STUDIES OF RADON IN IRELAND: OUTDOOR LEVELS, DETECTOR INTERCOMPARISON AND SCHOOL REMEDIATION METHODS

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For Robbie, for everything

DECLARATION

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SUMMARY

Although Ireland has a comprehensive radon strategy in place and an active research community, the rate of testing is still low. The work in this thesis aims to reduce knowledge gaps by making outdoor radon measurements, aid the introduction of radon testing for conveyancing purposes and evaluate the necessity of remediation follow-up programmes.

Long term outdoor radon measurements across Ireland were recorded using CR-39 track etch detectors. For this study, a measurement protocol was designed that was optimised for the relatively low radon concentrations expected outdoors. This protocol included pre-etching the detectors before exposure to allow radon tracks to be more easily distinguished from background. The average outdoor radon concentration for the Republic of Ireland was found to be 5.6 $\pm$ 0.7 Bq/m^3 which equates to an annual dose of 0.05 mSv to the Irish population.

Prior to this study, a value of 6 Bq/m^3 was estimated to be the outdoor concentration for Ireland and this value was extrapolated from a Q-Q (quantile-quantile) plot of indoor log-normal radon concentration. The correlation of this theoretical value with the measured value from this study verifies that this method of extrapolating is correct, and should be considered to predict the outdoor concentration in other countries where indoor data exists.

A statistically significant difference between inland and coastal radon concentrations was evident but no difference between mean radon concentrations on the east coast and those on the west coast was observed.

Currently in Ireland radon testing is not a requirement for conveyancing or for dwelling rental and one perceived obstacle for its inclusion is the recommended 3 month measurement period. A comparison study was therefore conducted of seven different types of radon detectors (Atmos 12 dpx, RAD7, RStone, Sun Nuclear 1028, Ramon 2.2, Canary and CR-39) for exposure periods of 1, 2, 3 and 4 weeks. The comparison

was performed under two conditions: (a) in a purpose-built radon chamber with an average radon concentration of 2,560 Bq/m^3 (b) in a home environment with a radon concentration of 57 Bq/m^3, in both cases measured by the reference detector (Atmos 12 dpx) with a ±10% uncertainty range. In (a) 5 out of 8 detectors recorded radon concentrations within the Atmos uncertainty range and all detectors recorded within ±15%; in (b) 3 out of 9 detectors recorded within the Atmos uncertainty range and 6 out of 9 measured within ±20%, for a 4 week measurement. The results from this study show that radon surveys can be conducted for shorter periods than the recommended 3 months where a rapid indication is needed of whether the radon concentration is above the reference level, such as when assessing the concentration during and after remediation work.

In Ireland, during the years 1998 to 2004 the Department of Education and Skills (DES), with the assistance and guidance of the Environmental Protection Agency - Office of Radiological Protection (EPA-ORP), undertook the National Survey of Radon in Irish Schools. This was a large scale project to measure the radon levels in primary and post-primary schools in Ireland. Rooms which were found to have a concentration greater than 200 Bq/m^3 had some form of remediation installed to reduce the radon concentration. To determine the durability of the remediation methods and study the importance of structured follow-up programs, radon re-tests were performed in 16 of the schools that had some form of remediation implemented. The results showed that 26% of the rooms had risen to above the reference level of 200 Bq/m^3 again and 10% to above 400 Bq/m^3 , leading to the prediction that currently 790 rooms are $>$ 200 Bq/m^3 and 80 rooms are $>$ 400 Bq/m^3 in the 3,826 schools originally tested.

Through case studies and a questionnaire, the radon awareness in schools was observed to be correlated to an ability to maintain radon concentrations below the reference level. Only 15% of the schools in this study regularly checked or serviced their remediation systems. Testing for radon is crucial in indicating where remediation methods are less effective or have failed.

PUBLICATIONS

1. G. A. Gunning, D. Pollard and E. C. Finch. An outdoor radon survey and minimizing the uncertainties in low level measurements using CR-39 detectors. *Journal of Radiological Protection*, 34(2):457–467, 2014.

2. G. A. Gunning, M. Murray, S. C. Long *et al.* Inter-comparison of radon detectors for one to four week measurement periods. *Journal of Radiological Protection,* 36(1):104–116, 2016.

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1

MOTIVATION AND THESIS OUTLINE

1.1 THESIS MOTIVATION

Radon gas is sometimes referred to as the 'Silent Killer' and although it is a dramatic slogan aimed at gaining the public's attention, it is true. Radon is a naturally occurring gas which is exhaled at varying rates from the rocks and soil. As it is colourless, odourless and tasteless, its presence can only be detected by dedicated monitors. In the outdoor environment its concentrations are influenced mainly through climatological parameters, such as wind, rain, pressure and temperature, but in the built environment it can accumulate to dangerously high concentrations, which over long exposure periods can significantly increase the risk of lung cancer.

The link between radon and lung cancer was suspected in the early 20th century and radon was subsequently confirmed as a carcinogen in 1988 by the International Agency for Research on Cancer (IARC), the cancer research institute for the World Health Organization (WHO) [1]. Since then many journal publications, reports, conferences, working groups and regulatory bodies have been dedicated to contributing to and expanding the knowledge of this radioactive gas. These contributions bring awareness and conclusions to what is an ongoing battle to reduce the radiation dose associated with radon. It is estimated that the worldwide mean annual dose to the general population, due to radon, is approximately 40-50% of the total mean annual dose. Sources such as cosmic radiation, thoron, medical, food and gamma radiation, contribute to the remainder of the dose.

Radon is the second highest cause of lung cancer and in Ireland it is estimated that up to 250 lung cancer deaths per year are linked to radon [2, 3]. Also, the mean indoor radon concentration for Ireland is high (estimated value of 89 Bq/m^3) in comparison with the mean worldwide indoor concentration which is estimated to be 40 Bq/m^3 [4, 5].

The National Radon Survey predicts that approximately 7% of homes in Ireland are above 200 Bq/m^3; the reference level at which remedial work is recommended [6]. These and other findings are the result of continual research initiatives without which we would not know the scope of the radon problem in Ireland.

This thesis aims to build on the current knowledge of radon, with particular reference to Ireland but with relevance to all countries who are reviewing their current radon programmes or implementing new radon programmes. By continually contributing to radon research, it is hoped to raise awareness and reduce the number of lung cancer deaths due to radon.

1.2 THESIS OUTLINE

Chapter 2

Chapter 2 will introduce the topic of radon. It will give some background information on what radon is, how it was discovered and why it is a cause for concern. Details will be outlined on how these health concerns are converted in to measurable risks and quantified doses, along with the different conversion methods being implemented by different organisations. The sources of radon will also be outlined and the factors that affect the concentration of radon in different environments.

The discussion will then concentrate on radon in Ireland, the current legislation and how this legislation is implemented by the Environmental Protection Agency - Office of Radiological Protection (EPA-ORP or hereafter simply the EPA) and other state organisations. The EPA are involved in many research collaborations, including the work in this thesis, and some of EPA's large radon projects are also summarised in this chapter.

Thoron is also briefly mentioned. It is not part of the research in this thesis but it has many similarities to radon and worthy of discussion, even if only to contrast with radon.

Chapters 3, 4 and 5 all have project specific introductions and reviews of the relevant literature.

Chapter 3

The main objective of Chapter 3 is to quantify the average outdoor radon concentration to which the Irish population are exposed; previous to this study an extrapolated figure from indoor data was used. Eighteen metrological sites dispersed around the Republic of Ireland were used to house CR-39 detectors for a measurement period of 12 months. CR-39 detectors are ordinarily used to measure indoor radon concentrations which are usually at least an order of magnitude greater than that expected outdoors. Therefore, the measuring protocol had to be examined and adapted to minimise the uncertainties in using CR-39 for the outdoor measurements.

The data for the 18 sites were then analysed to determine whether inland and coastal concentrations, and also east and west coast concentrations, were statistically different from each other.

Also, in order to investigate the dependence of outdoor radon concentration on climate, a RAD7 continuous radon monitor recorded outdoor radon concentrations every 6 hours over a 2 month period. Using hourly temperature, pressure and rainfall data from a nearby weather station, it was attempted to correlate the radon variations with these climatological parameters.

Chapter 4

In Chapter 4, seven different types of radon detectors were compared for exposure periods of 1, 2, 3 and 4 weeks. The comparison was conducted under two conditions: (a) in a purpose-built radon chamber with an average radon concentration of 2,560 Bq/m^3 (b) in a home environment with a radon concentration of 57 Bq/m^3. This study was completed in order to determine whether representative radon measurements could be carried out for shorter than the recommended 3 month measurement and to determine the accuracy of some commonly used radon monitors.

Shorter measurements would be advantageous in two situations: 1) during or following remediation work shorter measurements would be beneficial to indicate whether the remediation was successful or if further remediation is necessary 2) during the process of conveyancing where there is a time urgency factor to complete the transaction. Con-

veyancing is the process of legally transferring property from one person to another. Although there is currently no requirement to conduct radon measurements for conveyancing in Ireland, it is expected that radon queries will be included in the conveyancing process in accordance with the government's National Radon Control Strategy. A shorter measurement period would aid the inclusion of radon measurement in the future.

Chapter 5

Chapter 5 describes the final experimental project which re-visits 16 schools that had some form of remediation installed in rooms due to elevated radon concentrations (greater than 200 Bq/m^3). The initial survey and installation of remediation was completed 10-17 years prior to this study as part of the large scale 'National Survey of Radon in Irish Schools', which was conducted by the Department of Education and Skills (DES) and the EPA. The main aim of this project is to determine the durability of the remediation method and subsequently look at the value of structured follow-up programmes.

Through case studies and a questionnaire, the study also looks at the level of radon awareness in Irish schools and whether retrofitting has any effect on the radon concentrations.

Chapter 6

Chapter 6 summarises the main conclusions from the work detailed in this thesis and outlines recent progress in the research field. The thesis is concluded with some reflections.

2

INTRODUCTION

The current body of information and publications on the subject of radon is growing enormously; it is a well researched area with many conclusive results but, as with many areas of knowledge, some inconclusive. With this wealth of radon data and reviews, the world of science is attempting to raise awareness of radon with the objective of reducing the associated radiation dose. The objectives of this chapter are to give an explanation of what radon is, a background account of the discovery of radon, the associated health effects and radiation dose, together with an overview of the sources of radon and a comparison of radon with thoron.

2.1 RADON

Radon-222, commonly known as 'radon', is a radioactive noble gas emitted in the decay of radium-226, a member of the uranium-238 decay series. With a density of 9.7 kg/m^3 it is one of the densest gases and is approximately eight times denser that air (1.2 kg/m^3 at sea level). Uranium is ubiquitous in soils and rocks and consists of ~99.3% uranium-238 which has a half-life of 4.5 billion years; this means that radon will be in steady supply to our environment for essentially an indefinite period of time. Table 2.1 details the associated half-lives, decay type and the emitted energies of the U-238 progeny [7]. Radon has a half-life of 3.8 days and ultimately decays to the stable isotope lead-206, through the progeny illustrated in figure 2.1, Z is the atomic number and A is the mass number.

The abundance of radon in air is quantified as the number of disintegrations per second (becquerel) per unit volume (m^3) and the becquerel is named after the French Physicist, Henri Becquerel who discovered radioactivity in 1896. For an introduction to

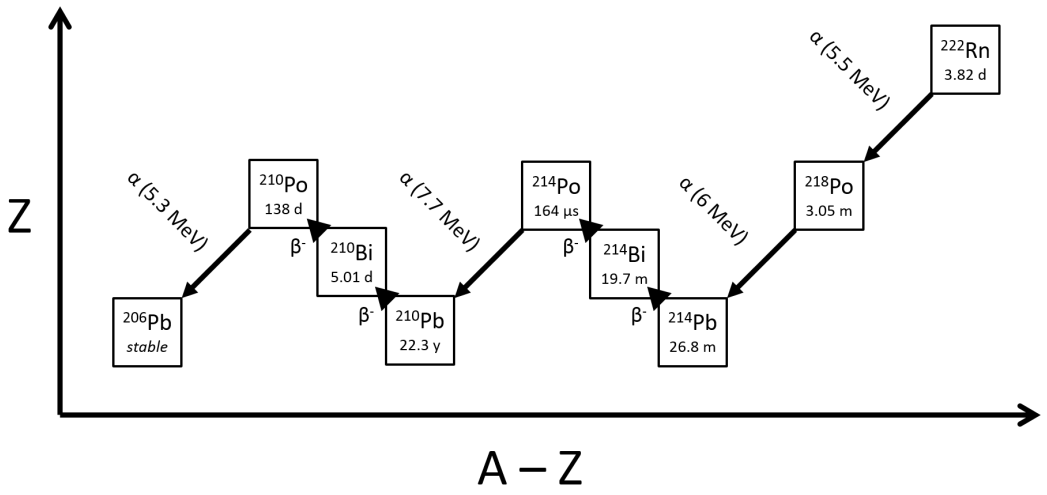


Figure 2.1: Radium-226 decay chain.

the concepts of radioactive decay and decay types, 'Modern Physics' by Kenneth Krane is recommended [8]

2.2 LITERATURE REVIEW

There are several good accounts and historic reviews of radon and its discovery [9–13]. Though it is accepted that radioactivity was discovered by Henri Becquerel and that Marie and Pierre Curie discovered radium in 1898, there are some conflicting accounts whether it was Ernest Rutherford or Friedrich Ernst Dorn who discovered radon gas. McLaughlin stated in his overview that Rutherford himself accredited Dorn with the discovery and this is evident in the associated reference [13, 14]. At the time, *circa* 1900, several investigators were conducting research with the mineral pitchblende and extracting the elements radium, thorium and actinium. Each element was found to exhale an associated gas which at the time, and for a long time afterwards, were referred to as radium emanations, thorium emanations and actinium emanations respectively but are now known as radon, thoron and actinon. In the subsequent years, radon had the most research emphasis placed on it. It has a longer half-life than thoron and actinon and is therefore more abundant in the environment, creating a larger health concern.

Radionuclide (historical name in parentheses)	Half-life	Energy (MeV)		
		Alpha[a]	Beta (max.)[b]	Gamma photons[b] (transition probability in parentheses)
$^{238}_{92}$U	4.47×10^9 a	4.20	—	—
$^{234}_{90}$Th	24.10 d	—	0.20, 0.11	0.093 (0.06), 0.063 (0.05)
$^{234m}_{91}$Pa	1.17 min	—	2.27 (99.84%)[c]	1.001 (0.008), 0.766 (0.003) Internal transition (0.16%)[c]
$^{234}_{92}$U	2.46×10^5 a	4.77	—	—
$^{230}_{90}$Th	7.54×10^4 a	4.69	—	0.068 (0.004)
$^{226}_{88}$Ra	1600 a	4.78	—	0.186 (0.036)
$^{222}_{86}$Rn	3.824 d	5.49	—	—
$^{218}_{84}$Po (RaA)	3.05 min	6.00 (99.98%) [c]	Energy not known (0.02%)[c]	—
$^{214}_{82}$Pb (RaB)	26.8 min	—	0.67, 0.73	0.352 (0.38), 0.295 (0.19), 0.242 (0.074)
$^{214}_{83}$Bi (RaC)	19.9 min	5.5 (0.02%)[c]	3.27, 1.54, 1.51 (99.98%)[c]	0.609 (0.46), 1.764 (0.15) 1.120 (0.15)
$^{214}_{84}$Po (RaC′)	1.64×10^{-4} s	7.69	—	—
$^{210}_{82}$Pb (RaD)	22.3 a	—	0.017, 0.064	0.047 (0.043)
$^{210}_{83}$Bi (RaE)	5.01 d	—	1.16	—
$^{210}_{84}$Po (RaF)	138.4 d	5.30	—	—
$^{206}_{82}$Pb (RaG)	Stable	—	—	—

[a] Only the energy of the most intense alpha is listed.
[b] Only the most prominent gammas or betas are listed.
[c] Indicates branching — the percentage in parentheses is the proportional decay by the indicated mode.

Table 2.1: Radionuclides in the uranium-238 decay chain.

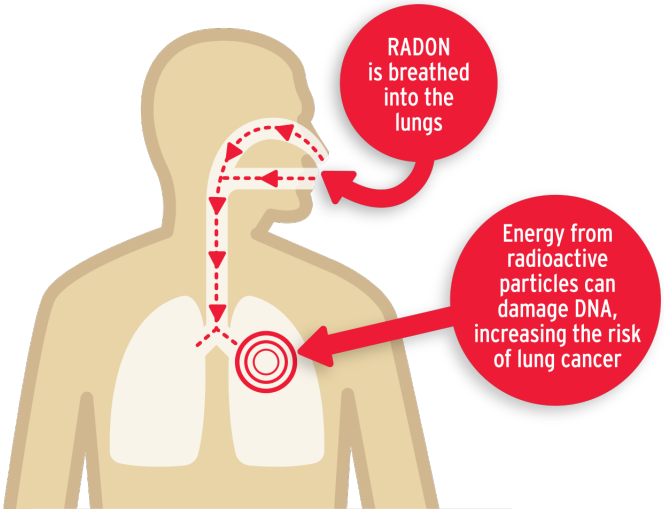


Figure 2.2: Radon entry to the body (image courtesy of the EPA).

It is of interest to note that there is also an Irish connection to this early period of radium advances. John Joly, an Irish geophysicist, carried out many experiments with radium. In connection with the work carried out in Chapter 3 of this thesis, he examined the exhalation rate of 'radium emanations' from the ground and he correlated changes in the rate with weather conditions [15]. More profoundly he utilised the medical benefits that could be achieved from radium and pioneered the 'Dublin method' in radiotherapy, whereby instead of using the expensive and dangerous radium to treat tumours, radium emanations i.e. radon was used. The radon gas was collected in glass capillary tubes which were inserted into or in close proximity to the tumour [16].This treatment method was used for subsequent decades.

Jacobi gives a concise and interesting historical review on how the link between radon and lung cancer was realised [10]. As early as the 16th century, unusually high occurrences of lung disease in underground miners were observed and reported by Paracelsus in his book *Uber die Bergsucht und andere Bergkrankheiten* (About the 'Bergsucht' and other Miners' Diseases), but it wasn't until after the discovery of radium that assumptions were made on the connection between radon and lung cancer. In Schneeberg, Germany, radon measurements taken in mines in 1936 reported concentrations of 70-120 kBq/m^3 and this is where the name for the disease originated, Schneeberger Lungenkrankheit (Schneeberger Lung Disease) [10].

Following numerous epidemiology studies in the 1950s and 1960s of mine workers in the United States of America (USA), Germany and Czechoslovakia, the connection between radon and lung cancer in miners was confirmed, but it wasn't until the late 1970s that the health effects of radon in domestic indoor air were seriously considered [11]. In 1988, radon was first classified as a human carcinogen by the IARC, the cancer research institute for the WHO [1]. In the same year, Nero [17] published an overview of the increasing US available data and reported an annual arithmetic mean of 55 Bq/m^3 for US homes. The publication also estimated an associated 0.4% lifetime risk of lung cancer at this mean concentration.

The BEIR VI Report, '*The Health Effects of Exposure to Indoor Radon*', was published by the Committee on Biological Effects of Ionizing Radiation (BEIR) of the National Academy of Sciences (NAS), USA, in 1999. It is a very extensive report detailing the discovery of radon, compiling epidemiology studies and ultimately estimating the lung cancer risk associated with radon inhalation.

2.2.1 *Dose due to radon*

As previously indicated, radon gas decays by emitting alpha particles and because of this it has an associated dose. Radon is not an external hazard as alpha particles cannot penetrate the outer layer of skin, but since radon decay products are electrically charged, they readily become attached to particles in the air. When inhaled they can be deposited onto lung tissue. Also, some of the radon gas inhaled will stay in the lungs and then decay. Thus the radiation dose from radon gas is delivered primarily to the lungs and it is two of the decay products, polonium-218 and polonium-214, that are the main contributors to the radiation dose and not the radon gas itself [18]. In the decay of polonium-218 and polonium-214 high energy alpha particles are emitted with energies of 6.00 MeV and 7.69 MeV respectively, which can cause damage to the lung tissue cells and potentially cause cancer.

The gray, Gy, and sievert, Sv, are the SI units (Système International d'Unités) of measurement for doses due to radiation exposure. The gray is a measure of the absorbed

dose i.e. the amount of radiation energy deposited in a medium such as human tissue or organs (joule/kilogram). The sievert is a weighted measure which in addition takes account of the type of radiation, and therefore is applicable to all types of ionising radiation, see table 2.2 [19]. Conversion of the radon concentration (Bq/m^3) into a dose (sievert, Sv) can be expressed in different ways depending on the derivation of the dose conversion. Epidemiology studies use previous studies of population exposures to radon and observe the subsequent health effects. Data can then be extrapolated to estimate the dose received in order to see these health effects. Another approach is a dosimetric approach whereby the absorbed dose is derived through analysis of the aerosol characteristics (unattached fraction and size distribution), the breathing rate, mucous clearance rate and the target cells.

The International Commission on Radiological Protection (ICRP) derived its conversion using epidemiology studies and stated a 'risk coefficient' of 2.83 x 10^{-4} per WLM (working level month, see table 2.2) [20]. This risk coefficient is the risk of lung cancer per unit exposure or per unit dose to the lungs. In a more recent publication, the ICRP revised this risk coefficient and raised it to 5 x 10^{-4} per WLM, almost double the previous value [21]. The revision was based on a review of epidemiology studies of miners and pooled residential studies in Europe, China and North America [22–24]. In the same publication, the ICRP stated that they intend to publish, in the near future, a dose coefficient for radon based on biokinetics and dosimetric models, and at a recent ICRP meeting a figure of 12 mSv per WLM was indicated [25]. This would mean that for the particular case of a dwelling, with an exposure time, t, of 7000 hours per year (80% occupancy) and equilibrium factor of 0.4 (for indoors, see table 2.2), a radon concentration, R_n, of 200 Bq/m^3 would result in an annual effective dose of 10.4 mSv.

UNSCEAR (United Nations Scientific Committee of the Effects of Atomic Radiation), on the other hand, recommend a dose conversion factor, D_f, of 9 nSv per ($Bq.h/m^3$) for converting radon concentration to effective dose (as opposed to ICRP's model of reporting the equivalent dose risk to the lungs).

Based on the same exposure time and equilibrium factors used in the ICRP conversion, the UNSCEAR model equates a radon concentration of 200 Bq/m^3 in a dwelling to 5 mSv per year [26] using the following equation,

$$H_{eff} = R_n * E_i * t * D_f \tag{2.1}$$

where H_{eff} is the annual effective dose and$E_i$is the equilibrium factor for indoors.

The UNSCEAR report also summaries a number of major dosimetric studies of the lung dose due to radon and states that values for dose conversion factors range from 6 to 15 nSv per (Bq.h/m^3) from other epidemiology studies. Chen gives a useful explanation and comparison of the ICRP, UNSCEAR and other proposed dose conversion models [27]. For convenience, the concepts and dose conversions from the last two paragraphs are summarised in table 2.2.

Smoking is the leading cause of lung cancer and radon is the second. In the USA, it is estimated that 10 – 15% of all lung cancers are caused by radon and in Ireland up to 250 cases per year (13% of all lung cancer cases) are linked to radon [2, 3, 11]. Many studies have been conducted to estimate the risk of lung cancer due to residential radon exposure. Three studies, in Europe, North America and China, pooled data in order to create a larger cohort for reliable analysis [22, 24, 28]. The Europe, North America and China studies estimated an 8%, 10% and 13% increase of lung cancer per 100 Bq/m^3, respectively [21].

The European collaboration combined data from 13 studies and analysed individual data for 7,148 people with lung cancer and 14,208 people without lung cancer. Information including smoking habits, demographic, lifestyle and radon exposure for at least 15 years was required for candidate selection. Figure 2.3 is an illustration from the published analysis and shows the cumulative risk of death by lung cancer by age 75 due to long term exposure to a range of 'usual radon' concentrations, for smokers and lifelong non-smokers [28]. 'Usual radon' refers to the estimated long term average concentration as opposed to the measured radon concentration. The latter has uncertainties due to (a) differences in concentrations measured in the same dwelling but in different years and (b) some homes being measured indirectly. The areas of the circles in the figure are proportional to the numbers of controls with usual radon levels in ranges < 200, 200-399, 400-599, and ≥ 600 Bq/m^3. The publication states that the risk of death from lung cancer by the

age of 75 is 0.4%, 0.5%, 0.7% and 0.9% for a lifelong non-smoker and 10%, 12%, 16% and 22% for a smoker, for long term exposure to radon concentrations of 0, 100, 400 and 800 Bq/m^3, respectively.

The relationship between radon and lung cancer has been extensively researched and documented but exposure to radon may also have other health effects. By combining data from 11 studies of over 64,000 underground miners, Darby *et al.* investigated the risks of cancers other than lung cancer due to radon [29]. The publication observed increases in the mortality rate for cancers of the stomach, liver and for leukaemia but could not conclude that radon was the cause of these deaths.

Kendall *et al.* detailed the dose due to radon and it's decay product on several organs through ingestion, inhalation and deposition on the skin, making references to other studies in the same field [30]. Calculations of the dose to the skin, due to plate-out (deposition) of the radon progeny, were relatively high and it was noted that this could result in melanoma skin cancer. More epidemiology studies are needed to confirm this relationship though. The same publication also reported that the stomach receives the highest dose from ingesting radon. It calculated that by ingesting 600 l of water, with a radon content of 1000 Bq/l, an annual dose of 50.4 mSv is received. The publication recommends designing an epidemiology study to demonstrate the risk of stomach cancer due to drinking water with a high radon content but noted that it can be difficult to assemble a sufficient cohort.

Several studies have also been undertaken to state conclusively whether a link between expose to radon and childhood leukaemia exists [31–33], and there are other publications that summarise these finding in order to compare the conclusions [34]. Factors such as the long latency period of cancer and inability to determine the exact radon exposure of a sufficiently large cohort of children make it difficult to state with certainty that children are more susceptible to the health effects of radon. Currently it remains inconclusive.

Quantity	Definition	Unit	Value
Attached/Unattached fraction	Fraction of radon decay products attached/not attached to aerosols	Dimensionless	Measured [35]
Working level	Short-lived radon progeny in 1 litre of air resulting in 1.3x10^5 MeV of potential alpha energy	WL	1 WL = 1.3 x 10^5 MeV/m^3 = 2.08 x 10^{-5} J/m^3
Potential alpha energy	Total alpha energy emitted during the decay of an atom to a stable isotope	MeV	19.2 MeV (radon) 20.9 MeV (thoron) [36]
Joules	Unit of energy	J	1 J = 6.242 x 10^{12} MeV
Working level month	1 WL exposure for working month of 170 hrs	WLM	1 WLM = 3.54 x 10^{-3} Jh/m^3
Occupancy	Number of hours assumed occupied per year	hours/year	Dwelling: 7000 hrs/y Workplace : 2000 hrs/y
Equilibrium factor	a measure of equilibrium between radon and its short-lived decay products	Dimensionless	Indoor: 0.4 Outdoor: 0.6
Radon concentration in air	number of disintegrations per second per unit volume	Bq/m^3	1 Bq/m^3 = 4.4 x 10^{-3} WLM (at home) 1 Bq/m^3 = 1.26 x 10^{-3} WLM (at work)
Risk coefficient	Increase of risk per unit exposure/dose	per WLM / per Sv / per 100 Bq/m^3	5 x 10^{-4} per WLM (ICRP 115)[21]
Does coefficient/conversion factor	Conversion value that converts radon concentration to effective dose	Sv per WLM / Sv per (Bq/m^3 h)	ICRP 65: 4 mSv per WLM (ICRP proposed 12 mSv per WLM) UNSCEAR: 9 nSv per (Bq.h/m^3)
Absorbed dose	Energy, E, deposited in tissue by radiation	gray, Gy (or J/kg)	Absorbed dose, $D = E/M$, M is the mass of tissue
Equivalent dose	Dose related to single organ/tissue (absorbed dose weighted by harm of radiation type)	Sv	Equivalent dose, $H = D \times W_F$, W_F is the weighting factor (e.g. for γ and β, $W_F = 1$; for α, $W_F = 20$)
Effective dose	Dose related to whole body (sum of tissue-weighted effective doses)	Sv	Effective dose, $H_{eff} = \sum_T W_F H_T$, $H_T$is the equivalent dose for each organ

Table 2.2: Summary of units and conversion coefficients.

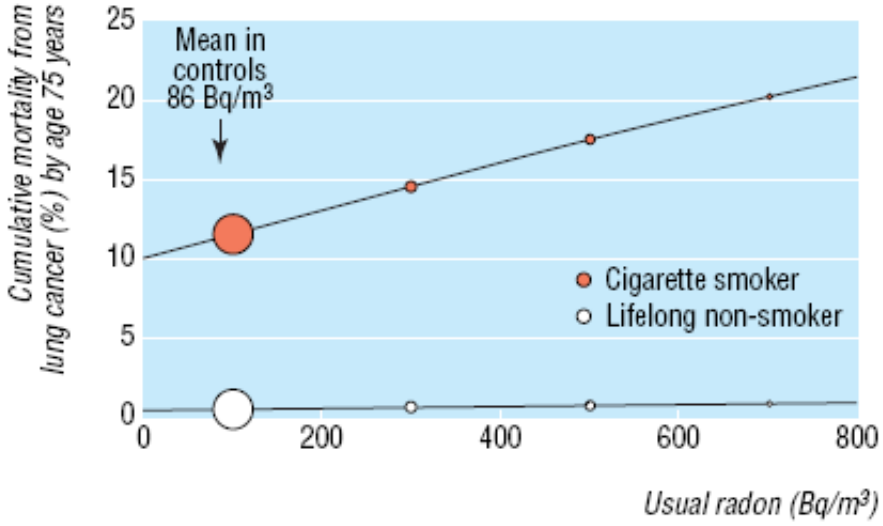


Figure 2.3: Cumulative risk of death from lung cancer by age 75.

2.2.2 *Sources of radon*

2.2.2.1 *Geology*

Naturally occurring uranium consists of the isotopes U-238 (~99.3%), U-235 (~ 0.7%) and a trace amount of U-234. It is U-238 that is relevant to this study as it produces radon gas in its decay series, see table 2.1. U-238 is found in the earth, soil and rocks underneath, and particular types of bedrock are associated with higher levels of uranium, for example granite, phosphatic rocks and organic rich shale. Rock types associated with lower uranium content are basalts, marine quartz sandstones, non-carbonaceous shale and siltstone, silica-poor metamorphic and igneous rocks, as listed by Gundersen [37].

Knowledge of local rock type and soil permeability can be exploited to map radon prone regions. Many countries, including Ireland, have employed a technique of measuring radioactivity using gamma spectrometers installed on low flying aeroplanes to record radioactivity emission from uranium-238 and radium-226 [38–42]. Often the data is combined with soil sampling and available indoor data. A study by Appelton *et al.* found that 25% of the total variation in indoor radon concentration could be explained by the mapped bedrock geology [43]. This method of radon prediction can be used to predict areas that are at higher risk to elevated radon concentrations due to their locality, but cannot give the relative risk for individual buildings because the spatial resolution is

not high enough and because it does not take into account the building type or the occupancy patterns.

2.2.2.2 *Outdoor environment*

The concentration of radon in the outdoor environment is fundamentally due to the underlying bedrock and the permeability of the soil and rock, but climatological factors such as wind, temperature, rainfall and air pressure also affect the rate of radon emission from the ground and subsequently the concentration in outdoor air. The reliance of outdoor radon concentration on climate will become more apparent in Chapter 3, where the mean outdoor radon concentration for Ireland was measured over a 12 month period and the climate conditions are correlated with a 2 month radon measurement.

The typical outdoor radon concentration is about 10 Bq/m^3 but this has been observed to range from 1 Bq/m^3 to over 100 Bq/m^3 [4]. This is relatively low in comparison to the figures that are reported for indoors and may be one of the reasons that country-wide research on outdoor levels is limited [44–46]. Measurements have concentrated on areas of suspected high radon concentrations such as mines and regions of volcanic or earthquake activity. One such study was conducted in Kobe, Japan and it found that ground-water radon concentrations increased fourfold nine months prior to and tenfold nine days prior to, relative to the initial concentration, an earthquake hitting the area [47]. Ghosh *et al.* document a comprehensive summary of a number of studies showing a similar phenomenon [48]. In a high number of these studies, emphasis is placed on measurements prior to any seismic activity with significantly less emphasis on post-seismic measurements. However, Einarsson *et al.* reported that radon concentrations returned to the pre-seismic concentrations 3 months after an earthquake in South Iceland [49].

2.2.2.3 *Indoor environment*

Radon gas concentrates in the built environment. If it enters buildings and accumulates, it can increase the risk of lung cancer to occupants. Radon can enter a house from the ground below through cracks in the foundation or walls, suspended floors or gaps around service pipes. This is why radon concentrations are higher in the basement and ground

floor than on upper floors (figure 2.4). Lack of sufficient air exchange, along with the relatively long 3.8 day half-life of radon, facilitates radon gas accumulation.

The factors affecting the outdoor radon concentration were previously listed as wind, temperature, rainfall and air pressure. The indoor environment, as well as being affect by the aforementioned, is also affected by the building type, building materials, age of building and ventilation. There is thus an accumulation in the number of factors that determine the concentration of radon in the indoor environment.

The temperature and air pressure difference between the inside and outside environment have the most significant affect on the indoor radon concentrations. In the cooler winter climate, buildings are heated and the air pressure inside is lower than outside causing radon in soil to be drawn in [26, 50]. Combined with the tendency for houses to be less well ventilated during winter, radon concentrations are higher compared with summer.

The UNSCEAR report of 2000, estimates the mean worldwide indoor concentration of radon to be 40 Bq/m^3 but it states that variations within and between countries can be significant [4]. For example, table 2.3 shows the mean indoor concentration for a selection of countries; the UK and Ireland have a similar geographic location and climate but their mean indoor concentrations are 20 $Bq/m^3$and 89 Bq/m^3respectively. Geology, building type, building materials and ventilation would contribute to this difference in means. Chambers and Zielinski compiled indoor radon data from a number of sources and produced a world indoor radon map, figure 2.5 [4, 51–53]. The map gives an insight to the mean radon concentrations in a number of countries and it is an indicator of the countries that are missing data, but it does not show the large variations in concentrations within small proximities.

Among the world's highest recorded indoor concentrations are 49,000 Bq/m^3 in Castleisland, Ireland [54], 50,000 Bq/m^3 in Norway [55] and 84,000 Bq/m^3 in Sweden [4], all in dwellings. Many countries have set an acceptable concentration of radon for different types of buildings known as a reference (or action level). Above this level, measured in Bq/m^3, some specified remedial action is recommended to reduce the exposure to the occupants. Different types of remediation methods are referred to in Chapter 4, where their effectiveness and durability are detailed.

