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Time-integrating radon gas measurements in domestic premises: comparison of short-, medium- and long-term exposures

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Abstract

To identify the most applicable technology for the short-term assessment of domestic radon levels, comparative assessments of a number of integrating detector types, including track-etch, electret and activated charcoal were undertaken. Thirty-four unremediated dwellings in a high-radon area were monitored using track-etch detectors exposed for one-month and three-month periods. In parallel, one-week measurements were made in the same homes at one-month intervals, using co-located track-etch, charcoal and electret detectors exposed simultaneously, while three of the homes were also monitored by continuous-sampling detectors at hourly intervals over extended periods. Calibration of dose-integrating devices against each other and against continuous-monitoring systems confirmed good responsivity and linearity. Although track-etch, charcoal and electret devices are suitable in principle for one-week measurements, zero-exposure offset and natural radon variability cause many one-week results to be equivocal, necessitating repetition of the measurement. One-week exposures can be

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reliable indicators in low-radon areas or for new properties, but in high-radon areas, the use of three-month exposures is indicated. This analysis also established confidence limits for short-term measurements.

Keywords: Radon; Track-etch; Electret; Activated charcoal; Continuous sampling; Comparative assessment; Health risk; Domestic properties

1. Introduction

Radon is a naturally-occurring radioactive noble gas, having variable distribution in the geological environment as a decay product of uranium found, in differing degrees, in a wide range of rocks and soils and in building materials incorporating or manufactured from these sources. There are three naturally occurring isotopes of radon, ^{222}Rn, a direct product of ^{226}Ra in the ^{238}U decay-series with a half-life of 3.8 days; ^{220}Rn, a decay product of ^{232}Th, with a half-life of 55.6 s; and ^{219}Rn, a decay product of ^{235}U, with a half-life of 3.6 s. Radon has high mobility, enabling it to move out of underlying rocks and ground-water into caves, mines and the built environment. Of the three isotopes, ^{222}Rn is the most significant, its relatively long half-life enabling it to migrate quite significant distances within the geological environment before decaying. Although radon dissipates rapidly once in outdoor air, it can concentrate in the built environment. For UK dwellings, the mean radon level is around 20 Bq m^{-3}, compared to 4 Bq m^{-3} in outside air (Wrixon et al., 1998) but levels up to 17,000 Bq m^{-3} have recently been found in residential properties (NRPB, 2004).

Ionising radiation is well known to have adverse health effects, and inhalation of radon and its progeny ^{218}Po and ^{214}Po adsorbed onto atmospheric particulates is currently believed (Darby et al., 2005; Krewski et al., 2005) to provide the majority of the dose to the respiratory system. This results in damage to the sensitive inner lining of the lung, increasing the risk of cancer, and it is further estimated that the annual mortality from exposure to radon in buildings represents 9% of all deaths from lung cancer, and 2% of all cancer deaths, in Europe (Darby et al., 2005). The total annual mortality from this type of cancer in the UK is between 30,000 and 35,000 (UK Dept. of Health), suggesting that between 1800 and 2100 deaths annually are caused by exposure to radon and its progeny.

Indoor radon levels are subject to a number of variations. In addition to the natural daily cycle, other longer temporal and spatial cycles are evident, related to occupancy, weather conditions and seasonal factors, indoor radon levels being generally higher in winter than in summer. As the risk of lung cancer increases with increasing radon exposure, the preferred measure of this risk is the long-term average radon level, the current UK recommendation being the use of three-month measurements in conjunction with the application of a Seasonal Correction Factor (Pinel et al., 1995; Gillmore et al., 2005). In some circumstances, however, particularly during the house-sale process or when confirming that safety measures in new homes are satisfactory, a measurement extending over three months is impractical or inappropriate. The question then arises as to whether short-term measurements, although probably less reliable, have sufficient value to be of use. We explore here the issue of the viability of short-term measurements in detail, highlighting issues relating to detectors and protocols.

Following a description of the project methodology, results will be presented from a year-long study of radon levels in homes in the County of Northamptonshire, in the United Kingdom, utilising a number of types of radon detector. This area, identified in the map in Fig. 1 (Miles and Appleton, 2005), was designated as a radon Affected Area in 1992 by the UK National Radiological Protection Board (now part of the Health Protection Agency), and has a mean annual radon concentration of 70 Bq m^{-3}, with around 5600 homes in the county anticipated to exhibit radon concentrations in excess of the UK Action Level of 200 Bq m^{-3} (Green et al., 2002). Some aspects of this study have already been reported (Phillips et al., 2004; Denman et al., 2004a,b; Denman et al., in press; Gillmore et al., 2005). The implications of the results are discussed and recommendations arising from them, particularly the value and reliability of a short-term testing protocol, are made.

2. Method

2.1. Experimental procedure

To define a 'reference' technology for relating short-term to long-term exposure results, dose-integrating detectors were assessed by making direct comparison with instantaneous radon data for corresponding exposure periods obtained from two Durridge RAD-7[1] systems. These were operated in three properties for extended periods spanning the full year of the project, and had been calibrated prior to commencement of the study. To ensure reliable correlation, short-term exposures of detectors were carried out in the immediate vicinity of the RAD-7 systems.

