

 Table 3: Pearson correlation matrix for radon and climatic parameters

In summary, mean monthly radon concentration shows:

- Minimal or zero correlation with any of the three Temperature parameters
- Minimal or zero correlation with Atmospheric Pressure or Wind-speed
- Minimal (negative) correlation with Rainfall
- Minimal or zero (negative) correlation with Relative Humidity.

In contrast, the UK 1-month SCF set shows:

- Strong correlation with all three Temperature parameters
- Strong correlation with wind-speed and RH
- Medium correlation with Rainfall
- Minimal or zero (negative) correlation with Radon

#### 4 Discussion

Soil-gas radon, generated by  $\alpha$ -decay within radium-bearing soil and bedrock, can enter a building through any opening, e.g. cracks in solid floors, gaps in suspended floors or apertures around service entry points, in contact with the ground. Since indoor air is generally warmer, and therefore less dense, than outdoor air, the consequent pressure differential acts as driving force (Stack effect) for soil-gas and any entrained radon, with magnitude generally dependent on the heating level and the outdoor temperature. Previous attempts to identify relationships between indoor radon concentration and climatic influences have produced inconsistent outcomes. Postendörfer et al. [31] concluded that, in a building with relatively stable air temperature, windspeed was the principal factor determining indoor radon concentrations. In contrast, an extended (some 3000 daily samples in three near-continuous groups) Japanese study [32] failed to identify such a correlation, although the power spectrum of the radon progeny ratio was shown to be similar to that of wind-speed. As noted, Miles [24] demonstrated good correlation between mean radon concentration and monthly temperature, but only while outdoor temperature is lower than that indoors, as required by the indoor underpressure model [5] postulated as responsible for radon-bearing soil-gas advection. Climent et al. [33] showed that radon concentration correlated inversely with air temperature and soil temperature at 300 mm soil depth, and directly with humidity, rainfall and wind direction, in each case without time delay. No correlation was evident with soil temperature at 100 mm depth, with wind speed or with seismic activity within a 100 km range. Dolejs and Hulka [34], investigating the effects of climatic parameters on the difference between short- (weekly mean) and long-term (annual mean) radon outcomes, showed that while the effects of atmospheric pressure and rainfall were not significant, external weekly mean temperature was a significant factor. Finally, our own previous investigations into the effect of climate on radon concentration [6], also in undisturbed basement situations, failed to demonstrate rigorous causality, although weak correlations with rainfall and mean daily temperature were identified.

The results reported here are not incompatible with these varied conclusions. As shown in Figure 2, certain climatic parameters show marked annual periodicity; external air temperature exhibits maximum and minimum values in August and February respectively, with wind-speed and relative humidity approximately in anti-phase to temperature. As Table 3 shows, radon concentration correlates best with rainfall (|r| = 0.36); external temperature (|r| = 0.24) and temperature difference (|r| = 0.23) show poorer correlation. Radon is essentially uncorrelated with atmospheric pressure (|r| = 0.07), the remaining correlation coefficients being in the range (0.16 - 0.19).

While this paper was in preparation, Florea and Duliu [35] reported eighteen years (January 1993 – December 2010) of continuous observation of atmospheric radon, using a twice-daily continuous aspiration and activity-counting procedure. Although addressing atmospheric rather than indoors radon, this study showed features common with those observed here, including annual periodicity of radon concentration, and negative correlation with precipitation, temperature and, to a lesser extent, with wind-speed.

The intrinsic thermal stability of the investigation site offers the opportunity for a realistic test of the various approaches to the definition of Seasonal Correction Factors. If indoor radon concentration at the study site is dominated by the stack-effect, then wind-speed, together with the temperature difference between interior and exterior environments, would be expected to contribute significantly towards determining radon ingress. It could be argued, however, that in its partly-subterranean location with no openings to the external environment, the investigation site is effectively decoupled from any stack-effect influences. In this situation, radon ingress, being therefore effectively determined by soil-gas radon content alone, will demonstrate a corresponding seasonal dependence. As soil characteristics are influenced by changes in meteorological conditions, temporal variations of soil-gas radon concentration are widely observed [36].