Reference levels are set by individual countries on the advice of organisations such as the International Commission on Radiological Protection (ICRP) [20], the International Atomic Energy Agency (IAEA) [7] and the WHO [56]. The majority of countries with reference levels have set them in the range 200 - 400 Bq/m^3 for domestic dwellings and between 400 - 1000 Bq/m^3 for the workplace, see table 2.3 for the range of reference levels in countries around the world along with the estimated mean indoor concentration. Usually reference levels for the workplace are enforceable, and for domestic dwellings are advisable but some countries, for example Switzerland, Latvia and Ecuador, have set compulsory levels for dwellings, which require action. The WHO recommend setting reference levels as low as reasonably possible and suggest 100 Bq/m^3 or, depending on the country's specific conditions, should not to exceed 300 Bq/m^3. The organisation also makes the point that reference levels do not specify a rigid boundary between safety and danger, but define a level of risk from indoor radon that a country considers to be too high if it continues unchecked into the future [56]. An annual average of 300 Bq/m^3 in a domestic dwelling equates to an annual dose of 10 mSv based on the WHO conversion model but equates to an annual dose of 7.5 mSv based on the UNSCEAR mode [26, 56].

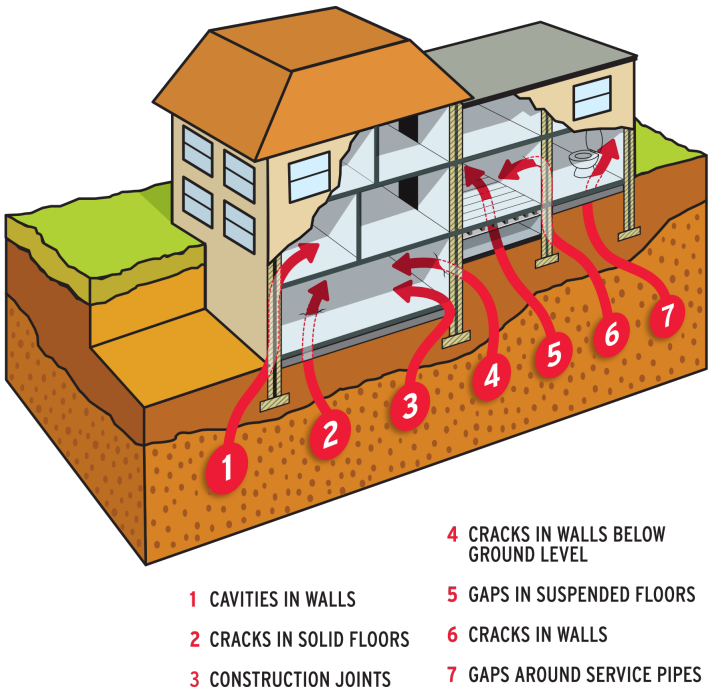


Figure 2.4: Radon entry to the indoors (image courtesy of the EPA).

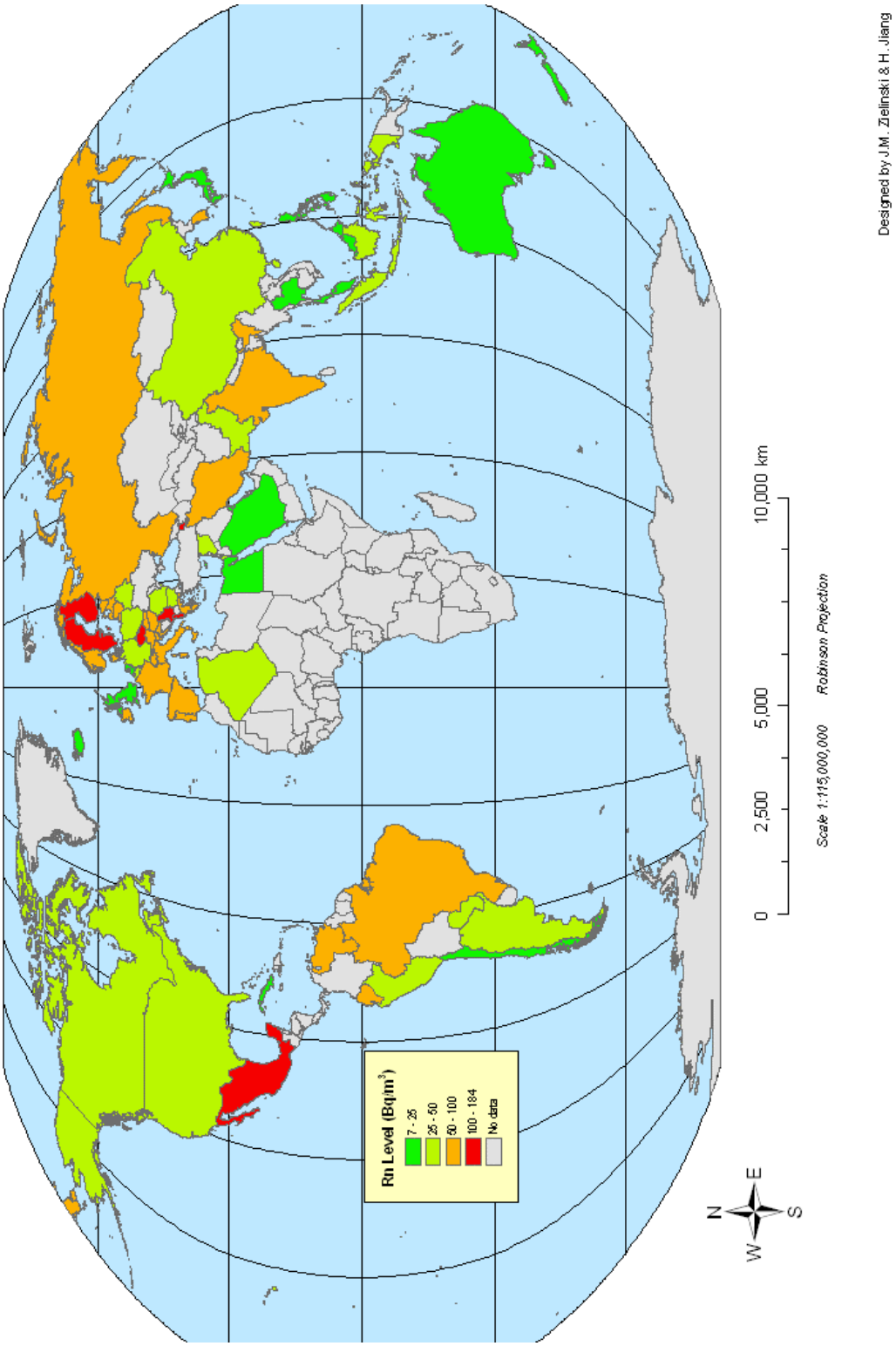


Figure 2.5: Mean indoor radon map (based on data up to 2007).

Table 2.3: Reference levels in countries around the world.

Country	**Dwelling reference level**	**Workplace reference level**	**Indoor estimated annual mean**	**References**
Argentina	200 Bq/m^3 (new build) 400 Bq/m^3(existing)	400 Bq/m^3	34 Bq/m^3	[51, 57, 58]
Austria	200 Bq/m^3 (new build) 400 Bq/m^3(existing)	200 Bq/m^3 (new build) 400 Bq/m^3(existing)	97 Bq/m^3	[51, 52, 57, 59]
Belgium	200 Bq/m^3 (new build) 400 Bq/m^3(existing)	800 kBq/m^{3}h/year.	48 Bq/m^3	[51, 57, 59]
Bulgaria	500 Bq/m^3	-	-	[51]
Canada	200 Bq/m^3	200 Bq/m^3	45 Bq/m^3	[51, 60]
China	400 Bq/m^3	1000 Bq/m^3	44 Bq/m^3	[51]
Croatia	-	-	68 Bq/m^3	[52]
Cyprus	-	-	19 Bq/m^3	[52]
Czech Republic	200 Bq/m^3 (new build) 400 Bq/m^3(existing)	400 Bq/m^3 (new build) 1000 Bq/m^3(existing)	140 Bq/m^3	[51, 52, 57, 59]
Denmark	200 Bq/m^3 (minor intervention) 400 Bq/m^3(robust intervention)	400 Bq/m^3	53 Bq/m^3	[51, 52, 59]
Ecuador	100 Bq/m^3(minimum) or 400 Bq/m^3 (compulsory)	-	94 Bq/m^3	[51, 61]
Estonia	200 Bq/m^3 (new build) 400 Bq/m^3(existing)	1500 Bq/m^3	60 Bq/m^3	[52, 57]
Finland	200 Bq/m^3 (new build), 400 Bq/m^3(existing)	400 Bq/m^3	120 Bq/m^3	[51, 52, 59]

Country	Dwelling reference level	Workplace reference level	Indoor estimated annual mean	Reference
France	-	400 Bq/m^3 (public buildings)	63 Bq/m^3	[51, 52, 59]
Georgia	200 Bq/m^3	-	-	[51]
Germany	100 Bq/m^3	200 Bq/m^3(existing), 400 Bq/m^3(new build)	50 Bq/m^3	[51, 52, 59]
Greece	200 Bq/m^3 (new build), 400 Bq/m^3(existing)	200 Bq/m^3 (new build) 400 Bq/m^3(existing)	55 Bq/m^3	[51, 52, 57, 59]
Ireland	200 Bq/m^3	200 Bq/m^3 for schools or long stay institutions, 400 Bq/m^3 workplace	89 Bq/m^3	[2, 5]
Italy	No	400Bq/m^3	70 Bq/m^3	[52]
Japan	No	No	16 Bq/m^3	[51, 62]
Korea	-	-	62 Bq/m^3	[63]
Kyrgyzstan	$<$ 200 Bq/m^3	-	-	[51]
Latvia	200 Bq/m^3, 600Bq/m^3(compulsory)	1000 Bq/m^3	70 Bq/m^3	[51, 57]
Lithuania	400 Bq/m^3	400 Bq/m^3	32 Bq/m^3	[51, 52, 57]
Luxembourg	150 Bq/m^3	-	115 Bq/m^3	[52, 57]
Malta	-	-	40 Bq/m^3	[52]
Netherlands	-	-	23 Bq/m^3	[52]
Norway	200 Bq/m^3	200 Bq/m^3 (new build) 400 Bq/m^3(existing)	89 Bq/m^3	[51, 52, 57]
Peru	200-600 Bq/m^3	1000 Bq/m^3	-	[51]

Country	**Dwelling reference level**	**Workplace reference level**	**Indoor estimated annual mean**	**Reference**
Poland	400 Bq/m^3 (existing)	20 mSv per year (underground worker)	49 Bq/m^3	[52, 57, 59]
Romania	400 Bq/m^3	4 WLM (underground)	45 Bq/m^3	[51, 52, 59]
Russia	200 Bq/m^3 (simple mitigation) 400 Bq/m^3 (upper action level)	-	50-60 Bq/m^3	[51]
Slovakia	400 Bq/m^3(existing)	1000 Bq/m^3	108 Bq/m^3	[52, 57]
Slovenia	400 Bq/m^3	400 Bq/m^3 (schools and kindergartens), 1000 Bq/m^3(companies)	87 Bq/m^3	[51, 52]
Spain	200 Bq/m^3	-	90 Bq/m^3	[52, 59]
Sweden	200 Bq/m^3	400 Bq/m^3	108 Bq/m^3	[51, 52]
Switzerland	400 Bq/m^3, 1000 Bq/m^3 (compulsory)	3000 Bq/m^3	77 Bq/m^3	[51, 52, 59]
Turkey	400 Bq/m^3	-	56 Bq/m^3	[64]
UK	200 Bq/m^3	400 Bq/m^3	20 Bq/m^3	[51, 52]
USA	150 Bq/m^3	150 Bq/m^3	48 Bq/m^3	[51, 57, 65]

2.2.2.4 *Radon in drinking water*

Uranium is dispersible in water, and therefore can be evident in water sourced from lakes and rivers, and in higher concentrations from water that is sourced from wells and bore holes. The concentration of radon in water is measured in becquerels per litre (Bq/l), and liquid scintillation counting is the most common method for measuring radon in water. Exposure to radon in drinking water can be through ingestion where the stomach cells are exposed to increased levels of radiation and also through inhalation, as the radon is released from the water when heated or agitated, for example, boiling water or from a shower. A European Commission recommendation report published in 2001 states that an annual effective dose caused by a concentration of 1000 Bq/l is comparable to an indoor radon concentration of 200 Bq/m^3 [66]. The report also recommends setting 1000 Bq/l as the reference level whereby remedial action should be taken to lower the concentration, on radiological protection grounds. Ireland currently has set the reference level for public water supplies as 500 Bq/l and for private supplies as 1000 Bq/l but this will soon change to 500 Bq/l for both sources, due the introduction of a new drinking water directive [67].

2.2.2.5 *Radon from building materials*

Uranium, thorium and potassium have long-lived radioisotopes occurring naturally in the environment. Together with their decay products they are referred to as NORM, Naturally Occurring Radioactive Materials. Specifically these three isotopes, and their progeny, are referred to as terrestrial NORM as they are found in the Earth's crust. Cosmogenic NORM and TENORM (Technologically Enhanced Naturally Occurring Radioactive Material) are other classifications; the former refers to radioactivity found naturally in the Earth's atmosphere and the latter refers to NORM that has been processed, resulting in elevated levels of radioactivity.

Many building materials are taken from the ground and it is an obvious deduction that varying amounts of NORM will subsequently be transferred to the built environment. Alpha, beta and gamma radiation can then be emitted and the radioactive gases thoron and radon exhaled from the walls, floors and ceilings indoors. The radiation dose due to

NORM is usually less significant than the radon in air concentration. From a radiological protection view, many studies have been undertaken to determine the levels of exposure due to the quantities and concentration in different building materials [68, 69]. In the European Union, a guidance document states that doses exceeding 1 mSv/y should be taken into account. The document also contains typical activity concentrations of common building materials and other reference material [70]. Lee *et al.* conducted a study of NORM in building materials in Ireland and reported that the levels were unlikely to result in any significant radiation exposure [71].

2.2.3 *Radon in Ireland*

The Radiological Protection Act 2014 is the primary legislation governing the use of ionising radiation in Ireland. Statutory Instrument no. 125 of 2000 (S.I. 125/2000) sets out the obligations of employers, the protection of workers and members of the public in relation to ionising radiation [72, 73]. The EPA is the national authority in Ireland for regulating and implementing legislation for radiological protection. Formerly known as the Radiological Protection Institute of Ireland (RPII), the establishment was merged with the Irish Environmental Protection Agency to become the 5^{th} office of the EPA, the Office of Radiological Protection, in 2014. The EPA regulate the use of radiation sources, ensure Ireland's emergency preparedness, monitor air quality and provide advise and information to the public, stakeholders and Government.

Ireland has a history of radon problems. The first indoor measurements were conducted in 1983-1984 for a 12 week period using CR-39 detectors in 250 homes [74]. The results from this early study indicated a median value of 43 Bq/m^{3}with a maximum value of 700 Bq/m^3. Several year later, this preliminary study set in motion a large scale survey, the 'National Radon Survey', of over 11,000 houses, which found that the average indoor concentration was in fact 89 Bq/m^3 [6].

In a survey of 39 OECD (Organisation for Economic Co-operation and Development) countries, this indoor average of 89 Bq/m^3 appears as the eighth highest [51]. Also, two dwellings in Co. Kerry were identified as having radon concentrations of 49,000 Bq/m^3

and 37,000 Bq/m^3 which equate to annual doses of 1.2 Sv and 1.0 Sv, respectively [75]. Previous geological mapping of this area had identified a limestone bedrock overlain with Namurian shales. Limestone is a highly permeable rock and Namurian shale in some areas was found to have a high uranium content [42].

Because of these concerns, the EPA has an active involvement in radon research through participation in national, European and international initiatives. The EPA has representatives and advisers on many influential committees and working groups, and as a result they have many publications in all aspects of radon. The projects detailed in this thesis are a contribution to these collaborations and publications.

Two of the EPA's large scale radon projects are the 'National Survey of Radon in Irish Schools' and 'Radon in Dwellings: The National Radon Survey' [6, 76]. Between 1998 and 2004, the EPA measured 38,531 ground floor rooms in 3826 primary and post-primary schools in Ireland to assess the distribution of radon in Irish schools. The project was called the National Survey of Radon in Irish Schools (hereafter referred to as the National School Survey or the NSS) and the results are detailed and expanded on in Chapter 5.

The National Radon Survey entailed completing measurements in approximately 11,000 houses between 1992 and 1999, from which the average indoor radon concentration of 89 Bq/m^3 was deduced. The study also estimated that 7% (or currently 110,000) of dwellings in Ireland had concentrations in excess of the national reference level of 200 Bq/m^3. The data from the survey were also used to compile a radon prediction map for Ireland (figure 2.6). The map is split in to 10 km grids, with a five-band colour scheme representing the predicted percentage of dwellings with radon concentrations above the reference level of 200 Bq/m^3. The bands are set as: <1%, 1-5%, 5-10%, 10-20% and >20% and percentage bands >10% are defined as high radon areas. A small number of grids predicated as many as 50% of dwellings with radon concentrations > 200 Bq/m^3 [6].

As stated, the reference level for indoor radon in Ireland is 200 Bq/m^3 for a domestic dwelling and 400 Bq/m^3 for the workplace. The reference for a dwelling is advisory and so there is no legal enforcement capabilities but the reference level for a workplace is guided by legislation in the 'Safety, Health and Welfare at Work Act 2005' and S.I.

no. 125 of 2000. This Act requires employers to identify all hazards in their workplace, including radon, to assess the risk to health and safety from these hazards and to put in place measures to eliminate or reduce the risk. Also, indoor workplaces located in high radon areas (located at ground floor or basement level) must be measured for radon.

In a 2014 report, the EPA detailed the doses received by the Irish populations through the various exposure routes, and indicated that radon is the largest contributor (55.1%) to the total 4 mSv annual dose [75]. Figure 2.7 represents the distribution of the radiation dose to the Irish population. Ireland currently uses the UNSCEAR model for converting radon concentration to effective dose whereby the reference level of 200 Bq/m^3 equates to a dose of 5 mSv per year. If the new ICRP guidelines were adopted (see section 2.2.1) then the dose due to radon would be more than doubled (200 Bq/m^3 would result in a dose of 10.4 mSv) and the updated dose distribution would be as illustrated in figure 2.8. Note that the pie chart assumes no change in the dose conversion for thoron.

The National Radon Control Strategy was launched by government in 2014 with the aim to reduce the number of radon related lung cancer cases through preventions, reduction, awareness and regulation. The strategy set out 48 recommendations on how to achieve these aims, along with a timeline and this is being implemented by a cross-government co-ordination group [2].

2.3 THORON

Also included in figure 2.7 is the percentage of annual dose due to thoron, 8.7 % (0.35 mSv). The work detailed in this dissertation does not investigate thoron but because it is often mentioned in association with radon, a short synopsis of information will be given.

Thoron (Rn-220) is an isotope of radon (Rn-222). Like radon, it is also a naturally occurring noble gas but it is formed in the decay chain of thorium-232. Thoron has a half-life of 55.6 seconds and decays through the progeny listed in table 2.4 to the stable isotope lead-208. Unlike radon, which has long-lived progeny (Pb-210: 22.3 years, Bi-210:

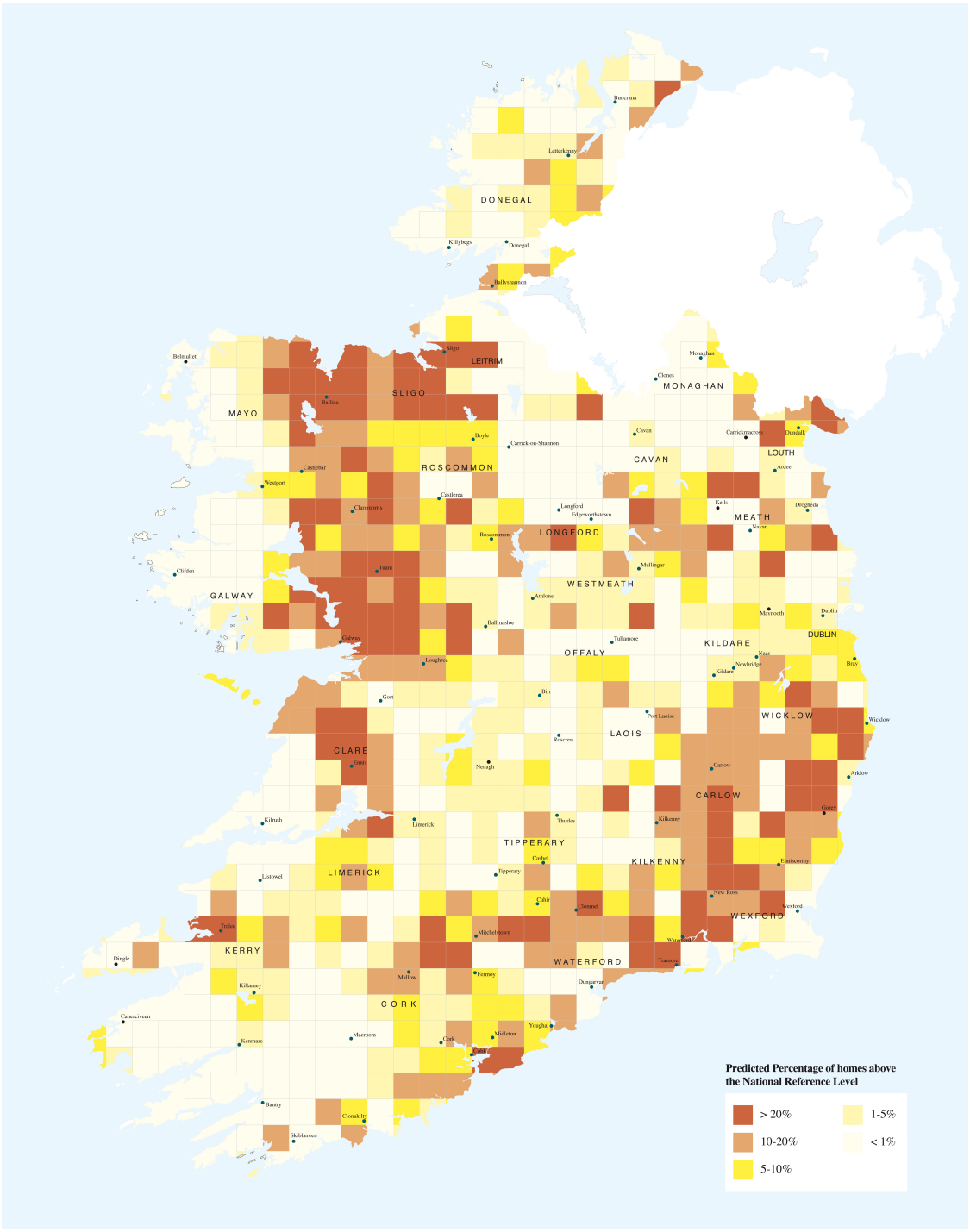


Figure 2.6: Radon map of Ireland (image courtesy of the EPA)

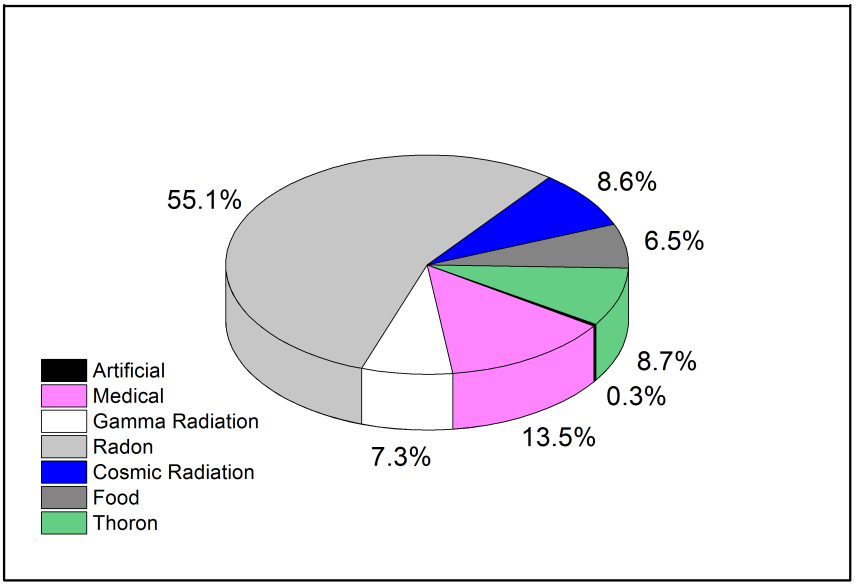


Figure 2.7: Distribution of dose to the Irish populations, 2014

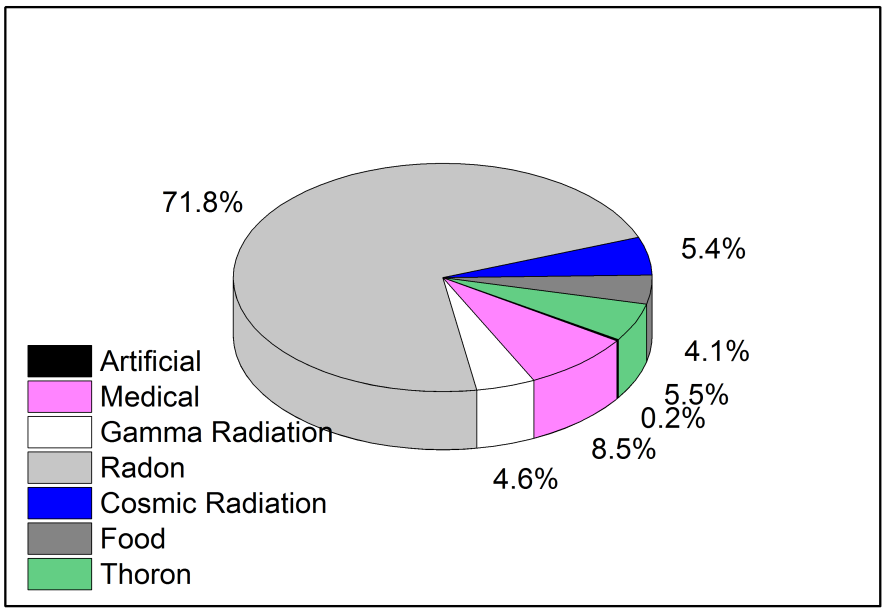


Figure 2.8: Distribution of dose to the Irish population as per proposed update to ICRP dose conversion

5 days and Po-210: 138 days), thoron progeny are short-lived with Pb-212 having the longest half-life of 10.6 hours [7].

Due to thoron's short half-life it migrates only a short distance from its source, with a diffusion length of typically 0.1 cm to 5 cm in building materials, and Urosevic *et al.* stated that its concentration decreases exponentially from the source [36, 77]. Radon, in contrast, has a typical diffusion length of 0.5 cm to 100 cm in building material [36]. Due to radon's longer half-life, and with the aid of diffusion and air currents, it has a more homogeneous distribution in indoor air than thoron. The main source of thoron indoors is the building materials.

Nikezic *et al.* found that the concentration of thoron in indoor air is normally up to 10% of that of radon and this can cause radon detectors to overestimate the radon concentration [78]. Many manufacturers recommend placing the radon detector at least 15 cm from the wall as this will reduce the contribution of thoron to the measurement.

The dose coefficient for thoron is 40 nSv per (Bq/m^3 h) which is over 4 times larger than the dose coefficient for radon (9 nSv per (Bq/m^3 h)), and therefore the dose due to exposure to thoron can not be neglected [79]. Pb-212, the decay product of thoron, has a half-life of 10.6 hours and even though thoron may be in small concentrations in indoor air, Pb-212 concentrations and its decay products can be significant. McLaughlin *et al.* conducted measurements of radon, thoron and thoron progeny in approximately 200 dwellings and found the arithmetic mean to be 78.5 Bq/m^3, 21.9 Bq/m^3 and 0.47 Bq/m^3 respectively [80]. Further analysis estimated the mean dose due to radon as 1.9 mSv/y and as 0.35 mSv/y due to thoron progeny. While radon contributes the majority of the annual dose, as expected, the contribution due to thoron is a substantial 18% of the reported radon dose.

For further reading, a review by Porstendörfer is recommended. The review gives a very comprehensive report of radon and thoron relative to each other, comparing many of their properties in relation to decay, emanation and transport in materials, deposition on surfaces, characterisation in outdoor and indoor air, and the dose and exposure relationship. [36].

Radionuclide (historical name in parentheses)	Half-life	Energy (MeV)		
		Alpha[a]	Beta (max.)[b]	Gamma photons[b] (transition probability in parentheses)
$^{232}_{90}$Th	1.405×10^{10} a	4.01	—	0.064 (0.0026)
$^{228}_{88}$Ra	5.75 a	—	0.039	0.013 (0.02)
$^{228}_{89}$Ac	6.15 h	—	1.16, 1.73	0.911 (0.26), 0.969 (0.16)
$^{228}_{90}$Th	1.91 a	5.42	—	0.084 (0.012)
$^{224}_{88}$Ra	3.66 d	5.69	—	0.241 (0.041)
$^{220}_{86}$Rn (Tn)	55.6 s	6.29	—	—
$^{216}_{84}$Po (ThA)	0.145 s	6.78	—	—
$^{212}_{82}$Pb (ThB)	10.64 h	—	0.34, 0.57	0.239 (0.43)
$^{212}_{83}$Bi (ThC)	60.55 min	6.05 (36%)[c]	2.25 (64%)[c]	0.727 (0.066)
$^{212}_{84}$Po (ThC′)	2.99×10^{-7} s	8.78	—	—
$^{208}_{81}$Tl (ThC″)	3.05 min	—	1.80, 1.29, 1.53	2.615 (0.36), 0.583 (0.30)
$^{208}_{82}$Pb (ThD)	Stable	—	—	—

[a] Only the energy of the most intense alpha is listed.

[b] Only the most prominent gammas or betas are listed.

[c] Indicates branching — the percentage in parentheses is the proportional decay by the indicated mode.

Table 2.4: Radionuclides in the thorium decay series

3

AN OUTDOOR RADON SURVEY

3.1 INTRODUCTION

The estimated population-weighted average outdoor radon concentration globally is 10 Bq/m^3 [79]. This is substantially lower than what we would expect to see indoors. This could explain why there is a wealth of world indoor radon data, but research on outdoor concentrations has focused on areas of suspected high radon concentrations, for example mines and regions of volcanic activity. Publications on country-wide radon surveys are limited, and usually either short term measurements in a number of locations or long term measurements in a single locations, are presented. Single location measurements have been completed: in Colorado, where a mean concentration of 18 Bq/m^3 over 2 years was found; outside Munich, Germany where a mean concentration of 8.6 Bq/m^3 over 10 years was found; in Milan, Italy where a mean concentration of 10 Bq/m^3 over 4 years was found; and in Bathinda, India where a mean concentration of 5.5 Bq/m^3 over 6 months was found [81–84]. These values are similar to the world average outdoor figure.

Although some outdoor studies limited the number of measurement locations (such as Milan, Botswana and Hong Kong), they were able, through the use of continuous radon monitors, to extend their analysis to look at: daily variations, seasonal variations, and the effect of meteorological conditions on radon concentration. Both Sesana *et al.* and Murty *et al.,* along with many other studies, observed daily variations with a trend of highest concentrations in the early morning and lowest in the afternoon [36, 79, 84–86]. This trend is due to air turbulence and a meteorological phenomenon known as a temperature inversion layer. During the day, solar heating warms the ground and sets up air turbulence enabling the radon to be readily transported upwards and dispersed away from the ground. During the night however, with no solar heating, the ground can rapidly cool and a layer of colder air is trapped under a layer of warm air; a

thermal inversion is set up. The warm layer acts as a cap, trapping radon and other air pollutants close to the ground.

Sesana *et al.* and Oikawa *et al.* took hourly measurements over longer periods, and hence they were both able to observe seasonal trends of higher concentrations in winter than in summer, as illustrated from the data of Sesana in figure 3.1 [46, 84]. Oikawa *et al.* reported lower concentrations in the summer months from July to September and higher concentrations from October to December; this was attributed to air pressure, with a high air pressure resulting in a lower radon concentration [46].

Wind, temperature, rainfall (soil water content) and air pressure all affect the rate of radon exhalation from the ground. The correlation between rainfall and radon concentration is not straightforward. Kullab *et al.* observed that radon decreased during rainfall and explained that this is due to the porosity of soil [87]. Porosity is a measure of the void space of a material, and for radon it is a particularly important factor which can enable or prohibit radon emanation though the soil or rock. Rainwater can fill this void area and thus decrease the route that radon can emanate to the atmosphere above. The opposite effect was been observed by Paatero, who attributed the increase in radon during rainfall to a phenomenon called 'radon washout'. This is where during heavy rainfall the radon is removed from the air by the falling rain and deposited on the ground below, resulting in higher radon concentrations at ground level [88, 89]. Winds and air currents move and disperse exhaled radon while temperature affects the air pressure. An area of high pressure air suppresses radon gas diffusing out of the ground due to a relative pressure gradient into the soil while an area of low pressure has the inverse effect [90].

Anomalies can arise with studies where the climate is temperate. In Cyprus, outdoor radon concentrations of 11 $\pm$ 10 Bq/m^3 were found to be higher than those indoors (7 $\pm$ 6 Bq/m^3) but only a total of 33 measurements (12 outdoor and 21 indoor measurements) of duration approximately 1 hour were taken [91]. Due to the large uncertainties and the fact that the measurements were completed in August when houses are well ventilated due to the hot climate, it can be deduced that the outdoor and indoor concentrations were in equilibrium. Kenawy *et al.* published a paper citing the range of radon measured in 3 cities in Egypt but failed to clearly differentiate between indoor and outdoor [92].