2.2. Radon detector selection

Dose-integrating radon detectors were procured from suppliers who had submitted detectors to the UK National Radiological Protection Board (NRPB) Inter-Comparison of Passive Radon Detectors (Howarth and Miles, 2002). The final detector inventory included 1400 track-etch (Alter and Fleischer, 1981) devices from two different suppliers, 600 activated charcoal detectors (George, 1984) and 50 electrets (Kotrappa et al., 1988) (reusable devices that were deployed for a total of nearly 1000 exposures). Best practice was followed at all times in respect of detector handling and analysis. The majority of detectors were placed and collected by trained personnel, ensuring

[1] Durridge Co., 7 Railroad Avenue, Bedford, MA 01730, USA.

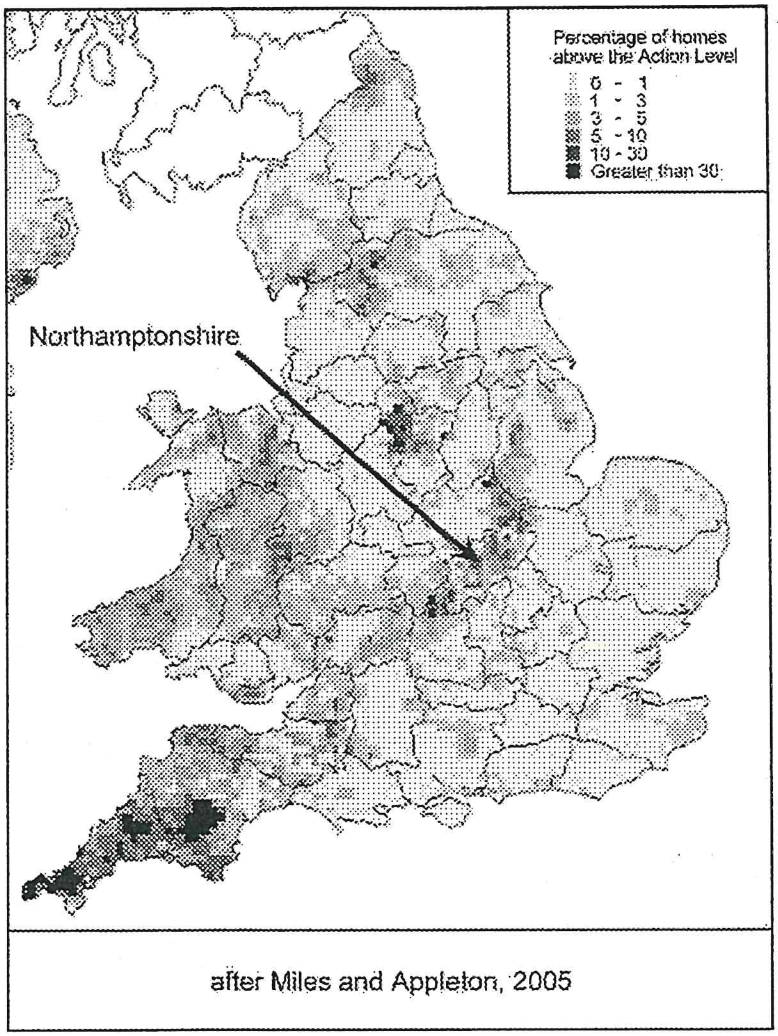

Fig. 1. Radon map of England, showing location of Northamptonshire (after Miles and Appleton, 2005).

data rigour and resulting in a return rate close to 100%, much higher than that achieved in many postal based projects, which depend on occupier compliance.

2.3. Property selection

Starting from a pre-existing confidential database of unremediated domestic properties with known elevated radon levels in a high-radon area around Northampton, U.K., a short-list of 60 potential candidate dwellings was identified. Following further assessment, a sub-set of 37 dwellings was identified (34 of which subsequently successfully completed the monitoring programme) as suitable for long-term monitoring, which took place during the period April 2002–May 2003. The dwellings were selected to include a variety of type, construction era and construction/furnishing features, as indicated in Tables 1–3, respectively.

Northamptonshire is a county of essentially sedimentary rocks, mainly of the Lower and Middle Jurassic age (Gillmore et al., 2005). Highest radon levels are found on Northampton Sand (Thompson, 1897; Hains and Horton, 1969), where more than 20% of homes can be over the Action Level, while the lowest levels occur on Oxford Clay and Blisworth Clay, where less than 1% of homes are over the Action Level. Radon Potential data for the dwellings taking part in the study were obtained from the British Geological Survey, whose assessments are derived from geologically-based interpretation of radon measurements in dwellings, provided by the NRPB without prejudicing confidentiality undertakings to householders and Government (Miles and Appleton, 2001). Ninety-four percent of the dwellings were situated on a common geology (Northampton Sand), 91% were in areas where more than 30% of dwellings were estimated to exhibit radon levels in excess of the Action Level and 41% were located in a compact area (radius 400 m).