Using the available data, four surrogate SCF series were derived:

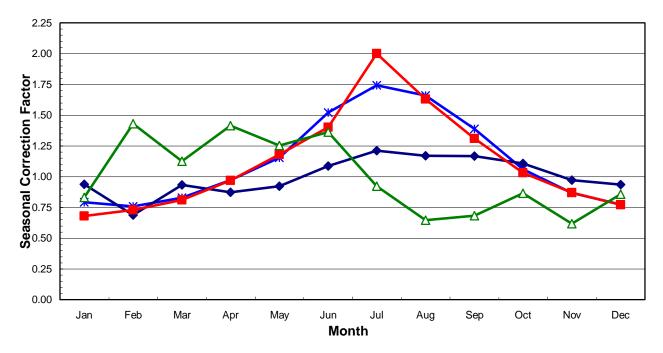
- 1. Directly from the mean monthly radon concentration at the investigation site, by normalisation of each monthly mean against the annual mean.
- 2. Using the HPA formula in conjunction with the mean monthly external temperature at Pitsford Meteorological station.
- 3. Using the HPA formula in conjunction with the mean monthly temperature difference between the Pitsford Meteorological station and the investigation site.
- 4. Using the HPA formula in conjunction with the mean monthly internal temperature at the investigation site.

Plots of the first three of these, together with the current UK 1-month SCF set, are shown in Figure 3

Each set was cross-correlated in turn with all of the other sets and with the UK 1-month SCF set, the results being shown in Table 4.

SCFs derived from the observed radon concentration data show relatively poor correlation with SCFs derived from either the external temperature (|r| = 0.18), the internal temperature (|r| = 0.15) or the UK HPA 1-month SCFs (|r| = 0.17) but show significantly enhanced correlation (|r| = 0.59) with the SCFs derived from the temperature difference. This indicates firstly, that the relative temperature difference between the test site and the external environment provides a major component of the forces driving radon ingress, and secondly that in applying the HPA formula shown in Equation 2, better representation of the actual seasonal correction to be applied in any particular case might be achieved by using the temperature difference between the test site and the external environment rather than the absolute external temperature.

It is apparent that an SCF set derived using mean monthly external temperature data provides near-perfect correlation (|r| = 0.97) with the existing recommended SCF set. This observation suggests that, particularly for month-long time-integrating radon determinations, the complexity of deriving SCFs from temperature data is outweighed by the convenience of using the existing HPA SCF set. Slightly poorer correlation (|r| = 0.79) exists between SCFs derived from the internal-external temperature difference. Correlation between SCFs derived from the local radon concentration and the ambient UK 1-month set is poor (|r| = 0.17), supporting previous observations in the Northamptonshire area [10].



#### Figure 3: Seasonal Correction Factors

Open triangle – Radon at investigation site Star – HPA formula using Northants temperature data Closed square – UK 1-month SCF set Closed diamond – HPA formula using temperature difference

	UK 1-month	Radon-derived	Temperature, using Eq. 2			
	SCF set	SCF	External Temperature	Temperature Difference	Internal Temperature	
HPA 1-month SCF	1.00	-0.17	0.97	0.79	0.35	
Radon-derived SCF	-0.17	1.00	-0.18	-0.59	0.15	
External Temperature	0.97	-0.18	1.00	0.82	0.32	
Temperature Difference	0.79	-0.59	0.82	1.00	0.20	
Internal Temperature	0.35	0.15	0.32	0.20	1.00	

#### 5 Conclusions

Radon concentration and ambient temperature were monitored at hourly intervals in an environmentallystable, rarely-visited basement in a public-service building. Sampling periods totalled 1025 days during the period between June 2003 and March 2008 (1766 days), the longest continuous sampling period and interval between sampling periods being 335 and 271 days respectively. Mean daily, monthly (calendar) and annual radon concentrations and internal temperatures were calculated, averages over corresponding months of at least three separate years being available for each calendar month. Corresponding meteorological parameters were generated from data collected at a weather station 10 km from the measurement site. The mean and standard deviation over the whole radon data-set are 73.0 Bq.m<sup>-3</sup> and 62.7 Bq.m<sup>-3</sup> respectively, corresponding mean and SD internal temperatures being 26.2 C and 1.7 C, respectively.

Monthly mean radon concentration shows minimal or zero cross-correlation with the three temperature parameters, atmospheric pressure and wind-speed; there is limited negative correlation with rainfall and possible negative correlation with relative humidity.

Surrogate SCF sets derived using the HPA temperature-based formula indicate relatively good correlation (|r| = 0.59) between radon seasonality and the internal-external temperature difference, but poorer correlation

(|r| = 0.18) with absolute external temperature, the parameter implied by use of that formula. In contrast, the HPA 1-month set correlates very well (|r| = 0.97) with the SCF set derived from external temperature.

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