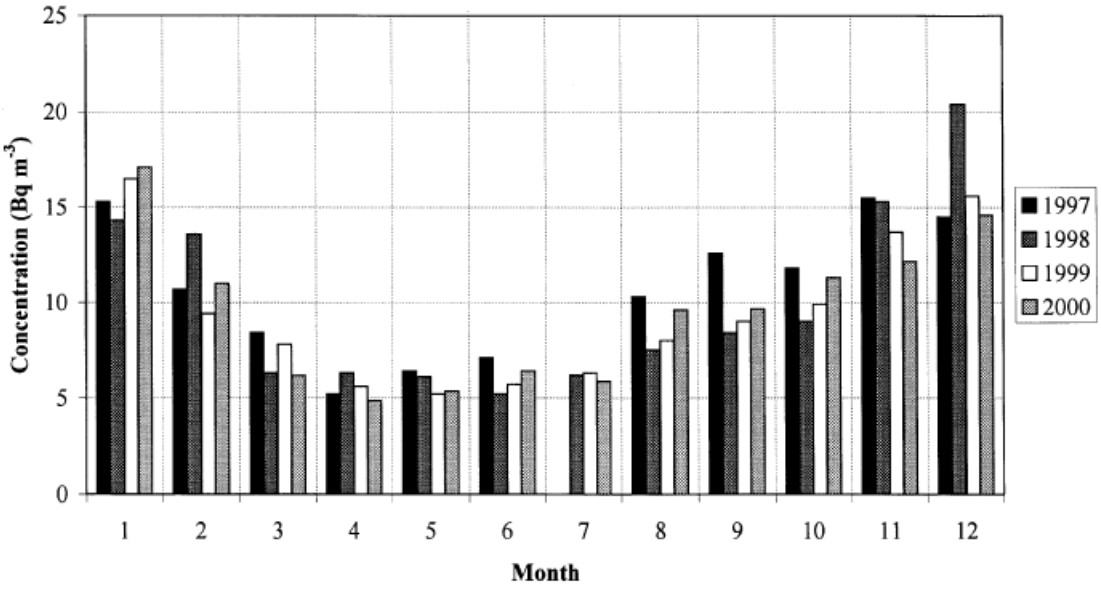


Figure 3.1: Average monthly radon concentrations for a 4 year period in Milan.

Three studies in particular are very good models for wide scale outdoor radon studies; these were conducted in North America, Japan and the Netherlands. Japan completed its outdoor study by taking measurements in 700 locations for 1 year and found the mean radon concentration to be 6.1 Bq/m^3. In combination with the average indoor radon concentration (15.5 Bq/m^3) the resultant effective dose to the general public was reported as 0.45 mSv/y [46].

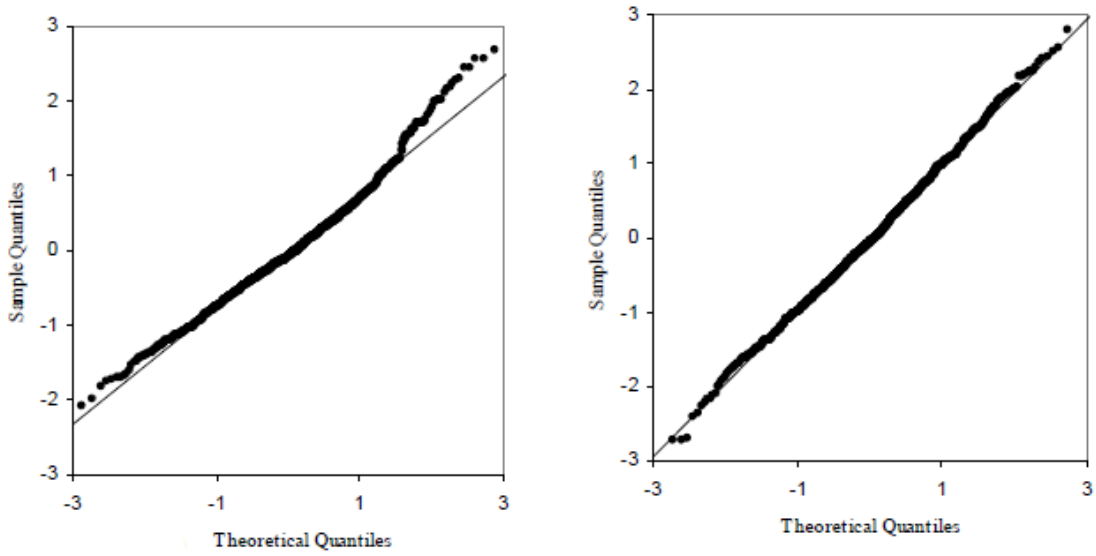
Another large scale study was completed in the Netherlands in 1985, where the radon concentration was measured at 200 locations [44]. Two detectors were *in situ* at each site; one remained for a 12 month period and the other was replaced at 6 months to gain an insight into climatological influence. The average radon concentration was found to be 3.7 Bq/m^3 and the concentration was 1.9 Bq/m^3 higher from July to January than from January to July. This was attributed to climatological parameters. Soil concentrations were also investigated by placing detectors in canisters just below the surface. The number of sample locations was not specified but concentrations 1000 times greater than those at 1.5 - 2 m were seen. This substantial difference is also reported in several other countries [52].

One of the most comprehensive environmental studies undertaken was part of a large-scale epidemiology study, the Iowa Radon Lung Cancer Study, which started in 1993 [45].

The exposure to outdoor radon was viewed as an important factor in the accumulation of dose to the participants. 111 locations in Iowa and 64 locations in Minnesota were monitored using track etch detectors at a height of 1.5 m and for a duration of 1 year. The study reported a geometric mean outdoor concentration of 25 Bq/m^3 [93]. The study also looked at other variables. Five sites had a repeat year-long measurement to see the variation from year to year, but no significant difference was seen (<15%). In Iowa, the monitoring sites were uniformly separated by 40 km, while in Minnesota small and long distances were sampled to look at spatial variations. Smaller variations were observed in the closer spaced detectors than the larger spaced detectors. At 4 sites, detectors were placed at 1 and 2 m to investigate variation of radon concentrations with height; no significant variation was seen.

In a survey of 39 OECD (Organisation for Economic Co-operation and Development) countries Ireland was found to have the eighth highest indoor average radon concentration [51]. Because of the high average indoor concentrations in Ireland, the work presented in this Chapter was undertaken in order to quantify the contribution made by the outdoor radon concentration and increase the accuracy of known exposure to the Irish population [54]. Apart from Joly's early exploratory work (section 2.2), the first Irish outdoor radon measurements were conducted in 1947-1948 by Burke and Nolan, where an ionisation chamber was used to measure the 'Radium A' (radon) content of air [94]. The results were reported as the number of radium A atoms per unit volume of air, and a normal monthly value of '10 Ra A atoms x 10^{-5}/cm^3' was noted. Observations on the dependence of meteorological conditions were also made, and wind was reported to have the most effect on decreasing the concentration. Further studies of outdoor radon in Ireland were conducted in 1966 by McLaughlin and Nolan [95]. In this study a photomultiplier tube was used, mainly at a location in Dublin, to count scintillations produced by radon decays. 24 hour measurements were taken over a four month period and the average radon concentration was found to be 12 x 10^{-14} Ci/l (4.4 Bq/m^3), with a minimum value of 3 x 10^{-14} Ci/l (1.1 Bq/m^3) and a maximum value of 86 x 10^{-14} Ci/l (31.8 Bq/m^3).

Other than these early measurements, no country wide outdoor study has been conducted in Ireland. A value of 6 Bq/m^3 has been estimated by extrapolation from the results of the indoor National Radon Survey on the basis that (R_I - R_O) is log-normally



(a) Q-Q plot of raw indoor radon concentrations.

(b) Q-Q plot corrected for theoretical 6 Bq/m^3 outdoor concentration.

Figure 3.2: Q-Q plots to determine baseline outdoor radon concentration

distributed, where R_I is the indoor radon concentration and R_O is the outdoor radon concentration [6, 96].

A quantile-quantile plot (Q-Q plot) is a probability plot used to test whether a dataset follows a given distribution. Indoor radon concentrations are known to follow a log-normal distribution and Fennell *et al.* noted that a Q-Q plot of a dataset from the National Radon Survey also followed this log-normal distribution, but was skewed at the higher and lower concentrations (figure 3.2a) [6]. Gunby *et al.* also noted this in their UK data and concluded that a baseline outdoor radon concentration was causing the deviations [97]. The Irish dataset was examined for this baseline correlation and when an outdoor concentration of 6 Bq/m^3 was subtracted from the data, the log-normal probability improved (figure 3.2b) [6]. Fennell *et al.* extrapolated that 6 Bq/m^3 was the average outdoor radon concentration for Ireland.

In this study, passive track etch detectors were used to make long term time averaged measurements of radon concentration. As stated previously, outdoor radon concentrations vary seasonally but not significantly from year-to-year. Therefore an exposure period of 12 months was chosen for this outdoor study as an average annual value would include the seasonal variations [93, 98]. In order to measure accurately the low radon concentrations

expected outdoors, the measurement protocol has been specifically optimised for outdoor conditions.

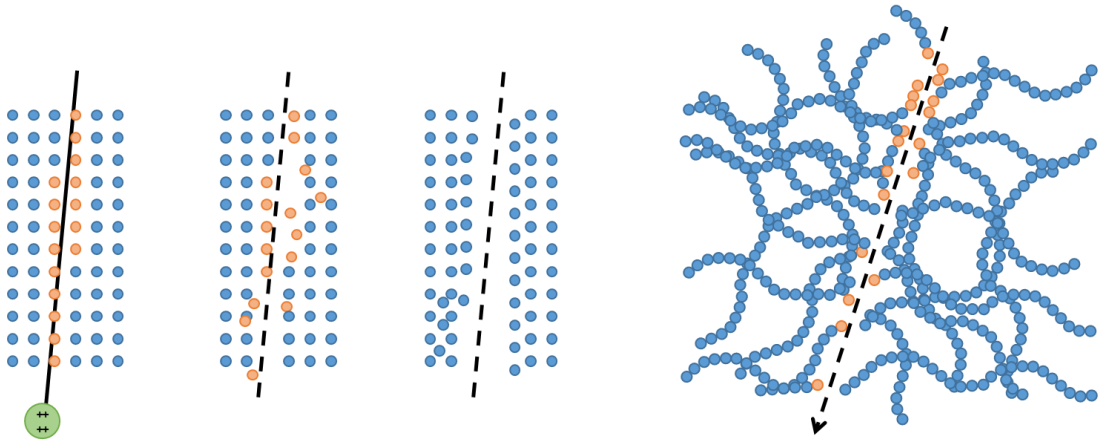
3.2 EXPERIMENTAL METHOD

3.2.1 *Track-etch detectors*

When a charged particle impinges on a dielectric material, a trail of continuous damage – typically on the nanoscale – is left along the trail of the particle. This trail is called a 'latent track'. These latent tracks were first observed in mica by Silk and Barnes [99], where the latent tracks were created by fission fragments. The width of the fission-induced tracks in mica was estimated to be in the order of 6 – 15 nm [100], far below the resolution of an optical microscope.

The mechanism of the radiation damage in dielectric materials depends on the nature of the material (figure 3.3). If the material is a crystalline structure, as shown in figure 3.3a, then the passage of the charged particle through the material will leave a trail of ionised sites. This ionisation is unstable and will result in ions being ejected into the solid away from the path of the incident particle. This migration will lead to the formation of vacancies and interstitials in the material surrounding the particle's path. The presences of these vacancies and interstitials stresses the material, which relaxes elastically by straining the surrounding material, thus leaving the path, observable on the nanoscale. Note that this type of latent track formation would not be formed in conducting or semi-conducting materials because both of these would be able to release the initial induced Columbic energy by charge transfer, rather that ion ejection. Also, the continuous path of damage along the particle's trajectory precludes a model of particle-atom collision, which would only show damage at the end of the particle's track.

If the material is an insulating polymer (figure 3.3b), the mechanism of damage is supplemented by polymer scission by secondary electrons. The secondary electrons are produced by the interaction of the impingent charged particle with atoms. Fast electrons



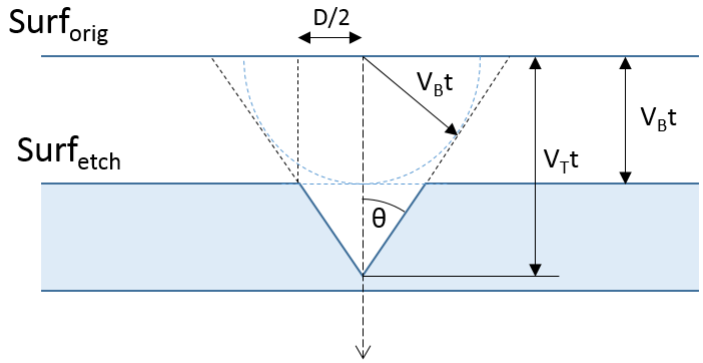
(a) Track formation - solid crystal (b) Track formation-plastic

Figure 3.3: Track formation

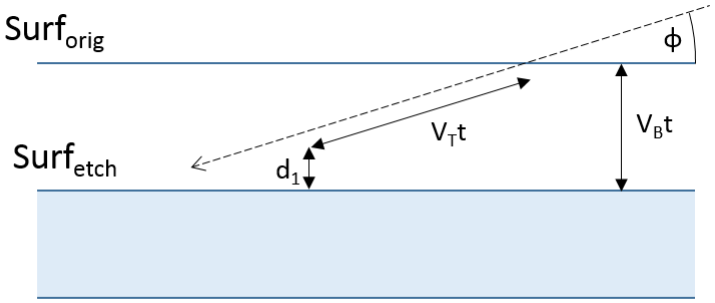
are ejected from the atoms, with enough energy to 'cut' the polymer chains into shorter chains. Furthermore, excitation and de-excitation can also directly lead to breakage of the long-chain polymers. All of these mechanisms can lead to damage in the polymer, forming a latent track.

In order for the latent track to be more easily observed, chemical etching can be used to make the track observable with a microscope. This process is dependent upon the property that the etch rate along a latent track, V_T, is greater than the etch rate of the bulk material, V_B. This is a reasonable assumption, given that the material along the track will be more readily etched due to the damage in the structure.

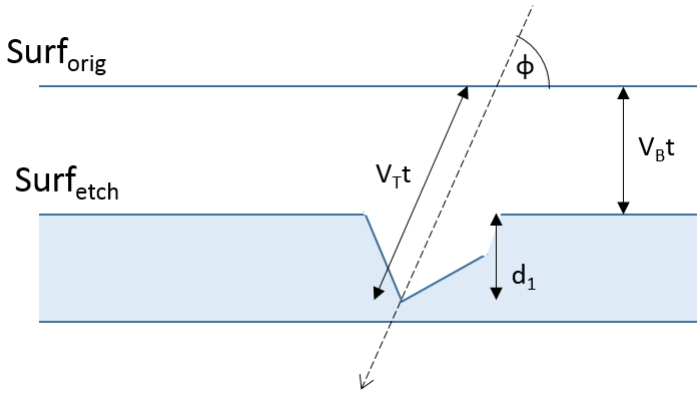
The formation of the optically visible track is described in figure 3.4a for a normally-incident particle. Upon etching, the surface will be etched at a rate of V_B, leading to a recession ($\text{Surf}_{\text{etch}}$) of the original surface ($\text{Surf}_{\text{orig}}$) by a distance of $V_B t$, where t is the etch duration. The track with have been etched by a rate of V_T, and thus a distance of $V_T t$ from the original surface. A cone will form around the track, given the linear variation of the combination bulk and track etch rates along the track. If a particle is incident at an angle, Φ, less than a critical angle ϑ, where $\theta = arcsin(V_B/V_T)$, then the normal component of etching along the track will be overtaken by the bulk rate, and thus no track will appear (figure 3.4b). Above the critical angle, the normal component of track etching will be faster than the bulk etch, and a track will appear (figure 3.4c).



(a) Geometry for the formation of an optically visible track.



(b) Track formation when the angle of incidence, Φ, is less that the critical angle.



(c) Track formation when the angle of incidence, Φ, is great that the critical angle.

Figure 3.4: Formation of optically visible tracks by chemical etching.

In this study CR-39 (Columbia Resin number 39) track etch detectors were used to detect radon. This is a polyallyl diglycol carbonate (PADC) plastic. The dimensions of the PADC plastic are 31 mm x 12 mm x 1 mm and a 6 digit number is engraved on each individual detector for identification. It is seated in a two-part polypropylene holder, which acts as a simple radon diffusion chamber (figure 3.5). This chamber excludes dust, radon daughter products and limits the access of moisture but allows the entry of radon gas. Alpha particles from decay of radon and its daughter products strike the plastic producing latent tracks. The track density can be related to the radon in air concentration, R_C (Bq/m^3), using the following equation,

$$R_C = 1000\left(\frac{C-B}{St}\right) \tag{3.1}$$

where C is the average track density count (tracks/cm^2), B is the background density count (tracks/cm^2), t is the exposure time (hours) and S is a sensitivity factor (tracks.m^3/cm^2.kBq.h). The sensitivity is defined as the average track density (corrected for background counts) per unit exposure and is derived by exposing a sample of detectors to a known radon concentration. Sheets of PADC can vary in their sensitivity, due to variability in the quality of the plastic.

The detectors arrive in pre-numbered heat sealed, radon proof aluminium bags. Each bag contains approximately 110 detectors which are cut from the same sheet of PADC plastic.

As radon and its decay products are electrically charged when formed, electrostatic charges inside the holder can affect where the solid decay products plate-out (get deposited) on the detector. The material of the black holders contains graphite and therefore acts as a conducting medium. However, there can still be a build-up of charge on the CR-39 and this can affect the distribution of alpha particles. Variations in track density in the region of 10-12% have been observed in different detectors exposed under the same conditions due to the interaction of charges [101, 102]. To alleviate this issue the detectors are washed in an anti-static surfactant (lipsol) on receipt from the supplier. Durrani *et al.* explains how this washing method creates a conducting layer on the

detector surface which allows charge to 'leak away' and also recommends washing the detector housing for the same reason [103].

Following exposure the detectors are chemically etched, for 8 hours in 6.25N Sodium Hydroxide (NaOH) at 75°C [6, 104, 105]. The track density is measured either manually by counting tracks in a known area under a microscope, or automatically using image analysis software.

An automated computer programme is ordinarily used to count the number of tracks on detectors. A digital camera takes a digital image of an area (0.5 cm^2) of the detector and Quantimet Image Analysis System (Qmet) is used to count the number of alpha tracks on the image, to calculate the track density. The Qmet identifies and accepts tracks based on the size and roundness.

Manual counting is carried out periodically for quality control and also to investigate high background detectors, when a detector is flawed, when automatic counting reports results $>$ 10,000 tracks/cm^2 and when a new set of control detectors is prepared for the Qmet. A Nikon Optiphot microscope (x 10 magnification) is used with a grid graticule fitted to define an area (0.2 cm^2), within which the number of tracks is counted. From this the track density (number of tracks per cm^2) is calculated, incorporating a correction factor for the graticule area. The objective lens is calibrated using a certified slide from Graticules Ltd., England, at the beginning of a study.

These measurement protocols for CR-39 radon track etch detectors were developed and optimised to measure the radon concentrations indoors by the EPA in accordance with 'ISO 17025' [106]. Accreditation is awarded by a national accreditation body which accredits in accordance with the relevant International Organisation for Standardisation (ISO) standards and guides. In Ireland the national body is the Irish National Accreditation Board (INAB) and for radon testing ISO 17025 for homes and workplace is awarded.

Indoor radon concentrations are typically at least an order of magnitude greater (the Irish reference level being 200 Bq/m^3) than that expected outdoors. As stated previously, the current accepted outdoor radon concentration in the Republic of Ireland is 6 Bq/m^3. When measuring such low radon concentrations the measurement uncertainties become crucial. In order to obtain a credible result, some investigation into minimising the

Figure 3.5: CR-39 detectors (image courtesy of the EPA).

uncertainties in using CR-39 was warranted. Sheets of PADC were examined to observe how the background count varies in each, to determine the accuracy of manual counting versus automatic counting and to investigate if longer etching times can 'etch-out' the background tracks.

3.2.2 *Development of an optimised measurement protocol*

To develop a measurement protocol which was optimised for the lower outdoor concentrations, the following issues were addressed:

- minimising the background track density on the unexposed CR-39 detectors
- the choice of method used to measure track density (manual or automated track counting)
- minimising the uncertainty in the background track density

During the production of CR-39 detectors a certain number of alpha tracks may be laid down; these are called background tracks. The detectors are manufactured and cut from sheets of PADC, and the background track density has been found to vary both within sheets and between sheets [107]. Furthermore the manufacturer (TASL Ltd., UK) states that the background track density is different for the top and bottom surface of a detector, so the top (identified by the 6 digit serial number) is always used when counting tracks. The background track density and the associated uncertainty ultimately

	8 hr etch	**16 hr etch**	**24 hr etch**
Manual counting			
ATD[1](tracks/cm^2)	12.4	20.0	27.0
Standard error in count, %	± 11.4	± 8.9	± 7.7
Limit of detection (tracks/cm^2)	41.9	47.6	67.4
Automatic counting			
ATD (tracks/cm^2)	33.9	43.3	64.8
Standard error in count, %	± 6.8	± 6.1	± 4.9
Limit of detection (tracks/cm^2)	79.3	79.3	137.9

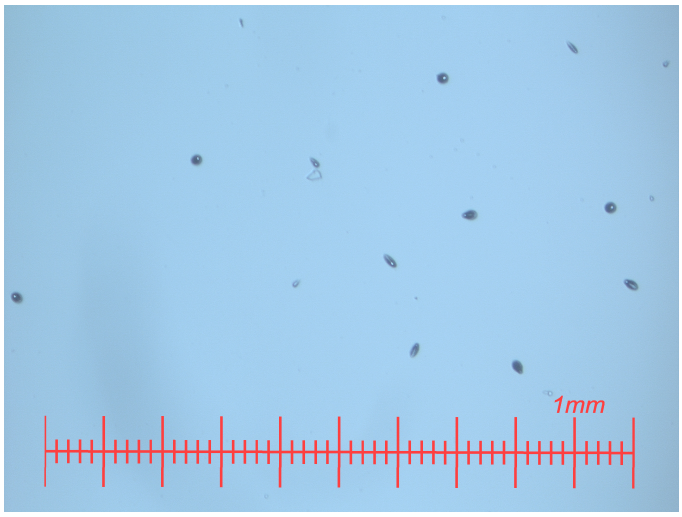
Table 3.1: A comparison of background counts as a function of counting method and etch time.

[1] Average track density

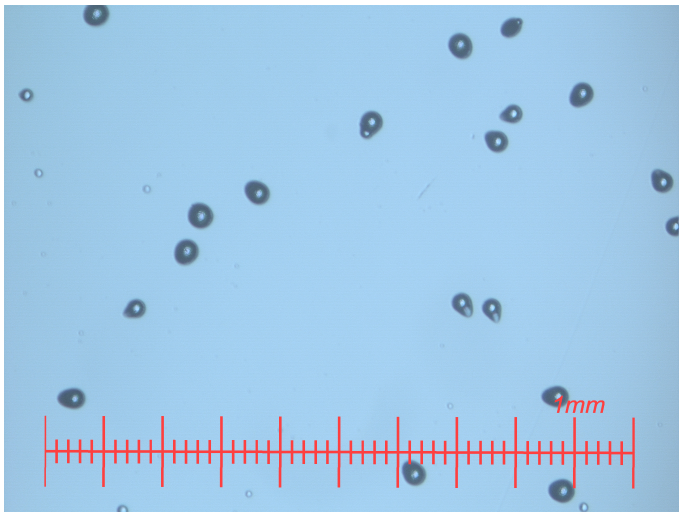
limit the lower detection limit of the detectors. Detectors with a low background count were requested for this project from the manufacturer. These detectors are manufactured in a low radon environment (<5 Bq/m^3) and only briefly exposed while they are being taken out of the oven in which they are cured. They are then wrapped up in radon-proof bags for storage prior to being sent to the customer [108].

The limit of detection (LoD) of a detector (CR-39 in this study) is the lowest quantity of a substance that can be recorded reliably or with a statistical certainty. It is typically deduced by measuring blank samples and calculating the LoD as the mean + (2 x standard deviation) [109]. In this study a more conservative statistical approach was taken and the LoD was calculated as the mean + (3 x standard deviation), corresponding to a 99.7% confidence in the calculation. To investigate the effect of pre-etching on the detection limit and determine if it is possible to 'etch-out' background tracks through a longer etch time, 30 detectors were etched in sets of 10 for 8 (single etch), 16 (double etch) and 24 (triple etch) hours. The detectors were then counted both automatically and manually to investigate which was the more appropriate method; the results are shown in table 3.1. Figure 3.6 depicts the change in alpha track size with increasing etch time.

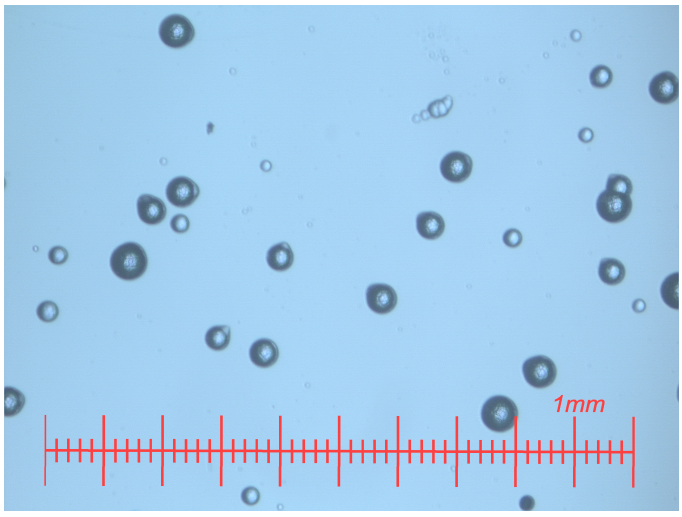
Manual counting for all etch times gave a lower background count. The larger background automatic count is due to aberrations in the plastic which the automated system counted as alpha tracks. The longer etch time did not etch-out background tracks but



(a) 8 hour etch.



(b) 16 hour etch.



(c) 24 hour etch.

Figure 3.6: Change in track size with etch time.

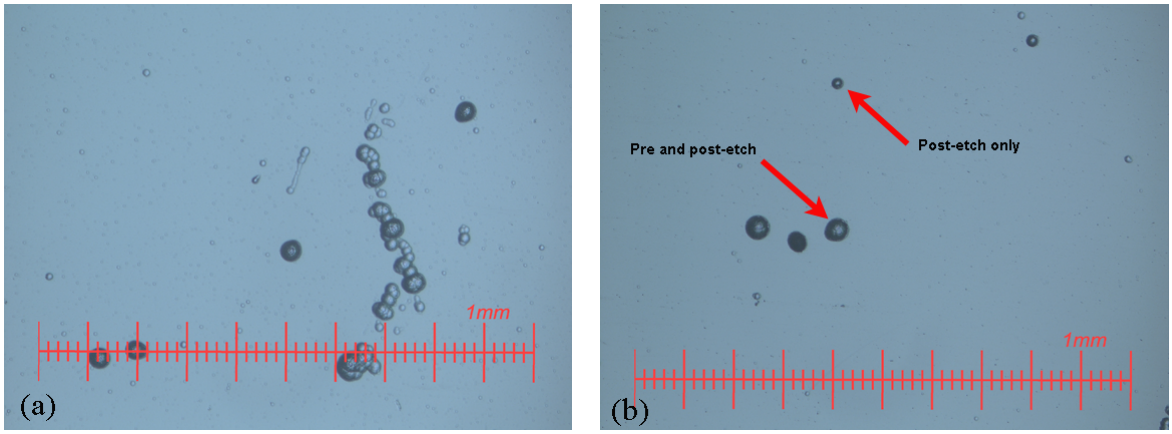
instead, along with enhancing alpha tracks, enhanced any aberrations which can be indistinguishable from true tracks (figure 3.8a). This gave rise to the increase in track density with etch time. The quoted standard errors in the counts are ultimately due to Poisson counting statistics. As can be seen from table 3.1, for all etch times the manual method gave a lower detection limit; hence it was decided to use the manual counting method for both the remainder of the uncertainty minimisation study and the outdoor study. Along with these 30, now pre-etched, detectors, an additional 30 un-etched detectors were exposed to a known radon concentration for 19 hours in a walk-in radon chamber.

This chamber consists of a purpose-built stainless steel, walk-in room filled with radon from a radium-226 source in which the radon concentration is about 2,500 Bq/m^3 (figure 3.7). The radon chamber is located at the EPA, Dublin, Ireland, and it is used to calibrate their CR-39 detectors and for research projects. It has a glass observation window and entry to the chamber is by an airlock two-door system. It can take one month for the radon concentration in the chamber to build up from zero; therefore the source is normally left unshielded and the radon gas is in equilibrium. The radon concentration in the chamber is monitored using an Atmos 12 dpx; this continuous monitor is the EPA's traceable reference detector and is detailed in Chapter 4.

An exposure time of 19 hours was chosen because an exposure to about 2,500 Bq/m^3 for 19 hours, equates to approximately the same track density (140 tracks/cm^2) that an exposure to 5 Bq/m^3 over 12 months would yield. 5 -10 Bq/m^3 was the expected outdoor radon concentration range. Following exposure, the previously etched 8, 16 and 24 hours sets of detectors were etched for a further 8, 16 and 24 hours respectively and the previously un-etched detectors were etched in groups of 10 for 8, 16 and 24 hours.

At the longer etch times of 16 hours and 24 hours, distinguishing between true alpha tracks and aberrations in the plastics became difficult (figure 3.8a). Also, as the longer etch time did not etch-out background tracks, an 8 hour pre-etch and an 8 hour post-etch time were used as the optimised conditions for this study. A pre-etch gave the advantage of easy distinction between background tracks and tracks laid down following the pre-etch (figure 3.8b) and, by counting the detectors after the pre-etch, a background concentration could be defined for each individual detector.

Figure 3.7: EPA radon chamber.



(a) Aberrations in a 24hr etch.

(b) 8 hour pre and post-etch.

Figure 3.8: Microscope images of alpha tracks following etching.

	8 hr etch	**16 hr etch**	**24 hr etch**
Pre & Post-etch			
Sensitivity (tracks.m^3/cm^2.kBq.h)	4.5	4.7	4.3
Post-etch			
Sensitivity (tracks.m^3/cm^2.kBq.h)	4.4	5.3	4.8

Table 3.2: Sensitivities for various etch times.

The effect of the longer etch time on the sensitivity factor was also investigated by exposing 6 sets of 5 detectors (3 sets pre-etched and 3 sets not) in the radon chamber for 1 week and the results are reported in table 3.2. The detectors are most sensitive for a 16 hour post-etch, however, the difference in sensitivity over an 8 hour etch was small and did not outweigh the advantage of the better specificity between tracks and background at the shorter 8 hour etch time.

3.2.3 *Fading and ageing*

The sensitivity of CR-39 detectors reduces with time due to fading and ageing. If the damage caused to the detector by the impingent charged particle, ie. the latent track, is repaired before any etching is performed, then the track will appear to have 'faded'. While the exact nature of the atomic processes that govern latent track repair is beyond the scope of this work, it has been observed that the latent tracks can be removed by annealing the detector [110]. Furthermore, the rate of fading can be modelled with the Boltzmann equation where t, the time needed for total track fading, is given by $t = A\,exp(E_{act}/kT)$, where k is Boltzmann's constant, E_{act} is the activation energy for track repair, T is the temperature and A is a constant. The determination of the activation energy can be used to approximate the exact atomic or molecular processes that control this repair. For further information on this topic please refer to 'Nuclear tracks in solids: principles and applications' by Fleischer *et al.* [100]. Ageing, the decrease in sensitivity due to storage in air before exposure to charged particles, has not to date been explained by a commonly-accepted theory.

As measurements for outdoor radon concentration would be taken over 12 months, the ageing and fading effects on the CR-39 detectors becomes important. Although many studies exist on the effects of ageing and fading on CR-39 detectors [110–112], here it was deemed necessary to quantify the ageing and fading effects specific to our detectors, due to the outdoor environmental conditions to which they would be subjected over the 12 months. This section details how the reductions to sensitivity of the detectors due to ageing and fading were calculated, and how the correction factors were applied to the outdoor radon measuring detectors.

For this study, 10 sets of 5 detectors were used. By exposing five sets together in a walk-in radon chamber (radon concentration ~ 2500 Bq/m^3) and storing them outdoors for 3, 6, 9 and 12 months (the fifth set, '0 months', was used as a control), the fading of exposure tracks with time was observed. Similarly, four additional sets were stored outdoors for 3, 6, 9 and 12 months (again a fifth set, '0 months', was used as a control) and then, exposed together in the radon chamber to analyse the change in sensitivity due to ageing of the detectors (figure 3.9).

During the interim months the detectors were stored at -30^{o}C to suspend the ageing and fading effects [111]. This facilitated post-etching all the detectors in one batch, so as to eliminate where possible batch-to-batch variations in the sensitivity. Miles demonstrated that a small change in the etch temperature can cause a substantial change in the sensitivity of the detector, 15% for 1^{o}C. This is more of an issue with an automatic counting system because it uses track size as one of the parameters to count tracks, but the procedure was included in this study as a cautionary approach [113] .

In a manner consistent with the treatment proposed for the outdoor measuring detectors, the ageing and fading detectors were pre-etched and the background tracks counted manually. To ensure the detectors did not accumulate any other radon tracks, apart from when exposed in the radon chamber, each set was heat sealed in radon-proof bags.

The quality of the radon-proof bags was tested by exposing five CR-39 detectors heat-sealed in a bag and five not in a bag, in the walk-in radon chamber. Before exposure, the detectors were first etched for 8 hours in 6.25N Sodium Hydroxide (NaOH) at 75 °C and manually counted for background tracks. Following exposure in the radon chamber

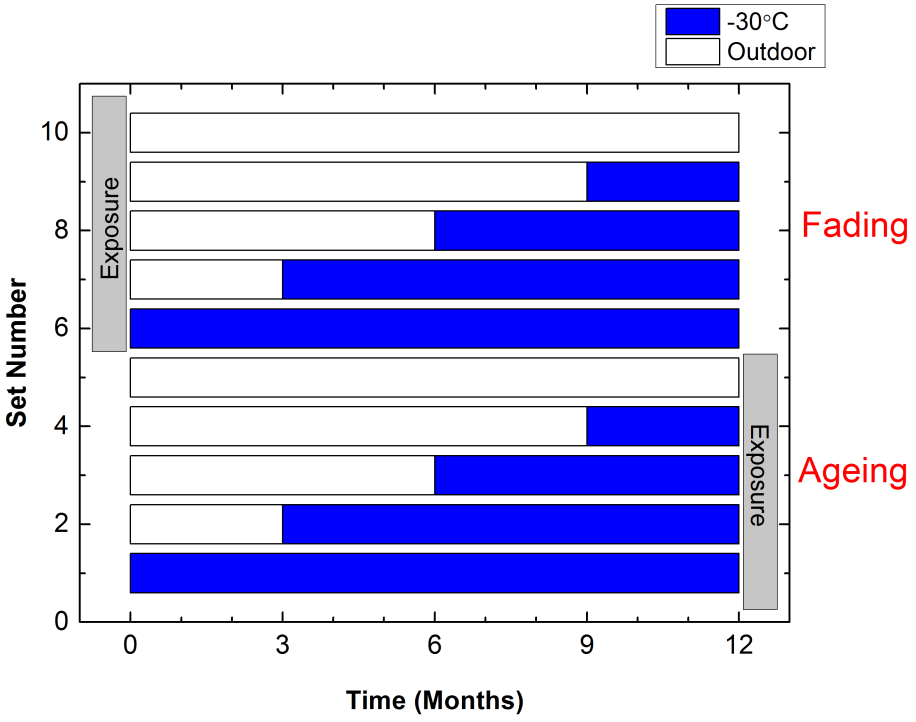


Figure 3.9: Ageing (sets 1-5) and fading (sets 6-10) schedule.