Of the 34 properties completing the programme, 20 were monitored for four consecutive three-month periods and simultaneously for 12 consecutive one-month periods; the remaining properties were monitored for periods of nine months (four properties), six months (six properties) or three months (four properties). In addition, one-week measurements were made in all properties at approximately one-month intervals, using co-located sets of track-etch, activated charcoal and electret detectors exposed simultaneously. In a small number of properties, further comparisons were made with continuous-monitoring techniques over periods of up to six months.

2.4. Detector management

Detectors were exposed according to the NRPB protocol (Wrixon et al., 1998), care being taken to avoid areas of high relative humidity, e.g. bathrooms or kitchens. This protocol uses two detectors, one placed in the main living room (generally at ground level) and one in the main bedroom (usually on the first-floor). The protocol calculates a weighted average of the two readings, the bedroom being assigned a weighting of 0.55, the living room 0.45, reflecting their relative occupancies.

Table 1
Dwellings completing the project: type

Type	Total	Percentage
Detached	17	50
Semi-detached	9	26
Link	4	12
Bungalow	4	12
Total	34	

Table 2
Dwellings completing the project: construction era

Date of initial construction	Total	Percentage
Pre-1950	17	50
1950–1970	5	15
1971–1980	7	20
1981–1990	5	15

A Seasonal Correction Factor (Pinel et al., 1995) is then applied, depending on the start month of the exposure. Within each three-month period, each dwelling had:

- Three-month (90-day) track-etch exposures in living room and main bedroom.
- Three consecutive one-month (28-day) track-etch exposures in living room and main bedroom.
- A number of 7-day, track-etch, activated charcoal and electret exposures in living room and main bedroom.

The 7-day exposures were managed to ensure that detector exposure was 168 ± 2 h, with 28-day exposures similarly managed to ensure exposure was 672 ± 2 h.

On completion of the required exposure, detectors were sealed in accordance with the manufacturer's specifications, further sealed in double plastic bags and mailed as quickly as possible to their respective manufacturers/suppliers for processing. This latter requirement was particularly pressing for activated charcoal devices, since the adsorbed radioactivity continues to decay once the exposure is completed. Electrets were measured in-house using proprietary equipment supplied by the manufacturer.

2.5. Ethical issues

The work reported here was carried out in accordance with the participating institutions' ethical codes of practice. All householder details were anonymised,

Table 3
Dwellings completing the project: construction and furnishing features

Feature	Total	Percentage
Construction materials		
Brick	14	40
Brick/breeze	16	47
Stone	4	12
Cellar	10	29
Suspended wood floors	14	40
Fitted carpets		
Full	20	58
Partial	5	15
Double glazing	29	85
Central heating	34	100
Open fireplace (lounge)	23	68

any necessary geographical identification being maintained by the use of UK postcodes.

3. Results

3.1. Reference technology

Fig. 2 shows typical short-term radon concentration variability reported by the RAD-7 systems, showing the diurnal order-of-magnitude range typically experienced. Fig. 3 similarly shows typical mean weekly radon concentrations derived from RAD-7 monitoring in three properties, normalised to the overall mean for each property to facilitate comparison.

Fig. 4 shows the results from one-week exposures, plotted against the average radon levels determined from RAD-7 measurements in the respective properties over the periods corresponding to the short-term exposures, together with the line of unity slope, while Table 4 summarises linear regression parameters from these plots, together with comparable data from one-month and three-month exposures.

Analysis of the one-week regression parameters leads to the following conclusions:

- Track-etch detectors represent extremely well the time-averaged real-time radon (gradient = 1.01) and exhibit extremely good linearity (correlation coefficient = 0.99) with a moderate background offset (35 Bq m^{-3}).
- Electret detectors represent relatively well the time-averaged real-time radon (gradient = 1.07), with good linearity (correlation coefficient = 0.98) and a significant background offset of 105 Bq m^{-3}.
- Activated charcoal detectors have the smallest background offset, 1.5 Bq m^{-3}, with good linearity but significantly more scatter, particularly at higher radon levels, than exhibited by the other classes of detector (correlation coefficient = 0.96). The raw results from the processing of exposed detectors, provided by the detector supplier, indicated a constant gradient of 1.22, suggesting that these detectors respond 'faster' to radon than the other types of detectors. Subsequent discussion with the supplier, however, confirmed that a "margin of error" correction, equal to +22.2% had been included in the processing protocol provided by the detector manufacturer (Ahern, 2003). Once this is taken into account, the regression slope for charcoal detectors against time-averaged radon determinations reduces to essentially unity, with background of 1.24 Bq m^{-3}, as indicated in Table 4. This corrected value is used in all subsequent discussion.

One-month and three-month results must be treated with more caution than the one-week data due to the smaller numbers of data-points. However, these data confirm the basic features of the one-week data. Specifically:

- Linearity remains good for all types of detector (correlation coefficients close to unity).

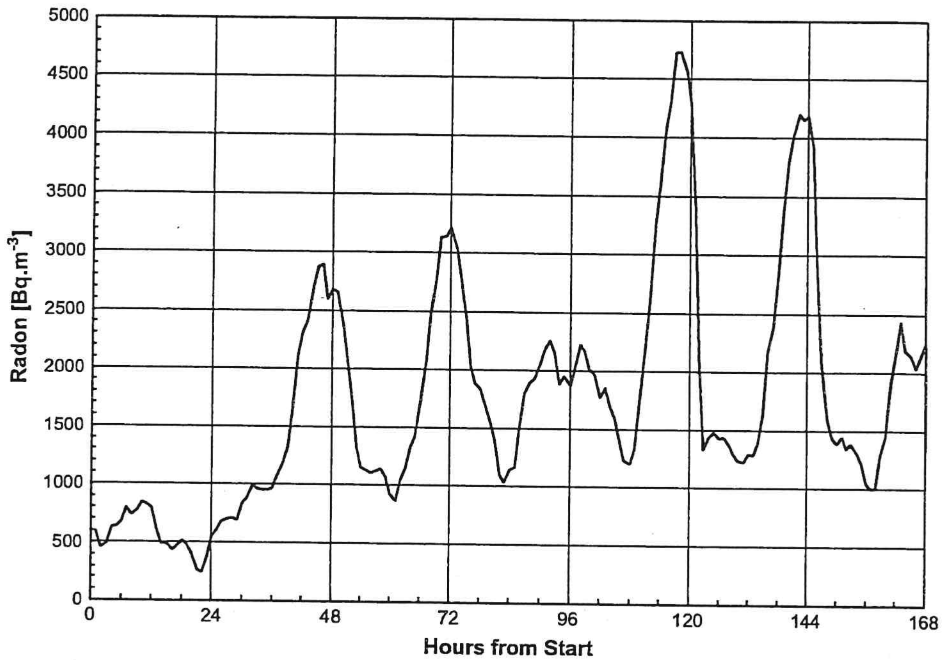

Fig. 2. Typical short-term radon concentration variability, as monitored using RAD-7 system.

- Track-etch detectors represent reasonably well the time-averaged real-time radon, with gradients close to unity.
- The decreasing background observed as exposure time increases (Table 4) reflects the increased precision associated with extended measurements and is consistent with the regression model obtained from the one-week results.

3.2. *One-week (7-day) measurements*

On the basis of the foregoing results, it was concluded that the activated charcoal detector, with its good linearity and almost negligible background offset, represented the best option for relating short-term (i.e. one-week) and long-term measurements statistically. Results from activated charcoal detectors were therefore used as reference for assessing the results from other short-term detectors in the majority of properties, where the limited number of RAD-7 systems available had precluded real-time monitoring.

All sites showed strong week-on-week variability. Using one-week activated charcoal outcomes from each property as reference, the accuracy with which these were represented by track-etch and electret detectors was explored. Results are shown in Fig. 5, for track-etch detectors exposed for periods of one week, one month and three months, and in Fig. 6 for electrets exposed for 7 days, in each case with the line of unity slope superimposed for convenience of reference.

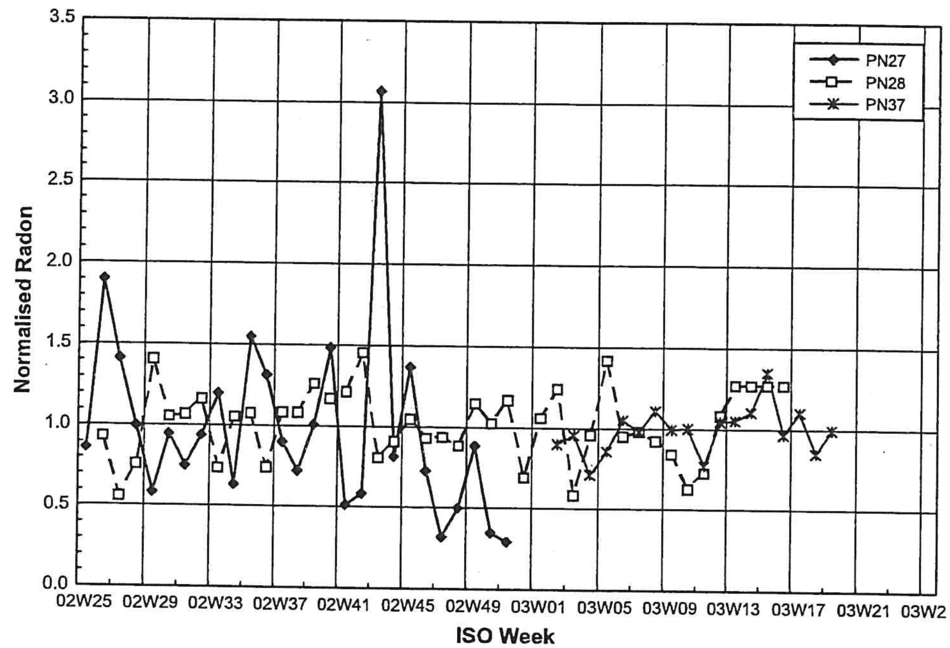

Fig. 3. Weekly mean radon levels in three properties determined using RAD-7 systems. Solid diamonds: Property A, open squares: Property B, star: Property C, continuous line: unity slope.