	Radon concentration before exposure, Bq/m³	**Radon concentration after exposure, Bq/m³**
Sealed detectors	59 ± 12	82 ± 12
Exposed detectors	80 ± 10	2480 ± 35

Table 3.3: Comparison of detectors sealed in a radon-proof bag and exposed detectors.

for 1 week, the detectors were then etched under the same conditions, manually counted and the results compared. Table 3.3 indicates that the bags appear to be resistant to radon gas entry to a satisfactory degree.

After 12 months, the fading and ageing detectors were removed from -30 °C storage. The fading detectors were immediately etched and the ageing detectors were exposed in the radon chamber, to a known radon concentration, for 7 days and then etched. All detectors were manually counted to find the track density and a sensitivity for each detector.

A linear least-squares fit was applied to the plot of normalised (to 1) radon sensitivity against the time exposed outdoor for the fading and ageing detectors (figures 3.10 and

3.11). The R^2 value reported in these figures is a measure of the linear relationship between the x-axis and y-axis, a value of 0 indicates no linear relationship and a value of 1 indicates a perfect correlation. From figure 3.10, no definite overall trend could be deduced for the fading data and a change in sensitivity due to fading was therefore not reported.

An ageing effect on the radon track sensitivity was evident from a similar plot for the ageing effect on detectors (figure 3.11). A slope of -0.0052 ± 0.0013 per month was reported. This equates to a decrease in sensitivity of 0.5% per month and can become significant when reporting on longer detection periods, as in this study.

To calculate the effect of ageing on the detectors it was assumed that the radon concentration was constant over the 12 months. The normalised sensitivity due to ageing, S_{ageing}, at a time t (months) is therefore

$$S_{ageing} = -0.0052t + 0.97 \tag{3.2}$$

Hardcastle and Miles integrated their corresponding result to give the average decrease in sensitivity over the 12 months. However, in this study there was no fading effect and as the ageing effect can be taken as linear, the number of months was simply multiplied by the monthly decrease factor (0.5%) [112] . After 12 months the sensitivity of the detectors had decreased to 94%. In applying this correction factor to the outdoor radon concentrations it was assumed the tracks were laid down uniformly over the 12 months.

3.2.4 *Survey of outdoor radon*

Outdoor radon was measured over a 12 month period (August 2008 - August 2009) at 18 locations. The detectors were placed in Stevenson screens (figure 3.12) located at meteorological observing stations operated by the Irish national weather service, Met Éireann. Stevenson screens are standardised white, louvred shelters used to house meteorology equipment. The screens shield against rain and direct heat radiation, from the sun or other sources, but allow air to circulate freely, making them ideal shelters for

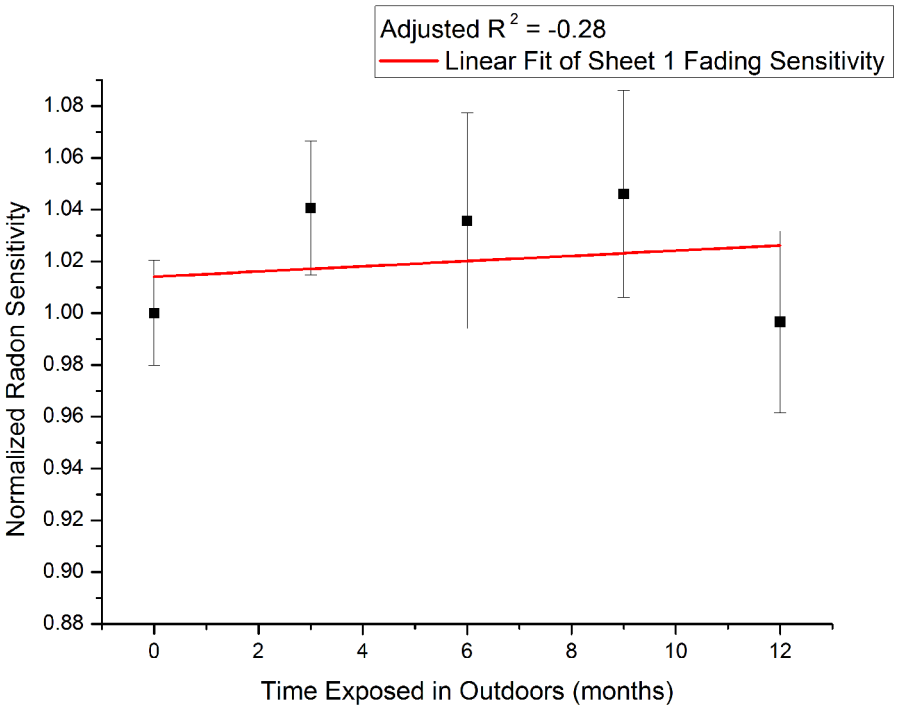


Figure 3.10: Outdoor fading effect on radon track sensitivity.

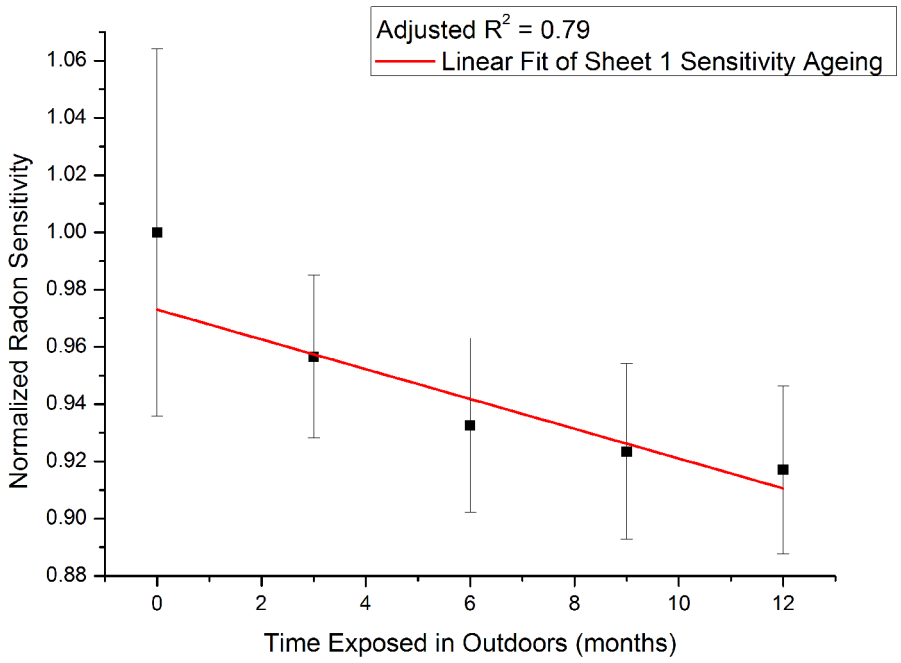


Figure 3.11: Outdoor ageing effect on radon track sensitivity.

Figure 3.12: An opened Stevenson screen with detectors *in situ*.

radon measurements. A set of five detectors was stationed at each site and were first etched and manually counted to determine the pre-existing background count on each, as per the developed optimised protocol.

Since the detectors were sent by post to each measurement site, radon-proof bags were used in the event of a delay in placing the detectors in the Stevenson screen.

Due to the time it takes to manually count detectors and, as there were over 170 detectors in total, the sites were divided into 3 batches and each was delivered by post a week apart. The same method (as below) was used for each batch. The sensitivity was calculated for each batch due to the 3 week length of the dispatch procedure. This was done by pre-etching and counting an extra set of 5 detectors with each of the 3 outdoor detector batches. The extra set was then exposed in the radon chamber, post-etched and counted to calculate the sensitivity for each batch.

After pre-etching, any tracks laid down on the detectors would be counted as outdoor tracks, and this could lead to an inaccurate outdoor radon concentration. To ensure minimal time between pre-etching the detectors, counting the background tracks and

posting to the site, precautions were taken during this period to minimise this risk. The detectors were stored in a charcoal chamber overnight to minimise the background tracks being laid down. Charcoal adsorbs radon in the air and so creates a low radon environment [114]. Also, the counting was carried out in a well-ventilated, first floor room to minimise background alpha tracks being laid down on the detector. Upper floor rooms have a lower radon concentration than ground floor rooms, mainly because radon emanates from the ground and disperses [87].

Five detectors were set together on a strip of nylon (as used in cable ties) and labelled with the intended site name and relevant contact details. The sets were heat-sealed in a radon-proof bag and, along with instructions, the detectors were posted to the sites. Two days later, each monitoring site was contacted to confirm delivery and retrieve the time placement details.

After 12 months, the detectors from the 18 outdoor sites were returned by post over a period of 12 days. As they were received they were sealed in radon-proof bags and stored at -30^{o}C until etched. The detectors were etched for 8 hours under the standard conditions and counted to gain the post-exposure track density. From the sensitivity calculated for each batch of detectors, the outdoor radon concentrations in air were calculated.

3.3 RESULTS AND ANALYSIS

3.3.1 *Outdoor radon concentrations*

Table 3.4 details the outdoor radon concentrations in the Republic of Ireland, before and after adjusting for ageing. The adjusted concentrations are also shown my location on the map in figure 3.13.

During long term studies, unexpected incidents can arise and impact on the result uncertainty. At one of the sites in this study (Ballyhaise), the time of removal of detectors was not noted, giving up to a 10 hour uncertainty in the exposure time of 358 days. This

equates to an uncertainty of approximately 0.1% which is negligible in comparison to the reported statistically uncertainty.

At another site (Malin Head), which is a particularly windy location, it was reported that the detectors fell out of the screen during a storm but were reinstated after approximately an hour. No unusual etching patterns or damage was seen on the detectors from this site.

The mean of the averaged measurements for each of the 18 sites is 5.6 Bq/m^3. This agrees well with the extrapolated value of 6 Bq/m^3. This was previously derived from the National Radon Survey data and is in line with values reported from other studies from comparable regions around the world. The standard error of the mean (SEM) for the 18 site concentrations is 0.29 Bq/m^3. To obtain a 99% confidence interval, the SEM was multiplied by a 'Z-score' value of 2.576; this gives a rounded value of 0.7 Bq/m^3. (The Z-score value for a stated confidence interval is listed in a 'Z-table' which is derived from a normal distribution.) Thus the national mean, with 99% confidence can be stated to be 5.6 $\pm$ 0.7 Bq/m^3.

Site location	**Radon conc., Bq/m^3**	**Standard error**	**Radon conc.,Bq/m^3 (corrected for ageing)**	**Standard error**
(1) Roches Pt., Co. Cork	4.0	± 0.3	**4.2**	**± 0.3**
(2) Sherkin Island, Co. Cork	4.1	± 0.7	**4.3**	**± 0.7**
(3) Malin Head, Co. Donegal	4.1	± 0.6	**4.4**	**± 0.7**
(4) Ballyhaise, Co. Cavan	4.2	± 0.9	**4.5**	**± 0.9**
(5) Mt Dillon, Co. Roscommon	4.3	± 0.6	**4.5**	**± 0.7**
(6) Belmullet, Co. Mayo	4.3	± 0.5	**4.6**	**± 0.5**
(7) Mace head, Co. Galway	4.4	± 0.6	**4.7**	**± 0.6**
(8) Valentia, Co. Kerry	4.5	± 1.2	**4.8**	**± 1.3**
(9) Newport, Co. Mayo	4.6	± 0.4	**4.9**	**± 0.4**
(10) Johnstown Castle, Co. Wexford	5.0	± 1.1	**5.3**	**± 1.0**
(11) Merrion Sq., Dublin	5.2	± 0.6	**5.5**	**± 0.7**
(12) Moore Park, Co. Cork	5.7	± 0.4	**6.0**	**± 0.4**
(13) Claremorris, Co. Mayo	6.2	± 1.0	**6.6**	**± 1.1**
(14) Dunsany, Co. Meath	6.4	± 0.5	**6.8**	**± 0.6**
(15) Mullingar, Co. Westmeath	6.5	± 1.0	**6.9**	**± 1.1**
(16) Casement, Co. Dublin	6.9	± 1.0	**7.2**	**± 1.1**
(17) Oak Park, Co. Carlow	7.1	± 0.3	**7.5**	**± 0.3**
(18) Gurteen, Co. Tipperary	7.3	± 0.9	**7.7**	**± 0.9**

Table 3.4: Outdoor radon concentrations in Ireland: the measured concentration and the concentration correct for the ageing effect on CR-39 detectors.

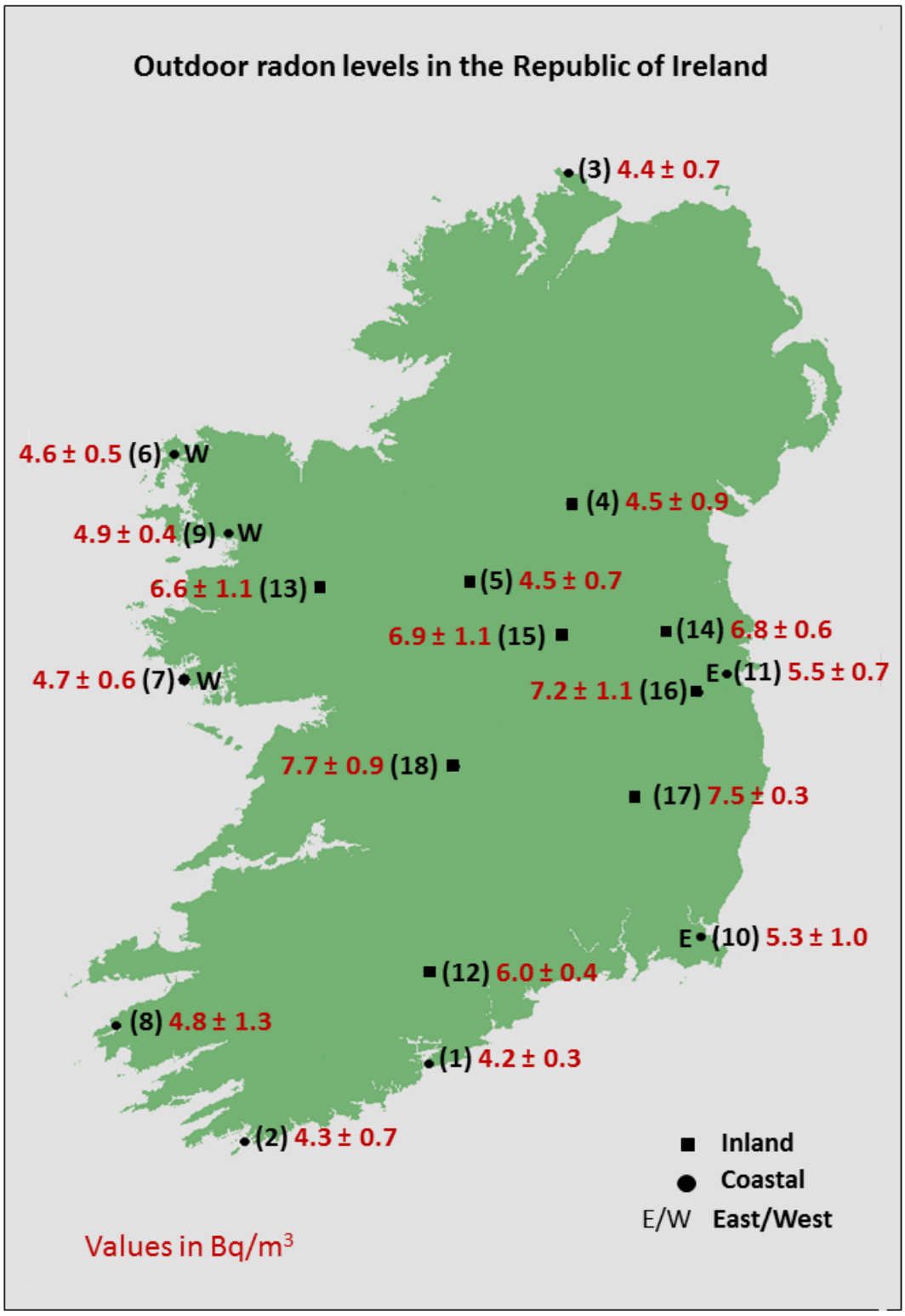


Figure 3.13: Outdoor radon concentrations in the Republic of Ireland.

3.3.2 *Variations in radon concentrations*

3.3.2.1 *Outdoor radon concentrations*

To study the diurnal variation of outdoor radon concentration, continuous measurements were conducted for 2 months at the EPA offices, Co. Dublin using a RAD7 radon monitor (see Chapter 4 for monitor details). The RAD7 recorded the radon concentration every 6 hours at a height of 1.2 m from the ground. For the period from mid July to mid September, a mean radon concentration of 3.7 ± 0.1 Bq/m^3 was reported, with a minimum and maximum concentrations of 0 Bq/m^3 and 26 Bq/m^3. The radon concentration was also recorded at the same site for 12 months using CR-39 detectors. The same optimised protocol was employed as in the outdoor survey and a concentration of 5.6 ± 0.7 Bq/m^3 was reported (it is purely coincidental that it is the same figure as the average outdoor figure for Ireland!). The difference between the 12 month measurement and the 2 month measurement can be explained by seasonal variation: a lower concentration in warmer, drier months is expected and has been reported in other studies with a similar climate to Ireland [84, 115]. The CR-39 detectors at this site were housed in a Stevenson screen but as this screen was set at ground level, the data were not used in calculating the annual average outdoor radon concentration for Ireland; the standard height of Met Éireann Stevenson screens is 1.2 m above ground level.

A nearby (7 km distance) weather station, Phoenix Park, recorded hourly rainfall, relative humidity and air temperature data for the same period and was provided to this study by Met Éireann. The outdoor radon concentration data with the associated climatology parameters for the same period are presented in figures 3.14, 3.15, 3.17 and 3.16.

Diurnal radon variations were observed with a maximum radon concentration occurring generally in the early morning and a minimum concentration occurring in the early evening, as per figure 3.14 and more clearly in figure 3.15. As explained in section 3.1, this cyclical variation is due to air turbulence dispersing the radon during the day and a thermal inversion layer trapping the radon close to the ground during the night. As the

radon concentrations were recorded only every 6 hours, the exact time of the diurnal maximum and minimum values may not be represented.

Figures 3.16 and 3.17 illustrate the variations in radon concentration with associated variations in rain and pressure respectively. The last week of the data illustrates larger variations in the radon concentration (up to a maximum of 26 Bq/m^3). For the same interval of data there is no recorded rainfall. During periods of little or no rain the porosity of the soil is high which facilitates increased exhalation of radon from the ground. This is evident at particular periods in figure 3.16: July 31st, August 7th and September 12th, where there is still diurnal variations but the maximum radon concentrations have increased. The effect of rising and falling pressure did not have as dramatic a correlation with radon concentration as temperature and rainfall did. For Ireland, Met Éireann report that the smallest pressure gradients occur in spring and summer, the time period for the measurements in figure 3.17, and therefore it would be expected that pressure would have less influence on radon for this time period. In contrast, and as discussed in section 3.1, pressure is more influential on seasonal variations with a lower pressure in winter months resulting in a higher emanations from underground.

3.3.2.2 *Variations in site radon concentrations*

To determine whether any variance in radon concentrations between Irish coastal and inland sites and between east and west coast concentrations is significant, a statistical analysis test called a Student's t-test was used. This is a hypothesis test used to assess whether two groups of data are statistically different from each other (within a stated confidence interval), or if the difference is purely a chance find. The t-test was first devised in Dublin in 1908 by William Sealy Gosset, who published the work under his pen name, Student.

The statistical t-test compares the difference in the mean values of the 2 groups (independent groups or sample/populations groups), in order to ascertain whether the null hypothesis (mean1 – mean 2 = 0) is rejected or not rejected. There are several types of t-tests relating to the type of data and the type of comparison required (1-sample, 2-sample or paired), but they all use the means, standard deviation and number of samples to calculate a t-value and the degrees of freedom, in order to reject or fail

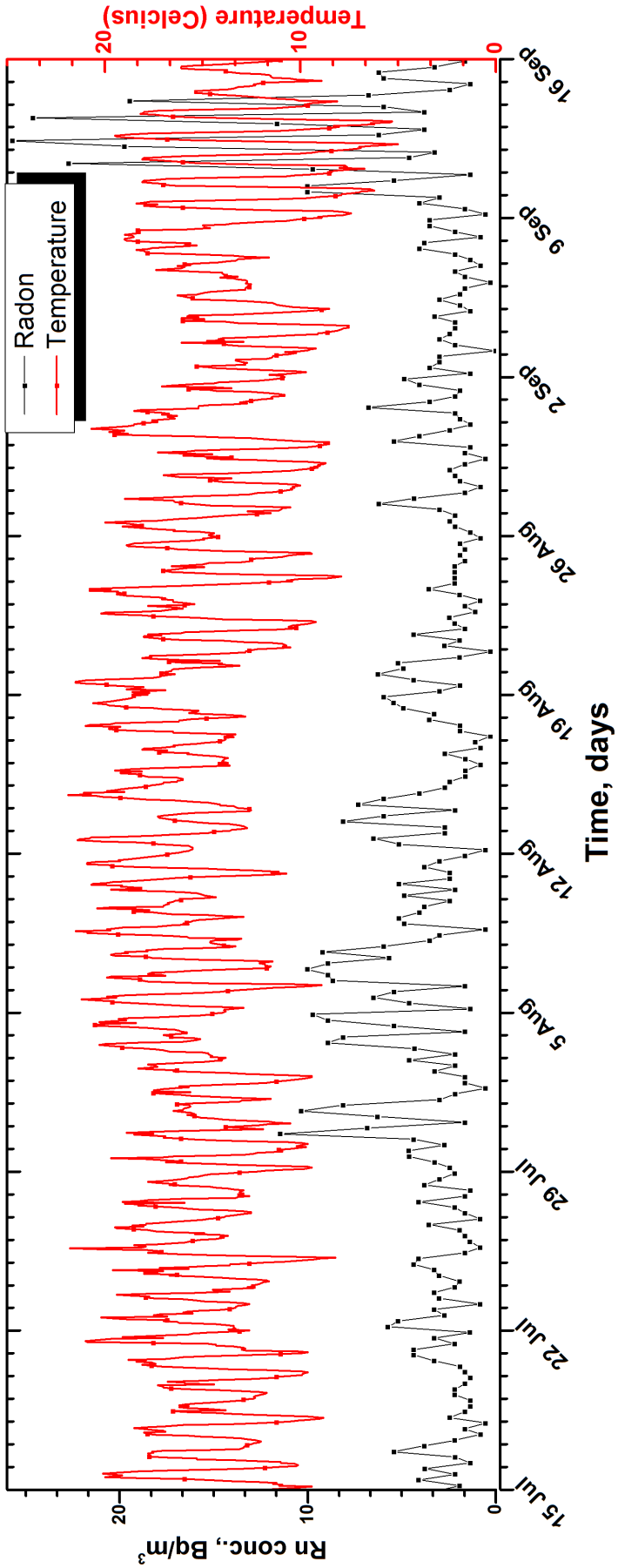


Figure 3.14: Radon concentration variation with temperature.

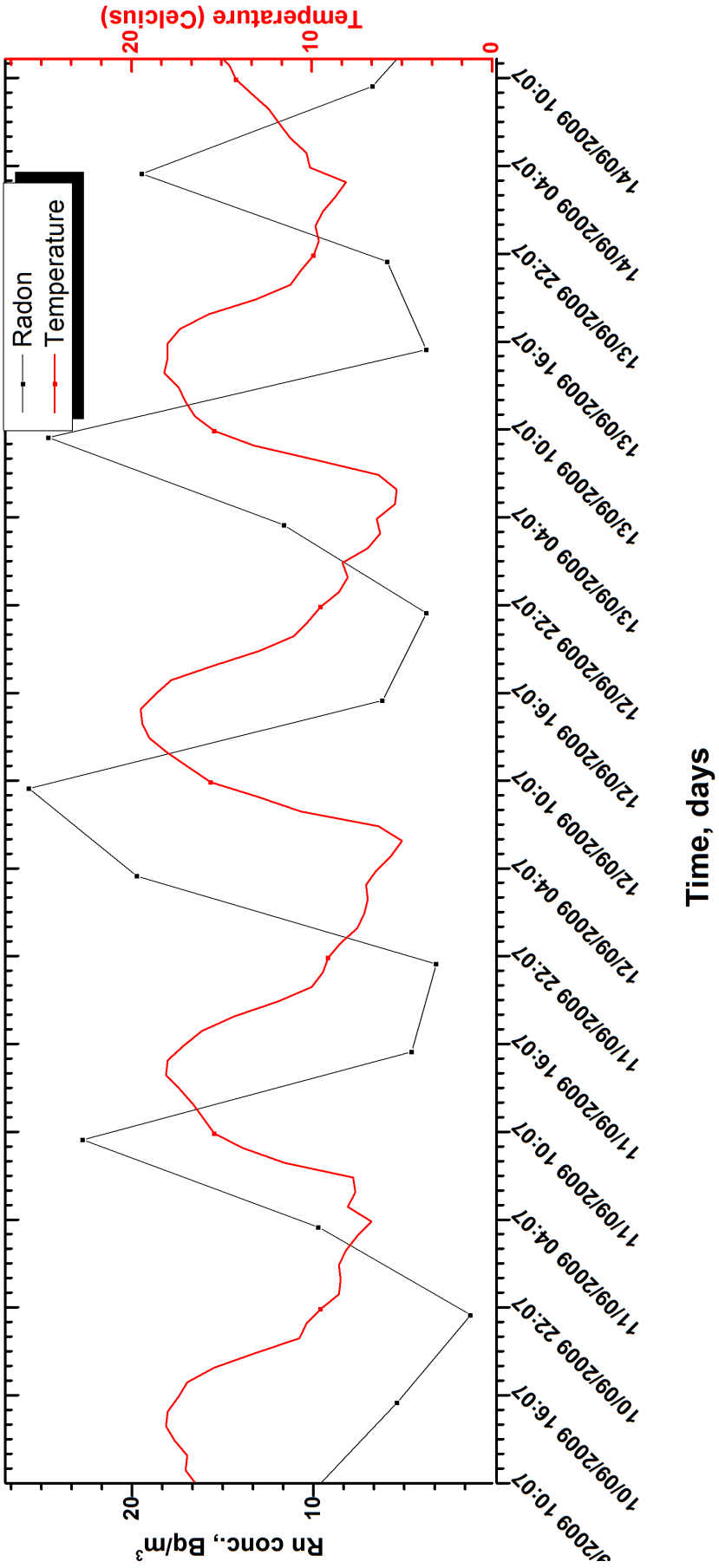


Figure 3.15: Radon concentration variation with temperature (zoom of figure 3.14).

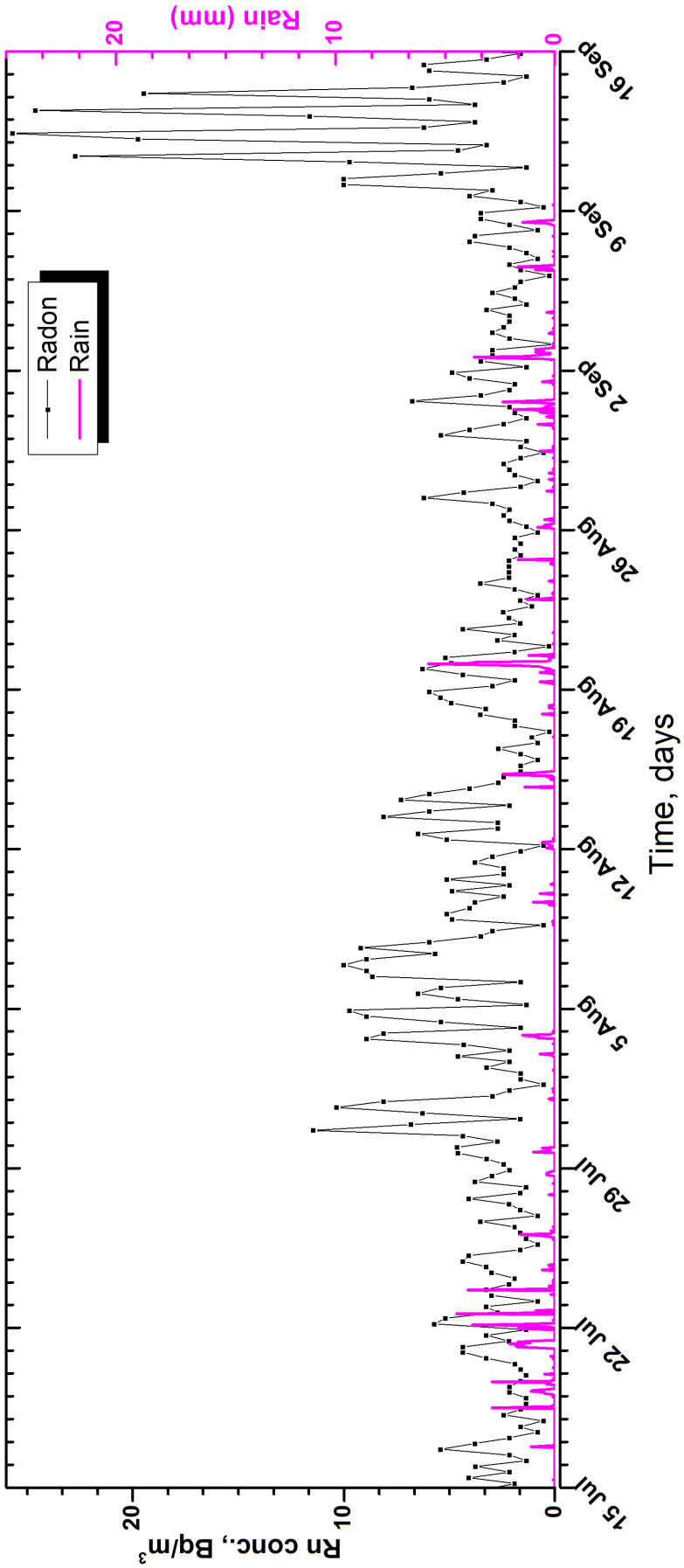


Figure 3.16: Variation of radon concentration with rainfall.

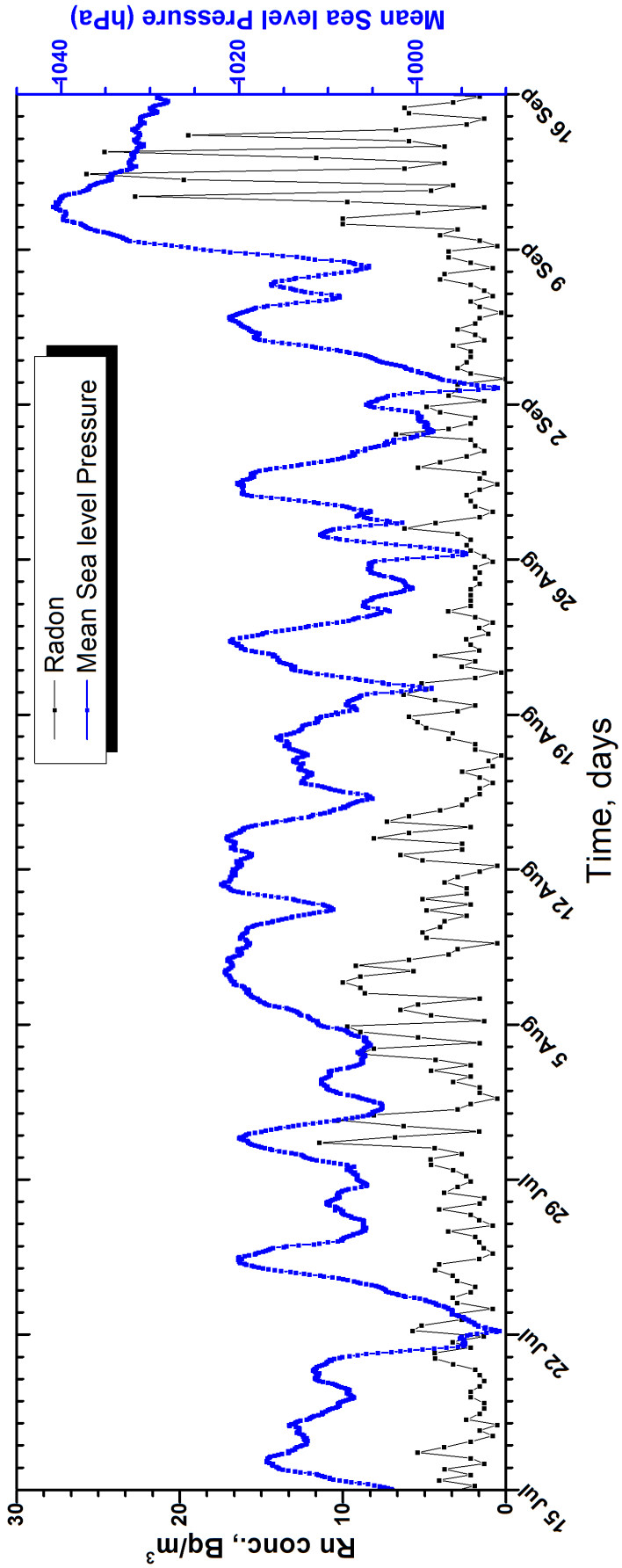


Figure 3.17: Variation of radon concentrations with pressure.

to reject the null hypothesis for a stated p-value. The t-value (or t-statistic), t_v, for a 1-sample group is defined as,

$$t_v = \frac{\bar{x} - \mu}{s/\sqrt{n}} \tag{3.3}$$

where $\bar{x}$ is the mean of the sample, μ is the mean of the parent population and s is the sample standard deviation of n samples. It is is a measure of the difference between two groups normalised by the standard deviations of the sample values.