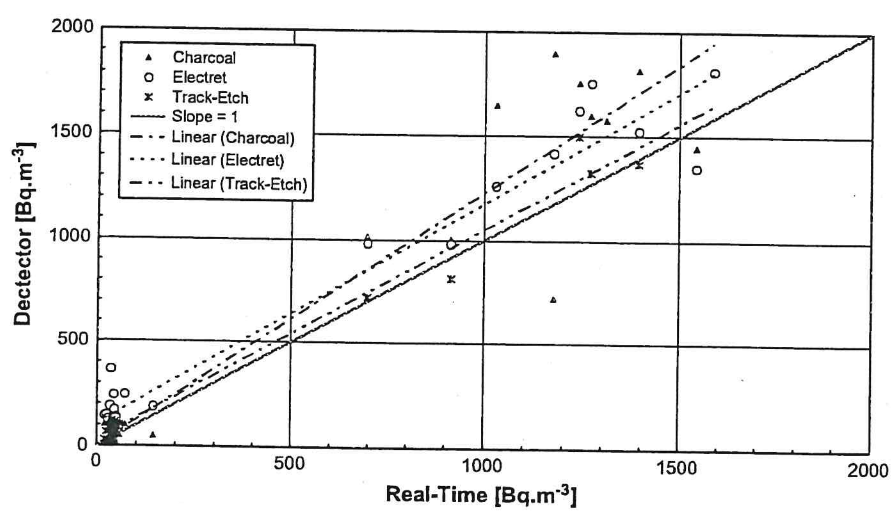

Fig. 4. Radon levels reported from one-week exposures in four classes of detectors vs. mean real-time levels determined by RAD-7 measurement for the corresponding exposure periods. Closed triangle: charcoal, open circle: electret, star: track-etch, continuous line: unity slope.

Table 4
Regression parameters for dose-integrating detectors vs. RAD-7

	Track-etch One-week	Charcoal One-week	Electret One-week	Track-etch One-month	Track-etch Three-month
Slope	1.01	1.22	1.07	0.93	1.00
Intercept	35.6	1.52	104.8	4.3	1.35
Correlation coefficient	0.99	0.96	0.98	0.99	1.00

Table 5 summarises the principal conclusions of the one-week comparison against activated charcoal, presenting regression parameters for track-etch and electret detectors against the corrected activated charcoal outcomes. Both track-etch and electret systems show good linearity against charcoal, with gradients of 1.01 and 0.93, respectively, with corresponding correlation coefficients, of 0.89 and 0.79. However, these are poorer than in the correlation against RAD-7 results summarised in Table 4, reflecting the uncertainties in the comparative values obtained using the measurement techniques.

3.3. One-month (28-day) measurements

Fig. 7 shows the seasonal variation in outcomes from one-month exposures, calculated as the mean normalised (relative to the annual mean for each dwelling) radon level for all properties taking part, referred to the month of commencing the exposure, together with RAD-7 results from one dwelling (Property A). Also shown is the Seasonal Correction Factor for one-month exposures. Over the period

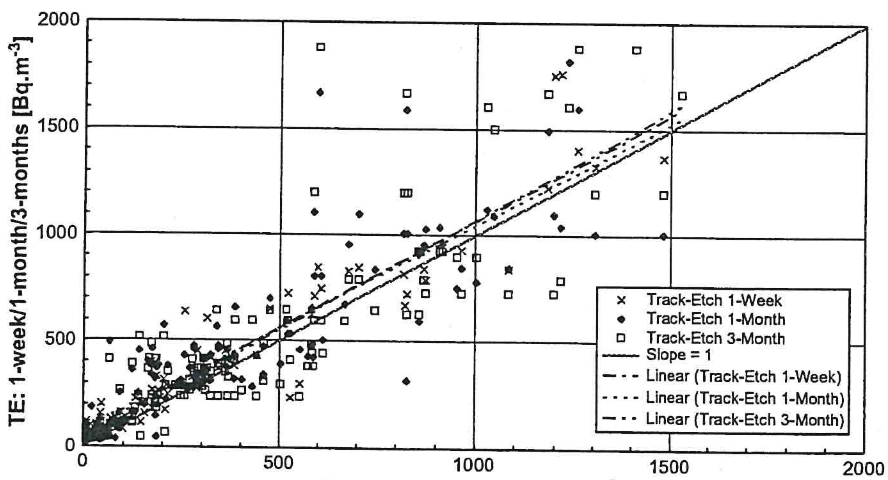

Fig. 5. Radon levels reported from one-week track-etch exposures vs. radon levels reported from simultaneous one-week activated charcoal exposures. Cross: one week, closed diamond: one month, open square: three months, continuous line: unity slope.