The degrees of freedom, DF, is the number of values in a statistical calculation that are free to vary, while keeping the same result. For example, in a mean calculation of n samples, $n-1$ values can vary but the n^{th} value is then set in order to maintain the same mean result, therefore $DF = n-1$. The p-value is the level at which it can be confidently stated that a difference between the two means is not a chance find. For example, a p-value of 0.01 would indicate a 99% confidence interval that the null hypothesis is rejected, or not rejected depending on the outcome. For most scientific disciplines, a p-value of 0.05 (or 95% confidence interval) or less is an indicator of significance and so was taken as the level of significance for this study.

In this study, it is required to determine whether 2 sets of radon concentrations are significantly different, therefore an independent two sample t-test is required. In the specific case (as is here) where the 2 datasets have unequal sample size or unequal variance, a Welch's t-test is used to calculate the t-value, t_W,

$$t_W = \frac{\bar{x}_1 - \bar{x_2}}{\sqrt{\frac{SD_1^2}{n_1} + \frac{SD_2^2}{n_2}}} \tag{3.4}$$

where $\bar{x}_1$ and $\bar{x}_2$ are the means of the 2 datasets, SD_1 and SD_2 are the standard deviations and n_1 and n_2 are the sample sizes. For the same conditions, the degrees of freedom are calculated using,

$$DF = \frac{(SD_1^2/n_1 + SD_1^1/n_1)^2}{(SD_1^2/n_1)^2/(n_1 - 1) + (SD_2^2/n_2)^2/(n_2 - 1)} \tag{3.5}$$

The t-value is then compared with the test difference of the null hypothesis (here it is 0) and, along with the associated degrees of freedom, are looked up in a t-table of significance, to test whether the ratio is large enough to state if the difference between the groups is not likely to have been a chance finding [116].

A data analysis and graphics software package (OriginPro) was used to perform the calculations on the radon data measured in this study (using individual detector values from each site rather than the site mean). Table 3.18 shows the reported results of a t-test analysis for the coastal and inland radon concentration (the site locations are indicated in figure 3.13). The difference in means is -1.7. This difference and the t-value are negative in this instance, simply because the second mean is larger than the first. The upper and lower limits in the confidence levels refer to the separation of the means and since this interval does not include 0, i.e. at no point does mean1 - mean2 = 0: the null hypothesis is rejected. Therefore, at a p-value of 0.05, the difference of the population means is significantly different with the test difference or, in other words, it can be stated that with 95% confidence, the difference between coastal and inland radon concentrations is not a chance find.

Application of the same statistical analysis to the east and west coast values (the site locations have been indicated in figure 3.13) returned the data in table 3.19. A difference in the means of 0.7 is stated and at all 3 confidence levels the interval includes 0, ie., at some point mean1 – mean 2 = 0: the null hypothesis can not be rejected. Therefore it can be stated, with 95% confidence, that this difference in means is actually a chance find and there is no significant difference between east and west coast radon values.

It should be noted that a t-test is applied to a sample when the parent population distribution is normal. The sample will follow a t-distribution, but as the sample size increases it will approach to the normal distribution of the parent population. A normal distribution is the continuous probability distribution of measurements around the mean, whose mathematical equation is given by

	Sample Number	Mean	Standard Deviation	SEM
Coastal	45	4.74491	1.59311	0.23749
Inland	45	6.42544	2.02016	0.31155
Difference		-1.68053		0.21046

t value	DF	
-4.38182	83.464	(Welch Correction)

Null Hypothesis: mean1-mean2 = 0

Alternative Hypothesis: mean1-mean2 <> 0

At the 0.05 level, the difference of the population means is significantly different with the test difference (0)

Conf. Levels in %	Lower Limits	Upper Limits
90	-2.31808	-1.04298
95	-2.4427	-0.91836
99	-2.69029	-0.67077

Figure 3.18: Statistical t-test of inland and coastal radon data.

	Sample Number	Mean	Standard Deviation	SEM
East Coast	10	5.40263	1.95341	0.61772
West Coast	20	4.73811	1.61212	0.36048
Difference		0.66452		0.66971

t value	DF	
0.92913	15.33139	(Welch Correction)

Null Hypothesis: mean1-mean2 = 0

Alternative Hypothesis: mean1-mean2 <> 0

At the 0.05 level, the difference of the population means is NOT significantly different with the test difference (0)

Conf. Levels in %	Lower Limits	Upper Limits
90	-0.47474	1.80379
95	-0.70732	2.03636
99	-1.18606	2.5151

Figure 3.19: Statistical t-test of east and west coast radon data.

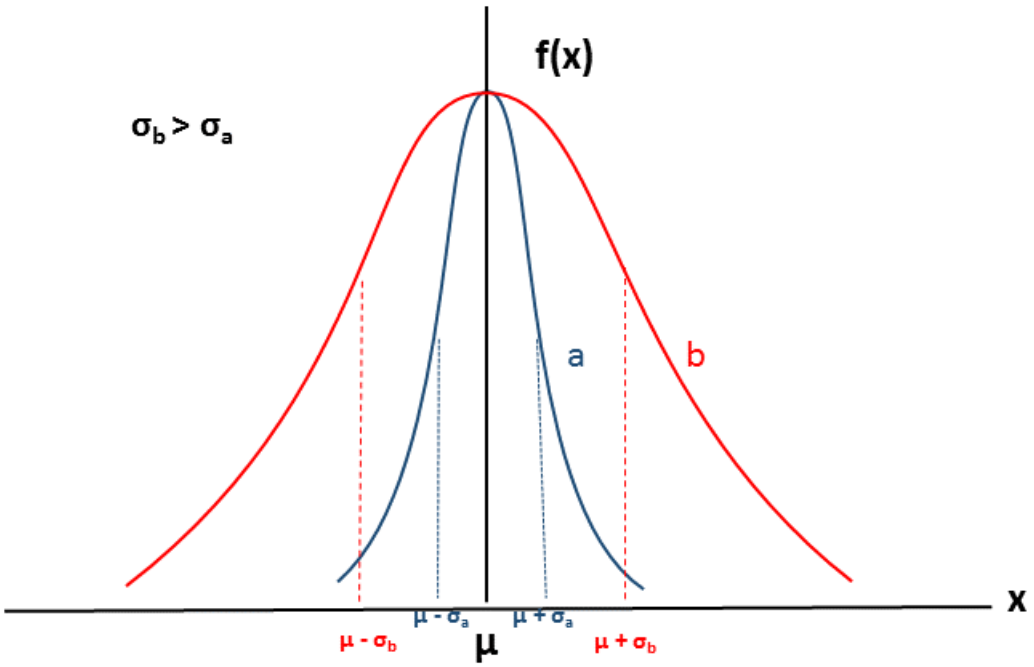


Figure 3.20: Normal or bell-shaped curve which represents a normal distribution with mean, μ, and standard deviation, σ.

$$f(x) = \frac{1}{\sqrt{2\pi\sigma}} e^{-(x-\mu)^2/(2\sigma^2)} \tag{3.6}$$

for $-\infty > x < \infty$. Graphically, the distribution follows a bell-shaped curve with the mean indicated by the peak and the width of the curve dictated by the standard deviation, σ, as per figure 3.20. In this figure, the distributions have the same mean, μ, but different standard deviations: $\sigma_b > \sigma_a$ which results in a larger spread in data points in b.

Figures 3.21 and 3.22 are histograms in bins of 1 Bq/m^3 and serve as a graphical analogue to the tabular form of the t-test data. The curves superimposed on figures 3.21 and 3.22 are the most likely fit of a normal distribution to the data. The mean of each set of results is seen at the peak of the curve. In comparing the 2 graphs, a difference in

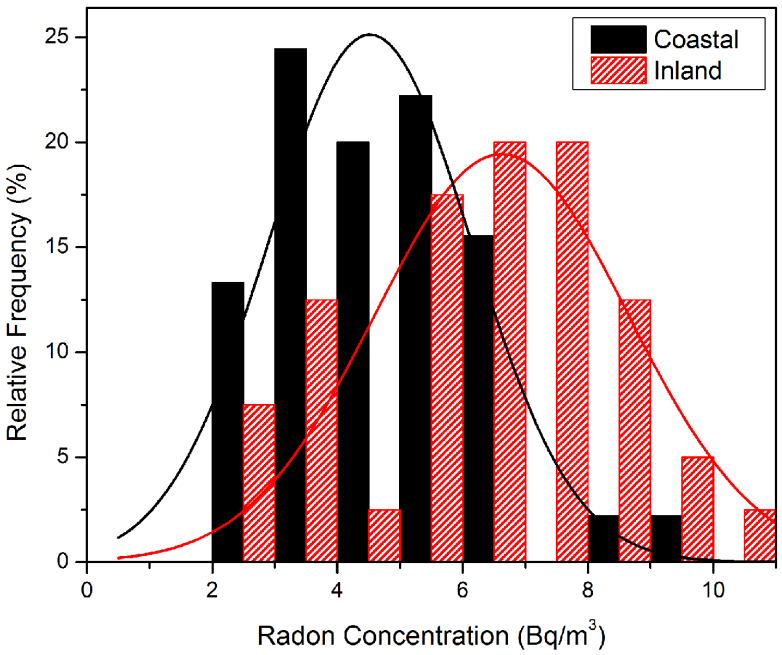


Figure 3.21: Histogram and normal distributions of inland and coastal radon concentration.

the means is seen in both graphs but the volumes under the bell-shaped curve overlap considerably in the east/west coast graph (figure 3.22).

3.4 CONCLUSION

The mean outdoor radon concentrations of 18 sites in Ireland was measured to be 5.6 ± 0.7 Bq/m^3 which is in close agreement with the previously derived value of 6 Bq/m^3. The updated figure has been noted in the 2014, EPA report 'Radiation doses received by the Irish population' [75, 79]. Using equation 2.1, an outdoor equilibrium factor of 0.6 and exposure time of 1760 hours, an annual outdoor mean radon concentration of 5.6 Bq/m^3 equates to an annual dose of 0.05 mSv to the Irish population

As stated, 6 Bq/m^3 was previously estimated as the outdoor radon concentration for Ireland. This was extrapolated from a Q-Q plot of indoor log-normal radon concentration. The correlation of this theoretical value with the measured value in this study verifies

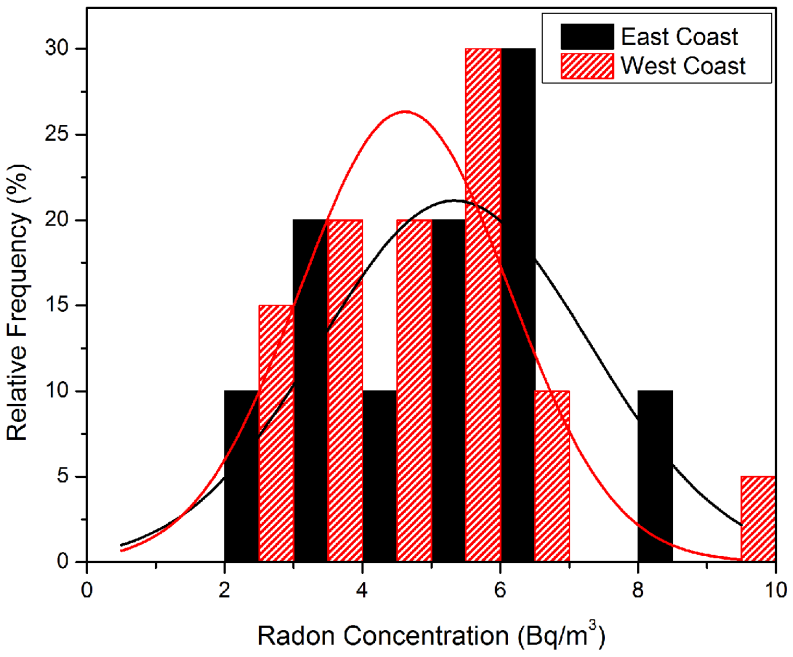


Figure 3.22: Histogram and normal distributions of east and west coastal radon concentration.

that this method of extrapolating is correct and should be considered to predict the outdoor concentration in other countries where indoor data exists.

Using the measurement protocol derived from this study, it was found that low level outdoor concentrations can be measured with good precision using CR-39 track etch detectors. This protocol included pre-etching the detectors before exposure to allow radon tracks to be more easily distinguished from background.

Fluctuations in radon concentration are multi-factorial with regard to metrological parameters but, from a 2 month continuous monitoring of rain, temperature, pressure and radon concentration, clear observations were made. Daily trends of high and low concentrations were observed in the early morning and later afternoon respectively, and were attributed to air turbulence and a thermal inversion layer. Rainfall also had a notable influence on the outdoor radon concentrations. During dry periods the radon concentration rose due to the increase of porosity of soil. As continuous data only recorded a two month summer period, radon variations were not directly correlated with

pressure. It is predicted that a correlation between pressure and radon would be more evident in a 12 month measurements, where seasonal variation could be analysed.

The difference between inland and coastal radon can be correlated with the variation of air radon concentrations over land and over seawater. Radon gas emanates from land mass and therefore it would be expected that radon concentrations in air over the sea would be lower; this might be expected to result in lower concentrations in coastal areas. On average over a year, this study shows that coastal sites have lower radon concentrations than inland sites. In Ireland, the weather systems and prevailing wind generally come from the west but this, however, does not result in a detectable difference between average radon concentrations on the west and east coasts.

3.5 FUTURE WORK

Many of the results from this study were conclusive and quantitative (outdoor radon concentration, the correlation with weather variations and site location). One conclusion presented that is worthy of further investigation, is the relationship between indoor and outdoor radon. This study verified that the method of extrapolating a mean outdoor radon concentration from probability plots of indoor data was correct. It would be beneficial to test the concept of extrapolating in other countries as it could become an import tool for predicting the outdoor concentrations in countries where the mean indoor value is high and for countries that are developing a radon programme. It would facilitate estimating the dose to the population more accurately, especially as many countries have noted that radon is the largest contributing factor to the annual mean population dose.

As an extension of the work completed in this chapter, it would also be beneficial to examine available indoor radon data measured in close proximity to these outdoor sites.

4

INTER-COMPARISON OF RADON DETECTORS

4.1 INTRODUCTION

It is internationally recognised that the measurement of indoor radon concentrations is important, as is the implementation of a method of remediation if the concentrations are found to be above the reference level [117]. However, the percentage of houses measured is still low in most countries [2, 52]. The inclusion of radon testing in the conveyancing process is one possible method of increasing the rate of testing. Some countries, such as the UK and USA, have initiated this process by promoting a strong recommendation for measurement during conveyancing. Norway, which requires measurement and remediation for rental properties, and Sweden, which requires a radon measurement for the energy declaration (as outlined in their 'Energy Declaration of Buildings Act', 2006) when buildings are constructed, sold or rented, are among the few countries with legislation to enforce testing in dwellings.

A perceived obstacle to the inclusion of radon testing during the conveyancing process is the recommended measurement period. Radon concentrations vary over time and especially from season to season, and a 12 month test therefore gives the best indication of the annual average. As a 12 month testing period is not always practical, the approved standard length of test in many countries is 3 months [51]. A seasonal correction factor is typically applied to allow for the seasonal variations [118, 119].

The objectives of the research detailed in this chapter are (a) to carry out an inter-comparison of several radon detectors in order to explore their performance, (b) to determine whether radon measurements can be undertaken for shorter than the recommended 3 month period. If the performance of a monitor is sufficiently accurate for shorter measurement periods, then it can potentially be used to indicate whether the

radon concentrations differ significantly from the reference level and thus warrant further investigation, for applications such as during conveyancing or following remedial work.

4.1.1 *Radon detectors*

There are numerous types of detectors available on the market to measure radon concentration. Generally they can be classified in 2 categories: passive and active. Most radon detectors convert the alpha particles from radon and its progeny to a measurable physically quantity, except for the activated charcoal canisters which collects radon; the collected radon concentration in this case can then be analysed by liquid scintillation counting (alpha detection) or gamma ray spectrometery (gamma detection).

4.1.1.1 *Passive detectors*

Passive detectors are low cost and do not require a power supply for operation, but require some kind of post-exposure analysis to determine the exposure levels. These have the advantages of being relatively cheap and not reliant on external power sources. The integrating nature of the passive detector, however, removes any time-dependency of the exposure longer than the measurement time, which tends to be longer than in active detectors. The most common type of passive detectors are:

- Charcoal absorption canisters. Activated charcoal is used to adsorb radon and radon progeny during the exposure period and then analysed by liquid scintillation or gamma ray spectrometry to determine the radon concentration. This type of detector is usually used for short term measurements, 2 - 7 days, and require prompt analysis within the subsequent days as the radon can also desorb.

- Electret detectors. these detectors consist of electrostatically charged teflon mounted in a chamber. The detector operates on the basis that changes in the charge on the detector surface are caused by the collection of ions from radon decay. By measuring the charge on the electret before and after exposure, the change in charge is used to calculate the radon concentration.

- Alpha-track detectors. CR-39 (as detailed in Chapter 3) and LR-115 detectors use polyallyl diglycol carbonate and cellulose nitrate plastics respectively, to calculate the radon concentration in air. Alpha particles from the decay of radon and its progeny strike the plastic, leaving tracks in it. The tracks are enhanced by chemical etching and then counted using an image analysis software. The detectors can be open-faced or housed in a holder to exclude dust, radon daughter products and limits the access of moisture [104, 105].

4.1.1.2 *Active detectors*

Active detectors on the other hand require a power supply and are more expensive than passive detectors. They can be split in to two main groups: direct conversion and indirect conversion detectors. A direct conversion detector has atoms or molecules which are ionized by incident radiation, causing a proportional quantity of mobile charges which can be measured; ionization chambers and solid-state detectors are direct conversion detectors. An indirect conversion detector has atoms or molecules which are excited to meta-stable states by incident radiation. These meta-stable states then decay by light-emission, which is converted to a proportional and measurable charge in a second step; scintillation detectors are indirect detectors.

- Scintillation detector: A scintillation detector is comprised of a scintillation medium and a photo-multiplier tube. The scintillation material is an organic or inorganic material of any phase than can be excited to a meta-stable state upon incident radiation. The relaxation of this meta-stable state is achieved by the emission of light. This photon can then be collected by a photo-multiplier tube. The photo-multiplier tube consists of a photo-cathode, a series of dynodes and an anode. The photo-cathode converts an incident photon to an electron via the photo-electric effect. The resulting electron is focussed on to the dynodes, which multiply incident electrons, causing a cascade of electrons to eventually fall on the anode. A continuous photon flux thus can produce a measurable current proportional to the incident radiation intensity.

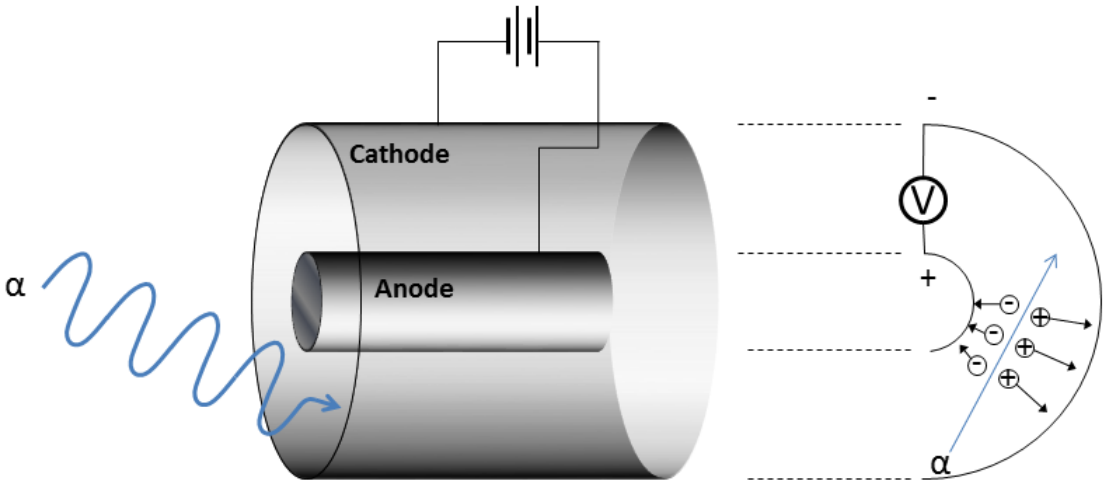


Figure 4.1: A schematic drawing of an ionisation chamber.

- Ionisation chamber: In this type of detector, a gas or liquid medium is enclosed between two electrodes (an anode and a cathode). As charged particles (incident radiation) move through the chamber they ionise the atoms or molecules of the gas and create ion pairs. An applied potential difference to the anode and cathode electrodes causes the negative ions to move to the anode while the positively charged ions are drawn to the cathode. The movement of ions results in a current being set up, the magnitude of which indicates the intensity of the radiation. Control electronics process the current signals and output information on the incident radiation.

- Semiconductor detectors: The semiconductor detector is essentially a form of ionisation chamber, with the chamber medium being a solid-state, semiconducting material. In this type of detector a semiconductor material (eg. silicon or germanium) converts incident radiation to electron-hole pairs via the photovoltaic effect, in a similar manner to the creation of an ion pair in the ionization chamber. The electron-hole pair is created when the incident radiation causes a promotion of electrons from the valence band (VB) of the semiconductor, the energy levels of electrons involved in the atomic bonding of the crystal. This electron is promoted across a range of energies (the band gap) to the conduction band

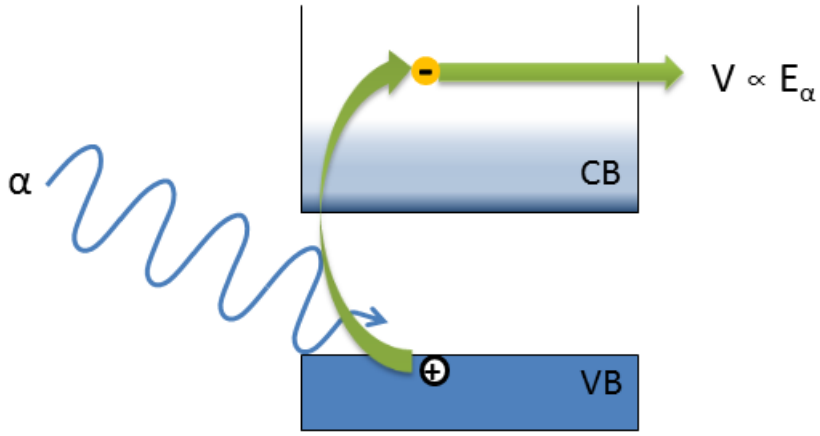


Figure 4.2: A solid state detector. The incident alpha particles promote electrons from the valence band (VB) to the conduction band (CB).

(CB) (figure 4.2). When promoted to the conduction band, the electron is free to move through the material and the vacancy left behind by the promotion - the hole - is also mobile. A potential across the semiconducting material sweeps the electrons to an anode and the holes to a cathode, creating a measurable current. The number of electron-hole pairs created (and thus the magnitude of the current) is proportional to the particle energy, E_a, and the pulse rate is proportional to the rate of incident particles. In common applications, the signal is amplified through electronic circuits and with the accumulation of many signals, an energy spectrum is produced which can identify the isotope of the incident radiation. This enables the detector to distinguish between the different decay products of radon.

There are many different configurations of the semiconductor detector, with the main distinguishing factor being the method by which the driving potential, to sweep the charge carriers out for measurement, is established over the material. In a silicon surface barrier detector, this can be done by depositing thin layers of aluminium and gold on opposite sides of the semiconductor, which act as collecting electrodes for the generated charge carriers. In other devices, a p-n junction is established. When a semiconducting material is pure, it is a poor conductor owing to the existence of the band gap and the material is said to be intrinsic. There

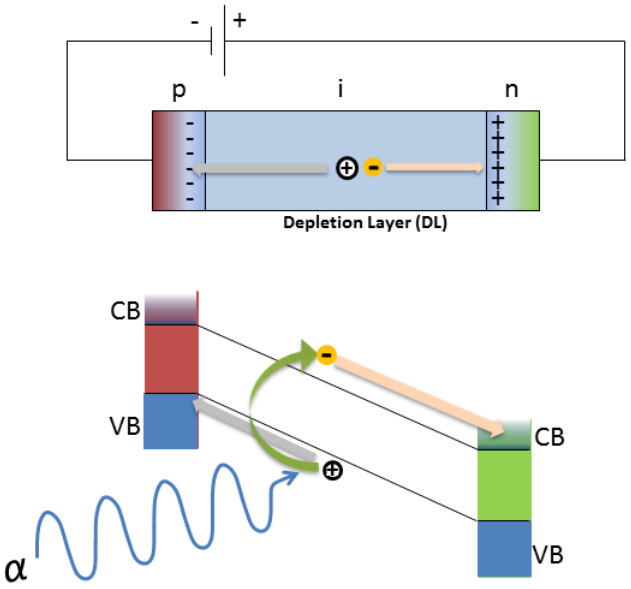


Figure 4.3: Schematic of a PIN photodiode indicating the p-type layer (p), intrinsic layer (i) and the n-type layer (n).

are equal numbers of electrons and holes in the material. The addition of small amounts of atoms from neighbouring groups of the periodic table can cause an excess of either electrons or holes, making the material n- or p-type, respectively. In the n-type material, electrons are now the majority charge carriers and holes are the majority charge carriers for a p-type material. This process is called doping. Sandwiching an n- and p-type material together establishes a p-n junction. At the interface, the oppositely-charged majority carriers from each side drift into the neighbouring material and recombine, creating a region where no free charge carriers are present - the depletion region. An external reverse bias sets up an electric field across this depletion layer, which can be used to separate electron-hole pairs generated in the semiconductor for measurement. These p-n junctions are used in devices such as the passivated implanted planar silicon (PIPS) detector using modern semiconductor manufacturing methods to create p-n junctions on silicon wafers. If larger depletion layers are required for more penetrating radiation, a PIN junction can be used. This includes an intrinsic layer between a p-type and an n-type material (figure 4.3) which can be achieved using higher purity semiconductor materials or by drifting lithium in to the silicon to artificially make

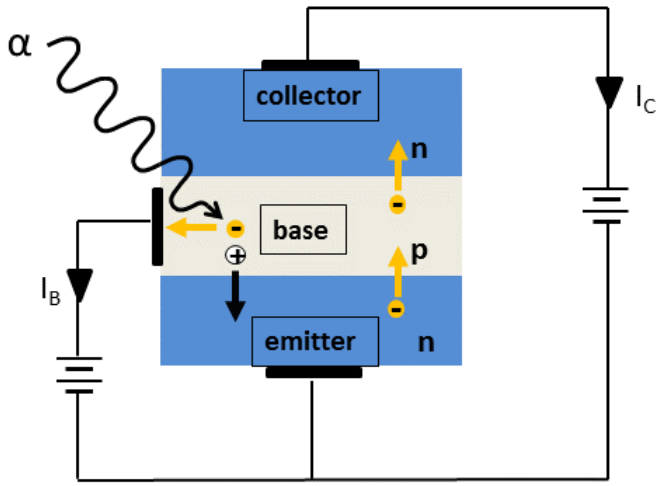


Figure 4.4: Bipolar junction transistor.

the material intrinsic [120–122]. Another configuration of the p-n junction can be used with bipolar junction transistor technology, where two p-n junctions are joined together in a p-n-p or an n-p-n configuration. The three regions are called the collector, emitter and base (figure 4.4). Looking at a n-p-n configuration, if a positive potential is applied between the collector and emitter, the base and collector are in reverse bias and the width of the depletion layer is increased so there is little or no current. When an alpha particle is incident on the base layer electron-hole pairs are created and positive hole carriers flow to the emitter, setting up a small base current. If the base layer is very thin and the emitter is heavily doped then electrons drift from the emitter to the collector, setting up an amplified collector current. External electronics record the α particle arrival time and the charge in real-time. External amplification is not required as the signal has already been amplified by the bipolar junction transistor itself [123, 124].

Monitors can also be categorised as being either continuous or time integrated. Continuous monitors record radon concentrations at periodic intervals for the duration of the exposure period, anything from 10 minutes to daily intervals. Time integrated measurements calculate the radon concentration at the end of the exposure period based on the length of the exposure.

Method	Measurement type	Detector
Preliminary test for radon	Short-term sampling	Charcoal absorption, continuous monitor, electret
Assessment of exposure	Time integrating	Alpha track, continuous monitor, electret
Remediation testing	Continuous monitoring	Continuous monitor

Table 4.1: Primary methods and devices for residential radon measurements.

Table 4.1 summarises the type of monitor used for the radon measurement required [56]. Continuous monitors are seen to be the most versatile but they are also the most expensive and may not be practical for conventional 3 month dwelling measurements.

Other important aspects of radon measurements are the protocol accreditation and detector calibration. As referred to in Chapter 3, accreditation is awarded by a national accreditation body which accredits in accordance with the relevant ISO standards and guides. In Ireland, the national body is the INAB and for radon testing ISO 17025 for homes and workplace is awarded.

Calibration of passive radon detectors is usually carried out in the providers' lab whereby several of the detectors are periodically exposed to a known radon concentration or other alpha radiation source. Active monitors are calibrated by the manufacturer or by a reference laboratory and a calibration certificate is issued to state the correction factor, the uncertainty in measurement, the background count and the date of calibration. The frequency of active monition calibration is carried out on the advice of the manufacturer and is typically annually or semi-annually.

In addition to accreditation and calibration, many manufacturers and suppliers regularly partake in intercomparison schemes facilitated by research institutes or reference laboratories [125–127].

4.2 EXPERIMENTAL METHOD

Figure 4.5: Radon detectors (l-r): Atmos 12dpx, RAD 7, Sun Nuclear 1028, RStone Pro, Ramon 2.2, Canary and a CR-39 (images courtesy of the supplier/manufacturers).

The radon detectors used in this study were (figure 4.5) [128]:

- An Atmos 12 dpx, which is based on ionisation chamber technology (figure 4.1) [129, 130]. The Atmos has an internal CPU (central processing unit) memory which stores the data automatically and can be read via the LCD (liquid crystal display), by connecting to a device running the Atmos32 software package or printing directly for producing reports and time distributions of the radon concentration. The Atmos has a measurement range from 1 Bq/m^3 to 1 x 10^5 Bq/m^3.

- A RAD 7, which uses Canberra PIPS (Passivated, Ion-implanted, Planar, Silicon) alpha detector, a type of PIN diode to detect alpha particles [131]. Of the detectors in this study, the RAD7 is the second most expensive after the Atmos in retail price. Additional accessories facilitate measurements in water and below ground and it has a measurement range from 4 Bq/m^3 to 7.5 x 10^5 Bq/m^3.

- A Sun Nuclear 1028, which has accompanying PC software for printing reports and enables read-only time and date results to 'prevent tampering' [132].

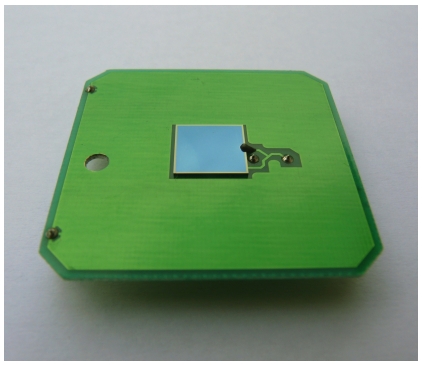


Figure 4.6: PIN diode used in the Ramon 2.2 detector (image courtesy of GT-Analytic SARL).

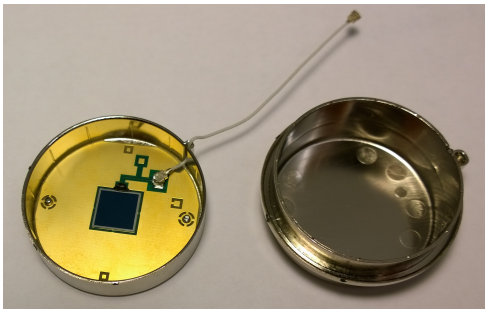


Figure 4.7: Canary PIN diode with protective cover to eliminate light (image courtesy of Corentium AS).

- Two RStone Pros. The RStone Pro working principle is founded on bipolar junction transistor technology [123, 124, 133]. It has wireless connection making it possible to program and manage remotely. The data can be converted to most spreadsheet applications for further data analysing.

- A Ramon 2.2 (figure 4.6), which utilises photodiode semiconductor technology in order to detect radon [134]. The Ramon counts all incident alpha particles and cannot distinguish between alpha particles from radon and those from the decay products (most notably from Polonium-218 and Polonium-214).

- Two Canary monitors (figure 4.7). The Canary is battery operated for ease of transport and for measurement placing. The LCD screen displays the average daily, weekly and long term radon concentrations. In contrast to the Ramon, the Canary uses algorithms to distinguish between the alpha particles based on the amplitude of the voltage [135].

- CR-39 detectors: both suppliers of the CR-39 detectors in this comparison study sourced the PADC plastic from Track Analysis Systems Ltd., (Bristol, UK) [136].

All the detectors except the CR-39 detectors are active digital radon monitors giving a real-time or time-averaged radon concentration. They range in complexity, price, accuracy and level of information recorded (table 4.2).

It was decided to run the comparison for exposure periods of 1, 2, 3 and 4 weeks and in two environmental conditions: an occupied home ("real world" conditions) and a radon chamber (with an artificially elevated radon concentration). The occupied home is a terrace house with two storeys in County Dublin. The concentration was measured several years previously with a CR-39 detector for a period of 3 months and reported to be 39 Bq/m^3.

The two settings contrast not only in radon concentrations but also in environmental conditions. Although the chamber is subject to pressure, humidity and temperature variations, a home environment is also subject to the occupant's lifestyle habits (hours occupied, temperature settings, ventilation), the building specification (insulation, age, building materials) and the location (rock type, coastal/inland, local weather conditions).

The digital detectors were set up on a bench in the radon chamber, at a height of 88 cm and a distance of 160 cm from the Ra-226 source. The Atmos was set to take readings every 10 minutes. However, the RStone, Sun Nuclear, Canary and Ramon have minimum sampling intervals of 1 hour. In the case of the RAD7, sampling intervals of 2 hours are recommended for measurements lasting a week or more, but for this study it was set to 1 hour, in line with the RStone, Sun Nuclear, Canary and Ramon.

Two hours prior to the start of the measurement, eight CR-39 detectors (two for each of the measurement periods) were removed from storage in a freezer [111] and brought to room temperature. Following this, they were then placed in the radon chamber and the digital monitors were reset to commence the inter-comparison.