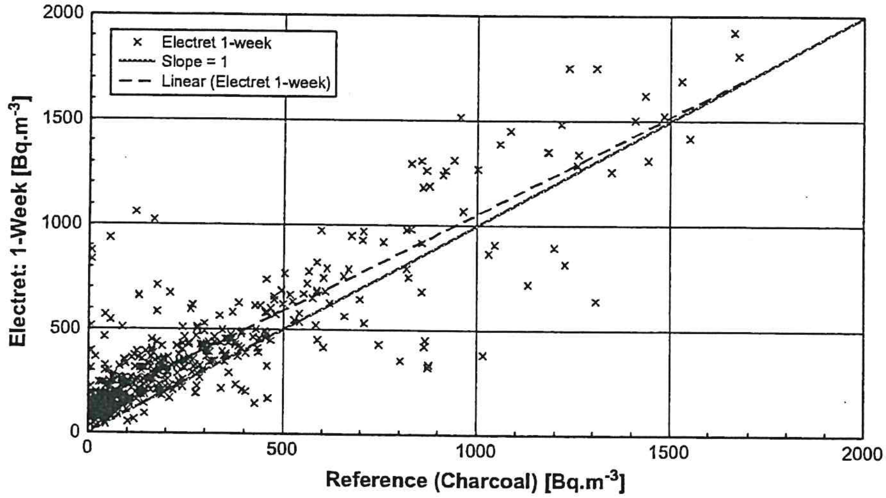

Fig. 6. Radon levels reported from one-week electret exposures vs. radon levels reported from simultaneous one-week activated charcoal exposures.

May 2002–April 2003, the combined one-month seasonal variation in track-etch outcomes does not match the outcome from RAD-7 monitoring of Property A over the corresponding period, and shows evidence of a discontinuity between April (of 2003) and May (of 2002). Unfortunately, the available RAD-7 data do not span the full 12-month period. Moreover, the one-month seasonal variation observed in the track-etch outcomes over the period May 2002–April 2003 does not match the cyclical behaviour of the Seasonal Correction Factor.

3.4. Three-month (90 day) measurements

Fig. 8 shows the seasonal variation in outcomes from three-month exposures, calculated as the mean normalised (relative to the annual mean for each dwelling) radon level for all properties taking part, referred to the month of commencing the exposure. Also shown is the Seasonal Correction Factor for three-month exposures, again plotted against the month of commencement.

As with the one-month outcomes, the three-month seasonal variation observed in the track-etch outcomes over the period May 2002–April 2003 does not match the behaviour expected by application of the Seasonal Correction Factor, and the

Table 5
Regression parameters for track-etch and electret vs. activated charcoal as baseline

	Track-etch	Electret	Track-etch	Track-etch
	One-week	One-week	One-month	Three-month
Slope	1.01	0.930	0.97	1.04
Intercept	56.37	125.54	77.32	32.49
Correlation coefficient	0.89	0.79	0.77	0.78

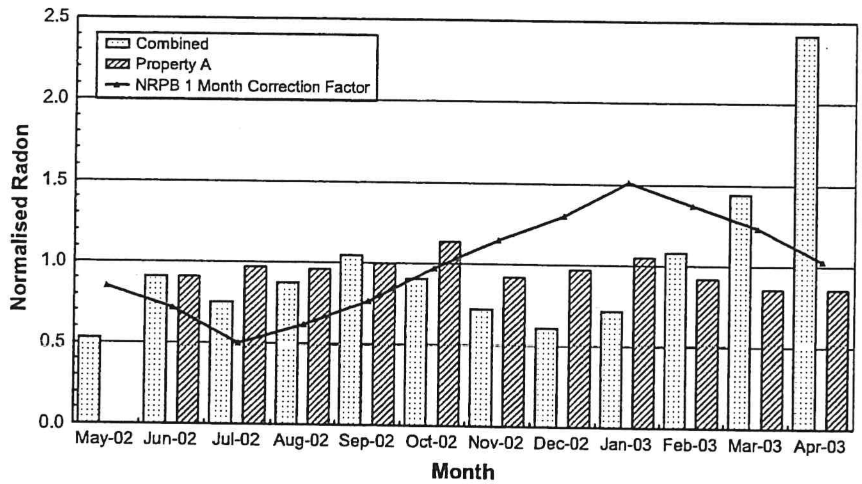

Fig. 7. Combined data for one-month exposures, normalised to annual average, by start month of measurement, with superimposed Seasonal Correction Factor for one-month exposure and mean one-month radon determined from RAD-7 measurements in Property A. Hatched bars: Property A, solid bars: cll properties combined, line/closed squares: Seasonal Correction Factor.

experimental results again show evidence of a discontinuity between April (of 2003) and May (of 2002).