At weekly intervals, two CR-39 detectors were removed from the radon chamber and the time-averaged radon concentrations from the Canarys and the Ramon were manually noted (they do not have a data memory function). The CR-39 detectors were sealed in a

radon-proof bag and returned to the issuing company for analysis. Figure 4.8 summarises the inter-comparison schedule.

With regard to detector calibration, it is recommended by the manufacturers to calibrate the Atmos, RAD7, Sun Nuclear and RStone annually. However, according to the manufacturers the Canary and Ramon do not need annual calibrations. Calibration is carried out at an accredited facility where detectors are exposed to a reference radon atmosphere which is traceable to a primary radon gas standard [125, 137]. The detectors are also exposed to a radon-free atmosphere (aged air, nitrogen or outside air) to determine the background count of the detector [51]. This background count is due primarily to long-lived radon progeny (Pb-210 has a half-life of 22 years) which build up over time on the detection area. These progeny arise from earlier plateout of short-lived products in the radon decay chain.

It was noted during the 4 week chamber study that the RAD7 and the Sun Nuclear had not been calibrated in a number of years (3-5). Subsequently, the RAD7 and Sun Nuclear were sent to BfS (Bundesamt für Strahlenschutz), Germany for calibration and the 4 week cycle was restarted under the same conditions to incorporate a further comparison of calibrated and uncalibrated monitors. The Atmos had a valid calibration certificate issued by the Swedish Radiation Authority (SSM), and the RStones were calibrated by the manufacturer, R Sens Srl, Italy, a few months before this study commenced.

Once the measurements in the radon chamber were complete, the detectors were removed and set up in the home environment under the combined guidance laid out in the manufacturers' instructions: 10 cm from walls, 1 m from doors, vents, fireplaces, windows, draught, not in direct sunlight and not moved during measuring. Two additional sets of CR-39 detectors were contributed to this section of the study, both were from the same supplier but calibrated using different Atmos detectors: only one Canary detector was available. The measuring intervals of the monitors were kept as before.

As in the radon chamber inter-comparison, the CR-39 detectors were removed from freezer storage 2 hours prior to commencing the measurements. After the 2 hours, they were put *in situ* and the digital monitors were reset.

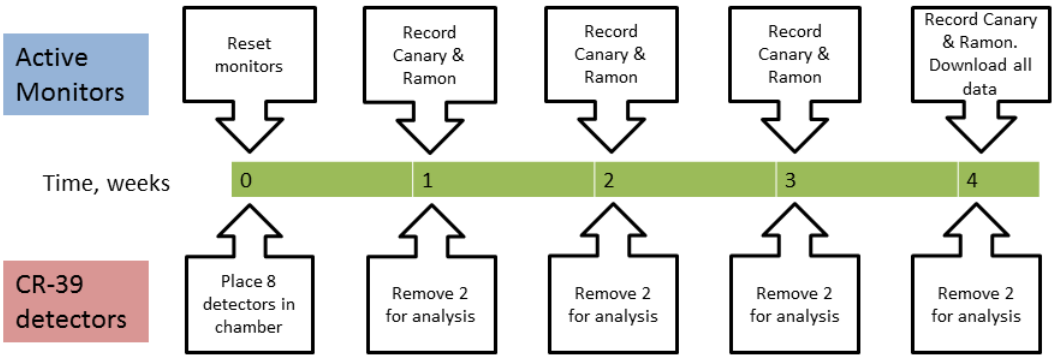


Figure 4.8: Timeline for inter-comparison of radon detectors in the radon chamber.

For 4 weeks, the radon concentrations from the Ramon and the Canary were recorded daily and at weekly intervals two CR-39 detectors from each supplier were sealed in radon-proof bags and returned for analysis.

4.3 RESULTS AND ANALYSIS

The Atmos, RAD7, Sun Nuclear and RStone detectors display data in the format of the radon concentration (Bq/m^3) as measured at the set time interval (10 or 60 minutes). A rolling mean of these individual radon concentrations was calculated. The CR-39, Canary and Ramon report the radon concentration as a mean for the specified lapsed time interval.

BfS issued calibration certificates for the RAD 7 and the Sun Nuclear which stated that calibration factors of 1.08 and 1.14, with uncertainties in calibration of 0.08 and 0.10, should be applied to measurements from these monitors respectively. Figures 4.9 and 4.10 depict the mean radon concentrations for each detector against time for the radon chamber environment and the home environment respectively; the calibration factors are included.

For this study, the Atmos (being the EPA's reference detector) was set as the benchmark in order to make comparisons between the detectors. It cited an average radon concentration over the 4 week period of 2,560 Bq/m^3 in the radon chamber and 57 Bq/m^3 in the home environment, with an associated uncertainty of $\pm 10\%$ as stated

Detector name (manufacturer)	**Detection system**	**Quoted accuracy**	**Approximate retail price (2015)**	**Information recorded**	**Additional features**
Atmos 12 dpx (Gammadata Instruments AB)	Ionisation chamber	± 10% at 800 Bq/m^3 after 10 min	€10,750 + VAT	Time, date, temp, radon conc. and uncertainty, energy spectrum	1 month data storage, Atmos 32 software, inbuilt humidifier, sniff test
RAD7 (Durridge Company Inc)	PIN photodiode	± 5% absolute accuracy, 0-100% RH[a]	$6900 + VAT	Time, date, temp., relative humidity, radon conc. and uncertainty, energy spectrum	999 cycle data storage, Capture software, sniff test, thoron measurements, radon in soil and water.
Sun Nuclear 1028 (Sun Nuclear Corporation)	Diffused junction photodiode	± 25% or 37 Bq/m^3 after 24 hrs[b]	€900 + VAT	Time, date, radon conc.	720 cycle data storage, Sun Nuclear software, disturbance sensor
RStone Pro (RSens)	Bipolar junction transistor	6% after 48 hrs at 150 Bq/m^3	€1600 + VAT	Time, date, temp., relative humidity, radon conc. and uncertainty	Radon Pro software, remote monitoring and measuring through wireless network.
Ramon (GT-Analytic SARL)	PIN photodiode	4.5% after 1 mth at 100 Bq/m^3	€230 + VAT	Time-averaged radon conc.	Short-term (7 day) average and long-term average displayed
Canary (Corentium AS)	PIN photodiode	<20% after 7 days and <10% after 1 mth (at 100 Bq/m^3)	€200 + VAT	Time averaged radon conc.	1 and 7 day average and up to 12 month long measurement average
CR-39 (PADC from Track Analysis System Ltd. used in this study)	Track etch detector	Typically 10-25% at ~ 200 Bq/m^3	€40-60	Alpha tracks on PADC plastic – radon conc. calculated by analysis system	TASL, UK state a lower limit of detection of 5 $Bq/m^3$and an upper limit detection of 15 MBq/m^3

Table 4.2: A summary of the features of the radon detectors used in this study.

a RH – Relative humidity
b Whichever ever is greater

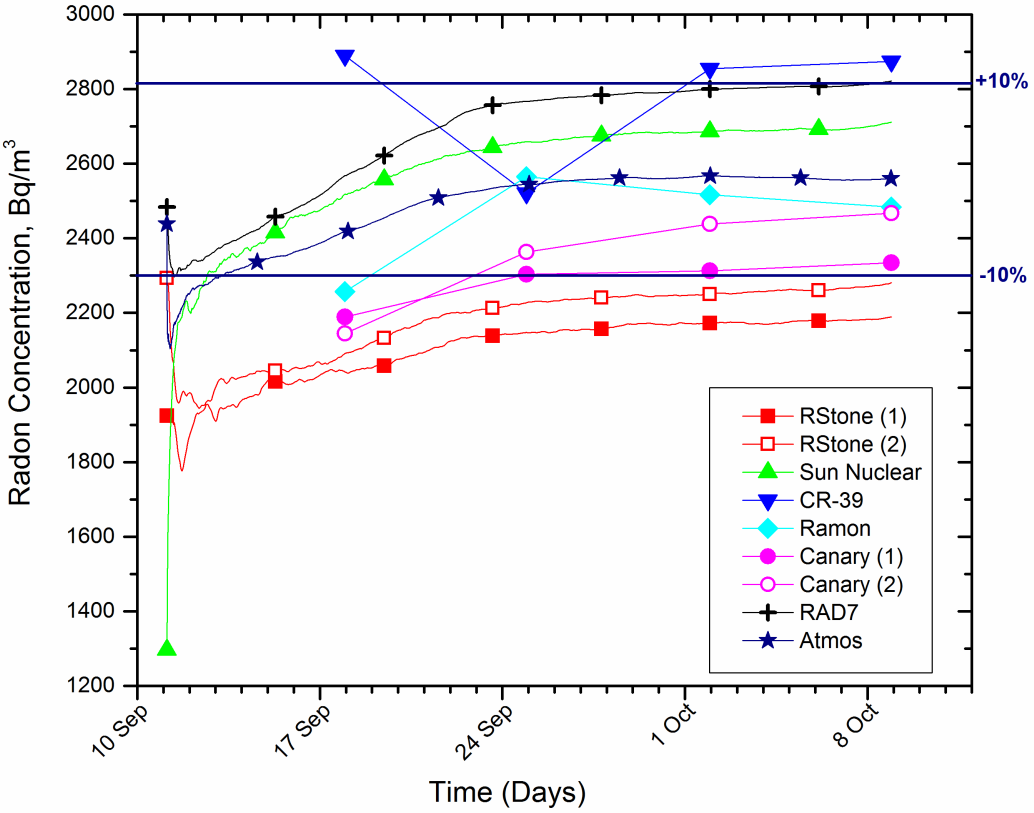


Figure 4.9: A plot of the mean radon concentration against time for radon detectors in the radon chamber. Data are indicated by symbols for the CR-39, Ramon and Canary detectors but data for the remaining detectors are indicated by lines and symbols are used only for clarification: similarly for figures 4.10 and 4.14.

in the most recent calibration certificate. The ±10% reference lines in figures 4.9 and 4.10 highlight which detectors fell within the Atmos uncertainty limits of the average radon concentration in each environment. At the higher radon concentration, most of the detectors fell within ±10% after just 2 weeks, with only the CR-39 detectors and RStone detectors remaining outside the limits after 4 weeks. At the lower radon concentration, reference lines of ±20% are also included as fewer detectors (RAD7, Canary and RStone (2)) fell within ±10% at 4 weeks. In fact, CR-39 (1), CR-39 (2) and the Ramon detectors remained outside ±20%, although the Ramon is trending towards the Atmos line, suggesting that at lower concentrations a larger integration time is needed to achieve stability.

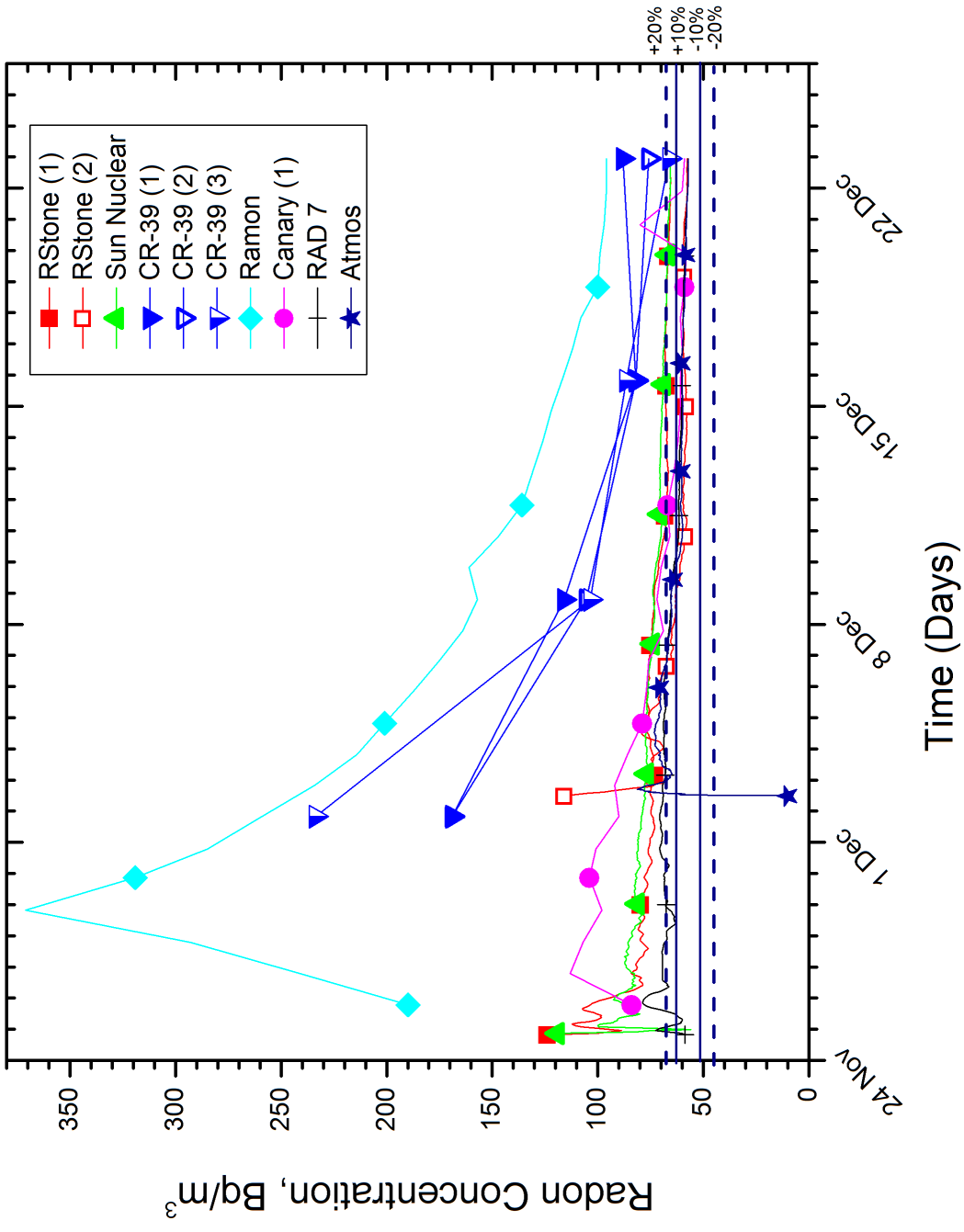


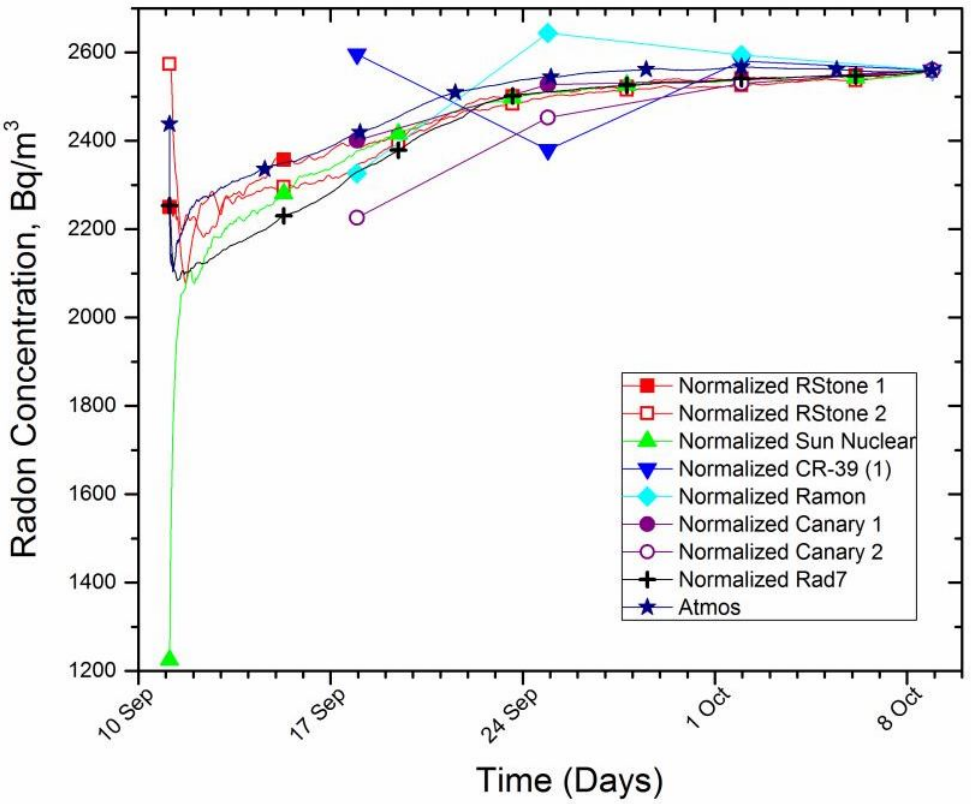
Figure 4.10: A plot of the mean radon concentration against time for various radon detectors in the home environment.

It was observed, particularly from figure 4.9 that the radon concentration for each monitor tends to follow the same trend line. Figures 4.11a and 4.11b display the data from the radon detectors normalised to the average radon concentration recorded by the Atmos in order to view how quickly each detector aligns with Atmos trend line. In the radon chamber data (figure 4.11a), by week 3 all the detectors apart from the Ramon and CR-39 (1) have almost converged to the Atmos trend line and in the home data (figure 4.13) after just over 2 weeks the majority of the continuous monitors have converged to the Atmos but the Ramon and CR-39 detectors converge closer to 4 weeks.

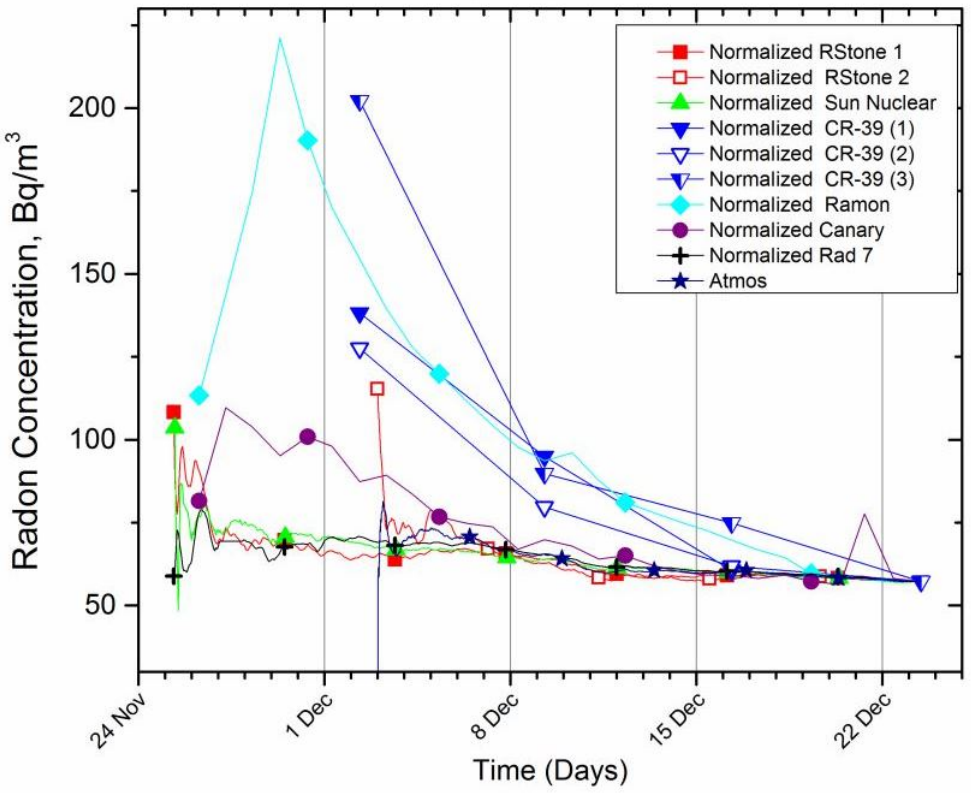
Figures 4.12 and 4.13 detail how each detector varied as a percentage from the Atmos for each period (1, 2, 3 and 4 weeks). The bar graphs depict the larger variation in percentage terms from the Atmos at the lower concentration. This is to be expected due to the statistical nature of counting but the method of detector calibration also contributes. Note that week 1 of the Atmos data in figure 4.13 did not download correctly due to a software error. The RAD7 was used as the reference detector for this week as it is in very close agreement with the Atmos for the following 3 weeks. The RStone (2) is also missing data for this week due to a fault in the reset function.

As mentioned, the calibration certificates from the RAD7 and Sun Nuclear stated calibration factors for each monitor. To show the significance of applying the calibration factors to the Sun Nuclear and RAD7, the 'before and after' calibration data were plotted alongside the Atmos data, as shown on figure 4.14. The ±10% uncertainty in the Atmos readings is indicated by the error bars in figure 4.14. At higher radon concentrations both the RAD7 and the Sun Nuclear remained within the error bars of the Atmos, but at lower concentrations the Sun Nuclear measured outside the Atmos error bars. This result may be explained by the fact that the calibration performed on both the Sun Nuclear and the RAD7 was a 'one-point' calibration. This is where instruments are calibrated at only one radon concentration, and in this instance the concentration was at 2,060 ± 140 Bq/m^3, as reported in the calibration certificate. The RStone and Atmos also had 'one-point' calibrations performed at 4,138 and 1,437 Bq/m^3 respectively.

Table 4.3 summarises for each detector the mean radon concentrations with their associated uncertainties for the measurement periods 1, 2, 3 and 4 weeks and in both environments. Though the software programs for the Atmos, RAD7 and RStone display



(a) Radon concentrations recorded in the radon chamber normalised to the Atmos.



(b) Radon concentrations recorded in the home normalised to the Atmos.

Figure 4.11: Normalised radon concentrations.

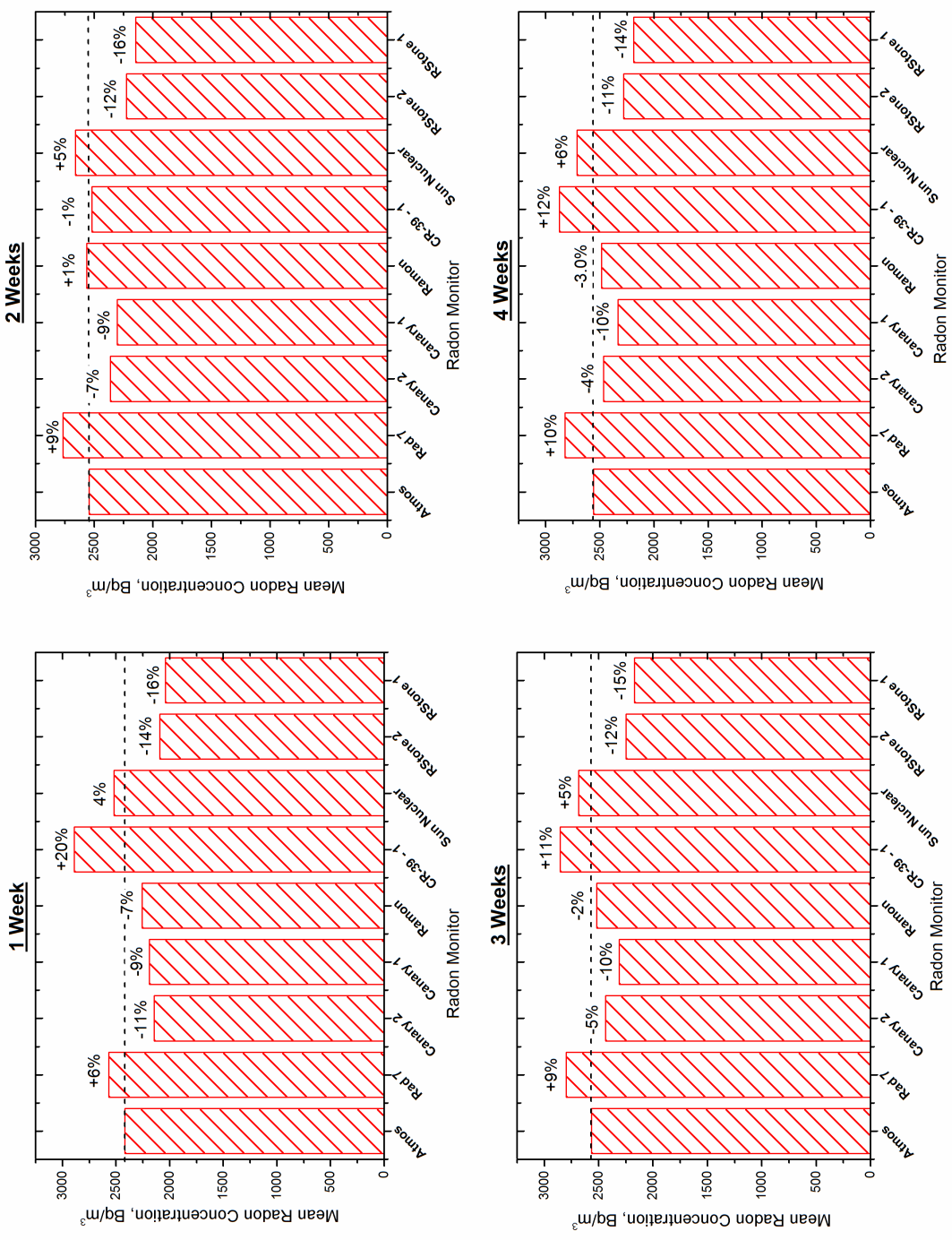


Figure 4.12: Comparison of radon detectors as a percentage difference from the Atmos for a 4 week measurement in the radon chamber.

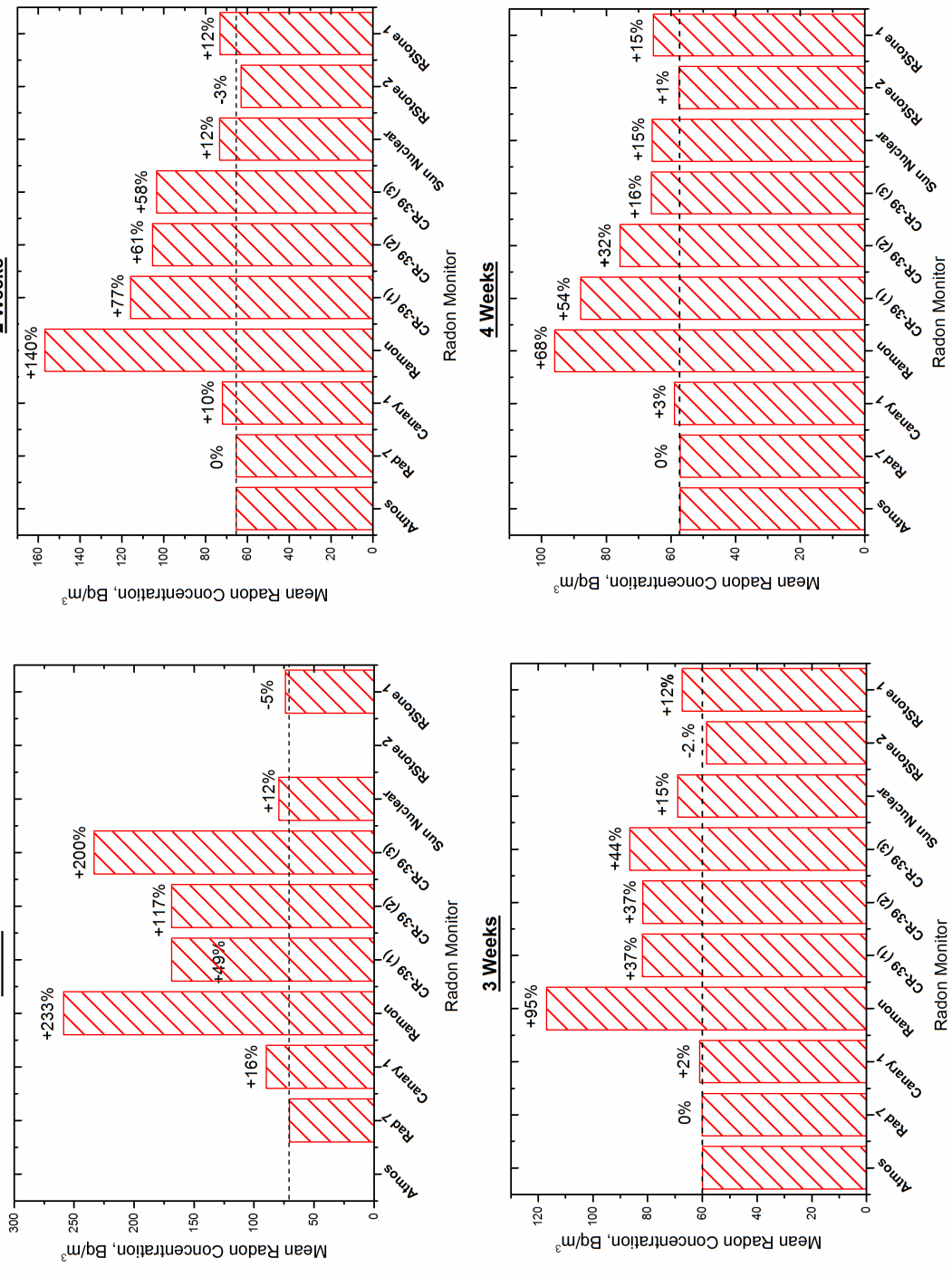


Figure 4.13: Comparison of radon detectors as a percentage difference from the Atmos for a 4 week measurement in a home.

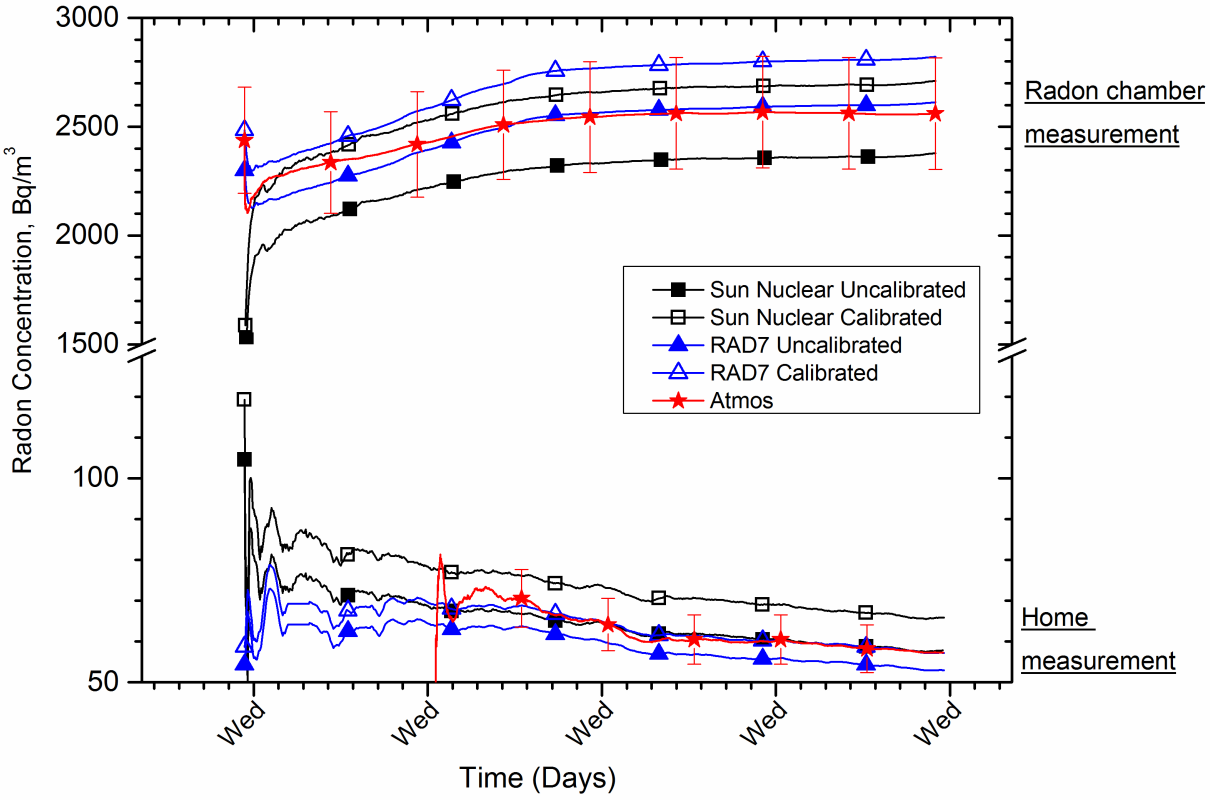


Figure 4.14: The effect of applying calibration factors on radon measurements.

the uncertainty in their measurements, the reported figures were very small and possibly underestimated. For example, the RAD7 ignores systematic bias due to uncertainty in calibration and traceability to national standard which can be up to 5%. According to the manufacturer's manual, the Atmos software uses 'one standard deviation' in the average radon concentration and is updated every 5 seconds'. Also, when detectors are calibrated by their manufacturers the software is updated to reflect any changes, but if the monitor is calibrated by another accredited facility the software is not usually updated. The accredited facility will include the background count of the monitor, the uncertainty in their calibration source and the uncertainty in their reference monitor. For these reasons, the uncertainties reported in the calibration reports were used for table 4.3. The uncertainties of the other detectors were calculated based on either guidance from the suppliers (Ramon and CR-39 detectors) or the stated accuracy in their manuals.

In the home environment, the 3 sets of CR-39 detectors recorded concentrations of 88±19 Bq/m^3, 76±19 Bq/m^3 and 66±17 Bq/m^3 for a 4 week period (table 4.3). Several years previously, the concentration in the same room and in a similar position was measured to be 39 Bq/m^3, also using a CR-39 detector but for a 3 month period. This indicates that short-term measurements of radon concentrations apply to that period only (within the stated uncertainty) and not to an annual average. However, when results are interpreted with caution, shorter measurements may be useful as a guide to whether a building exceeds the reference level. Rochford *et al.* aimed to correlate 1 month and 3 month measurements using CR-39 detectors but found that concentrations falling between 89 Bq/m^3 and 235 Bq/m^3 require a 3 month measurement to provide a sufficiently accurate value for a mean annual concentration [138]. As counting statistics are low for shorter periods, Hansen *et al.* investigated whether the measurement errors for CR-39 detectors could be reduced by using a larger surface area or by adjusting the PADC composition to increase sensitivity to low-angle alpha particles [139]. They concluded that, although increasing the surface showed no improvement, changing the composition showed potential and warranted further investigation. Furthermore, the percentage error could be reduced by almost 20% at 100 Bq/m^3 by increasing the measurement from 7 to 10 days.