Of the properties studied, a significant number had previously been tested for radon by NRPB using the standard three-month protocol. Analysis of the three-month outcomes revealed noticeable changes in radon over the period between NRPB

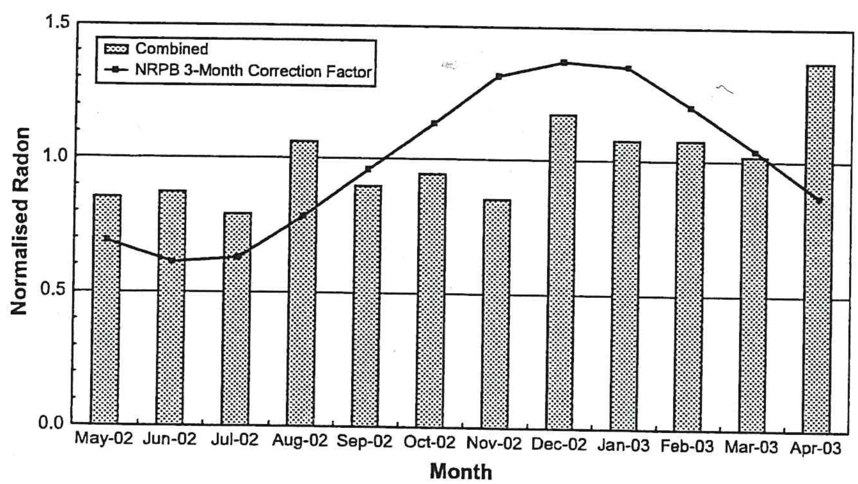

Fig. 8. Combined data for three-month exposures, normalised to annual average, by measurement start month, with superimposed Seasonal Correction Factor for one-month exposure. Solid bars: combined three-month outcomes, line/closed squares: Seasonal Correction Factor.

assessment and the present study, with some 65% of the properties surveyed experiencing increased radon levels.

3.5. *Estimation of mean radon levels from one-week, one-month and three-month outcomes*

For each property and exposure period, the ratios of one-week and one-month outcomes to the corresponding three-month outcomes were calculated, giving results for one-week and one-month track-etch, one-week electret, and one-week activated charcoal, all vs. three-month track-etch. Fig. 9 plots the distribution of ratios of activated charcoal and electret one-week results to the corresponding three-month track-etch outcomes, together with log-normal fits to these data. Comparable plots were obtained from the other comparisons.

Using mean and standard deviation for each dataset, 95% confidence levels were derived representing the probability that one-week or one-month radon levels were within 5%/10%/20% of the three-month radon level, summarised in Table 6. Derived from this analysis, Table 7 indicates the threshold levels above/below which there can be 95% confidence that the indicated annual level is greater/less than the Action Level of 200 Bq m^{-3}.

4. Discussion

4.1. *Reproducibility*

In evaluating the performance of any method of radon measurement, it is critically important to use some form of primary standard. Although the study reported

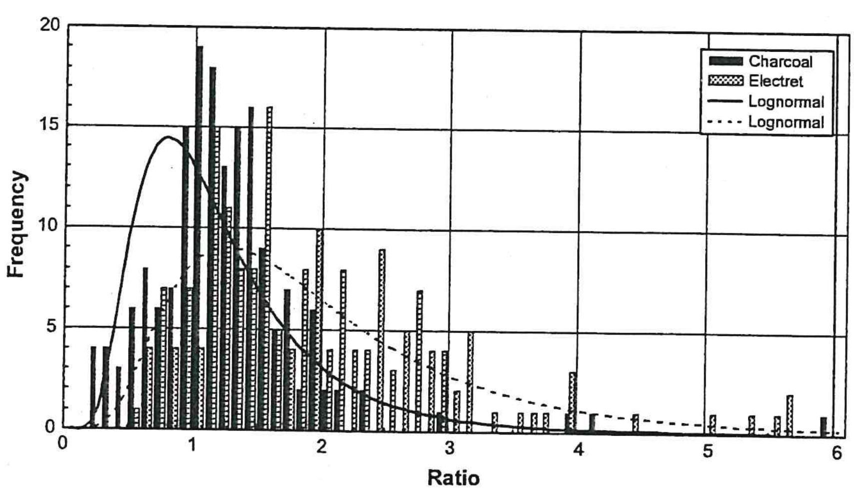

Fig. 9. One-week/three-month ratios – activated charcoal and electret vs. Gamamdata three-month. Solid bars: charcoal, solid line: charcoal log-normal fit, hatched bars: electret, broken line: electret log-normal fit.

Table 6
Probabilities that one-week and one-month outcomes represent three-month track-etch outcomes

Required accuracy (%)	Charcoal (%)	Electret (%)	Track-etch (%)	Track-etch (%)
	One-week	One-week	One-week	One-month
5	6.6	4.9	5.7	11.1
10	13.2	9.9	11.4	22.0
20	26.5	19.9	22.8	42.1

here was essentially comparative, based largely on single-exposure detectors procured from a number of validated suppliers, one element, namely the two RAD-7 systems deployed throughout the study, had been calibrated immediately prior to the study, and were therefore considered to be reliable secondary standards. While the possibility is acknowledged that even a perfect set of comparative measurements obtained using two methods might simply result from the two methods being equally biased, in practice this is unlikely, and the correlations found here are believed to be real.

4.2. Action Level indicators

All detector systems exhibit good linearity with mean radon level during exposure and all appear intrinsically suitable for use in Domestic and Workplace applications. Short-term (i.e. 7-day) exposures are possible but a greater proportion of results will be equivocal than if one-month or three-month exposures were employed, necessitating repeat exposures. Statistical analysis confirmed that for individual one-week track-etch, activated charcoal and electret outcomes less than 75, 68 or 59 Bq m^{-3}, respectively, the annual average is guaranteed (95% confidence) to be below the UK domestic Action Level (200 Bq m^{-3}). These outcomes are in good agreement with the NRPB recommendation (Miles et al., 2004) that "if a well-conducted charcoal measurement yields a result of 75 Bq m^{-3} or less, it can be taken as very likely that the true annual average does not exceed the Action Level of 200 Bq m^{-3}". Similarly, the upper level is set at the point where there is 95% confidence of exceeding the 200 Bq m^{-3} level.

Using the known percentage of homes in the counties of Cornwall, Northamptonshire and Buckinghamshire (areas of the UK with high, moderate and low numbers, respectively, of properties with radon levels above the Action Level (Green et al., 2002)), calculations were made of the proportion of measurements with each of the methodologies resulting in positive indications of radon levels above and below

Table 7
Threshold confidence limits (95%)

Confidence level (95%) (Bq m^{-3})	Charcoal	Electret	Track-etch	Track-etch	NRPB advice
	One-week	One-week	One-week	One-month	Three-month
Lower	68	59	75	109	130
Upper	522	667	518	478	300

the Action Level, and of the proportion of results which will be equivocal. Results are summarised in Table 8, which indicates the ranges of measurement over which short-term radon concentrations derived from the various detector technologies and exposure periods can be regarded as definitive (95% confidence) indicators of mean annual levels below or above the Action Level of 200 Bq m^{-3}. Results falling between these bounds must be regarded as equivocal, necessitating a repeat determination for resolution. It is evident that 7-day measurements are noticeably more useful in areas with low and medium numbers of properties with radon levels in excess of the Action Level, where the majority of results will be reliable indicators. In an area, such as Cornwall, with a high proportion of properties with radon concentrations in excess of the Action Level, the majority of 7-day results will be equivocal, with only around 5% of results being definitely abnormal. The improved accuracy offered by three-month determinations in this situation is a significant benefit. One-week exposures would also be suitable for newly-constructed houses with radon precautions where radon levels are expected to be low.

4.3. Seasonal Correction Factor issues

Application of a Seasonal Correction Factor implies repetition of the seasonal conditions affecting radon emanation with an annual periodicity, the assumption being made that this periodicity has a sinusoidal form (Pinel et al., 1995). The lack of cyclical continuity, demonstrated during the course of the present programme by the general disjoint in radon levels between May 2002 and April 2003, suggests that additional influences, possibly climatological, are driving radon emanation. Similar evidence has been reported elsewhere (Martz et al., 1991). Seasonal Correction Factors are derived from 'averaged' data collected from a variety of geologies, many of which are granitic or otherwise uranium-rich. Although geological influences on radon emanation have been noted in the literature (Gunby et al., 1993), the present study has been limited essentially to a single, non-granitic geology, viz. Northampton Sand.

Table 8
Action Level thresholds – statistics in high, medium and low-radon counties

Locality % above Action Level	Relative to Action Level	One-week track-etch (%)	One-month track-etch (%)	Three-month track-etch (%)	One-week electret (%)	One-week charcoal (%)	Three-month (NRPB) (%)
High	Below	36.2	54.3	55.4	30.2	35.5	61.3
Cornwall	Equivocal	56.2	40.3	35.1	66.3	60.0	25.9
23.3%	Above	4.6	5.4	9.5	3.5	4.50	12.8
Medium	Below	71.4	81.7	82.6	63.3	68.20	85.9
Northamptonshire	Equivocal	27.7	17.0	15.2	36.0	30.9	10.9
6.99%	Above	0.9	1.4	2.2	0.7	0.9	2.6
Low	Below	85.9	93.5	93.5	78.4	83.1	95.9
Buckinghamshire	Equivocal	14.1	6.3	6.3	21.6	16.9	3.8
1.19%	Above	0.0	0.2	0.2	0.0	0.0	0.3

The observed disparity between the present results and those expected from application of a conventional Seasonal Correction Factor raise a number of questions requiring resolution:

- Do individual geologies exhibit local seasonal variation?
- Does seasonal variations remain constant from year to year?
- Are seasonal variations in individual dwellings similar?

5. Conclusions

In order to identify the most applicable technology for short-term domestic radon measurements, comparative assessments of track-etch, electret and activated charcoal detectors were undertaken. Thirty-four unremediated dwellings in a high-radon area were monitored using track-etch detectors exposed for one-month and three-month periods. In parallel, one-week measurements were made at one-month intervals, using co-located track-etch, charcoal and electret detectors exposed simultaneously, while three homes were also monitored by continuous-sampling detectors at hourly intervals over extended periods. Calibration of dose-integrating devices against each other and against recently-calibrated secondary-standard continuous-monitoring systems confirmed good responsivity and linearity. Although track-etch, charcoal and electret devices are suitable in principle for one-week measurements, zero-exposure offset and natural radon variability cause many one-week results to be equivocal, necessitating repetition of the measurement. One-week exposures can be reliable indicators in low-radon areas or for new properties, but in high-radon areas, the use of three-month exposures is indicated. This analysis also established confidence limits for short-term measurements.

Acknowledgements

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