Detector	**1 Week mean R_C, Bq/m^3**	**2 Week mean R_C, Bq/m^3**	**3 Week mean R_C, Bq/m^3**	**4 Week mean R_C, Bq/m^3**
Radon Chamber				
Atmos	2418 ± 243	2545 ± 256	2568 ± 257	2560 ± 256
Rad 7	2568 ± 205	2767 ± 221	2800 ± 224	2822 ± 226
RStone 1	2042 ± 98	2148 ± 103	2173 ± 104	2189 ± 105
RStone 2	2091 ± 90	2228 ± 96	2250 ± 97	2281 ± 98
Sun Nuclear	2209 ± 221	2333 ± 233	2802 ± 280	2780 ± 278
Ramon	2257 ± 41	2565 ± 30	2517 ± 25	2484 ± 21
Canary (1)	2189 ± 438	2304 ± 461	2313 ± 463	2334 ± 467
Canary (2)	2145 ± 429	2363 ± 473	2438 ± 487	2467 ± 493
CR-39 (1)	2890 ± 156	2521 ± 227	2855 ± 113	2874 ± 147
Home				
Atmos	-	65 ± 7	60 ± 6	57 ± 6
RAD 7	71 ± 6	66 ± 5	60 ± 5	57 ± 5
RStone 1	74 ± 3	73 ± 3	67 ± 3	65 ± 3
RStone 2	-	63 ± 3	58 ± 2	57 ± 2
Sun Nuclear	79 ± 8	74 ± 7	69 ± 7	66 ± 7
Ramon	259 ± 14	157 ± 8	117 ± 5	96 ± 4
Canary	90 ± 18	72 ± 14	61 ± 12	59 ± 12
CR-39 (1)	169 ± 47	116 ± 27	82 ± 18	88 ± 19
CR-39 (2)	169 ± 42	106 ± 27	82 ± 21	76 ± 19
CR-39 (3)	234 ± 59	104 ± 26	87 ± 22	66 ± 17

Table 4.3: Mean radon concentrations (R_C) for 1, 2, 3 and 4 week measurement periods in two different environments.

4.4 CONCLUSIONS

With a 4-week measurement and at concentrations around 2,500 Bq/m^3, 5 out of the 8 detectors in this study fell within the uncertainty of the reference Atmos detector. At concentrations of around 60 Bq/m^3, 3 out of 9 detectors were within the ±10% uncertainty of the Atmos and 6 out of 9 were within ±20%. If the aim of a measurement is to determine whether the radon concentration is above or below the reference level (100-400 Bq/m^3), then most detectors show a good comparison (±20%). If the aim of a measurement is to determine the radon concentration at various stages of remediation implementation, where radon concentrations might be higher (>2000 Bq/m^3), then most detectors show an even better comparison at this concentration (±10%). These results highlight that a one-point calibration is reliable only if it is performed close to the reference level unless the detector has a proven linear response, and it shows the importance of a multiple-point calibration when recording a range of radon concentrations. In addition to quality assurance, calibration is vital for determining the measurement uncertainty, which can be particularly significant for shorter periods.

Of the more expensive detectors, the RAD7 proved to give readings closest to those of the Atmos at both higher and lower radon concentrations. Although the cost is a consideration of how practical it would be to place it or the Atmos in a home for 1-4 weeks, they could be used to carry out 1-2 week measurements in a secured home or to calibrate frequently the lower cost detectors. Of the lower cost detectors, the Canary gave readings closest to those of the Atmos; in fact it measured within 10% of the Atmos for almost all of the time periods. It is cost effective, user friendly and portable and could easily be shipped to a home with basic operating instructions.

For the application of short-term measurements, radon concentrations are generally higher in winter so it is advisable to perform shorter measurements during these months to err on the side of caution. Although the results in this study have not been seasonally adjusted, appropriate adjustment factors should always be applied in practice [119]. Nevertheless, bearing in mind the limitations set by the uncertainties it is clear from this study that shorter measurements can be used to assess the concentration during and

after remediation, as an indicator for conveyancing or to indicate if there is a possibility that the radon concentration is above the reference level.

4.5 FUTURE WORK

Barros *et al.* conducted a study that compared winter short-term measurements (7-10 days) with annual measurements [140]. They found that short-term measurements predicted annual measurements greater than the USA reference level of 148 Bq/m^3, 88% of the time and concentrations greater than 88 Bq/m^3, 98% of the time. This study was conducted in the US where it is an acceptable protocol to conduct short-term measurements (as little as 2-5 days), to indicate the radon concentrations for real estate transactions. The United States of America-Environmental Protection Agency (USA-EPA) recommend conducting these screening test, under 'closed-house' conditions, whereby windows and outside doors are kept closed as much as possible during the test and 12 hours prior to commencing the test [65].

A beneficial extension of the work done in this chapter would be to compare 1 - 4 week measurements, using the monitors in this study, with the standard 3 month CR-39 measurements and for a range of radon concentrations. By increasing the amount of local published data on short-term radon testing and testing procedures, it is hoped that the Irish EPA can put forward strong recommendations and guidelines for the inclusion of radon testing during conveyancing of dwellings in Irish legislation.

If radon testing were required for conveyancing in Ireland, then an accreditation scheme for monitors and testing facilitators would need to be carefully laid out. Also, the concern of tampering with monitors during measurement would need to be addressed, for example using motion sensors as currently inbuilt in the RStone and SunNuclear. Where short-term measurements indicated concentrations above the national reference level, then it would be recommended to undertake a standard 3 month measurement to give a more accurate result. In order not to delay the conveyancing process in this circumstance, a clause could be introduced to the contract of sale whereby a nominal fee or bond is retained by the purchaser. In the event that remediation is required, following

confirmation by the 3 month measurement, then the bond would be allocated to this work. This 'radon bond' is currently recommended for conveyancing in the UK and is regularly used successfully.

5

RADON REMEDIATION AND FOLLOW-UP PROGRAMMES

5.1 INTRODUCTION

As discussed in Chapter 2, many countries have set a threshold radon concentration for the home environment, the workplace and drinking water. It is recommend that about this level, some specified action should be taken in order to reduce the exposure to radon. The reference level is set by individual countries on the advice of organisations such as the ICRP, the IAEA and the WHO. A table of the reference levels set in different countries was also presented in Chapter 2 (table 2.3). Ireland has set the reference level for indoor radon as 200 Bq/m^3 in a domestic dwelling and 400 Bq/m^3 for the workplace.

When a building is found to have rooms with radon concentrations above the reference level then some form of remediation should be implemented to reduce the concentration.

5.1.1 *Remediation*

In general, there are two ways in which to reduce indoor radon concentrations: either by preventing the radon from entering the building or by diluting the indoor high radon air with radon free or low radon air. The following are some common remediation methods used to reduce radon concentrations:

- Ventilation: As we saw in Chapter 3, the outside radon concentration is generally substantially lower (details are available from national or local authorities) than the indoor concentration, so by continuously replacing indoor air with outdoor air then radon dilution should occur [105]. Modern building regulations lay out mandates to provide adequate ventilation to ensure that harmful pollutants and moisture do

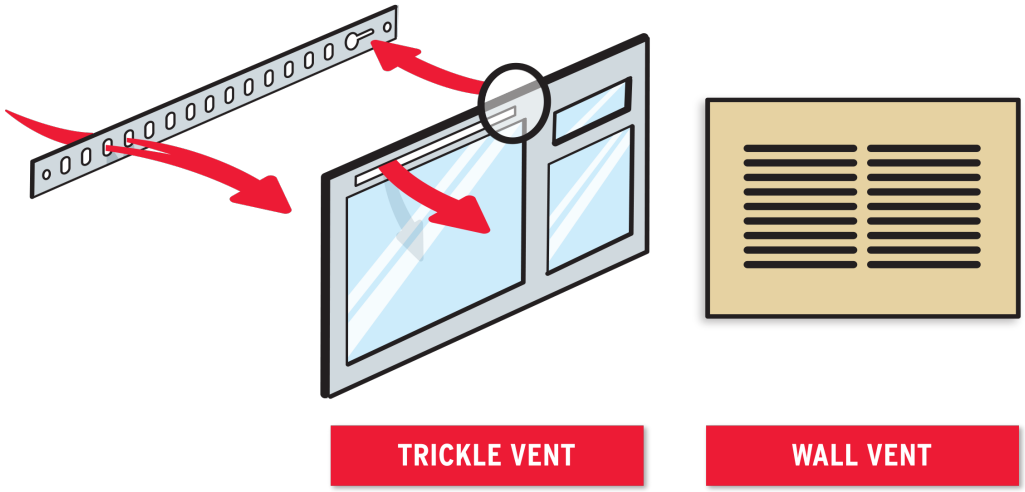
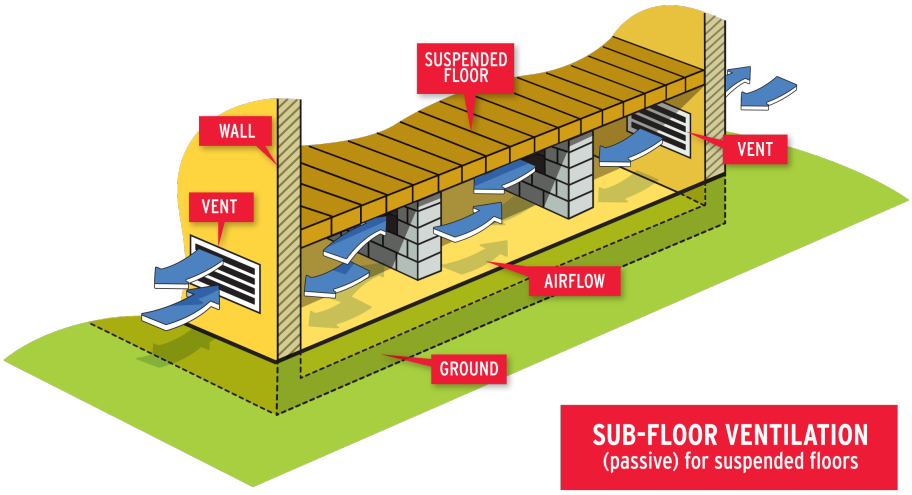


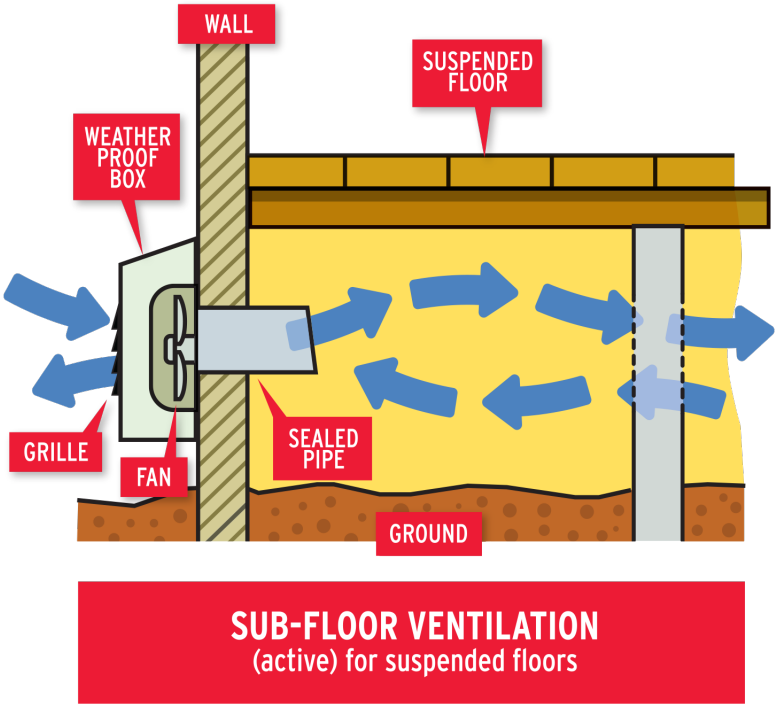
Figure 5.1: Passive ventilation (images courtesy of the EPA).

not build up inside buildings. There are different methods of ventilation which can be used to reduce the radon concentration indoors:

- Passive ventilation: Wall ventilation or window trickle ventilation can be installed to set up an air exchange (figure 5.1). Wall vents can be easily installed in existing buildings but they may not be a solution for larger buildings where some rooms have no outer wall. Passive ventilation is low cost and low maintenance and because of this, it is often used for slightly raised radon concentration or in combination with other remediation methods for higher radon concentrations.
- Underfloor ventilation: In buildings where the ground floor is suspended (timber or concrete floors) the use of airbricks or vents on outer, below floor level walls can set up an adequate underfloor ventilation, in order to prevent radon from entering the building and vent it directly to the outdoors instead (figure 5.2).
- Positive ventilation: The principle of this type of ventilation is to set up a positive pressure inside a building, thereby reversing the normal flow of radon into the building from the soil below. An intake of air from the outdoors would be drawn in to the attic space by a temperature (and hence pressure)



(a) Passive underfloor ventilation.



(b) Active underfloor ventilation.

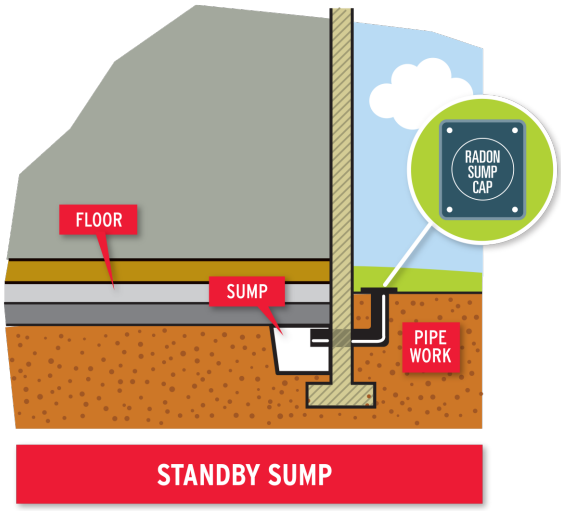
Figure 5.2: Underfloor ventilation (images courtesy of the EPA).

difference; in colder seasons this would be assisted by adding heating to the space. A fan in the attic is installed to create an airflow in to the rooms below via ceiling ducts and force the air to exit out of the building via gaps or openings in the doors, windows and service pipes.

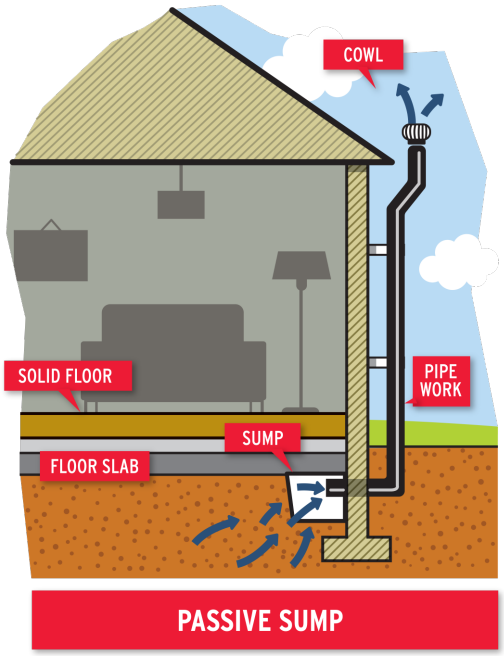
  - Balanced ventilation: This system of ventilation creates an air supply and an air extraction which when in equilibrium would prohibit radon being drawn up from the ground below. In warm climates, it can be achieved by opening two windows/doors in opposite sides of the building but in cold climates this is not practical so it can be achieved though mechanical systems with heating. Finnie *et al.* reported that, though this method of ventilation was less effective than other methods and some users reported condensation problems in colder climates, it was durable over several years [141].

- Sub-floor de-pressurisation (sump): A warm environment inside a house creates a pressure difference between it and the colder ground below; this draws radon from the soil/rocks in to the building. As the temperature difference is generally larger in winter, the pressure difference is larger, creating a higher radon concentration indoors. To counteract this pressure difference and alleviate the flow of radon to the indoors, a cavity can be created inside the foundation wall and linked to a mechanical (figure 5.3c and 5.3d) or passive extractor fan (figure 5.3b) on the outer wall through piping. The radon soil gas is vented to the outdoors and quickly dissipates. Where the radon is vented at ground level, the vent will need to be positioned away from windows and doors so as to avoid it re-entering the house. This method of remediation is called sub-floor de-pressurisation or a sump. In Ireland, building regulations specify that dwellings built since 1998 must be fitted with a standby sump (figure 5.3a) which can be later activated in the event that elevated radon concentrations are found inside the dwelling [142].

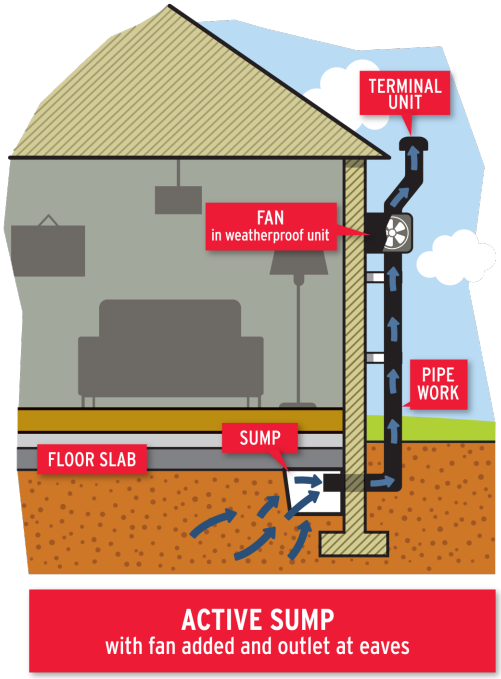
- Sealing: During the construction of new buildings it is favourable to lay a radon proof membrane which will prevent radon rising up through the foundations and into the building. In some countries, as is the case in Ireland, laying a radon membrane has been incorporated into the building regulations particularly in



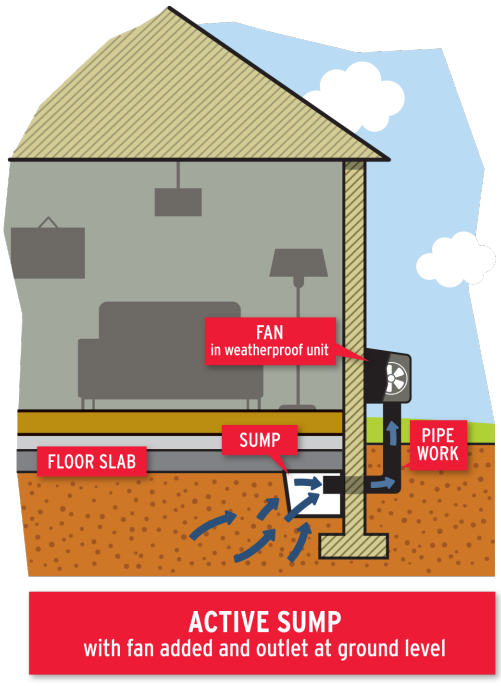
(a) Standby sump.



(b) Passive sump.



(c) Active sump (high vent).



(d) Active sump (low vent).

Figure 5.3: Sub-floor de-pressurisation or sump systems (images courtesy of the EPA).

high radon areas [142]. Other ways to seal a building are to fill cracks in the floors and walls or around entry points such as service conduits. Sealing is usually done in combination with another mitigation method as it can be difficult, almost impossible, to seal all entry routes.

Several publications detail the types and effectiveness of remediation methods [141, 143–147]. Hodgson *et al.* in particular give a very comprehensive report on the different types of remediation, their effectiveness when implemented alone and in combination, the effect of different house characteristics on remediation success, the cost of the remediation types and dose reduction calculations following remediation, for a dataset of approximately 2,400 dwellings [145]. Both this study and Finnie *et al.* examined the radon reduction for the different types of remediation methods [141]. They found sub-floor de-pressurisation to be the most effective, and sealing and passive ventilation to be the least effective.

Fewer publications look at how the remediation methods perform after several years [141, 148]. Finnie *et al.* looked at the radon concentration 7 - 10 years later in 224 homes and found the radon reduction was 50 - 80% following remediation, but 7 - 10 years later the methods were not as effective and the reduction was 30 - 80%. Rydock *et al.* conducted their study in a smaller sample of 12 homes and found that only 3 out of the 12 homes remained below the 200 Bq/m^3 reference level after 10 years.

Building upgrades and insulating improvements can have an impact on radon remedies. A UK modelling study predicted an increase in the radon concentration by up to 57% if the air tightness of the dwelling was increased [149]. An experimental and comprehensive Swiss study showed an average increase of 26% in 163 dwellings which were measured before and after thermal retrofitting, with a 35% increase in radon concentrations seen due to replacing of windows alone [150]. A study completed in Ireland also found that the type of retrofitting dictated whether the radon concentrations would increase. Wall cavity and loft insulation showed no impact on the radon concentration but replacing or sealing doors and windows had a greater effect [151].

The type of remediation implemented will depend on the radon concentration, the structure and the type of building. Often a radon adviser is contracted to determine which method is most suitable, and it is also common that a combination of methods is

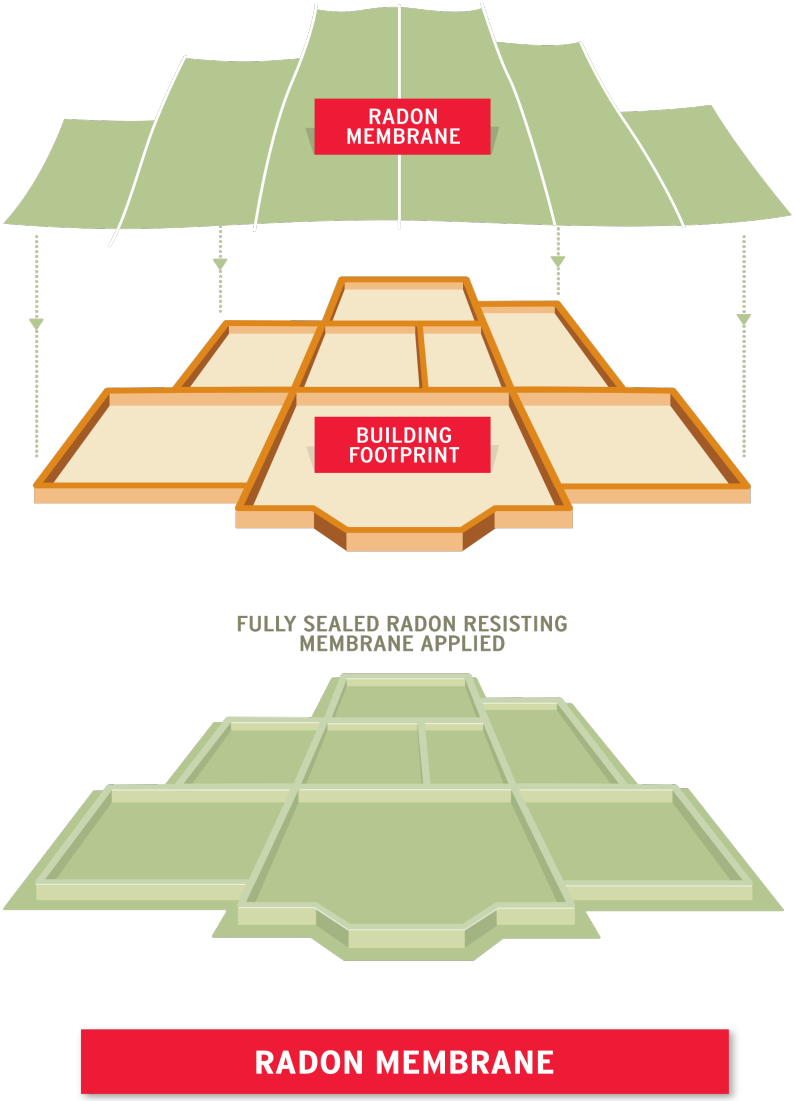


Figure 5.4: Radon membrane (image courtesy of the EPA).

installed. When remediation has been implemented, a re-test should be performed to confirm that it has been successful. Following this, the building occupier will need to maintain the radon system and, though not specifically recommended in Ireland, complete intermittent retests, the frequency of which will depend on the remediation. Passive ventilation or sumps need little maintenance but mechanical methods need a regular service programme because if there is a fan failure then the radon concentration will increase again. The lifetime of the mechanical unit would be stated by the manufacturer and this would dictate the need for servicing or replacement. The EPA recommend checking active sumps every 5 years: Rydock *et al.* also recommend this time frame [148].

This chapter describes a project which re-visits schools that have been remediated 10 - 17 years previously to determine the durability of the remediation method implemented and consequently looks at the value of structured follow-up programmes. The study also hopes to ascertain the level of radon awareness in Irish schools and to ascertain the effects of subsequent retrofitting on the radon concentrations.

5.1.2 *National survey of radon in Irish schools*

In Ireland, during the years 1998 and 2004 the DES, with the assistance and guidance of the EPA, undertook a large scale project to measure the radon concentrations in primary and post primary schools in Ireland called the National School Survey (NSS). In total, the radon concentration in 3,826 schools and 38,531 rooms was measured by the EPA using CR-39 detectors, and due to the size of the project it was completed over several phases. The average radon concentration was found to be 93 Bq/m^3; 3028 (7.9%) rooms had a radon concentration greater than 200 Bq/m^3 and 800 (2.1%) rooms had concentrations greater than 400 Bq/m^3 [76]. These results are summarised in figure 5.5 [76].

A radon remediation adviser from Cornwall County Council, UK, was contracted to implement a strategy for reducing the radon concentration in all rooms that were greater than 200 Bq/m^3 and local radon contractors implemented the strategy. The reference concentration of 200 Bq/m^3 was used because at the time the reference levels

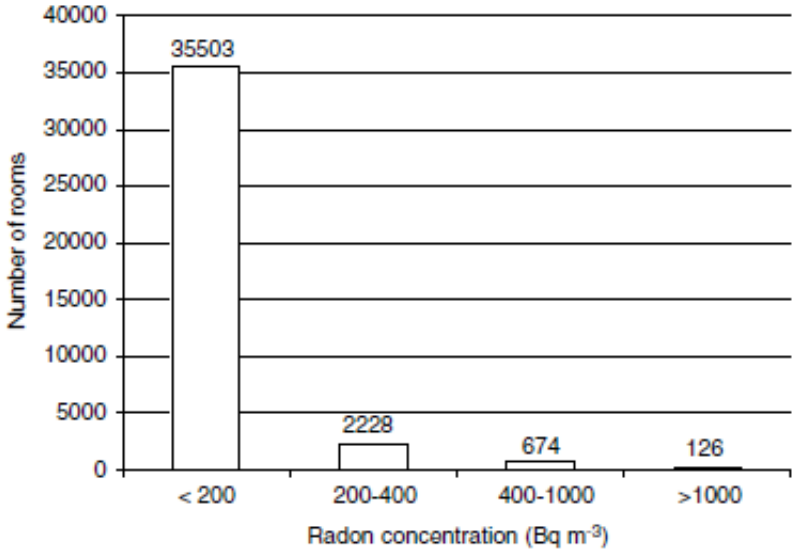


Figure 5.5: Distribution of room radon concentrations: results from the National School Radon Survey.

in Ireland were 200 Bq/m^3 for a home and 400 Bq/m^3 for a place of work, and the view was taken by the EPA that '*children should expect the same level of protection while at school as in the home*' [76]. In general, for concentrations $>$ 400 Bq/m^3 an active sump was installed, and for concentrations between 200 - 400 Bq/m^3 passive ventilation was implemented such as window or wall vents. Synnott *et al.* summarised the project and detailed the remediation methods which were put in place [76]. Many schools were re-tested to confirm if the remediation was successful, and a follow-up publication by Synnott *et al.* looked at the effectiveness of the remediation methods implemented [152]. Where the remediation was unsuccessful but was in the range 200 - 400 Bq/m^3 then no further remediation work was implemented as it was viewed as being below the reference level for a workplace, but in the instances where the concentration was greater than 400 Bq/m^3 then further remediation was implemented and the room was remeasured for confirmation.

Several countries have undertaken school radon surveys, and Clouvas *et al.* included a summary of the studies done in Europe when publishing their work on a Greek school survey [153]. The Irish survey is exceptional in its scale and comprehensiveness, and the volume of information obtained makes it ideal for the follow-up remediation study detailed in this chapter.

5.1.3 *Reference levels for schools*

As mentioned in section 2.2.1, several studies have been undertaken to state conclusively whether children are more susceptible to the health effects from radon than adults [31–33], and there are other publications that summarise these finding in order to compare the conclusions [34]. Currently it remains inconclusive, but as further research is carried out and if it becomes scientifically accepted that children are more susceptible, then a cautious and stringent approach would be advisable to ensure the concentrations are below 100 or 200 Bq/m^3 in classrooms.

The WHO advises setting the reference level as low as possible and proposed 100 Bq/m^3 [51]. In 2013, the EU published an updated directive, Council Directive 2013/59/Euratom, which stated that the reference level for a workplace should be set no higher than 300 Bq/m^3, unless 'warranted by national prevailing circumstances' [117]. The directive, which has an implementation date of February 2018, stated no explicit advice for setting reference levels for schools but instead, categorised them as a type of workplace or building with public access. Though the statutory reference level in the workplace in Ireland is 400 Bq/m^3, the cautious approach which the EPA adopted when setting the reference level of 200 Bq/m^3 for schools during the NSS was warranted, and in 2014 the National Radon Control Strategy recommended retaining this reference level of 200 Bq/m^3 for schools [2]. A radon concentration of 200 Bq/m^3 results in an equivalent dose of 5 mSv per year for a dwelling assuming an occupancy time of 7000 hrs [79]. For schools, the occupancy is assumed to be a minimum of 1000 hrs, which at a radon concentration of 200 Bq/m^3 would result in an effective dose of 0.7 mSv per year.

5.2 EXPERIMENTAL METHOD

The data from the original National School Survey (NSS) were supplied to this follow-up study (2014 - 2015) by the EPA and the DES.

For the follow-up study, 16 schools which had remediation implemented during the original NSS were revisited and tested for radon. 63% of the schools are located in high

radon areas as defined by the EPA's radon map for Ireland (figure 2.6) [6]. 77% of the rooms measured in this follow-up survey were originally greater than 200 Bq/m^3.

The follow-up survey was carried out in two phases. The first phase was a pilot survey, undertaken in six schools to highlight any problems that may arise. The six schools were chosen from the NSS data by the percentage of rooms which originally had radon concentrations $>$ 200 Bq/m^3, in order to obtain a sufficient data set to make conclusive comparisons of rooms with radon remediation.

It was decided to place and collect the detectors in person rather than use a postal service, as a site visit would enable an examination of the remediation methods to be carried out where possible and previous studies which used postal services reported a 26% non-return rate of detectors [76].

The schools were contacted and asked to partake in this radon follow-up study. Each school was then visited by the present researcher and a CR-39 detector was placed in each occupied ground floor room of the schools which remained *in situ* for 3 months, from March 2014 to June 2014. During the school visit, a short questionnaire was given to the principal (or a person assigned by the principal) for completion asking:

- Whether the school had a radon re-testing policy and if so to provide the results
- If any maintenance had been carried out on the currently installed remediation
- If any retrofitting had been done to the school
- If new extensions have been tested for radon or fitted with any remediation.

From the questionnaire, it was hoped to ascertain the level of radon awareness in the school and the extent of any radon programme present.

Following the 3 month detection period, the detectors were collected in person and analysed by the EPA in a similar protocol to that detailed by Fennell *et al.* [6], but a higher etch temperature of 95 ^{o}C and reduced time of 1 hour was used. The accuracy of these CR-39 detectors are $\pm$ 25%.

One of the issues highlighted in the visits to these six schools was the lack of consistency of room names from the NSS data and the current room name. In a number of schools, classrooms were traditionally named after the teacher occupying the room but 10 - 17

years later many teachers moved rooms, moved schools or retired. Correlating rooms listed in the NSS (1998-2004) with the current room names relied on the knowledge of staff members.

During the original NSS, the contracted radon adviser compiled a report for 93 of the remediated schools. This report detailed the radon concentration in each room with floor plans, the type of remediation implemented and photographs of the schools. For phase 2 of this follow-up study, the remaining schools were chosen from this report in order to simplify the task of correlating rooms from the two studies.

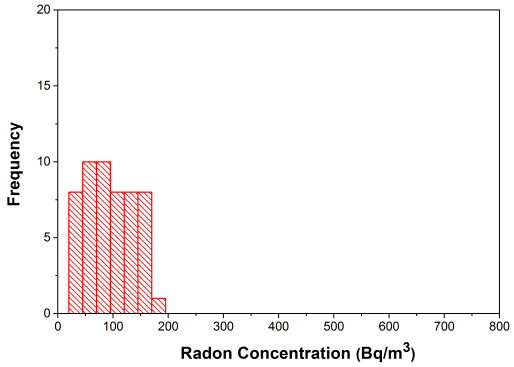
The second phase was conducted the following school year from January 2015 to June 2015, and again the schools were visited by the researcher and the CR-39 detectors placed in person. The remediation methods were also inspected where possible and a questionnaire completed by the principal or an assigned person.

In total, 276 rooms in 16 schools were measured for the follow-up study and the results were issued in a report to each school. In each report details were given of any remedial action needed. In the next section, the follow-up results are analysed and compared to the original NSS results.

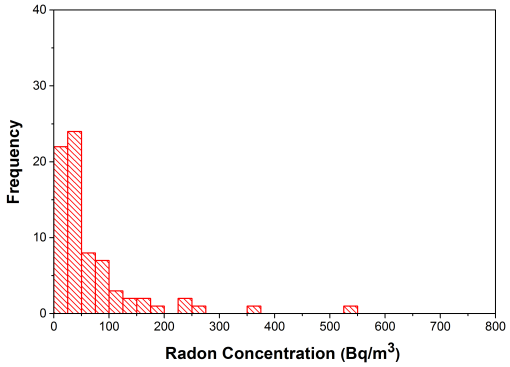
5.3 RESULTS AND ANALYSIS

Table 5.1 summarises the results from the follow-up study performed in 16 schools. No school reported a concentration less than 10 Bq/m^3 as this is the lower limit of detection of the CR-39 detectors supplied by the EPA [76].

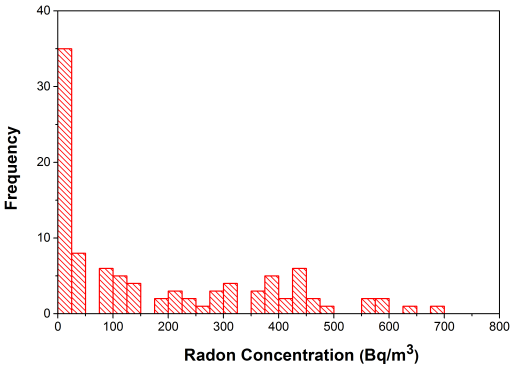
The arithmetic mean (AM), the geometric mean (GM) and the median are 3 different statistical methods of stating the centre of a data set, depending on what information needs to be presented. In table 5.1, the AM for Counties Galway and Wicklow are closely matched, Co. Dublin is slightly elevated and Co Kerry has a high AM which is approaching the Irish reference level for schools of 200 Bq/m^3. The arithmetic standard deviation (ASD) for Co. Kerry is 190 Bq/m^3 , indicating that the spread of values in this data set is large and possible outliers of high radon concentration are skewing the AM. This is confirmed by looking at the frequency distribution of the data for each county



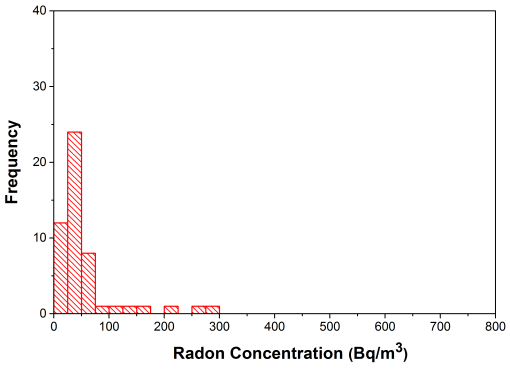
(a) Follow-up survey data for Co. Dublin.



(b) Follow-up survey data for Co. Galway.



(c) Follow-up survey data for Co. Kerry.



(d) Follow-up survey data for Co. Wicklow.

Figure 5.6: Histograms of frequency distribution for data of the follow-up survey in bins of 25 Bq/m^3.

in figure 5.6. Though the majority of the data for Co. Kerry (figure 5.6c) is below 100 Bq/m^3 there are several outliers $>$ 200 Bq/m^3 up to a maximum of 697 Bq/m^3. Stating the GM for each county gives a better indication of the radiation dose received by the majority of school occupants as this mean is not skewed by high value outliers to the same extent as the AM. It is, however, important to state the AM, as when it is quoted with the ASD data outliers can be identified to indicate where remediation methods have failed. The median for each county correlates quite closely with the respective GM.

	Co. Dublin	**Co. Galway**	**Co. Kerry**	**Co. Wicklow**	**Total**
No. of schools	3	6	4	3	16
No. of rooms	53	74	98	51	276
AM ± SE, Bq/m^3	94 ± 6	68 ± 10	182 ± 19	55 ±8	111 ± 8
ASD, Bq/m^3	43	87	190	58	136
GM, Bq/m^3	83	42	82	41	60
GSD, Bq/m^3	2	3	4	2	3
Minimum, Bq/m^3	23	10	10	12	10
Median, Bq/m^3	89	34	100	35	51
Maximum, Bq/m^3	181	541	697	293	697
Range, Bq/m^3	158	531	687	281	687

Table 5.1: Summary of the results from the follow-up school radon study in statistical format. AM is the arithmetic mean with standard error (SE) and arithmetic standard deviation (ASD), GM is the geometric mean and GSD is the geometric standard deviation.

5.3.1 *School level descriptive statistics*

As stated in section 5.2, results from the NSS were provided to this follow-up study by the EPA in order to make comparisons. These supplied results also included post-remediation measurements which were performed to confirm if the remediation implemented at the time was successful.

Table 5.2 presents the summary statistics for the three stages of the survey: the initial NSS, the post-remediation survey (completed between 1998 - 2004) and the follow-up survey completed in 2014 - 2015. Some rooms which were measured in the NSS were not remeasured post-remediation, particularly, if they previously measured < 200 Bq/m^3. Also, some additional rooms were measured in the follow-up study which were not measured in the NSS, possibly due to unreturned detectors in the NSS or additional rooms built in the interim. Synnott *et al.* [76] found that the nationwide AM in schools was 93 Bq/m^3, but here a higher AM of 520 Bq/m^3 is reported; this is due to the bias in choosing schools with elevated radon concentrations, but we can see that the AM post-remediation was 79 Bq/m^3 following successful remediation. In the follow-up survey, the AM has risen to 111 Bq/m^3 which indicates that either some remediation methods are not working as efficiently as when first installed or that some have failed. If the GM is compared for the 3 phases then it is seen that the post-remediation and follow-up values

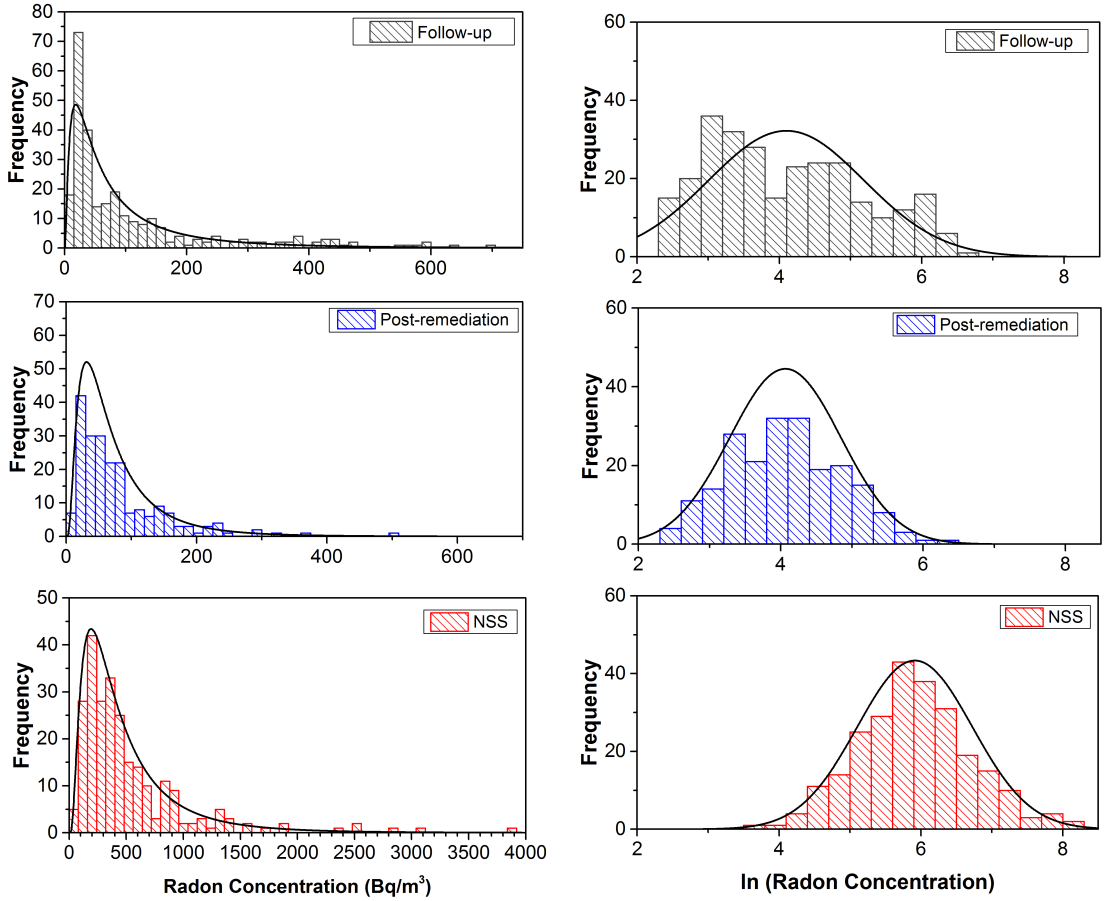
	National school survey	Post-remediation	Follow-up survey
No. of rooms	250	209	276
AM ± SE, Bq/m^3	520 ± 33	79 ± 5	111 ± 8
ASD, Bq/m^3	525	69	136
GM, Bq/m^3	370	58	60
GSD, Bq/m^3	2	2	3
Minimum, Bq/m^3	39	10	10
Median, Bq/m^3	356	57	51
Maximum, Bq/m^3	3848	499	697
Range, Bq/m^3	3809	488	687

Table 5.2: Statistical data for comparison of three surveys.

are almost equal; therefore it can be deduced that some of the remediation methods failed, resulting in outliers in the data.

The frequency distribution of indoor radon concentration generally varies log-normally [96, 154]. If the data from table 5.2 is represented in graphical format (figure 5.7a), it is seen that the frequency distribution of the NSS data follows a log-normal distribution as expected. The post-remediation data and the follow-up study data do not follow a log-normal distribution, due to remediation methods having been implemented. With a log-normal scaled abscissa, figure 5.7b depicts the frequency distribution of the log-normal radon concentration with a normal fit curve; again only the NSS data follows the fitted normal curve.

Often this log-normal relationship is used to predict how many homes are above the reference level or have high concentrations. This is done by plotting a log-normal probability plot of a sample dataset to obtain an indication for a larger cohort, perhaps a country [155]. Figure 5.8 shows the log-normal probability plots of the data for 16 schools in this study from the NSS, the post-remediation survey and the follow-up survey. Though this sample of data is biased because schools with radon concentrations > 200 Bq/m^3 were specifically chosen, the NSS data still follows the predicted reference line over most of the distribution with only small deviations at the upper and lower ends of the spectrum. The log-normal probability plots for the post-remediation data and the follow-up data deviate significantly in the lower and upper ends of the spectrum; this



(a) Frequency distribution of radon concentration with log-normal fit.

(b) Frequency distribution of the log-normal radon concentration with a Gaussian fit.

Figure 5.7: Lognormal distribution of radon concentration.

can be attributed to the remediation work implemented and also to the lower limit of detection of the CR-39 detectors (10 Bq/m^3).

5.3.2 *Room level descriptive statistics*

In total, 224 rooms from the NSS could be correlated with the rooms in this follow-up study. 52 new room measurements in the follow-up study could not be correlated with rooms in the NSS, this was due to the building of new rooms, the combining of rooms or rooms that were not measured in the NSS as a result of unreturned detectors. Table 5.3 compares the data from the NSS to the correlated and uncorrelated data from the follow-up survey with respect to three reference levels; 200 Bq/m^3 is the reference level

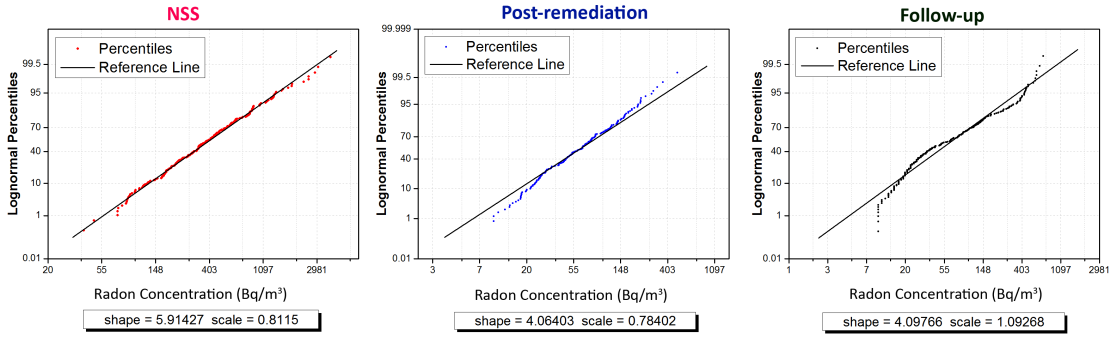


Figure 5.8: Probability of lognormal distribution.

for dwellings and schools in Ireland, 300 Bq/m^3 is the new workplace reference level set out in Directive 2013/59/ Euratom and 400 Bq/m^3 is the current Irish workplace reference level. Though the correlated and uncorrelated data are for different rooms, their close agreement confirms the trend for the percentage of rooms which are now above the three reference levels. The follow-up survey also highlighted 8 rooms which previously had no radon measurement, most probably due to unreturned detectors, but now measured $>$ 200 Bq/m^3 . Occupants of these rooms have been unknowingly exposed to these concentrations for possibly a long period of time.

		Follow-up survey	
Reference level	**Correlated NSS**	**Correlated rooms**	**Uncorrelated rooms**
$>$ 200 Bq/m^3	77%	16%	14%
$>$ 300 Bq/m^3	60%	8%	9%
$>$ 400 Bq/m^3	44%	5%	5%

Table 5.3: Percentage of rooms exceeding the reference levels in the NSS and the follow-up survey. The 'correlated rooms' column refers to rooms in the follow-up study which could be matched with the rooms from the NSS and similiarly for the 'uncorrelated rooms' column.

As explained in section 5.1.2, post-remediation measurements were not available for all rooms in this study because some schools did not carry out post-remediation testing and some rooms could not be correlated, so it can not be stated with certainty that the 224 correlated NSS and follow-up survey rooms were $<$ 200 Bq/m^3 following remediation. A smaller data set of 146 rooms could be correlated for the three stages: the NSS, the post-remediation survey and the follow-up survey, of which 129 were $>$ 200 Bq/m^3 during the NSS and had remediation implemented. Table 5.4 summarises the analysis of

Reference level	Correlated NSS, no. of rooms	Correlated PR survey, no. of rooms	Correlated FU survey, no. of rooms
> 200 Bq/m^3	129	5	34 (26%)
> 300 Bq/m^3	106	2	23 (18%)
> 400 Bq/m^3	79	0	13 (10%)

Table 5.4: Correlated data for 129 remediated rooms from the national school survey (NSS), the post-remediation survey (PR) and the follow-up survey (FU).

Reference level	National School Survey (1998-2004), no. of rooms	Prediction for 2015, no. of rooms
> 200 Bq/m^3	3028	790 (26%)
> 400 Bq/m^3	800	80 (10%)

Table 5.5: Extrapolation of the results of the follow-up survey to predict the number of rooms currently above 200 Bq/m^3 and 400 Bq/m^3 in 2015 in Irish schools.

these 129 rooms. Similar to the data in tables 5.2 and 5.3, it is seen that remediation was successful in reducing the radon concentration in all the rooms to below 400 Bq/m^3 and 97% to below 200 Bq/m^3. Measurements taken 10 - 17 years later in this follow-up survey indicate that 26% of the rooms are now above 200 Bq/m^3 and 10% of these rooms are now above 400 Bq/m^3 .

Synnott *et al.* reported that following the National Survey of Irish Schools, 3028 rooms were found to be greater than 200 Bq/m^3 and 800 were found to be greater than 400 Bq/m^3 [76]. If these data are used and the data from table 5.4 extrapolated, it can predicted that 790 rooms are currently above the Irish reference level for schools (table 5.5). This prediction does not take into account the 5% of schools that were not measured during the NSS or schools built since the NSS. Schools built since 1997 would adhere to the updated building regulations requirements for improved ventilation. More recently, the DES require that all new build schools must install a radon membrane, and once occupied must conduct radon tests [142].

5.3.3 *Radon awareness in schools*

From initial contact with the schools during the pilot survey, it was seen that the level of awareness and interest in radon varied greatly. To gain an overall perspective on the extent of awareness in the schools, the results of a simple questionnaire completed by a staff member at the school are summarised in table 5.6. 63% of the schools had re-tested for radon since the NSS programme ended, the majority of which (60%) are located in high radon areas of Co. Kerry and Co. Galway (figure 2.6). The level of awareness due to media attention and journal publications is likely to contribute to these schools implementing a re-testing programme [6, 54]. Only 15% of the schools had a protocol in place to maintain or check the installed remediation at regular intervals.

Of the schools which had built extensions, it was noted that a high percentage (83%) had installed a radon remediation system. Upon further investigation though, it was found that in each circumstance window trickle vents had been installed. These are a ventilation requirement set out in the building regulations, and therefore a remediation system was not intentionally installed [142]. Only two schools had measured the radon concentration in the new extension; again these schools were located in high radon areas of Co. Kerry, showing the influence of radon awareness on the protection against radon.

50% of the schools in this survey had some form of retrofitting done since the NSS, this was in the form of window replacement, wall insulation, roof replacement or heating system replacement. Of the seven schools which had raised radon concentrations in the follow-up study, four of them had retrofitting installed since the NSS. Due to the small dataset and the time lapse between post-remediation and the follow-up survey, it is difficult to state with certainty that these modifications had an effect on the radon concentrations. Also, remediation failure would have to be excluded as the cause of radon concentration increase before it could be attributed to the retrofitting.

5.3.3.1 *Case Studies*

In order to gain more insight into the influence that radon awareness and radon programmes have on schools, 3 schools are highlighted: case study 1 is in a high radon area with a limited radon programme (re-testing only), case study 2 is located in a

	No. of schools
Retrofitting installed since NSS	50%
Remediation system maintained	15%
Re-testing programme	63%
Remediation installed in extensions	83%

Table 5.6: Review of radon awareness questionnaire.

high radon area with a comprehensive radon programme (re-testing and remediation maintenance) in place and case study 3 is located in a low radon area with no radon programme. Figures 5.9, 5.10 and 5.11 are box charts for the three schools; the inner box depicts the mean radon concentration, the outer box is the standard error and the dash line is the median value. The whiskers define the 5th and 95th percentile range and the star represents the maximum and minimum radon concentrations.

- Case Study 1: This school is a large secondary school located in a high radon area of Co. Kerry. The Principal indicated that, to his knowledge, no maintenance had been carried out on the remediation methods (eight sub-slab pressurisation sumps as advised by the NSS radon contractor) but radon measurements had been carried out two years previously (2013) and the results were provided. The box graph (figure 5.9) indicates that two attempts were necessary in order to reduce the mean radon concentration to < 200 Bq/m^3 during the NSS, but the 2013 (Intermediate 1) radon measurements and the measurements for this survey (which are in close agreement) show that the mean radon concentration has risen to > 400 Bq/m^3. No explanation was given as to why no action was taken following the 2013 survey. Lack of radon awareness has given rise to staff and students at this school being exposed to a mean radon concentration of 433 Bq/m^3 and a median value of 405 Bq/m^3 for a minimum of two years.

- Case Study 2: This school is a medium size secondary school located in a high radon area of Co. Kerry. The principal stated that a local radon adviser was contracted by the school to maintain the remediation methods (seven sub-slab pressurisation sumps were advised by the NSS radon contractor) and that two radon surveys had been completed since the NSS; the results of both were provided to this survey.

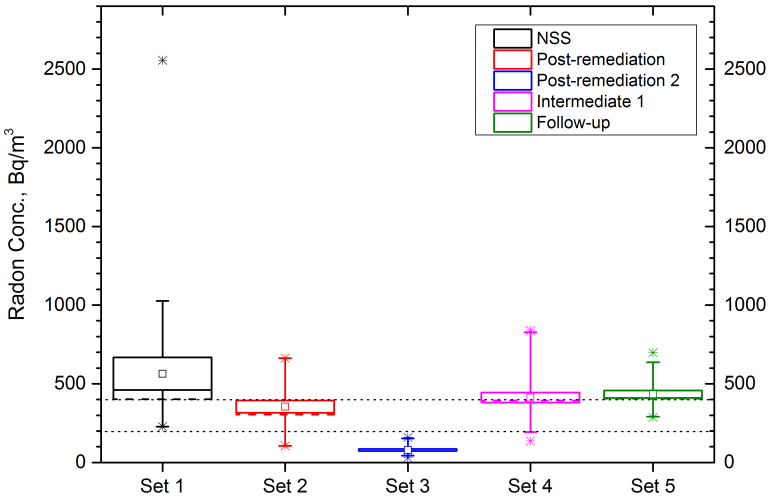


Figure 5.9: Case study 1 - Co. Kerry.

The box chart for this school (figure 5.10) indicates that rooms with high radon concentrations were identified in a retest ('Intermediate 1' dataset). Consequently, the school repaired the remediation system in the affected rooms and remeasured the radon concentrations. The 'Intermediate 2' dataset confirmed that the repair was successful and the mean radon concentration was reduced to < 200 Bq/m^3.

- Case Study 3: This school is located in a low radon area of Co. Wicklow. The Principal was not aware of the remediation methods installed in the school, so no maintenance was carried out and no intermediate testing was undertaken. According to the radon consultants' advisory report, three radon sumps should have been fitted to serve six rooms and four other rooms fitted with permanent trickle/window ventilation. These were confirmed at the time of detector placement, but one sump was found switched off and did not seem to be functioning (no noise when it was turned on). A new room had been added to the school and trickle vents were present in accordance with building regulations. From the box chart (figure 5.11), it can be seen that the mean radon concentration for the school is elevated in comparison with the post-remediation mean value and a sump failure has caused the radon concentration to increase to > 200 Bq/m^3 for the 2 rooms that it served, unbeknownst to the occupier.

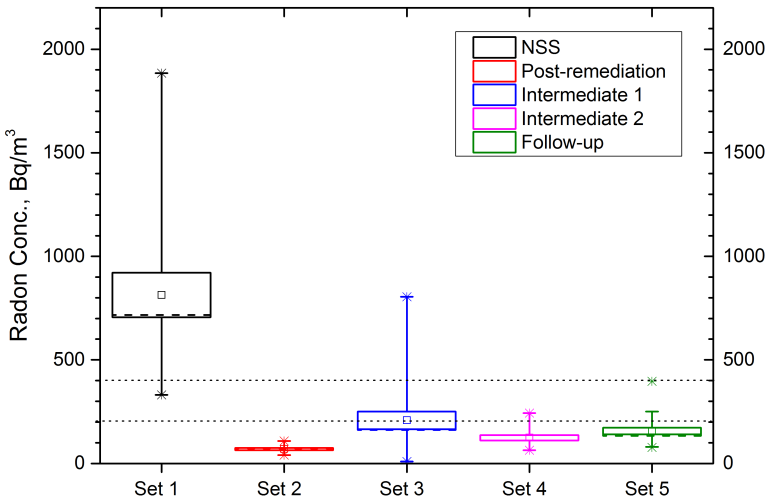


Figure 5.10: Case study 2 - Co. Kerry.

During visits to the school, most school principals were eager to learn about radon and to facilitate the follow-up programme. Case study 1 demonstrates the importance of schools being actively involved in maintaining radon remediation and responding to elevated radon measurements to avoid the staff and students being overexposed. Radon awareness and the presence of a radon programme are key to schools maintaining their radon concentrations below the reference level.

5.4 CONCLUSIONS

The results in this study show a trend of increasing radon concentration in schools which had remediation implemented 10-17 years previously. Table 5.4 is a significant summary of this trend as it highlights the consequence when remediation methods are not maintained and there is no follow-up programme in place. In rooms where remediation was installed, it is seen that 26% of the rooms have risen to above the reference level of 200 Bq/m^3 and 10% are now above 400 Bq/m^3. The results of this follow-up study lead to the prediction that currently 790 rooms are $>$ 200 Bq/m^3 and 80 rooms are $>$ 400 Bq/m^3 in the 3826 schools originally tested..

Through case studies and a questionnaire, the radon awareness in schools was observed to be correlated to an ability to maintain radon concentrations under the reference

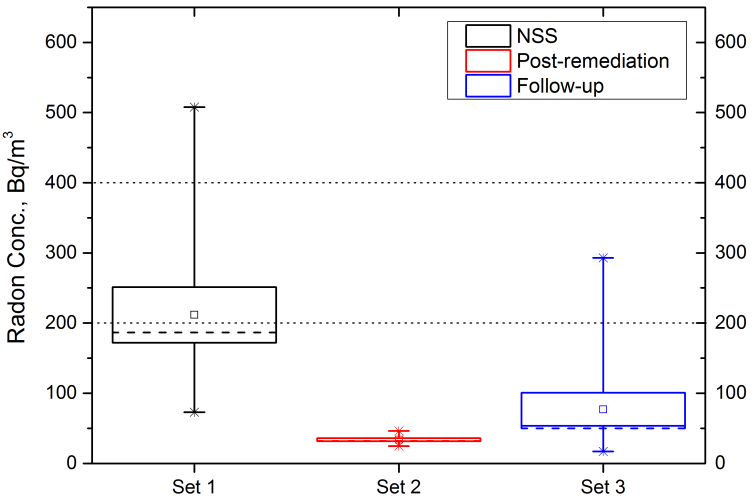


Figure 5.11: Case study 3 - Co. Wicklow.

level. Due to media attention and publications, high radon areas had a higher radon awareness and consequently a higher rate of retesting: a unique finding from this study. Media attention is not constant, and therefore it should not be relied on to create radon awareness.

Only 15% of the schools in this study regularly checked or serviced their remediation systems. When a remediation system fails, the radon concentration returns to an elevated level and the consequence of this was seen in Case Study 1. Students and staff in this school were exposed to a radon concentration in excess of 400 Bq/m^3 due to a lack of action in maintaining remediation systems. Testing for radon is crucial in indicating where remediation methods are less effective or have failed. A structured programme for all schools to re-test for radon and to service mechanical remediation systems at regular intervals should be incorporated in to the school's maintenance programme.

5.5 FUTURE WORK

5.5.1 *Implementing a radon programme in Irish schools*

Although the National Survey of Radon in Irish Schools is to be commended for its exhaustive completeness, a 'National School Follow-Up Programme' should also be

implemented to respond to the trend of rising radon concentrations reported in this follow-up study. Giving schools the knowledge and authority to manage their own remediation excludes the need for a repeat of the large scale NSS in the future. The following actions are recommended as a result of this study:

1. Schools with remediation systems currently installed should be prioritised and requested by the DES to have any occupied ground floor or basement rooms re-tested if not done in the last 5-10 years or since retrofitting works. If a radon test indicates an elevated reading then any mechanical remediation system associated with the room should be serviced or replaced. It is recommended that regular radon testing and remediation maintenance be included in the school's maintenance programme.

2. Most schools in this project were unaware of the remediation system installed and therefore, if possible, the original NSS report which detailed the remediation plan for the school should be reissued to the 93 schools.

3. The DES currently require newly built schools to install a radon membrane and to test for radon once occupied. If concentrations above 200 Bq/m^3 are found then the building contactor is obliged to install a remediation system to reduce the concentration. In such instances, it is recommended that regular radon testing and remediation maintenance be included in the school's maintenance programme.

4. All schools should be issued with radon awareness information and requested to measure their school following completion of any building retrofitting or if not measured previously, whether they are located in a high radon area or not.

5. It is recommended that a strategy be designed by the DES to educate school staff (especially the Principal and the Board of Management) on the basic principles of radon measurement and remediation to enable them to manage their own radon programme.

6. If the subject of radon were included in the education curriculum, a greater level of awareness of radon would be achieved. A module on indoor air pollutants as

part of the science course could incorporate not just radon, but also topics such as carbon monoxide, household products, mould, pollen etc.

5.5.2 *Radon measurements in pre-schools*

In 2014, the National Radon Control Strategy recommended that a reference level of 200 Bq/m^3 should be applied to all schools, including pre-schools which cater for children under 5 years old (some also cater for children up to 12 years old where an after-school care facility is provided). Regulations should therefore be introduced to require all pre-school facilities to measure the radon concentration in all ground level and below ground level occupied rooms. The time children spend at pre-care facilities (up to 10 - 11 hours) is generally longer than in school and, previous publications have found a tendency for higher radon concentrations in pre-school childcare facilities than in schools [156, 157]. Both Trevisi *et al.* and Vuchkov *et al.* suggested that these higher concentrations could be due to the number of floors in the school building, as generally pre-school and primary schools have only one floor with no basement. Vuchkov *et al.* also suggested that it could be due to reduced ventilation and stated that pre-schools tend to keep windows closed for longer periods. Fojtíková *et al.* looked at the effect of incorrect ventilation on radon concentration in a school classroom over a 1-week period (figure 5.12) [158]. The radon concentration rises during the night but, due to a lack of ventilation, the concentration only slowly decreases during the day. If the room was ventilated first thing in the morning, before school started, a decrease in radon would be immediately observed. The weekend period, where the school is closed and there is no ventilation, shows a steady increase in the radon concentrations.

In Ireland, TUSLA (the child and family agency in Ireland) currently inspect pre-school facilities under the 'Child Care (Pre-School Services) Regulations 2006' and it is recommended that radon reporting should be included in this inspection.

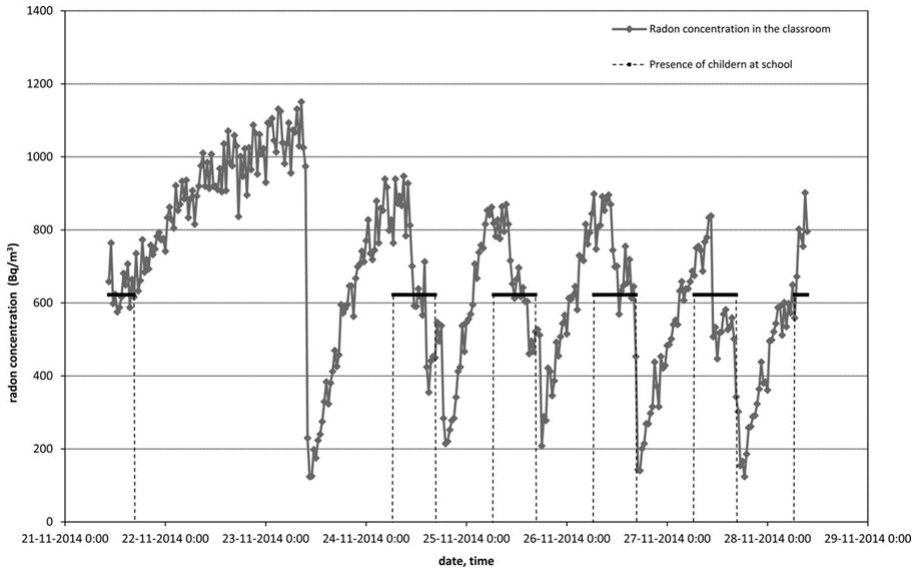


Figure 5.12: The influence of incorrect ventilation habits on radon concentration at school.

6

CONCLUSIONS

Several aspects of indoor and outdoor radon have been addressed in this thesis and the conclusions were detailed at the end of each experimental chapter. The aim of this chapter is to recapitulate on the primary conclusions, report on recent progress in the research field of radon in Ireland and to reflect on the work.

6.1 SUMMARY OF CONCLUSIONS

The mean outdoor radon concentration for Ireland was quantified as 5.6 ± 0.7 Bq/m^3 which is in close agreement with the previously derived value of 6 Bq/m^3. This agreement confirms that the extrapolation method, used to estimate the derived figure of 6 Bq/m^3, was correct. Therefore, the extrapolation method could become a useful tool for estimating the outdoor radon concentration in other countries. Causes in variations of the mean outdoor concentration were investigated and the influence of different climatology parameters and site location. Temperature and rainfall had the most notable effect on radon variations over a 2 months period and inland monitoring sites had higher concentrations than coastal sites.

With such a large choice of radon monitors available, which vary in complexity and price, Chapter 4 summarised the main features of 7 commonly used detectors. The accuracy of each was reported in comparison with a benchmark detector (Atmos 12 dpx) for exposure periods of 1 - 4 weeks. One of the lower priced monitors, the Canary, gave readings closest to those of the Atmos; in fact it measured within 10% of the Atmos for almost all of the time periods. The use of short term measurement (<1 month) is not intended to replace the standard 3 month test but it would facilitate the introduction of radon measurement for conveyancing and standardise measurements during or after remediation works.

Chapter 5 detailed the results of re-testing in 16 schools which had remediation implemented 10 - 17 years previously, during the NSS. In rooms where remediation was installed, it was seen that 26% of the rooms had risen to above the reference level of > 200 Bq/m^3 and 10% to above > 400 Bq/m^3. The results of this follow-up study lead to the prediction that currently 790 rooms are > 200 Bq/m^3 and 80 rooms are > 400 Bq/m^3 in the schools originally tested during the NSS. This highlights the need for remediation maintenance and follow-up programmes particularly in schools; a period of 7 - 10 years is recommended. A recommendation is strongly advised for implementing a radon programme for pre-school facilities in Ireland as this cohort was not included in the NSS. It is envisaged that it would be carried out in line with current inspections carried out by TUSLA.

6.2 RECENT PROGRESS

Naturally the research field is continuously developing, so it is important to highlight the latest progress in the field especially when it is directly related to or resulting from the work presented here.

Since the work in Chapter 3 was completed, a follow-up radon survey of Irish dwellings was carried out. An updated figure of 77 Bq/m^3 is now stated as the mean indoor radon concentration for Ireland [159]. Though this is a reduction from the previous mean concentration of 89 Bq/m^3 , it is still substantial in comparison to the estimated worldwide mean concentration of 40 Bq/m^3 [4].

The NRCS group report annually to government detailing progress and outlining the action plans for the forthcoming year [160, 161]. As a direct result of the work referred to in Chapter 5, the most recent NRCS report included a strategy to address radon in pre-school facilities. It is proposed that the EPA will provide training for TUSLA inspectors in aspects of radon so that radon queries become part of their inspection process. Also, as a result of the same study (Chapter 5), the NRCS report also states that the DES will devise a strategy to address re-testing and maintenance of remediation systems in schools.

In relation to the introduction of radon testing for conveyancing (Chapter 4), the latest NRCS report confirmed that questions relating to radon will shortly be included in the conveyancing contract [161]. Radon testing will not be compulsory but it is expected that the questions will be 'a powerful driver of action' [161]. Also, a registration scheme for radon measurement providers is due to be implemented in the coming year and this will assist in the quality assurance of radon testing.

6.3 REFLECTIONS ON WORK

Raising awareness about radon and encouraging action are important parts of the campaign against radon and the associated health effects. It is estimated that people spend 80% of their time indoors and in Ireland, it is estimated that only 7% of houses have been tested for radon. This low testing rate means that a high percentage of occupants may unknowingly be exposed to elevated radon concentrations. A Building Energy Rating (BER) certificate, which indicates the energy performance of the home, is required for all house rentals and conveyancing in Ireland yet a house could have a radon concentration of, say, 50,000 Bq/m^3 (equating to an annual dose of 1.2 Sv) and it would not have to be declared. Many occupants do not know the radon concentration that they are exposed to and although they may not take remedial action if they did, the information must be provided.

Given that the most harmful environmental factors have been proven to be more damaging to children than to adults, it would be prudent to adopt stricter limits and regulations for radon in buildings where children spend a high proportion of their time, even if it is questionable whether children are at a higher risk to the health risks specifically attributable to this 'Silent Killer'. The Safety, Health and Welfare at Work Act 2005 and S.I. no. 125 of 2000 dictate that radon testing is required for workplaces and therefore pre-schools, but only those located in high radon areas. As stated in the previous section, steps have been taken to query radon testing in pre-school facilities during inspections and it is hoped that these are the first steps toward compulsory testing in pre-school facilities.

It is evident, from the response to the work detailed in this thesis, that active research contributes to and improves national radon programmes. By continuing this work, improvements and strategies can be implemented to raise awareness, promote action and ultimately reduce the number of lung cancer cases due to radon.

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